

Low emissions from wood burning in an ecolabelled residential boiler

Maria Olsson, Jennica Kjällstrand*

Department of Chemical and Biological Engineering, Chalmers University of Technology, SE-41296 Göteborg, Sweden

Received 13 June 2005; received in revised form 27 October 2005; accepted 1 November 2005

Abstract

Emissions of organic compounds from wood burning in a modern ecolabelled residential boiler (30 kW) were studied. Smoke was collected in the chimney outlet at different times during the burning cycle for subsequent analysis by gas chromatography.

The studied ecolabelled wood boiler had high combustion efficiency, and the flaming phase emissions were very low. The greenhouse gas methane (CH_4) was determined in low concentrations of about a few mg m^{-3} and was the major volatile hydrocarbon emitted. The CH_4 emission factor was calculated to 0.04 g kg^{-1} dry fuel. Benzene, in the range $0.1\text{--}1 \text{ mg m}^{-3}$, was the predominant aromatic compound emitted. Other major aromatic compounds were methylbenzene, dimethylbenzenes and ethenylbenzene. The concentrations of the studied polycyclic aromatic compounds were generally low, except for naphthalene, which was the third most prominent aromatic compound. However, the total emissions of these health and environmentally hazardous compounds were low.

The already low emissions of most of the organic compounds decreased further towards the end of the burning cycle, although the concentration of carbon monoxide (CO) increased. This indicates that large CO emissions are not necessarily linked to large emissions of organic compounds. Relative to benzene, the concentrations of many of the aromatic compounds studied were higher in the glowing combustion phase, than in the flaming combustion phase.

The total environmental and health impact of the studied emissions from the ecolabelled boiler is considered to be low. This wood boiler can be recommended as an environmentally sound residential heating alternative.

© 2005 Elsevier Ltd. All rights reserved.

Keywords: Air pollution; Biofuel; GC-MS; Hydrocarbon; PAH

1. Introduction

In Sweden, as in many other countries, numerous homes are heated by small-scale wood burning. At

present, residential combustion of biomass accounts for more than 10 TWh of Sweden's energy supply (Gustavsson et al., 2001) and is a more common residential heating option than light fuel oil. Biomass is used for heating in about 600 000 Swedish homes. Sweden's total energy consumption in 2003 was 400 TWh (Swedish Energy Agency, 2004). Approximately 50 TWh of this was used for residential heating (Gustavsson et al., 2001). The

*Corresponding author. Tel.: +46 31 7722641; fax: +46 31 7722639.

E-mail address: jennica.kjallstrand@chalmers.se (J. Kjällstrand).

use of biofuels is expected to increase in accordance with the effort to achieve a sustainable energy supply without further adding to global warming. Emission data are essential for estimating the environmental and health effects of the present, and the future increased, small-scale biomass burning. Although it is desirable to increase the use of biofuels due to both environmental and economic reasons, it is also important to protect the local environment from emissions that are hazardous to health.

Today, all new wood boilers installed in urban areas in Swedish must comply with the emission limits set by the authorities. More than 90% of all boilers sold in Sweden today are environmentally approved for use in urban areas.

Several countries have developed an environmental labelling system to help consumers choose more environmentally friendly products. “The Swan” is the name of the Nordic ecolabel and includes criteria for wood boilers (Nordic Ecolabelling Board, 2004). The first ecolabelled wood boiler “Baxi Solo Innova” is one of the most popular models in Sweden today and almost 6000 units have been installed.

The present Swedish emission limits for wood boilers, set out in the regulations issued by the Swedish authority (Swedish Board of Housing Building and Planning, 2002), are listed in Table 1, together with emission limits laid down for the Nordic ecolabel The Swan.

Both the Swedish authorities and The Nordic Ecolabelling Board restrict the emissions of organic gaseous carbon (OGC, Table 1). This is a non-specific parameter, in which methane (CH₄) is included and represents a large proportion. The smoke constituents of OGC differ greatly from each

other in terms of their effects on the environment and health. Specific compounds have to be studied in order to estimate these effects.

Smoke from residential wood burning constitutes a complex mixture of organic compounds. Methoxyphenols and 1,6-anhydroglucose are primary thermal decomposition products from lignin and cellulose, respectively, which are released at inefficient burning in, for example, traditional devices such as open fireplaces or tiled stoves (Fine et al., 2004a, b; Kjällstrand, 2002; Simoneit, 2002). More efficient burning leads to a reduced total amount of organic compounds in the smoke. However, the proportion of benzene and polycyclic aromatic compounds increases (Alén et al., 1996; Kjällstrand and Petersson, 2001b).

Wood pellets are a relatively new heating fuel. The three different combustion alternatives for residential heating are pellet burners installed in boilers, pellet stoves and pellet boilers. Highly efficient combustion in correctly installed devices significantly decreases the emissions (Johansson et al., 2004; Olsson et al., 2003).

New technologies for residential burning of wood logs have been developed, with drastically reduced emissions (Johansson et al., 2004). The aim of this study was to investigate the emissions of organic compounds from a modern and efficient ecolabelled wood boiler and to show that it is possible to use biofuels in residential boilers with low emissions of organic compounds. The studied boiler is an example of such a new technology for low-emission and sustainable firewood combustion.

2. Methods

Birchwood was burnt in an ecolabelled residential wood boiler. Smoke samples were collected in the chimney outlet for subsequent analyses by gas chromatography. The methods used for sampling and analysis are shown in Fig. 1.

