

Emission characteristics of modern and old-type residential boilers fired with wood logs and wood pellets

Linda S. Johansson^{a,*}, Bo Leckner^b, Lennart Gustavsson^a, David Cooper^c,
Claes Tullin^a, Annika Potter^c

^a Department of Energy Technology, SP Swedish National Testing and Research Institute, P.O. Box 857, Borås 501 15, Sweden

^b Department of Energy Technology, Chalmers University of Technology, Göteborg 412 96, Sweden

^c IVL Swedish Environmental Research Institute Ltd., P.O. Box 4708, Göteborg 402 58, Sweden

Received 27 November 2003; received in revised form 30 March 2004; accepted 20 April 2004

Abstract

Emissions from commercial residential boilers fired with wood logs and wood pellets, have been compared. Seven boilers, selected with respect to age, design, connection to heat storage tank, and type of biofuel, were included in the study, which also covers two oil-fired boilers in comparison. The measurements of gaseous emissions comprised carbon monoxide (CO), carbon dioxide (CO₂), oxygen (O₂), total organic carbons (TOC), nitrogen oxides (NO_x), polycyclic aromatic hydrocarbons (PAH), and 33 volatile organic compounds (VOC). Particle emissions were characterised by mass concentration, number concentration, and the corresponding particle size distributions. In general, old-type wood boilers caused considerably higher emissions than modern wood and pellet boilers. The mass concentration of particles was 180 times larger in the worst old-type case (a water-cooled wood boiler without heat storage tank) compared to the best modern case (wood pellets). The TOC emission was shown to be correlated to the CO emission, both ranging between very low values and up to 10 000 mg/MJ, depending on design and operation. The highest emissions of unoxidised compounds occurred at the highest excess air ratio, and oxygen was not the limiting parameter for poor combustion. Instead, high excess air can be suspected to cool the combustion chamber, resulting in high CO emissions. VOC was dominated by methane. Especially from an old-type boiler the methane emissions could be high and the effect on climate change then may become larger than that of an oil boiler. However, substitution of an old-type wood boiler with a modern wood boiler attached to a storage tank or with a pellet boiler, would reduce methane emissions by 8 to 9000 times and the efficiency would increase. Most emissions could be considerably lowered by connecting the old-type wood boiler to a heat storage tank, or by charging small (in relation to the combustion chamber) batches of wood.
© 2004 Elsevier Ltd. All rights reserved.

Keywords: Wood; Pellets; Domestic combustion; Emissions; Biofuel

1. Introduction

Biofuel combustion is an important source of renewable energy. Besides combustion in large heat and power plants, residential biofuel combustion is employed for

heating and cooking, especially in scarcely populated areas and developing countries. In cold areas, biofuel is burned for heating. This can be performed in two ways: in a stove heating the surrounding room, or in a boiler heating water to be circulated through a piping system to heat an entire house. Residential combustion of biofuel is carried out in simple wood stoves as well as in well-controlled devices, such as pellet burners and modern wood boilers, and the emissions of pollutants

*Corresponding author. Tel.: +46-33-165501; fax: +46-33-131979.

E-mail address: linda.johansson@sp.se (L.S. Johansson).

vary over a wide range. For example, particle emissions have been reported to be 50 times higher from a wood stove or a fireplace, i.e. from uncontrolled combustion devices, than from more controlled devices (Johansson et al., 2003), and for wood stoves used for house heating the carbon monoxide (CO) emission may vary between 18 (Ndiema et al., 1998) and 180 (McDonald et al., 2000) g/kg fuel. Consequently, it is important to pay attention to the kind of residential biofuel combustion device, especially as poor combustion not only affects its immediate surrounding but also can lead to substantial emissions of greenhouse gases (Zhang et al., 2000). In contrast to stoves, wood boilers can be connected to heat storage tanks (a water tank for storage of heat). This is an advantage from an emission point of view, because then the boiler can be run at a regular heat output instead of being intermittently operated to directly follow the heat load of the house. With a storage tank optimal combustion conditions are approached, leading to lower emissions and higher efficiencies. Pellet burners, on the other hand are designed for intermittent operation and the impact of a storage tank is not as great. Besides the potential of decreasing emissions from wood boilers by using storage tanks, the emissions can be reduced by favourable firing behaviour and new combustion technique. There has been an important technology development in the wood boiler area over the last decades.

The common types of residential combustion devices have been described by, for instance, Baxter et al. (2002). Modern wood boilers are usually designed for down-draught combustion and with a ceramic-lined burn-out zone (Fig. 1A), and are normally connected to storage tanks. In old-type wood boilers up-draught combustion is most common, and the boilers are water cooled. An example is shown in Fig. 1B. Up-draught combustion is also typically used in wood stoves. In general, development of wood boilers has led to combustion devices with

increased efficiency and decreased emissions. In Sweden 57% of totally 260 000 residential biofuel boilers are still old-type traditional boilers without storage tank, 4% are old-type boilers with storage tank, 27% modern wood log boilers, and the remaining part pellet burners/boilers (Swedish chimney-sweepers' statistics, 2001).

Except for some work on particle emissions (Wieser and Gaegauf, 2000; Gaegauf et al., 2001; Johansson et al., 2003), information on emission characteristics related to residential biofuel boilers is scarce. There is a need to compare emissions from different techniques of biofuel boilers to ensure an ecologically sustainable use of biofuels. Consequently, the present work focuses on comparing gaseous and particulate emissions from old-type and modern boilers by means of systematic variation of combustion device, fuel quality, firing behaviour, and the influence of heat storage tank. Unsystematic irregularities caused by user habits are avoided by performing tests in the laboratory. Instead, user habits are simulated in a schematic way. The emissions from two residential oil burners are presented as reference, because oil-firing is a common alternative for house heating in Sweden (domestic gas and coal combustion are generally not used). The purpose of the comparison is to determine the emission differences between old-type and modern residential biofuel boilers and to quantify emission characteristics of different kind of biofuel combustion.

2. Experimental

Two old-type water-cooled wood boilers, two ceramic-lined modern wood boilers, and three pellet burners were selected. The installations are presented in Table 1. Each letter in the table case corresponds to one measurement case. Firing behaviour, as well as the fuel quality, was varied. The wood boilers were heated up by

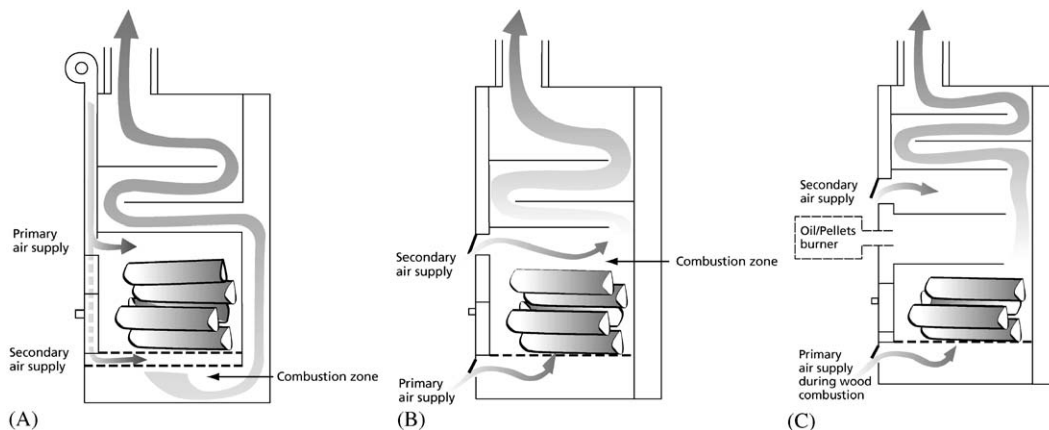


Fig. 1. A wood boiler designed for down-draught combustion (A), a wood boiler designed for up-draught combustion (B), and a multi-fuel boiler (C), which can burn wood, oil, or pellets.

