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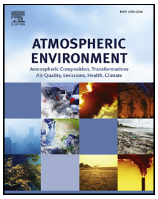

Methane emissions from small residential wood combustion appliances: Experimental emission factors and warming potential

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ABSTRACT

Methane emission factors (g/GJ) were determined testing residential heating biomass appliances (6–11 kW) under real-world operating conditions. User behavior for manually load appliances was simulated following a loading scheme starting from the cold start conditions, followed by two nominal batches and a final batch either with the nominal load of the appliance or by over loading the firebox (closing the air valves) and lasted until burn out. The results were analyzed both on batch-per-batch basis and for total combustion cycle from cold start to burn out in order to determine the critical situations causing high methane emissions. For comparison two automatic pellet appliances (8–25 kW) were also tested. Emission factors (EFs) for these automatic appliances are more than an order of magnitude lower with respect to batch-working room heaters. For the latter the average EFs ranged from 142 g/GJ to 238 g/GJ and showed both batch-to-batch and inter-appliance variability; however, many of the observed differences were not statistically significant. The results highlighted the importance of the user behavior to avoid high methane emissions. The climate relevance of methane emission levels has been assessed using global warming potential (GWP) taken from the literature, comparing CO_2equivalent emissions with that of N_2O and other near-term climate forcers (CO, NOx, VOC, black carbon) emitted by the same appliances. The results show that the warming impact of CH_4 is lower than that of BC and CO (compounds emitted in relevant levels in small appliances burning wood), but is still an important portion of the CO_2 avoided for the substitution of fossil fuels with biomass. Although the uncertainties associated with GWP are large and EFs are based on a limited number of appliances and fuel types, the results show that in the short term (i.e., 20-year period) CO_2eq for all the non-CO_2 forcers offset the CO_2 benefits of biomass use.

1. Introduction

Growing energy demand brought an increasing need for energy diversification. Several market factors such as the declining reserves and fluctuating prices of fossil fuels, national energy security, and high import costs of fossil fuels, have initially motivated the use of renewable energy sources worldwide. In recent years, the major driver for the substitution of fossil fuels with forms of bioenergy has been greenhouse gas (GHG) emission reduction. In this context, solid biomass appears an attractive alternative to fossil fuels. However, biomass combustion in residential heating appliances is known to impair local air quality given the high levels of particulate matter and volatile/semivolatile organic compound emissions (Corsini et al., 2017; Karagulian et al., 2015; Gianelle et al., 2013; Glasius et al., 2006). Nevertheless, assuming carbon neutrality, beneficial effects may be expected regarding the greenhouse gas (GHG) emissions because of net GHG emission savings substituting biomass for fossil fuels in various combustion processes (Caserini et al., 2010; Tsalidis et al., 2014), although the mitigation potential of bioenergy could be overestimated when biogenic-CO_2 flows and indirect effect as changes in the albedo are excluded (Cherubini et al., 2012; Holtsmark B., 2014; Giuntoli et al., 2016).

Methane (CH_4) is one of the GHG and it is the most abundant organic trace gas in the atmosphere. It has gained importance in the last decades for tropospheric and stratospheric chemistry, as well as a warming agent for the planet (Myhre et al., 2013). CH_4 is emitted in much less quantities with respect to carbon dioxide (CO_2) but is capable to trap more heat (per unit mass) in the atmosphere than CO_2. Besides, methane has an atmospheric lifetime of about 12 years, whereas a significant portion of CO_2 emitted remains in the atmosphere–climate system for many centuries (Solomon et al., 2009). This makes the CH_4 contribution to climate change much more important in the short run, whereas CO_2 is more important for the long "tail" of global warming in the long run. This implies that a reduction in methane emissions could be beneficial in the mitigation of climate change over relatively short timescales of a few decades or less.

Atmospheric CH_4 emissions are usually assessed by emission

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inventories; although uncertainties in the estimates of individual source strengths are large, it is generally thought that biomass burning is a relevant source, together with wetlands, livestock, rice cultivation, fossil fuel production (Yusuf et al., 2012; Karakurt et al., 2012). In the light of the fact that currently half of the solid biomass used in the EU is employed for household heating (EUBIA, 2014), methane emissions from these sources become potentially important. Nonetheless, there are few research studies on the emission factors (EFs) of methane from residential wood combustion sources as a basis for emission inventories (Paulrud et al., 2005; Robinson, 2011; Polglase et al., 2012). The Intergovernmental Panel on Climate Change (IPCC) Guidelines for National Greenhouse Gas Inventories (IPCC, 2006) and the related IPCC Emission Factor Data Base, provides only few data on CH_4 emission factor from wood combustion in stoves, mainly coming from US-EPA Clearinghouse for Inventories and Emissions Factors (US-EPA, 2005) and from biofuel stoves in rural areas (Zhang et al., 2000; Bhattacharya et al., 2002; Wang et al., 2009)

The present paper endeavors to give its contribution to fill in this gap of knowledge providing EFs from a wide selection of heating appliances and wood types. It reports methane EFs from six small scale (< 35 kW) residential wood combustion appliances and seven types of woody biomass fuel (five firewood types and two different qualities of commercial wood pellets). The study also discusses the operational factors that govern the methane formation in manually fed room heaters. Batch-to-batch results are also investigated to define the incidence of progressive conditions in the combustion chamber on the total emissions in order to determine where to act mostly to reduce emissions.