2.1. The wood boiler

Dry birchwood logs were burned in a Baxi Solo Innova residential wood boiler (30 kW), installed in the premises of the Swedish boiler distributor Baxi AB. The boiler was connected to a large hot-water storage tank and was intended for heating and hot-water production. The Baxi Solo Innova wood boiler is ecolabelled and was manufactured in 2001. The boiler is described in more detail in Section 3.6.

Table 1
Present limits for emissions to air from wood boilers (mg m⁻³ dry gas at 10% O₂) (Nordic Ecolabelling Board, 2004; Swedish Board of Housing Building and Planning, 2002)

Boiler effect	Swedish authorities < 50 kW	The Swan (ecolabel) < 100 kW
Organic gaseous carbon (OGC)	150	70
Carbon monoxide	—	1000 ^a /2000 ^b
Particles	—	70

^aAutomatically fed boilers.

^bManually fed boilers.

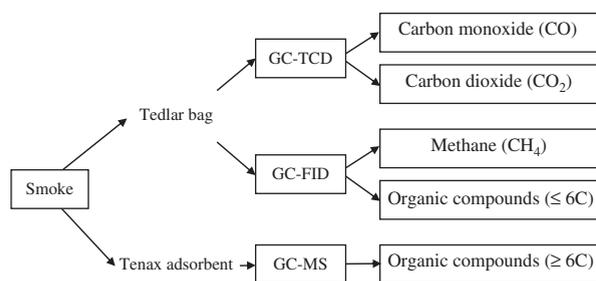


Fig. 1. Methods used for sampling and analysis of the different compounds and groups of compounds.

On four different sampling occasions, A, B, C and D, the boiler was loaded with 10–40 kg of dry birchwood. When using the largest volume (occasion D, 38 kg), the burning period exceeded 5 h.

2.2. Smoke sampling

Smoke samples were taken at the top of the chimney outlet in regular intervals during the combustion cycle (two to five samples per burning phase during the 2–5 h long combustion cycle). About 20 measurements of 25–35 different compounds were made.

Samples for subsequent analysis of organic compounds (≥ 6 carbon atoms) were taken using Tenax adsorbent cartridges (TA 60–80 mesh, 0.8 mm \times 30 mm) coupled to an air sampling pump (2–4 ml min⁻¹). The sampling volumes were deliberately varied between 5 and 40 ml.

Smoke was also collected in Tedlar bags for gas chromatographic analysis of carbon dioxide (CO₂), carbon monoxide (CO) and organic compounds ($\leq 6C$) including CH₄. The concentrations of CO₂, CO, nitrogen oxide and oxygen were also measured using an electrochemical cell instrument (Testo 300M, Nordtec Instrument AB).

During sampling, the smoke temperature in the chimney outlet was 50–60 °C. The gas velocities in the chimney outlet were measured using Testo 445 (Nordtec Instrument AB) and revealed an average value of 1.7 m s⁻¹. The length of the flue-gas pipe, from the boiler to the chimney outlet, was about 10 m.

2.3. Determination of organic compounds ($\geq 6C$)

Organic compounds ($\geq 6C$) were determined by gas chromatography-mass spectrometry (GC-MS). The adsorbent cartridges with the smoke samples were thermally desorbed at 220 °C in the injector of

a Varian 3800 gas chromatograph to the column (30 m \times 0.25 mm, Rtx 1701, Restek, 0.25 μ m phase layer). Helium (10 psi) was used as a carrier gas. The oven temperature was increased by 10 °C min⁻¹ from -50 to 50 °C and then by 5 °C min⁻¹ to 250 °C. The gas chromatograph was linked to a Varian Saturn 2000 mass spectrometer (ion trap at 120 °C, EI ionisation at 70 eV, mass range m/z 35–400).

The organic compounds were identified from their mass spectra, relative retention times and by comparisons with data from previous studies (Kjällstrand, 2002; Kjällstrand and Petersson, 2001b; Olsson et al., 2003, 2004). The concentrations were determined by integrating the peak areas in the total ion current chromatograms. The same concentrations of benzene were assumed for the GC-MS analyses as for the gas chromatography-flame ionisation detector (GC-FID) analyses. The response was set equal for all compounds studied by GC-MS.

2.4. Determinations of organic compounds ($\leq 6C$)

On sampling occasion D, organic compounds (≤ 6 carbon atoms, including CH₄) were determined by GC-FID. A sampling loop (3.23 ml) was filled with smoke from the Tedlar bags and injected to a Varian 3400 gas chromatograph. A PLOT aluminium oxide column was used (Al₂O₃/5% KCl, 50 m \times 0.32 mm i.d.). The oven temperature was kept at 0 °C for 5 min, increased 15 °C min⁻¹ to 110 °C and kept for 20 min, and finally increased by 10 °C min⁻¹ to 200 °C and kept for 30 min. Helium was used as a carrier gas (2 ml min⁻¹) and nitrogen as a make-up gas (20 ml min⁻¹). The compounds were identified from their retention times. The peak areas in the chromatogram were integrated and the concentrations of the total emissions were calculated. A reference gas mixture (BTEX reference gas, Scott Speciality Gases) with 10.5 ppm benzene and 10.5 ppm methylbenzene ($\pm 5\%$) was used for calibration.