Table 1
Measurement cases

Boiler/burner	Operation conditions, Power (kW)	Fuel type	Case	
Old-type wood boilers	Water-cooled multi-fuel boiler, up-draught combustion	Heat storage tank, 10 (100%)	Dry wood	a
		Large wood batches, 6	Dry wood	b
		Small wood batches, 6	Dry wood	c
	Water-cooled wood boiler, up-draught combustion	Heat storage tank, 24 (100%)	Dry wood	d
		Large wood batches, 7	Dry wood	e
		Small wood batches, 6	Dry wood	f
Modern wood boilers	Ceramic wood boiler with flue gas fan, down-draught combustion	Heat storage tank, 34 (100%)	Dry wood	g
		Heat storage tank, 30 (100%)	26% moisture	h
		Heat storage tank, 12 (100%)	38% moisture	i
	Ceramic wood boiler with natural draught, down-draught combustion	Heat storage tank, 28 (100%)	Dry wood	j
		Heat storage tank, 24 (100%)	26% moisture	k
		Heat storage tank, 23 (100%)	Briquettes	l
Pellet burners and boilers	Pellet burner with pilot flame	Nominal, 11 (100% load)	Wood pellets	m
		Intermittent, 6	Wood pellets	n
		Intermittent, 6 high draught	Wood pellets	o
		Intermittent, 3	Wood pellets	p
	Pellet burner with electrical ignition	Nominal, 22 (100% load)	Wood pellets	q
		Intermittent, 6	Wood pellets	r
		Intermittent, 3	Wood pellets	s
	Pellet boiler	Nominal, 16 (100% load)	Wood pellets	t
		Intermittent, 6	Wood pellets	u
		Intermittent, 3	Wood pellets	v
	Pellet burner with electrical ignition	Intermittent, 6	Bark pellets	w
Oil burners	Environmental branded oil boiler	Nominal, 18 (100% load)	Oil	x
	Multi-fuel boiler with oil burner	Nominal, 21 (100% load)	Oil	y

firing a batch of wood logs prior to measurements to avoid the influence of initial transients. Measurements started after the ignition of a second wood batch and lasted to the end of the burn-out of the third batch, defined as the time when the carbon dioxide (CO₂) concentration had decreased to 4%. As a burn-out phase ended, a new batch of wood was supplied. The pellet and oil burners were also heated up prior to measurements by automatic operation under the conditions investigated. Measurement during nominal output lasted for about 3 h, and during intermittent operation sometimes even longer. The intermittent operation was started and stopped in coordination with the operation cycles. The emissions are presented in milligrams pollutant or number of particles per MJ of fuel supplied, calculated from measured concentrations and fuel

analyses. All emission data are mean values for each case investigated.

2.1. Combustion devices and measurements

The combustion devices were adjusted for optimum air supply prior to measurements. The properties and operation characteristics of the four types of biofuel devices studied are shown in Table 1. Six combustion cases in old-type wood boilers (a–f), six cases in modern wood boilers (g–l), and eleven cases in modern pellet burners/boilers (m–w) were included. In the old-type wood boilers fired without storage tank two firing modes were simulated. The first mode represents a user that charges as large batches of wood (in relation to the combustion chamber) as possible and then throttles the

air supply (b and e) to allow combustion to proceed unattended during a considerable time. The second mode represents the user who is at home during daytime and attends the boiler by charging small batches of wood (c and f). Old-type wood boilers were fired with storage tank as well, which for modern wood boilers were used in all cases. The pellet burner (m–s and w) is a separate burner that can be installed in an optional boiler, while a pellet boiler (t–v) is a pellet burner integrated with a boiler unit (Baxter et al., 2002). In the present investigation, the pellet burners were installed in the water-cooled multi-fuel boiler used for firing wood logs (Fig. 1C). As a comparison, the emissions from two oil burners were measured. One of the oil burners (x) is integrated with a boiler. This unit is branded according to the Nordic Ecolabelling of oil burners (2000–2004). The other oil burners were installed in the water-cooled multi-fuel boiler (y), which also could fire wood logs and pellets (Fig. 1C).

2.2. Fuels

The chemical composition of the wooden fuels, pellets, briquettes and wood logs, is similar (Table 2), except that the moisture content was approximately 8% in the pellets and briquettes, and 15%, 26%, or 38% in the wood logs. Wood logs, of birch, with moisture content of 15% are considered dry, while higher moisture contents represent wet fuel. The ash content in the woody fuels is low compared to bark pellets, which also has a higher fraction of nitrogen. The length of the wood logs was about 500 mm. The cylindrical log was split once or twice with a resulting transversal size of 100 mm. Wood and bark pellets had a diameter of 8 mm and a length of 15–20 mm.

2.3. Measurement techniques

The measurement set-up is presented in Fig. 2. Particles were sampled isokinetically in two separate

lines, one for total mass concentration and one for number concentration and size distributions. In the first sampling line the flue gas was extracted and diluted in two steps. The first dilution took place under heated conditions (180°C) to avoid condensation and particle growth. Number particle concentration and size distribution were measured with an Electrical Low-Pressure Impactor (ELPI, Dekati Ltd.) and mass size distribution using a Dekati Low-Pressure Impactor (DLPI, Dekati Ltd.). Both instruments were calibrated by the manufacturer and measured concentrations and particle size distributions in the range of 30 nm to 10 µm. In the second sampling line, the total mass of particulate emission was measured gravimetrically according to a Swedish Standard method, SS028426, 1991. The third sampling line was used for continuous monitoring of CO, CO₂, oxygen (O₂), nitrogen oxides (NO_x) and total organic carbons (TOC). The measurement principles of the gas analysers were non-dispersive infra-red (CO, CO₂), paramagnetism (O₂), flame ionisation (TOC), and chemiluminescence (NO_x). Each gas analyser was calibrated with appropriate gas on zero and span points. The emissions of TOC are presented in methane-equivalents and the NO_x emissions as NO₂. Polycyclic aromatic hydrocarbons (PAH) were sampled according to ISO 11338 (2000). A number of representative grab samples were collected, which later were analysed as one sample. Blank PAH-samples were taken and corrected for in the results. The PAH samples was analysed with gas-chromatography/mass-spectrometry (GC/MS) analysis (a few weeks later). The PAH compounds analysed were: Naphthalene, 2-methyl-Naphthalene, 1-methyl-Naphthalene, 2,6-dimethyl-Naphthalene (Biphenyl), Acenaphthylene, Acenaphthene, 2,3,5-trimethyl-Naphthalene, Fluorene, Dibenzothiophene, Phenanthrene, Anthracene, 2-methyl-Anthracene, 1-methyl-Phenanthrene, 1-methyl-Anthracene, Fluoranthene, Pyrene, Retene, Benz(a)anthracene, Chrysene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(e)pyrene, Benzo(a)pyrene, Perylene, Indeno(1,2,3-cd)pyrene,