The paper aims also a first quantification of the relative warming impact of CH_4 emission measured, in the comparison with the climate effect of nitrous dioxide (N_2O), the third most important long-lived greenhouse gas, and the so called “near term climate forcers” (NTCF). These atmospheric pollutants such as black carbon (BC, called also elemental carbon EC) and organic carbon (OC), carbon monoxide (CO), nitrogen oxides (NOx) and non-methane volatile organic compounds (NMVOC), have been usually considered in the past only for their contribution to the degradation of air quality and subsequent impacts on health, but have been recently also studied for their property to interfere in the near term, via direct and indirect mechanisms, with the Earth's energy balance. The last IPCC reports (Myhre et al., 2013) provided for these pollutants a GWP basing on the work of Shindell et al. (2009), Fuglestvedt et al. (2010), Fry et al. (2012) and Collins et al. (2013), that allow to translate their warming potential in terms of CO_2-equivalent (CO_2eq) emissions.

The paper finally assesses whether the CO_2eq emissions associated with CH_4 and the other non-CO_2 climate forcers are comparable to the savings of GHGs due to the production of heat without fossil fuels in the same appliances; these CO_2eq emissions emitted by residential small appliances burning wood and pellet are then put in the context of the overall GHG emissions of Lombardy region, a highly industrialized region in northern Italy where the use of woody biomasses is widespread and documented (Pastorello et al., 2011).

2. Material and methods

2.1. Combustion tests and sampling

The experimental design and configuration are summarized in the following paragraphs. Further details are available in Ozgen et al. (2014), which reported CO, NOx, NMVOC and PM emission factors obtained during the same experimental campaign.

2.1.1. Tested residential heating appliances and fuels

The tested appliances were chosen among commercially available residential heating appliances commonly used in Italy. Table 1 provides a list of the technical details of the appliances (further information in Supplementary Material – SM1). Briefly, both closed fireplace and the advanced stove were equipped with manual primary and secondary (i.e., window flush) air regulation with dampers, whereas the open fireplace and the traditional stove had only manual primary air regulation damper. The open and the closed fireplaces were characterized by a low and wide combustion chamber with quite large combustion chambers that enabled mass of fuel/unit firebox volume ($M_{fuel}/V_{chamber}$) ratios of about 0.01–0.03 kg dm^{-3}. The traditional and advanced stoves on the other hand were distinguished by a slim and high configuration with small combustion chambers the former with particularly small combustion chambers ($M_{fuel}/V_{chamber}$ = 0.05–0.16 kg dm^{-3}, the higher value for the traditional stove).

Table 1
Main characteristics of the tested appliances.

Appliance	Rated heat output (kW)	Rated efficiency (%)	Air regulation
Open fireplace (OFP)	8	51	manual, 1^{ry}[a]
Closed fireplace (CFP)	11	82	manual, $1^{ry}+2^{ry}$[b]
Traditional stove (TS)	6	70	manual, 1^{ry}
Advanced stove (AS)	8	75	manual, $1^{ry}+2^{ry}$
Pellets stove (PS)	8	91	automatic, $1^{ry}+2^{ry}$
Pellets boiler (PB)	25	93	automatic, λ probe

[a] 1^{ry}: primary air.
[b] 2^{ry}: secondary (window purge) air.

Test fuels for batch-wise appliances consisted of five different types of locally available firewood (i.e., beech, false acacia, hornbeam, oak, and spruce). All the wood logs were fairly dry (fuel moisture $< 10\%_w$). The wood types differed mainly by fuel ash (higher for oak and false acacia), chlorine (higher for hornbeam and false acacia) and sulfur (higher for hornbeam, oak and false acacia) content. The dimensions of wood logs followed the appliance manufacturer instructions. The logs contained both bark and stem. Each batch was weighed before the test runs. The tests on automatic pellet appliances were run with DIN-Plus certified and non-certified pellets, differing mainly in fuel ash content, which is higher in the non-certified fuel. Table TSM1 in Supplementary Material reports the detailed fuel composition.

2.1.2. Experimental configuration

Flue gas was extracted from the dilution tunnel (Fig. 1, point 7) through a heated probe (160 °C) and methane was measured by a flame ionization detector (CAI 600 M-HFID) equipped with a catalytic cutter for non-methane hydrocarbons. The dilution tunnel with hood was constructed with the indications in the European Technical

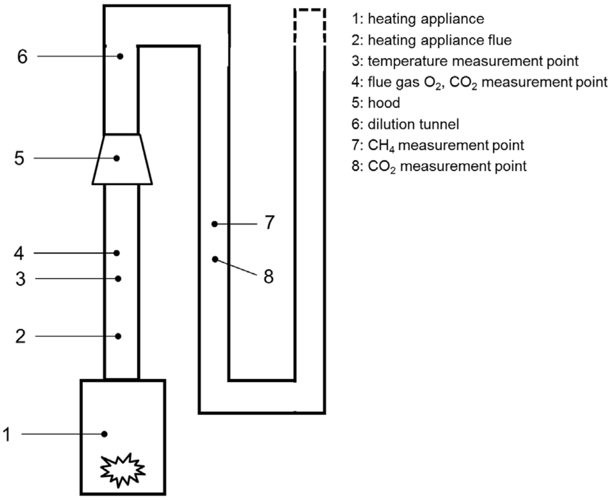

Fig. 1. Scheme of the experimental set-up.