2.5. Determination of carbon monoxide, carbon dioxide and methane

One gas chromatograph with a thermal conductivity detector (GC-TCD) was used to determine CO and CO₂, while one gas chromatograph with a flame ionisation detector was used to determine CH₄. The different instrumental conditions are described in Table 2. Gas sampling valves were

Table 2
Experimental conditions for the analytical determination of carbon monoxide, carbon dioxide and methane

	CO	CO ₂	CH ₄ ^a	CH ₄ ^b
Gas chromatograph	Perkin Elmer 3920b		Perkin Elmer 990	Varian 3400
Detector	Thermal conductivity detector ^c		Flame ionisation detector	
Carrier gas	He	He	N ₂	He
Column	Molecular sieve ^d	HayeSep Q ^e	Molecular sieve ^d	Al ₂ O ₃ /5% KCl ^f
Temperature (°C)	80	80	80	0–200
Injected volume (μl)	190	290	1200	3230

^aOccasion B and C

^bOccasion D

^c200 °C, 150 mA

^d13 ×, 1.7 m × 3.2 mm o.d.

^e1.9 m × 3.2 mm o.d.

^fPLOT, 50 m × 0.32 mm i.d.

used for injections. The instruments were calibrated by injecting pure CO, CO₂ and CH₄ from gas tubes.

3. Results and discussion

About 30 selected organic and inorganic compounds were determined in the smoke from the ecolabelled wood boiler at different times during the combustion cycle. The results are presented together with a discussion of the environmental impact and health effects of the various studied compounds, in addition to burning technology and sampling aspects.

3.1. Inorganic compounds and organic compounds (≤6C)

Average concentrations of some selected compounds in chimney smoke from the initial flaming combustion phase and the subsequent glowing combustion phase of the studied boiler are presented in Table 3. The duration of these two combustion phases is approximately equal.

Compared to glowing burning, flaming burning resulted in higher emissions of almost all compounds reported in Table 3. However, the CO emissions were higher during glowing burning. The greenhouse gas CH₄ constituted a large part of the organic emissions. Other major organic compounds (≤6C) were ethene, ethyne and benzene. Ethane and furan were not found in the smoke in detectable concentrations. Similar concentrations of CO₂, CO and CH₄ from the glowing combustion phase were measured on other sampling occasions. In a previous study in laboratory scale, birchwood

Table 3

Average concentrations (mg m⁻³) of carbon dioxide, carbon monoxide and organic compounds (≤6C) from the initial flaming phase and the subsequent glowing combustion phase (occasion D)

	Flaming ^a	Glowing ^b
Carbon dioxide	120 000	46 000
Carbon monoxide	200	5400
Methane	3.6	1.2
Ethene	1.7	0.04
Propene	0.05	<0.03
Ethyne	0.71	0.06
Benzene	0.36	<0.04
Combustion efficiency ^c	99.7	80.9

The values for organic compounds (≤6C) are based on GC-FID analyses.

^aAverage of four samples.

^bOne sample.

^cCO₂/(CO₂+CO), calculated on ppm basis.

embers were characterised by higher emissions of volatile hydrocarbons relative to CO and CO₂ (Olsson and Petersson, 2003).

The concentration of nitrogen oxide was about 100 ppm during the flaming phase and five times lower during the glowing phase of the burning cycle. The concentration of oxygen was 14% during the flaming phase and increased a few percent towards the end of the burning cycle. Acetic acid, formaldehyde and methanol are too polar to be determined by the equipment used, but are known to be present in smoke from incomplete burning at concentrations of a few percent of those of CO (McKenzie et al., 1995; Yokelson et al., 1999).

3.2. Aromatic compounds

Relative concentrations of some selected aromatic compounds determined by GC-MS are reported in Table 4. The relative concentration of benzene was arbitrarily set to 100 for both the flaming and the glowing combustion phase. The total concentrations of benzene were in the range 0.5 mg m^{-3} for flaming burning and 0.1 mg m^{-3} for glowing burning. Benzene, methylbenzene, naphthalene and dimethylbenzenes (three isomers) were the most prominent aromatic compounds.

Earlier studies indicate that, compared to most other compounds, the proportion of benzene increases steadily with combustion efficiency (Kjällstrand, 2002; Kjällstrand and Olsson, 2004). The concentration of benzene in smoke from the ecolabelled boiler was, however, very low. As an example, an earlier study of a residential wood stove from 1981 reported 30 times higher concentrations of benzene (Kjällstrand and Olsson, 2004). Another study of residential biomass combustion reported emission data for benzene from modern wood

boilers and pellet combustion appliances which were in the same range as data from this study (Johansson et al., 2004). Emission data for benzene from old-type wood boilers in the same study were about 100 times higher.

The smoke from the ecolabelled wood boiler contained two and three times as much benzene as methylbenzene, for flaming and glowing combustion, respectively. It is characteristic of wood smoke that the concentrations of benzene are higher than those of methylbenzene (Barrefors and Petersson, 1995; Olsson et al., 2003; Schauer et al., 2001). In contrast, the concentration of methylbenzene in urban air has been found to be approximately twice as high as that of benzene (Barrefors and Petersson, 1992), which is the same relationship as for vehicle emissions.

Relative to benzene, the concentrations of many compounds studied were higher from glowing combustion, for example dimethylbenzenes and methylphenols. In contrast, the relative concentrations of naphthalene and acenaphthylene from the flaming combustion phase were twice as high as from the glowing phase. For the other polycyclic aromatic compounds, the relative variation between the flaming and glowing phases was less evident. A chromatogram, showing the major aromatic compounds in the smoke from flaming burning, is presented in Fig. 2.