Table 2
Elemental analyses and heating values of the fuels

	Wood pellets	Bark pellets	Wood briquettes	Wood logs	Oil
Ash	0.5	3.7	0.3	0.3	0.6
Moisture	7.6	7.8	7.5	15/26/38	0
Lower heating value (MJ/kg)	19.0	20.1	18.9	19.0	43.1
Content					
Carbon	50.2	52.1	50.6	50.6	87.5
Oxygen	43.2	37.8	42.7	42.7	0
Hydrogen	5.9	5.9	6.4	6.4	12.7
Nitrogen	0.08	0.48	0.05	0.05	0.1
Sulphur	<0.01	0.03	<0.01	<0.01	0.09

Moisture—wt% wet fuel, other data wt% dry fuel.

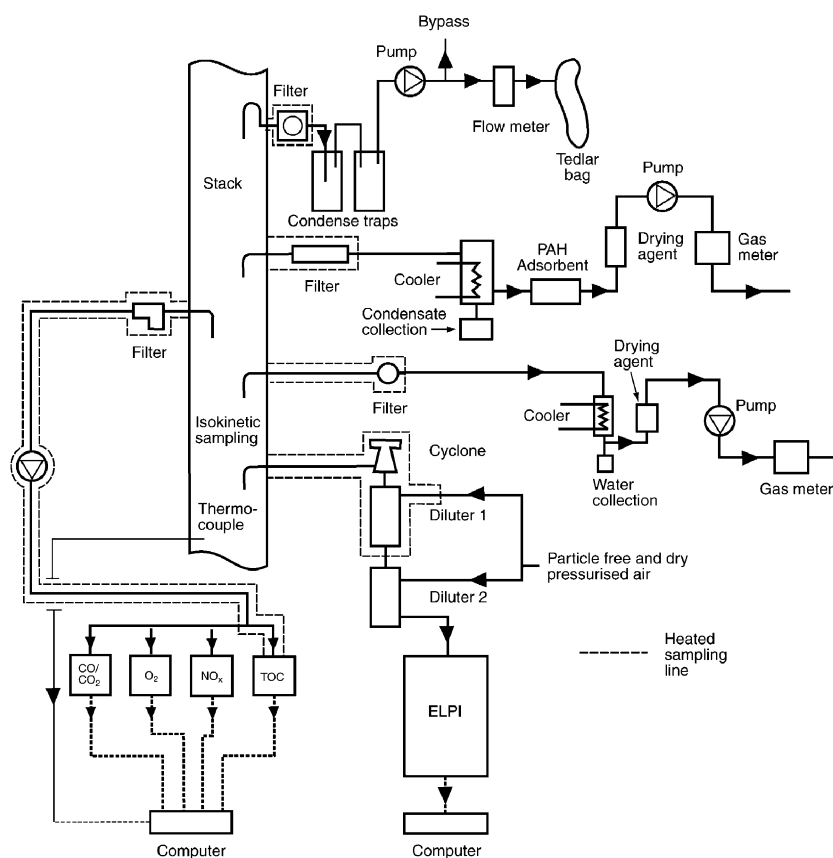


Fig. 2. Measurement set-up.

Dibenz(a,h)anthracene, Benzo(ghi)perylene. In the subsequent emission evaluation the sum of the PAH compounds analysed is presented. The final and fifth sampling line was for measurement of volatile organic compounds (VOC) by a number of representative grab samples, which later were analysed as one sample, subsequent analyses of compounds are specified in the result section. A filtered flue gas sample was cooled to room temperature, passed through an inert pump and collected in an 801 Tedlar bag. After the sampling period, fractions of the sample gas (1–20 ml) were drawn through Tenax (for C₆–C₁₄ hydrocarbons) and Carbo-pac (1,3-butadiene) adsorbent tubes for subsequent GC analysis. The remaining sample gas in the Tedlar bags was also analysed by GC within 24 h (for 18 light C₁–C₆ hydrocarbons). Blanks VOC-samples were taken and corrected for in the results.

3. Results

The emissions of CO, TOC, methane, non-methane VOC (NMVOC), PAH, mass and number concentrations of particles, and NO_x are presented in Table 3, and

specific NMVOC compounds are shown in the appendix (Tables 4 and 5). To simplify the results, the emissions from all the operational cases of Table 1 are sorted into groups and plotted against some related parameter, prior to presenting the influence from combustion device, firing behaviour, fuel quality, and connection of heat storage tanks. Three groups of emissions were measured: unoxidised gaseous compounds, particles, and nitrogen oxides. Unoxidised gaseous compounds, i.e. CO and hydrocarbons, are a consequence of imperfect combustion conditions, which can be explained by the well-known criteria for favourable combustion: long enough residence time at sufficiently high temperature with adequate mixing of air and combustible gases. In Fig. 3, the CO concentration is shown to increase with increased excess air. Consequently, oxygen was not the limiting parameter for CO oxidation during poor combustion conditions. Instead, high excess air can be suspected to cool the combustion chamber, resulting in high CO emissions. Fig. 4 shows that there are linear correlations between CO and TOC for wood boilers, pellet burners, and oil burners, and obviously TOC is also influenced by the air supply, just as CO, VOC (methane+NMVOC) and PAH. The