The concentrations of the selected polycyclic aromatic compounds were generally low, except for naphthalene, which was the third most prominent aromatic compound. Previous analyses of the smoke from a residential wood stove from 1981 reported 10 times higher concentrations of pyrene (Kjällstrand and Olsson, 2004). Another study reported more than 100 times higher emissions of polycyclic aromatic hydrocarbons from old-type wood boilers than from modern wood boilers (Johansson et al., 2004).

The polycyclic aromatic compounds assessed from the ecolabelled wood boiler, indene, naphthalene, acenaphthene, acenaphthylene, phenanthrene, anthracene, fluoranthene and pyrene, have previously been studied in smoke from conifer wood burning (Fine et al., 2001, 2004a, b; Freeman and Cattell, 1990; Jenkins et al., 1996; McDonald et al., 2000).

The low concentrations of, for example, phenanthrene and pyrene indicate efficient combustion at high temperatures, which corresponds to the boiler distributor's information that the combustion

Table 4

Relative concentrations (based on peak areas) and standard deviations (S.D.) of selected organic compounds ($\geq 6\text{C}$) from the flaming and glowing combustion phases

	Flaming ^a	S.D.	Glowing ^b	S.D.
Benzene	100	—	100	—
Methylbenzene	32	19	53	29
Ethylbenzene	4.2	3.1	7.6	4.8
Dimethylbenzenes (three isomers)	13	9	48	37
Ethynylbenzene	2.6	1.5	0.8	0.9
Ethenylbenzene	11	10	5.7	6.5
Benzofuran	9.6	7.3	8.1	5.9
Indene	3.9	3.6	1.9	2.2
Phenol	9.1	7.9	12	9
Methylphenols (three isomers)	6.2	7.8	19	18
Naphthalene	35	28	18	6
Methylnaphthalenes (two isomers)	9.0	8.4	9.0	6.9
Acenaphthene	2.1	1.8	1.4	1.0
Acenaphthylene	8.0	5.3	3.0	2.4
Dibenzofuran	4.2	3.3	3.6	2.2
Phenanthrene	10	6	11	11
Anthracene	2.2	1.1	1.2	0.8
Fluoranthene	4.9	4.6	4.6	5.6
Pyrene	9.5	7.6	9.5	10.8

The concentration of benzene was arbitrarily set to 100.

^aAverage of 10 samples.

^bAverage of nine samples.

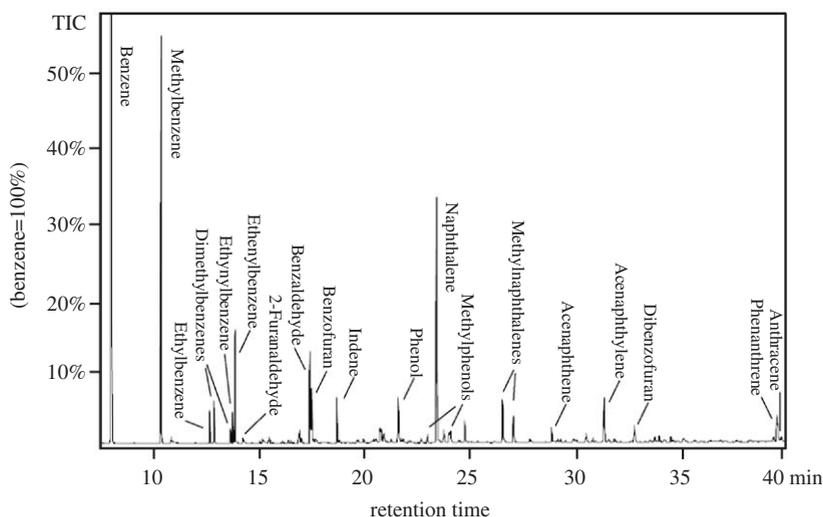


Fig. 2. Section of the total ion current chromatogram illustrating the main organic compounds ($\geq 6\text{C}$) as a percentage of the highest peak (benzene, 100%) of the flaming combustion phase of the wood boiler (40 min into the burning cycle, occasion C).

temperature is approximately $1000\text{ }^{\circ}\text{C}$. The emissions of polycyclic aromatic compounds are strongly influenced by the burning conditions (Alén et al., 1996; Jenkins et al., 1996). Polycyclic aromatic compounds are mainly formed at temperatures of between 700 and $900\text{ }^{\circ}\text{C}$, and their total yield increases with temperature and residence time in the furnace within this interval (Alén et al., 1996; Kjällstrand, 2002; McGrath et al., 2001). At higher combustion temperatures they are thermally decomposed (Kjällstrand, 2002; Olsson et al., 2003). The emissions of health hazardous aromatic compounds were much higher for a non-environmentally approved wood boiler developed in the 1970s (Kjällstrand and Petersson, 2001b).

Methoxyphenols (2-methoxyphenols and 2,6-dimethoxyphenols) were almost absent in the smoke from the Baxi Solo Innova. They are primary degradation products from wood lignin and are abundant in smoke from burning at combustion temperatures below $700\text{ }^{\circ}\text{C}$ (Alén et al., 1996; Kjällstrand, 2002). Examples are traditional burning devices such as open fireplaces and tiled stoves. Almost all methoxyphenols are thermally decomposed at the high combustion temperatures in modern wood boilers (Amen-Chen et al., 2001). Methoxyphenols have been assessed in smoke from pellet stoves in an earlier study, but not in emissions from pellet burners (Olsson et al., 2003). Newsprint is a known source of coniferyl alcohol (Kjällstrand and Petersson, 2000). During the study the boiler was set aflame using cardboard, instead of news-

print, which would result in a different smoke composition. However, at such high combustion temperatures, coniferyl alcohol is almost entirely decomposed thermally.