Table 3
CO₂, CO, TOC, particles and NO_x emissions

	Case	CO ₂ (%)	CO	TOC	CH ₄	NMVOC	PAH	Particles	Particles (1/MJ)	NO _x
Old-type wood boilers	a	8.4	4100	660	—	—	—	87	—	65
	b	4.7	5200	1300	—	—	—	350	—	72
	c	6.8	4800	1100	610	270	14	89	17 × 10 ¹³	71
	d	8.3	5900	1500	670	430	13	103	3.9 × 10 ¹³	67
	e	6.9	16400	4800	4800	2000	64	2200	200 × 10 ¹³	28
	f	5.6	8200	3000	—	—	15	—	2.8 × 10 ¹³	64
Modern wood boilers	g	12.2	707	14	1	1.9	0.21	27	4.5 × 10 ¹³	125
	h	11.5	507	33	0.8	1.3	0.14	25	2.4 × 10 ¹³	111
	i	5.1	3781	690	73	43	3.0	89	8.5 × 10 ¹³	101
	j	10.3	1300	89	14	14	1.1	32	3.6 × 10 ¹³	72
	k	9.1	770	63	9.2	7.8	0.44	23	6.4 × 10 ¹³	81
	l	10.2	880	28	4.3	3.9	0.33	18	2.0 × 10 ¹³	60
Pellet burners and boilers	m	9.5	36	4	0.76	1.2	0.32	22	1.4 × 10 ¹³	68
	n	6.0	350	78	2.7	3.3	0.26	—	1.7 × 10 ¹³	71
	o	4.8	290	31	—	—	0.12	28	1.3 × 10 ¹³	68
	p	3.7	960	250	14	23	0.27	65	7.4 × 10 ¹³	66
	q	13.0	120	3	<0.55	0.95	0.06	16	0.8 × 10 ¹³	70
	r	9.1	990	60	5.3	20	8.5	64	1.6 × 10 ¹³	64
	s	8.6	120	10	<0.84	1.7	0.55	15	0.8 × 10 ¹³	67
	t	11.7	30	1	—	—	—	13	—	—
	u	6.8	380	2	—	—	—	12	0.1 × 10 ¹³	62
	v	3.8	1100	92	—	—	—	51	0.2 × 10 ¹³	62
Oil	w	10.6	730	42	1.8	4.8	1.1	—	—	180
	x	12.2	2	1	0.46	0.64	0.17	12	0.01 × 10 ¹³	37
	y	10.6	9	32	0.52	0.38	0.006	6	0.1 × 10 ¹³	41

The measurement cases are explained in Table 1. Emissions are in mg per MJ, where not other units (% or number of particles per MJ) are indicated.

particles emitted contain both organic material, i.e. unburnt solid material, and inorganic material, i.e. ash particles. Regardless of combustion conditions, ash particles always remain as a by-product. In Fig. 5, it can be seen that the mass concentration of emitted particles was rather constant up to a concentration of about 100 mg/MJ TOC. At higher concentration the particle emission increased. The increase in mass concentration of particles during poorer combustion conditions agrees with earlier work, for example by Muhlbaler Dasch (1982) and Rau (1989). The enhancement of particle emission during poor combustion conditions is due to carbonaceous particles. The number concentration of emitted particles also increased for poor combustion conditions, as indicated by the dependence on TOC (Fig. 5).

Nitrogen oxides are formed from combustion air or from fuel nitrogen. The importance of fuel nitrogen is implied by a correlation between the NO_x emissions and the fuel nitrogen content (Fig. 6). As seen in Fig. 6, no clear effect of excess air on the NO_x emissions could be observed, probably because of a balance between the tendencies of excess air to increase the formation of NO and of the falling temperature to reduce the formation (of fuel-NO).

3.1. Combustion devices

In general, old-type wood boilers led to more emissions of particles and unoxidised compounds than modern wood boilers and pellet burners as can be seen from Table 3. The lowest emissions were measured from oil burners. It was noted that the pellet and oil burners were more equal in performance than the wood boilers. The lowest emissions from wood boilers were as low as from pellet burners and were obtained from combustion of dry wood logs in modern boilers connected to storage tanks. The emissions from an old-type wood boiler fired with large wood batches could be considerably decreased by connecting it to a storage tank.

The dominant volatile organic compound was methane, probably due to its relatively slow oxidation in comparison with other hydrocarbons (Glassman, 1996). The fraction of methane, based on the sum of methane and NMVOC, was 50 mass% for oil burners, 20–40 mass% in the pellet cases, 30–60 mass% for modern wood boilers, and 60–70 mass% for old-type wood boilers. The largest emissions of VOC also contained the largest fraction of methane. The methane concentrations measured during pellet combustion are

Table 4
Emissions of light VOC, to the left are the measurement cases explained in Table 1

	CH ₄	C ₂ H ₆	C ₂ H ₄	C ₃ H ₈	C ₃ H ₆	<i>Iso</i> - C ₄ H ₁₀	C ₄ H ₁₀	C ₂ H ₂	<i>Trans</i> -2- C ₄ H ₈	1-C ₄ H ₈	<i>Cis</i> -2- C ₄ H ₈	<i>Iso</i> - C ₅ H ₁₂	C ₅ H ₁₂	<i>Trans</i> -2- C ₅ H ₁₀	<i>Cis</i> -2- C ₅ H ₁₀	2- & 3- methyl- C ₅ H ₁₀	C ₆ H ₁₄
c	610	23	170	4.1	5.7	<0.9	<0.9	14	<0.86	0.95	<0.86	<1.1	<1.1	1.1	<1.1	<1.1	1.5
d	670	31	180	5	31	<0.7	<0.7	87	<0.7	<0.7	<0.7	<0.9	<0.9	<0.7	<0.9	<0.9	<1.3
e	4800	310	780	71	270	3	10	260	12	40	85	<1.1	3.1	5.6	4.1	2.5	1.5
g	1.0	0.03	0.2	<0.00	0.02	<0.01	<0.01	0.18	<0.01	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	0.01
h	0.80	0.02	0.20	<0.004	0.0	0.0	0.0	0.1	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
i	73	4.4	16	0.83	4.1	0.03	0.13	5.1	0.11	0.71	0.09	<0.01	0.04	0.03	0.02	<0.02	0.08
j	14	0.3	3	0.03	0.2	<0.01	<0.01	2.5	<0.01	0.03	<0.01	0.01	0.01	<0.01	<0.01	<0.01	<0.01
k	9.2	0.27	2.6	0.04	0.29	0.01	0.01	1.2	0.01	0.04	0.01	0.02	0.03	0.01	<0.01	0.01	0.01
l	4.3	0.19	1.2	0.03	0.14	<0.01	<0.01	0.5	<0.01	0.02	0.01	0.03	0.02	0.01	0.01	0.02	0.03
m	<0.76	<0.01	<0.03	<0.05	<0.01	<0.01	<0.01	<0.04	<0.01	<0.01	<0.01	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
n	2.7	<0.01	<0.05	<0.08	<0.02	<0.02	<0.02	<0.06	<0.02	<0.02	<0.02	<0.03	<0.03	<0.03	<0.03	<0.04	<0.03
p	14	3.2	5.3	0.9	2.8	0.1	0.2	1.3	0.2	0.6	0.2	0.2	0.1	0.2	0.1	0.2	0.1
q	<0.55	<0.02	<0.04	<0.04	<0.02	<0.02	<0.01	<0.08	<0.01	<0.02	<0.01	<0.06	<0.03	<0.05	<0.03	<0.02	<0.02
r	5.3	0.1	2.0	<0.05	0.1	<0.01	<0.02	5.34	<0.01	<0.01	<0.01	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
s	<0.84	0.03	0.2	<0.06	0.04	<0.02	<0.01	0.2	<0.01	<0.03	<0.02	<0.07	<0.03	<0.05	<0.03	<0.07	<0.05
w	1.8	0.1	0.5	<0.05	0.04	<0.01	<0.01	1.6	<0.01	<0.01	<0.01	<0.02	<0.02	<0.01	<0.01	<0.02	<0.02
x	<0.46	<0.00	<0.00	<0.00	<0.00	<0.01	<0.01	<0.00	<0.00	0.002	<0.00	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
y	<0.52	<0.003	0.02	<0.004	<0.004	<0.01	<0.01	0.02	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01

Emission values are in mg per MJ. In cases a, b, f, o, t, u, and v light VOC compounds were not measured.