1,6-Anhydroglucos and 2-furaldehyde were identified in the smoke. They are primary pyrolysis products of cellulose, hemicelluloses and starch (Edye and Richards, 1991; Simoneit, 2002). Their polar characteristics make them difficult to quantify in low concentrations on the used column. The estimated relative emissions of 1,6-anhydroglucos and 2-furaldehyde amounted to a few percent of the benzene emissions.

The standard deviation is high for some of the compounds in Table 4. This was expected, since the aim was to study the natural combustion cycle in a residential boiler as opposed to attempting to control or modulate the combustion. The samples were also taken at different times during the relatively long flaming and glowing phases.

3.3. Variation during the burning cycle

The variations in the concentrations of CO_2 , CO , CH_4 , ethene, benzene, methylbenzene, naphthalene and phenanthrene over time for one specific burning occasion (D) are illustrated in Fig. 3. The logarithmic diagram is based on five samples and indicates the changes in the concentrations of selected key compounds. The concentrations of most compounds decrease over the combustion cycle. The rapidly decreasing concentrations of CH_4 and

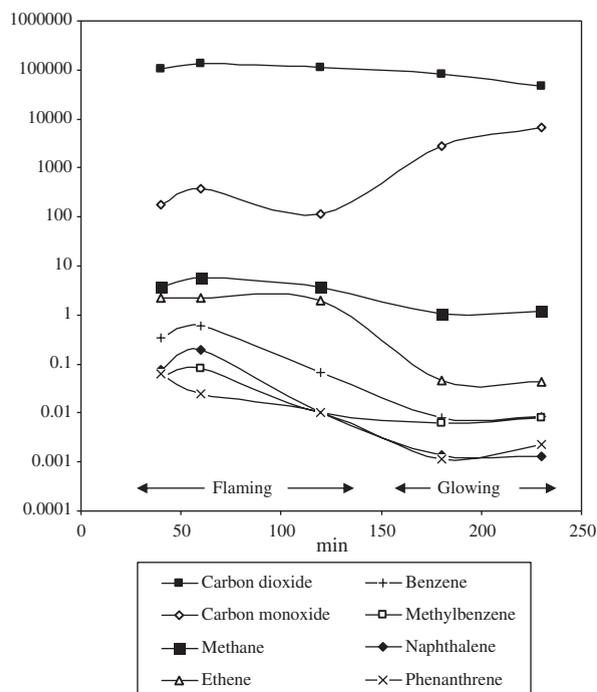


Fig. 3. Concentrations of selected compounds in smoke from the studied wood boiler during a full burning cycle (mg m^{-3}).

ethene after 120 min in Fig. 3 are considered to reflect the transition to the coke combustion phase. After 180 min only glowing combustion occurred and the total release of organic compounds was very low at the end of the burning cycle. These transition times depend on the amount of firewood.

CO was the only flue-gas component with increased concentration towards the end of the burning cycle. During the flaming combustion phase the concentration of CO was relatively constant at about 200 mg m^{-3} . During the subsequent glowing phase (coke combustion), the concentration increased to approximately 5000 mg m^{-3} .

The Nordic Ecolabelling Board's criteria include CO (Table 1). High concentrations of CO indicate incomplete combustion, but are not necessarily linked to large emissions of organic compounds (Fig. 3). It has earlier been suggested that high CO concentrations indicate emissions of organic compounds (McKenzie et al., 1995), but that is not supported according to the results in Fig. 3. Most organic compounds were emitted during the gas combustion phase during the first half of the burning cycle, when the CO concentration was relatively low. The emission of CO increased during the subsequent coke combustion phase, while the emissions of other compounds remained low.

3.4. The greenhouse gas methane

CH_4 is the predominant hydrocarbon in smoke from biomass burning. It is not hazardous to health, but contributes to global warming as it is a strong greenhouse gas compared to CO_2 . CH_4 emissions decrease in line with increasing combustion efficiency, implying great differences between various methods of burning. CH_4 emissions should therefore be one of the parameters considered in the environmental labelling of new combustion devices for small-scale burning of biomass. Both the Swedish authorities and The Nordic Ecolabelling Board restrict the emissions of OGC (Table 1). This is a non-specific parameter in which CH_4 is included and constitutes a large proportion.

The CH_4 emissions from the ecolabelled boiler were equivalent to an emission factor of about 0.04 g CH_4 per kg of dry fuel burned and are in the same range as the CH_4 emissions from a modern pellet burner (Table 5) (Kjällstrand, 2002; Olsson et al., 2003).

The CH_4 emission factor (EF_{CH_4} , g kg^{-1}) represents the proportion of carbon released as EF_{CH_4} during combustion. It was estimated based on an average carbon content of 45% of the dry wood (Delmas, 1994) and calculated as

$$\text{EF}_{\text{CH}_4} = ([\text{CH}_4]_{\text{plume}} - [\text{CH}_4]_{\text{ambient}}) / [\text{CO}_2] \times 0.45 \\ \times 1000 M_{\text{CH}_4} / M_{\text{C}} \times [\text{CO}_2] / ([\text{CO}_2] + [\text{CO}]),$$

where 1.7 ppm was used for $[\text{CH}_4]_{\text{ambient}}$ and all concentrations were expressed in ppm (Yokelson et al., 1999).

Table 5

Emissions of methane from different types of biomass burning (g methane per kg fuel) (Kjällstrand, 2002)

Fuel	Burning	Emission factor
Wood	Ecolabelled boiler	<0.1
Pellets	Burner	<0.1 ^a
Pellets	Stove	<0.1 ^a
Wood	Residential combustion appliances	0.02–10 ^b
Wood	Traditional cookstoves	0.5–10 ^c
Grassland	Fire	1–4 ^d
Forest	Fire	2–6 ^d

^a(Olsson, 2002; Olsson et al., 2003).

^b(Johansson et al., 2004; Kjällstrand and Olsson, 2004; Schauer et al., 2001).

^c(Bhattacharya et al., 2002; Ndiema et al., 1998).

^d(Ferek et al., 1998; Goode et al., 2000; Yokelson et al., 1999).

Small-scale burning in inefficient devices emits about 1–10 g CH₄ per kg fuel (Bhattacharya et al., 2002; Ndiema et al., 1998; Schauer et al., 2001). The burning of wood logs and pellets in modern devices lowers the CH₄ concentration in the smoke plume to the same occurrence as in ambient air.

The use of biofuels from sustainable ecosystems with continual regrowth does not contribute to an increased greenhouse effect with regard to CO₂ emissions. Overall, a change from fossil fuels to biofuels significantly reduces the emissions of greenhouse gases. Despite the CH₄ emissions, modern small-scale appliances from wood combustion, such as pellet appliances or the studied ecolabelled wood boiler, only contribute a few percent of the greenhouse effect arising from heating with fossil fuels, according to the emission factors in Table 5. However, since there are major differences between biomass burning devices, reducing the emissions of CH₄ from residential heating is a relatively cheap and easy way to decrease the emissions of greenhouse gases.

3.5. Health effects of residential wood burning

The emissions of the studied hazardous compounds in the smoke from the ecolabelled wood boiler were low. For example, the concentrations of ethene, propene, benzene and methylbenzene in the chimney outlet were even lower than those in the air in a short road tunnel (Barrefors and Petersson, 1992). Emissions at chimney level result in lower human exposure compared to ground level emissions.

The emissions of particulate matter have been found to be lower from modern wood boilers compared to older boilers (Johansson et al., 2004). Another study of the ecolabelled wood boiler showed particularly low emissions of PCDD/F and PCB (Hedman, 2005). The total health and environmental impact of the ecolabelled boiler can therefore be considered low.

Previous studies have shown that the emissions of organic compounds from traditional burning devices, such as open fireplaces and tiled stoves, are higher than those formed in the ecolabelled boiler. Furthermore, these emissions are to a larger extent composed of primary degradation products from lignin and cellulose, which are less hazardous (Kjällstrand, 2002). Methoxyphenols are one group of primary degradation products from traditional wood burning. They are antioxidants and might

therefore counteract the negative health effects of more hazardous compounds in the smoke from small-scale wood burning (Barclay et al., 1997; Kjällstrand and Petersson, 2001a; Ogata et al., 1997). Methoxyphenols were almost totally absent in the smoke from the ecolabelled wood boiler. Hence, this smoke has no antioxidant properties.

In a previous study, a traditional tiled stove was compared to a boiler developed in the 1970s and connected to a hot-water storage tank (Kjällstrand and Petersson, 2001b). The boiler was not environmentally approved. The emissions from the tiled stove were dominated by methoxyphenols. The boiler, on the other hand, had large emissions of secondary degradation products, such as benzene and polycyclic aromatic hydrocarbons, which are hazardous to health.

3.6. Burning technology for wood boilers

The Baxi Solo Innova wood boiler is one of the most popular models in Sweden today. In 2001, it was the first wood boiler to receive the Nordic ecolabel The Swan. The development of very efficient wood boilers has increased over the last 10 years (Strehler, 2000). Complete combustion of organic compounds requires a high temperature and sufficient residence time in the combustion area. Highly efficient residential boilers are constructed for reversed combustion and have ceramic combustion chambers.

A unit with reversed combustion has the advantage of longer burning intervals for one load of wood. The woodpile is burnt from below, and pieces of wood continuously drop down into the combustion chamber, leading to uniform heat development throughout the burning cycle. The function of the ceramic combustion chamber is to maintain a high combustion temperature during the whole burning cycle, resulting in decreased emissions during the flaming phase and an increased energy yield during the glowing combustion phase. Two-step air intake guarantees an adequate oxygen supply. The residence time is increased by long gas-paths. This is important for complete combustion of organic compounds and for efficient heat transfer from the hot smoke gases. Heat storage in a large water tank makes it possible to run the boiler at a high effect without over-heating.

Most wood boilers used for residential heating today are old and emit large quantities of organic compounds. The installation of a pellet burner and

the change to wood pellets normally leads to a marked decrease in emissions (Olsson et al., 2003). The production and use of wood pellets has increased steadily in Sweden over recent years (Hillring and Vinterbäck, 1998; Olsson, 2004). Although the change from wood logs to pellets significantly decreases emissions, considerable differences exist between pellet burners, boilers and stoves as well as between various combinations of pellet burners and boiler furnaces (Olsson et al., 2003).

3.7. Sampling aspects

Birchwood is a very common species of firewood used in Sweden and was therefore a natural choice. At very efficient combustion, as in the boiler used, the choice of tree species is not likely to alter the results.

It is only possible to measure the actual emissions to the atmosphere in the chimney outlet. Sampling closer to the combustion chamber may include less volatile compounds that normally condense onto the walls of the chimney, forming soot and tar for the chimneysweeper to remove. Chimney sampling at specific devices allows data from genuine, although not always perfectly installed, appliances.