Table 5
Emissions of heavier VOC (mg per MJ)

	1,3-C ₄ H ₆	C ₈ H ₁₈	C ₉ H ₂₀	C ₁₀ H ₂₂	C ₁₁ H ₂₄	C ₁₂ H ₂₆	C ₁₃ H ₂₈	C ₁₄ H ₃₀	C ₆ H ₆	C ₆ H ₅ - CH ₃	C ₆ H ₅ - C ₂ H ₅	<i>o</i> -C ₆ H ₄ - (CH ₃) ₂	<i>m, p</i> - C ₆ H ₄ - (CH ₃) ₂	1,3,5- C ₆ H ₃ - (CH ₃) ₃	1,2,4- C ₆ H ₃ - (CH ₃) ₃	1,2,3- C ₆ H ₃ - (CH ₃) ₃
c	—	0.10	0.14	<0.11	<0.07	<0.15	<0.15	<0.15	32	7.9	1.1	0.6	1.4	<0.15	<0.15	<0.15
d	—	<0.30	0.42	<0.30	<0.30	<0.30	<0.30	<0.30	66	12	1.3	1.2	1.6	<0.30	<0.30	<0.30
e	—	0.79	0.27	0.42	0.22	<0.16	<0.16	0.22	91	22	4.4	3.0	7.3	<0.16	1.4	0.8
f	—	0.75	0.19	0.40	<0.19	<0.19	<0.19	<0.19	46	16	2.3	1.8	5.2	<0.19	20	12
g	0.01	0.04	0.06	0.02	<0.02	0.53	<0.02	<0.02	0.46	0.10	<0.01	0.01	0.01	<0.02	0.03	<0.02
h	0.04	0.11	0.12	0.04	<0.04	<0.04	<0.04	<0.04	0.21	0.06	<0.01	0.03	0.02	<0.04	<0.04	<0.04
i	3.3	0.15	0.12	0.16	<0.13	<0.13	<0.13	<0.13	4.2	1.4	0.16	0.15	0.27	<0.13	<0.13	<0.13
j	0.21	0.14	0.13	0.15	<0.05	<0.05	<0.05	<0.05	5.8	0.84	0.12	0.07	0.12	<0.05	<0.05	<0.05
k	0.26	0.07	0.08	0.15	<0.05	<0.05	<0.05	<0.05	2.2	0.33	0.04	0.04	0.06	<0.05	<0.05	<0.05
l	0.08	0.09	0.14	0.03	<0.02	<0.02	<0.02	<0.02	0.99	0.16	<0.01	0.01	0.01	<0.02	<0.02	<0.02
m	0.04	0.12	0.14	0.01	<0.06	<0.06	<0.06	<0.06	0.11	<0.13	<0.01	<0.05	<0.03	<0.01	<0.01	<0.01
n	0.7	0.07	0.10	<0.02	<0.09	<0.09	<0.09	<0.09	0.9	0.4	0.04	0.07	0.07	<0.02	<0.02	<0.02
o	0.2	<0.07	<0.08	0.03	<0.11	<0.11	<0.11	<0.11	0.7	0.2	<0.05	<0.09	<0.04	<0.01	<0.01	<0.01
p	1.7	0.2	0.2	0.1	<0.15	<0.15	<0.15	<0.15	2.6	1.2	0.09	<0.12	<0.17	<0.02	<0.02	<0.02
q	0.01	0.05	0.06	0.01	<0.04	<0.04	<0.04	<0.04	0.1	<0.04	<0.01	<0.03	<0.01	<0.00	<0.00	<0.00
r	0.1	0.06	0.09	0.03	<0.06	<0.06	<0.06	<0.06	10.9	0.6	0.06	0.06	0.1	<0.01	<0.01	<0.01
s	0.04	0.05	0.06	<0.02	<0.06	<0.06	<0.06	<0.06	0.16	<0.06	<0.01	<0.05	<0.03	<0.01	<0.01	<0.01
w	0.03	0.11	0.17	0.05	<0.05	<0.05	<0.05	<0.05	1.43	0.26	0.04	<0.04	<0.04	<0.01	<0.01	<0.01
x	<0.004	0.05	0.07	0.06	<0.02	<0.02	<0.02	<0.02	0.01	0.02	<0.002	0.02	<0.004	<0.02	<0.02	<0.02
y	<0.004	<0.05	0.07	0.06	<0.02	<0.02	<0.02	<0.02	0.04	0.02	<0.002	0.01	0.04	<0.02	<0.02	<0.02

To the left are the measurement cases explained in Table 1.

in the range of an earlier study by Olsson et al. (2003). The remaining most frequent VOC compounds were ethane, ethene, ethyne, propene, benzene, and toluene (appendix). The largest PAH emission, 64 mg/MJ, was associated with old-type wood boilers and is higher than reported for wood stoves, i.e. 5.6 mg/MJ (Oanh et al., 1999) and 18 mg/MJ (Hedberg et al., 2002). PAH from

biofuel-combustion consisted, in average, of 70% Naphthalene, Phenanthrene, Fluoranthene, Pyrene, and Acenaphthylene, while oil combustion consisted, in average, of 70% Fluoranthene, Pyrene, and Phenanthrene. (The sequences of compounds are presented in order of concentration.) The highest mass concentration of particles, 2200 mg/MJ, was recorded during combustion of wood logs in the water-cooled boiler, not connected to a heat storage tank (Case e). Firing of wood logs in modern boilers with a ceramic-lined burn-out zone and connected to a heat storage tank (g and j), led to 70–80 times lower particle emission than highest measured concentration, levels that have been observed also by Gaegauf et al., 2001. Combustion of wood pellets (m and o-v) led to mass concentrations of particles in a range similar to modern wood boilers (g and j), while particle emission was lower from oil firing (x and y).

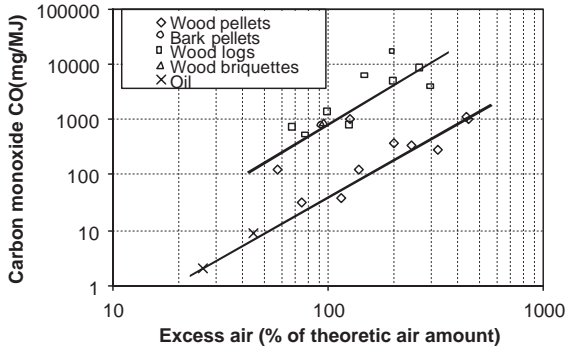


Fig. 3. Emission of carbon monoxide versus excess air for the cases in Table 1.