The use of Tenax cartridges and Tedlar bags permits storage and transportation of samples. Adsorbent sampling with Tenax additionally provides an option to identify both air borne and particle borne organic compounds, and allows detection of compounds in low concentrations, since larger sampling volumes are permitted. When sampling with adsorbent cartridges, it is important to be aware of the risk of breakthrough of the most volatile compounds. Decomposition of and reactions between compounds adsorbed by the Tenax adsorbent at high temperatures and in the presence of nitrogen oxides must also be considered.

Parallel measurements of CO and CO₂, using a portable electrochemical cell instrument, confirmed the results from the GC-TCD analyses.

4. Conclusions

The studied ecolabelled wood boiler has high combustion efficiency, with optimal energy use of the fuel when connected to a hot-water storage tank. The emissions of the greenhouse gas CH₄ were low, as were the emissions of the other studied compounds hazardous to health and the environ-

ment, for example benzene and polycyclic aromatic compounds. The total health and environmental impact of the ecolabelled boiler can therefore be considered low. This wood boiler is recommended as an environmentally sound residential heating option.

With regard to human health, future emission requirements for the ecolabelling of small-scale combustion appliances for wood logs and pellets should include benzene and a representative polycyclic aromatic compound. From a global environmental perspective, the greenhouse gas CH₄ should be included as well. Since the concentration of CH₄ in emissions from wood burning decreases with improved combustion efficiency, high CH₄ concentrations indicate poor energy management. These CH₄ concentrations should be related to the concentration of CO₂, in order to eliminate dependence on dilution.

Acknowledgements

Specialists from Baxi AB provided generous assistance and information. Associate Professor Olle Ramnäs at Chemical Environmental Science, Chalmers University of Technology, assisted with the analytical equipment. The financial support from the Swedish Energy Agency and the Adlerbert Research Fund is gratefully acknowledged.

References

- Alén, R., Kuoppala, E., Oesch, P., 1996. Formation of the main degradation compound groups from wood and its components during pyrolysis. *Journal of Analytical and Applied Pyrolysis* 36 (2), 137–148.
- Amen-Chen, C., Pakdel, H., Roy, C., 2001. Production of monomeric phenols by thermochemical conversion of biomass: a review. *Bioresource Technology* 79 (3), 277–299.
- Barclay, L.R.C., Xi, F., Norris, J.Q., 1997. Antioxidant properties of phenolic lignin model compounds. *Journal of Wood Chemistry and Technology* 17 (1–2), 73–90.
- Barrefors, G., Petersson, G., 1992. Volatile hazardous hydrocarbons in a Scandinavian urban road tunnel. *Chemosphere* 25 (5), 691–696.
- Barrefors, G., Petersson, G., 1995. Volatile hydrocarbons from domestic wood burning. *Chemosphere* 30 (8), 1551–1556.
- Bhattacharya, S.C., Albina, D.O., Abdul Salam, P., 2002. Emission factors of wood and charcoal-fired cookstoves. *Biomass and Bioenergy* 23 (6), 453–469.
- Delmas, R., 1994. An overview of present knowledge on methane emission from biomass burning. *Fertilizer Research* 37 (3), 181–190.

- Edye, L.A., Richards, G.N., 1991. Analysis of condensates from wood smoke. Components derived from polysaccharides and lignins. *Environmental Science and Technology* 25 (6), 1133–1137.
- Ferek, R.J., Reid, J.S., Hobbs, P.V., Blake, D.R., Liousse, C., 1998. Emission factors of hydrocarbons, halocarbons, trace gases and particles from biomass burning in Brazil. *Journal of Geophysical Research (Atmospheres)* 103 (D24), 32107–32118.
- Fine, P.M., Cass, G.R., Simoneit, B.R.T., 2001. Chemical characterization of fine particle emissions from fireplace combustion of woods grown in the Northeastern United States. *Environmental Science and Technology* 35 (13), 2665–2675.
- Fine, P.M., Cass, G.R., Simoneit, B.R.T., 2004a. Chemical characterization of fine particle emissions from the fireplace combustion of wood types grown in the Midwestern and Western United States. *Environmental Engineering Science* 21 (3), 387–409.
- Fine, P.M., Cass, G.R., Simoneit, B.R.T., 2004b. Chemical characterization of fine particle emissions from the wood stove combustion of prevalent United States tree species. *Environmental Engineering Science* 21 (6), 705–721.
- Freeman, D.J., Cattell, F.C.R., 1990. Woodburning as a source of atmospheric polycyclic aromatic hydrocarbons. *Environmental Science and Technology* 24 (10), 1581–1585.
- Goode, J.G., Yokelson, R.J., Ward, D.E., Susott, R.A., Babbitt, R.E., Davies, M.A., Hao, W.M., 2000. Measurements of excess O₃, CO₂, CO, CH₄, C₂H₄, C₂H₂, HCN, NO, NH₃, HCOOH, CH₃COOH, HCHO, and CH₃OH in 1997 Alaskan biomass burning plumes by airborne Fourier transform infrared spectroscopy (AFTIR). *Journal of Geophysical Research (Atmospheres)* 105 (D17), 22147–22166.
- Gustavsson, L., Tullin, C., Wrande, I., 2001. Small-scale biomass combustion in Sweden—research towards a sustainable society. In: *Proceedings of the First World Conference on Biomass for Energy and Industry*, Sevilla, Spain, June 2000. James & James (Science Publishers) Ltd., London, pp. 1553–1555.
- Hedman, B., 2005. Dioxin emissions from small-scale combustion of bio-fuel and household waste. Ph.D. Thesis, Umeå University, Umeå, Sweden.
- Hillring, B., Vinterbäck, J., 1998. Wood pellets in the Swedish residential market. *Forest Products Journal* 48 (5), 67–72.
- Jenkins, B.M., Jones, A.D., Turn, S.Q., Williams, R.B., 1996. Particle concentrations, gas-particle partitioning, and species intercorrelations for polycyclic aromatic hydrocarbons (PAH) emitted during biomass burning. *Atmospheric Environment* 30 (22), 3825–3835.
- Johansson, L.S., Leckner, B., Gustavsson, L., Cooper, D., Tullin, C., Potter, A., 2004. Emission characteristics of modern and old-type residential boilers fired with wood logs and wood pellets. *Atmospheric Environment* 38 (25), 4183–4195.
- Kjällstrand, J., 2002. Phenolic antioxidants in wood smoke. Ph.D. Thesis, Chalmers University of Technology, Göteborg, Sweden.
- Kjällstrand, J., Olsson, M., 2004. Chimney emissions from small-scale burning of pellets and fuelwood—examples referring to different combustion appliances. *Biomass and Bioenergy* 27 (6), 557–561.
- Kjällstrand, J., Petersson, G., 2000. Coniferyl alcohol from newsprint burning. *Nordic Pulp & Paper Research Journal* 15 (2), 98–100.
- Kjällstrand, J., Petersson, G., 2001a. Phenolic antioxidants in wood smoke. *The Science of The Total Environment* 277 (1–3), 69–75.
- Kjällstrand, J., Petersson, G., 2001b. Phenols and aromatic hydrocarbons in chimney emissions from traditional and modern residential wood burning. *Environmental Technology* 22 (4), 391–395.
- 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 (11), 2080–2091.
- McGrath, T., Sharma, R., Hajaligol, M., 2001. An experimental investigation into the formation of polycyclic aromatic hydrocarbons (PAH) from pyrolysis of biomass materials. *Fuel* 80 (12), 1787–1797.
- McKenzie, L.M., Hao, W.M., Richards, G.N., Ward, D.E., 1995. Measurement and modeling of air toxins from smoldering combustion of biomass. *Environmental Science and Technology* 29 (8), 2047–2054.
- Ndiema, C.K.W., Mpendazoe, F.M., Williams, A., 1998. Emission of pollutants from a biomass stove. *Energy Conversion and Management* 39 (13), 1357–1367.
- Nordic Ecolabelling Board, 2004. Ecolabelling of solid biofuel boilers (dominating source of heat). Criteria document, 14 December 2000–14 December 2006, version 1.4. Available at www.svanen.nu/DocEng/060e.pdf. SE-118 80 Stockholm, Sweden.
- Ogata, M., Hoshi, M., Shimotohno, K., Urano, S., Endo, T., 1997. Antioxidant activity of magnolol, honokiol, and related phenolic compounds. *Journal of the American Oil Chemists' Society* 74 (5), 557–562.
- Olsson, M., 2002. Wood pellets as low-emitting residential biofuel. Lic. Eng. Thesis, Chalmers University of Technology, Göteborg, Sweden.
- Olsson, M., 2004. Wood pellets—sustainable heating for residences. In: *Proceedings of the Annual Meeting of the World Student Council for Sustainable Development*, Chalmers, Göteborg, Sweden, March 2004. Also published in *Vision*, 2(1), 2004, available at www.wscsd.org/ejournal.
- Olsson, M., Petersson, G., 2003. Benzene emitted from glowing charcoal. *The Science of The Total Environment* 303 (3), 215–220.
- 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 (1), 51–57.
- Olsson, M., Ramnäs, O., Petersson, G., 2004. Specific volatile hydrocarbons in smoke from oxidative pyrolysis of softwood pellets. *Journal of Analytical and Applied Pyrolysis* 71 (2), 847–854.
- Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T., 2001. Measurement of emissions from air pollution sources. 3. C₁–C₂₉ organic compounds from fireplace combustion of wood. *Environmental Science and Technology* 35 (9), 1716–1728.
- Simoneit, B.R.T., 2002. Biomass burning—a review of organic tracers for smoke from incomplete combustion. *Applied Geochemistry* 17 (3), 129–162.
- Strehler, A., 2000. Technologies of wood combustion. *Ecological Engineering* 16 (S1), 25–40.

Swedish Board of Housing Building and Planning, 2002. Building regulations BBR—Mandatory provisions and general recommendations. BFS 2002:19 BBR 10. Available at www.boverket.se/publikationer/Boverketsbyggregler.htm. Box 534, SE-371 23 Karlskrona, Sweden.

Swedish Energy Agency, 2004. Annual Energy Review Sweden 2004 (In Swedish: Energiläget 2004). Box 310, SE-631 04 Eskilstuna, Sweden, available at www.stem.se.

Yokelson, R.J., Goode, J.G., Ward, D.E., Susott, R.A., Babbitt, R.E., Wade, D.D., Bertschi, I., Griffith, D.W.T., Hao, W.M., 1999. Emissions of formaldehyde, acetic acid, methanol, and other trace gases from biomass fires in North Carolina measured by airborne Fourier transform infrared spectroscopy. *Journal of Geophysical Research (Atmospheres)* 104 (D23), 30109–30125.