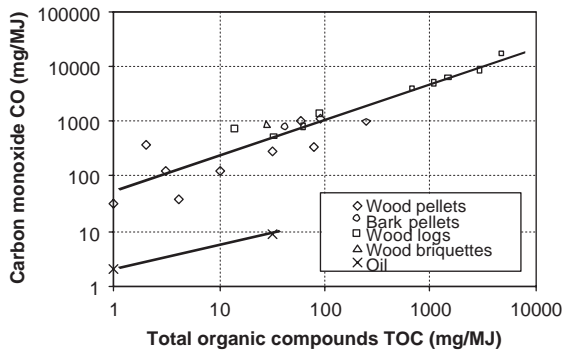


Fig. 4. Emission of carbon monoxide versus emission of total organic compounds (methane-equivalents) for the cases in Table 1.

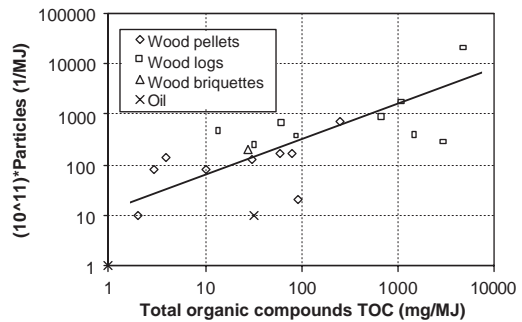
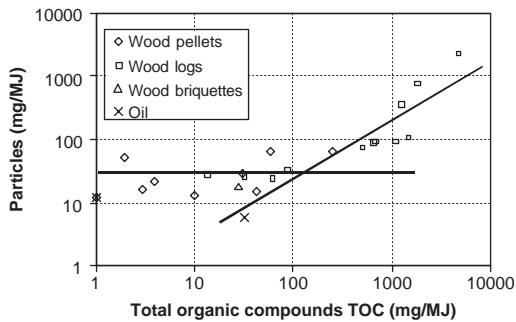


Fig. 5. Mass concentration of particles emitted versus total organic compounds for the cases in Table 1 (methane-equivalents) on the left-hand side and number concentration of particles emitted versus total organic compounds on the right-hand side.

In Fig. 7, emitted particle mass size distributions are presented for one modern wood boiler (j), one old-type boiler fired with small wood batches (c), nominal operation cases with the pellet burners (m and q), and combustion of bark pellets (w). During continuous firing of wood pellets, the mass size distributions of particles in the flue gas from the two pellet burners were very similar (m and q), showing a maximum at a particle size of around 130 nm, which is consistent with a previous investigation on fireplaces (Muhlbaler Dasch, 1982). The mass size distribution from the ceramic-lined boiler with natural draught (j) was similar to the pellet cases. The shape of the mass size distribution in the flue gas from the old-type water-cooled multi-fuel boiler fired with small fuel wood batches (c) is similar to that of the modern wood boiler (j) except for higher emission level.

Old-type wood-log firing (c–f) led to emission of a larger number of particles than from wood pellets (m–s, u and v) and modern wood-log firing, which in their turn emitted more particles than oil combustion. Number size distributions are presented in Fig. 8. In the flue gas from wood pellet combustion, the variation in

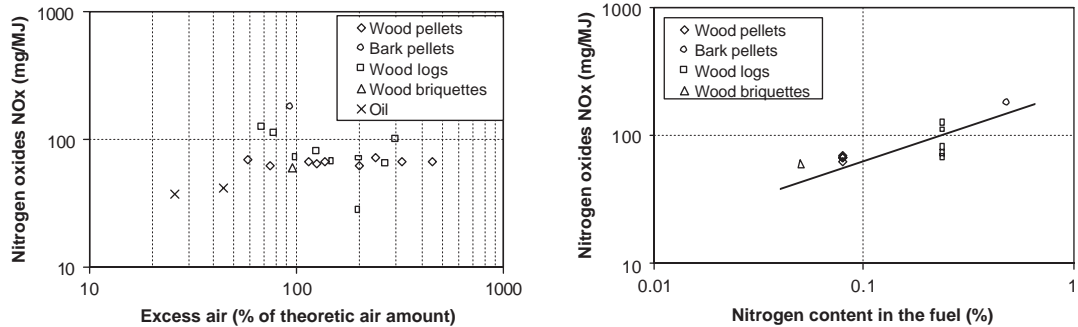


Fig. 6. Emission of nitrogen oxides (presented as NO_2) versus excess air for the cases in Table 1 to the left. Emission of nitric oxides (presented as NO_2) versus concentration of nitrogen in the fuel to the right. The data concern the range of excess air of $100 \pm 50\%$.

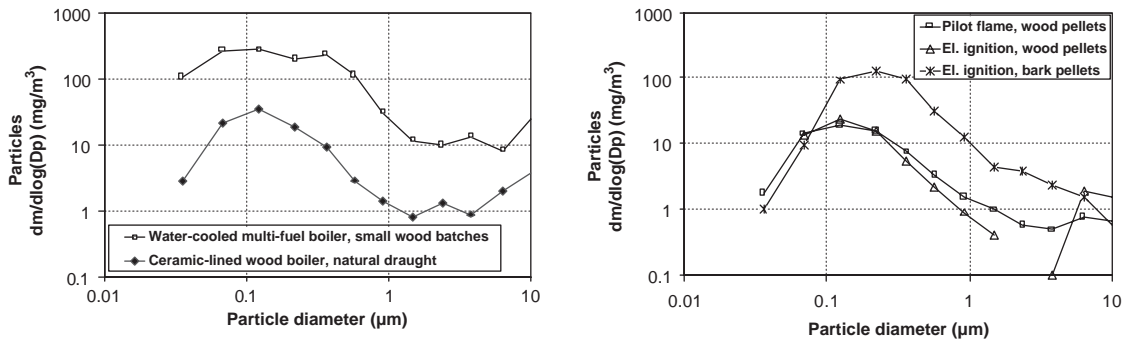


Fig. 7. Size distributions of particle mass in flue gas from wood boilers to the left (dry wood), and from pellet burners to the right (the concentrations are normalised to 10% O_2).

number size distribution was small, with maxima around 130 nm (m–o, q–s, u and v). However, the number size distribution in the flue gas from the pellet burner with pilot flame at 3 kW thermal outputs (p) had a somewhat increased emission without a distinct maximum and increased number of ultrafine particles (size < 100 nm). This case is more similar to combustion of wood logs. During combustion of wood logs, the number concentration increased with decreasing particle size (c–f and h–l), except for dry wood logs in one of the boilers with ceramic-lined burn-out zone (g), which resembles the pellet cases. As for wood logs, oil combustion (x and y) led to an increase in the number concentration as the particle size decreases. In the flue gas from oil burners there was a tendency of a supermicron maximum in the number size distribution.

3.2. Firing behaviour

Wood-log firing without a heat storage tank, typical for old-type wood boilers, is sensitive to firing behaviour. Emissions of unoxidised compounds and particles could be reduced by firing small wood batches and the particle emissions are similar to running the boiler

connected to a storage tank. It can also be noted that for wood boilers, the highest emissions of unoxidised compounds were obtained when the lowest NO_x concentrations were measured and reversibly, the lowest concentrations of unoxidised compounds were correlated to the highest NO_x values.

3.3. Fuel quality

Poor fuel quality yields high emissions also from new boilers. For instance, firing wet wood logs in a modern boiler (i) led to increased emissions of unoxidised compounds and particles. However, wood logs with moderate moisture content of 26% (h and k), as well as wood briquettes (l), 8% moisture content, had an almost similar effect on the emissions as dry wood (g and j), 15% moisture. Bark pellets (w) caused higher mass concentration of particles, which is probably explained by the higher ash content of this fuel compared to wood pellets. The maximum in particle mass size distribution for emissions from bark pellets was at a particle size around 220 nm, somewhat displaced towards larger particle size compared to wood pellets. The highest NO_x emission occurred during combustion of bark

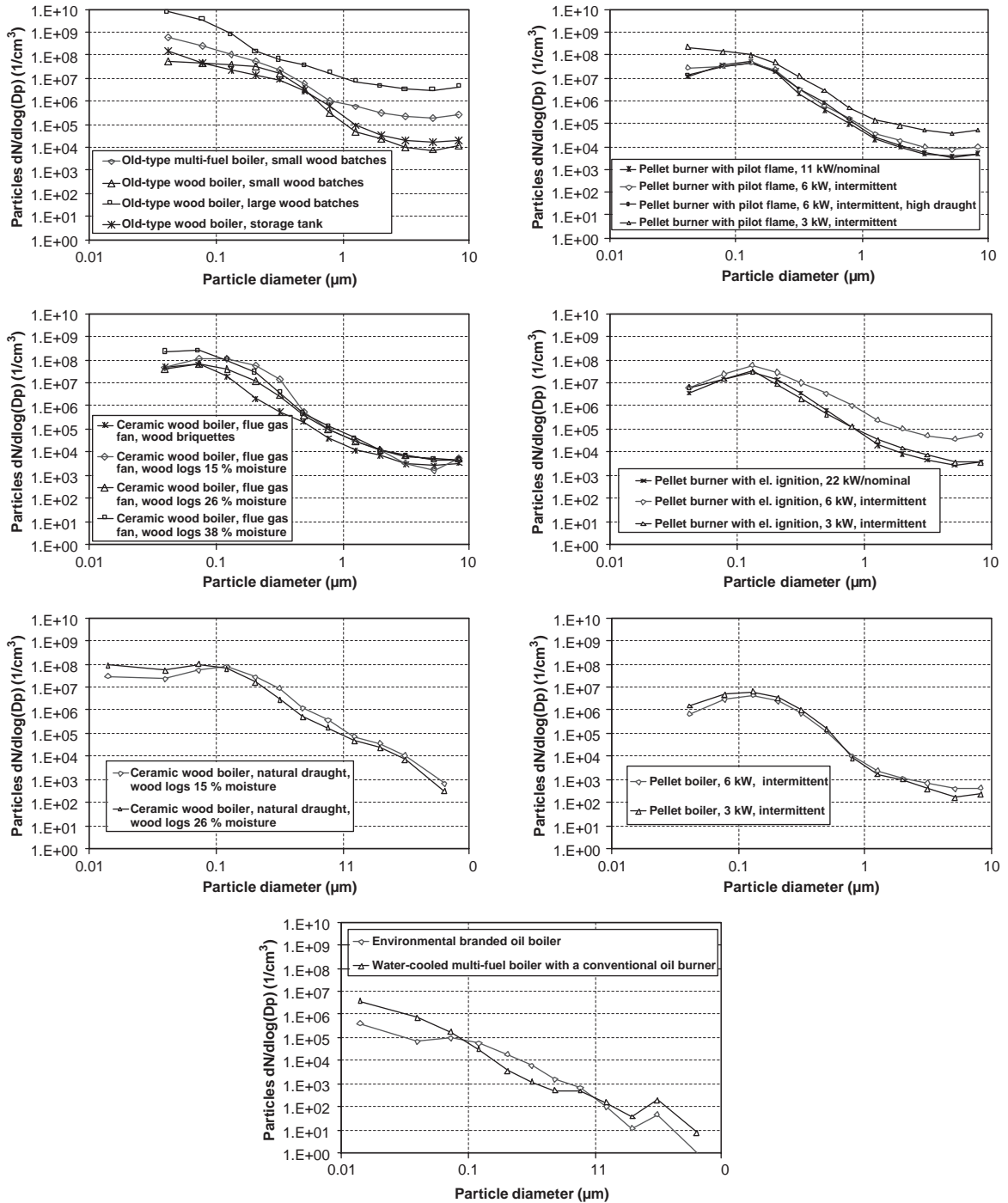


Fig. 8. Size distributions of particle number for wood boilers to the left, pellet burners to the right, and oil burners in the lower part of the figure (the concentrations are normalised to 10% O₂).

pellets, which was the fuel with the highest nitrogen content. In Fig. 6, the NO_x emission seems to be related to the nitrogen content in the fuel, which confirms earlier studies (Eskilsson et al., 2004).

3.4. Heat storage tank

By connecting a heat storage tank to an old-type wood boiler, the particle emission was reduced by up to

about 20 times (comparing d and e). The particle emission from a modern wood boiler was ca. 30% of those from an old-type wood boiler when both boilers were connected to heat storage tanks (d, g and j).

4. Discussion

The low emissions of particles and unoxidised gaseous compounds from oil burners were expected, since small-scale continuous combustion of a fluid fuel can be better controlled than batch-wise (wood boilers) combustion of solid fuel and semi-continuous combustion of pellets. However, it is possible to improve the control of batch-wise solid fuel combustion, as is shown in this study by comparing old-type wood boilers with modern wood boilers. This can probably be even better developed in the future. Pellet burners are new designs and can most likely be further improved to emission levels comparable to those of oil burners. Domestic pellet burners were developed in the 1980s, but not until the late 1990s these devices contributed to significant shares of the market in countries like Sweden, Denmark, and Austria (Baxter et al., 2002). Regardless of somewhat higher emissions of particles and unoxidised gaseous compounds from modern biofuel boilers compared with oil burners, modern domestic biofuel boilers are ecologically sustainable heating options as they do not contribute to the climate change. In contrast, domestic oil burners will always contribute to the climate change by emitting fossil CO₂.

Very large differences in emissions were observed between old-type and modern biofuel boilers. In particular, high emissions of the greenhouse gas methane were recorded for old-type wood boilers, which are the ones most frequently installed in houses today. The effect on climate change from an old-type wood boiler, because of high methane emissions, can be compared with residential oil burners emitting CO₂ by using the global warming potential (GWP). This parameter expresses the impact of a gas on the climate change in relation to CO₂. GWP for methane is 21 kg CO₂-equivalent/ kg methane (IPCC Guidelines for National Greenhouse Gas Inventories, 1996). The emission from the oil boilers was 74 000 mg CO₂ per MJ (calculated from case x and y) and from the worst old-type wood case (e) the methane emission was 4800 mg/MJ, which gives around 100 000 mg CO₂-equivalents/MJ. Besides multiplying these emissions with the GWP-factor, one should also consider that the efficiency of an old-type wood boiler is around 50%, whereas the efficiency of the oil boilers is typically 90%. Consequently, an old-type wood boiler may have more than twice as high an impact on climate change as an oil boiler, besides high emissions of particles and unoxidised gaseous compounds.

5. Conclusions

There are high emissions of unoxidised gaseous compounds and particles from old-type residential biofuel boilers. The measured VOC compound with highest concentration was methane in all cases, and as a consequence of methane emissions, the influence on climate change of an old-type wood boiler could be higher than that of an oil boiler. As an example, substitution of an old-type wood boiler with a modern wood boiler attached to a storage tank or with a pellet boiler could reduce methane emissions 8 (c and i) to 9000 (e and q) times, at the same time as the efficiency would increase. Installation of a heat storage tank in connection to an old-type wood boiler could reduce the methane emission seven times and the particle emission 21 times (d and e), thus improving the environmental performance to some extent. A similar improvement was observed when changing firing behaviour to charging smaller batches of wood logs. Modern wood boilers seem to maintain their low emission performance also in the case of degradation of the fuel quality in the form of increased moisture content. However, changing the fuel's composition from wood-based fuel like wood logs, wood pellets, and wood briquettes, to a fuel manufactured from a lower quality material, such as bark, results in higher emissions of particles and NO_x, because of higher ash and nitrogen content in bark.

Increased emissions of CO were found at high excess air ratios. This agrees with earlier studies on wood stoves (Nussbaumer, 2003), and is probably a result of cooling of the combustion process. The enhanced emissions of CO were accompanied by emissions of other unoxidised components: TOC, CH₄, NMVOC, and PAH. The mass concentration of emitted particles was rather independent of emission of unburnt gaseous compounds up to a concentration of about 100 mg/MJ TOC, after which the particle emission increased, probably because of high concentrations of unburnt particulate matter. The mass concentration of particles was 180 times larger in the worst old-type case (e) compared to the best modern case (u). The number of particles emitted increased with increased emissions of unoxidised gaseous compounds. From the number and mass size distributions it can be concluded that the emission of submicron particles, especially ultrafine particles (size < 100 nm) is enhanced by poorer combustion conditions.

Most biofuel boilers installed today are of the old type, and cause high emissions of particles and unoxidised gaseous compounds, as well as a climate change effect comparable with oil burners. It is urgent from both environmental and health point of view that they are replaced with modern residential wood boilers attached to storage tanks, or pellet boilers.

Acknowledgements

The financial support from the Swedish Energy Agency and from the Swedish Environmental Agency is gratefully acknowledged.

Appendix

Tables 4 and 5.

References

- Baxter, L., Coombes, P., Gifford, J., Guiu, G., Houmann Jakobsen, H., Koppejan, J., Livingston, W., Logie, R., Loo van, S., Nussbaumer, T., Obenberger, I., Oravainen, H., Schenkel, Y., Skreiberg, Ö., Tullin, C., 2002. Handbook of Biomass Combustion and Co-Firing. Twente University Press, Enschede, The Netherlands, pp. 93–110 (ISBN 9036517737).
- Eskilsson, D., Rönnbäck, M., Samuelsson, J., Tullin, C., 2004. Optimisation of efficiency and emissions in pellet burners. Biomass and Bioenergy, in press.
- Gaegauf, C., Wieser, U., Macquat, Y., 2001. In: Proceedings of the International Seminar of IEA Bioenergy Task 32: Aerosols from Biomass Combustion, Zürich, Switzerland. Address of responsible organization: Verenum, Langmauerstrasse 109, CH – 8006 Zürich, pp. 81–85.
- Glassman, I., 1996. Combustion, 3rd edition, Academic Press, San Diego, USA, p. 92.
- Hedberg, E., Kristensson, A., Ohlson, M., Johansson, C., Johansson, P.-Å., Swietlicki, E., Vesely, V., Wideqvist, U., Westerholm, R., 2002. Chemical and physical characterization of emissions from birch wood combustion in a wood stove. Atmospheric Environment 36, 4823–4837.
- IPCC Guidelines for National Greenhouse Gas Inventories Revised 1996 (IPCC/UNEP/OECD/IEA 1997), <http://www.ipcc-nggip.iges.or.jp/public/gl/invs1.htm>.
- ISO 11338, 2000. Stationary source emissions—Determination of mass concentration of polycyclic aromatic hydrocarbons.
- Johansson, L.S., Leckner, B., Tullin, C., Sjövall, P., 2003. Particle emissions from biomass combustion in small combustors. Biomass and Bioenergy 25, 435–446.
- McDonald, J.D., Zielinska, B., Fujita, E.M., Sagebiel, J.C., Chow, J.C., Watson, J.G., 2000. Fine particle and gaseous emission rates from residential wood combustion. Environmental Science and Technology 34, 2080–2091.
- Muhlbaler Dasch, J., 1982. Particulate and gaseous emissions from wood-burning fireplaces. Environmental Science and Technology 16 (10), 639–645.
- Ndiema, C.K.W., Mpendazoe, F.M., Williams, A., 1998. Emission of pollutants from a biomass stove. Energy Conversion and Management 39, 1357–1367.
- Nordic Ecolabelling of Oilburners and Oilburner/Boiler Combinations, Criteria Document, 5 October 2000–5 November 2004, Version 4; in: <http://www.svanen.nu/Eng/default.asp>.
- Nussbaumer, T., 2003. Combustion and co-combustion of biomass: fundamentals, technologies, and primary measures for emission reduction. Energy and Fuels 17 (6), 1510–1521.
- Oanh, N.T.K., Reutergårdh, L.B., Dung, N.T., 1999. Emission of polycyclic aromatic hydrocarbons and particulate matter from domestic combustion of selected fuels. Environmental Science and Technology 33, 2703–2709.
- Olsson, M., Kjällstrand, J., Petersson, G., 2003. Specific chimney emissions and biofuel characteristics of softwood pellets for residential heating in Sweden. Biomass and Bioenergy 24, 51–57.
- Rau, J.A., 1989. Composition and size distribution of residential wood smoke particles. Aerosol Science and Technology 10, 181–192.
- SS028426, 1991-12-04. Luftundersökningar - Utsläpp till luft - Bestämning av koncentration och mass-flöde stoft i gaskanaler - Manuell gravimetrisk metod. The Swedish Standardisation Commission.
- Swedish chimney-sweepers' statistics to the Swedish Rescue Services Agency, 2001.
- Wieser, U., Gaegauf, C., 2000. In: Proceeding of the Conference: First World Conference and Exhibition on Biomass for Energy and Industry, Sevilla, 805-808. Address of responsible organizations: Energia TA, Piazza Savonarola 10, I-50132 Florence, Italy, WIP, Sylvesteinstrasse 2, D-81369 Munich, Germany.
- Zhang, J., Smith, K.R., Ma, Y., Ye, S., Jiang, F., Qi, W., Liu, P., Khalil, M.A.K., Rasmussen, R.A., Thorneloe, S.A., 2000. Greenhouse gases and other airborne pollutants from household stoves in China: a data base for emission factors. Atmospheric Environment 34, 4537–4549.