

Emission Characteristic Identification and
CH₄ Emission Factor Development for Wood-fired Boiler

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Abstract

Wood-fired boilers, burning firewood as their main fuel, are used for residential heating in winter. As of 2013, wood-fired boilers were operated in approximately 45,447 households in Korea. However, there have been few investigations of the non-CO₂ emission factors of wood-fired boilers.

This study identified emission characteristics and developed the CH₄ emission factors for residential wood-fired boilers. Four surveys were conducted to collect exhaust gas from wood-fired boiler stacks. The CH₄, O₂, CO, and CO₂ of exhaust gases were analyzed in the laboratory, as were the fuels used in the wood-fired boilers.

As a result of identifying the CH₄ emission characteristics, CH₄ concentration appears to be low with the fan turned on because of the amount of combustion air being added to the furnace. Spearman's rho correlation analyses were performed to investigate the correlations between CH₄ concentrations and O₂, CO, and CO₂ among exhaust gases and temperatures in the furnace. In the result of spearman's rho correlation analysis, it can be seen that the higher the concentration of CO in the exhaust gases, the higher the concentration of CH₄. However, in case of between the CH₄ concentration and in-furnace temperatures it can be seen that the higher temperature in the furnace, the lower the concentration of CH₄.

CH₄ emission factors were estimated based on whether the fan was in operation. CH₄ emission factors were found to be 171.98 kg CH₄/TJ with the fan turned off, and 88.31 kg CH₄/TJ with the fan turned on. The difference between the CH₄ concentrations and emission factors is linked to the average furnace temperature and

the amount of combustion air added. The emission factors developed in this study were different than the IPCC default values. Comparison between wood stove and wood fired-boiler with fan turned off show that CH₄ emission factor developed in this study was slightly lower than wood stove. The difference between the CH₄ emission factors of this study and those of the IPCC are likely due to the specific combustion technology and the total moisture content of the fuel used.

Keywords: Greenhouse gas, Wood-fired boiler, CH₄ emission factor

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Chapter 1. Introduction

1.1 Background and purpose

The Paris Agreement was adopted at the 21st Conference of the Parties in December 2015, and took effect on November 4, 2016. As the Paris agreement goes into effect, developing countries as well as advanced countries must commit to reducing greenhouse gas emissions.

Korea plans to reduce its greenhouse gas emissions 37% from the business-as-usual (BAU) level by 2030. This is a very challenging goal for Korea because its greenhouse gas emissions have continued to increase, except IMF period in 1998.

In 2014, Korea's total greenhouse gas emissions were recorded as 690.6 MtCO₂eq of which 567.8 MtCO₂eq were emitted from fuel combustion (National Greenhouse Gas Inventory Report of Korea, 2016). Among greenhouse gases, CH₄ and N₂O account for 3.9% and 2.2%, respectively, which is much less than the amount of CO₂ which comprises 91.1% of Korea's greenhouse gas emissions. However, the global warming potential (GWP) of CH₄ is 21 times that of CO₂ and that of N₂O is 310 times higher (The IPCC Second Assessment Report; Climate Change 1995, 1995). The major agencies such as the Intergovernmental Panel on Climate Change (IPCC, 2006), World Resources Institute and the World Business Council for Sustainable Development (WRI/WBCSD, 2005) have set emissions and emission factors for not only CO₂ but also non-CO₂ as important indicators.

CO₂ emission factors depend on the carbon content of fuels while the non-CO₂ emission factors depend on technologies and conditions of combustion; these factors are not well known (IPCC, 2006).

Firewood is designated as a “carbon neutral” energy source. It is considered a key source to reduce greenhouse gas emissions. The Korea Forest Service and local governments have therefore provided firewood to rural households that installed wood-fired boilers.

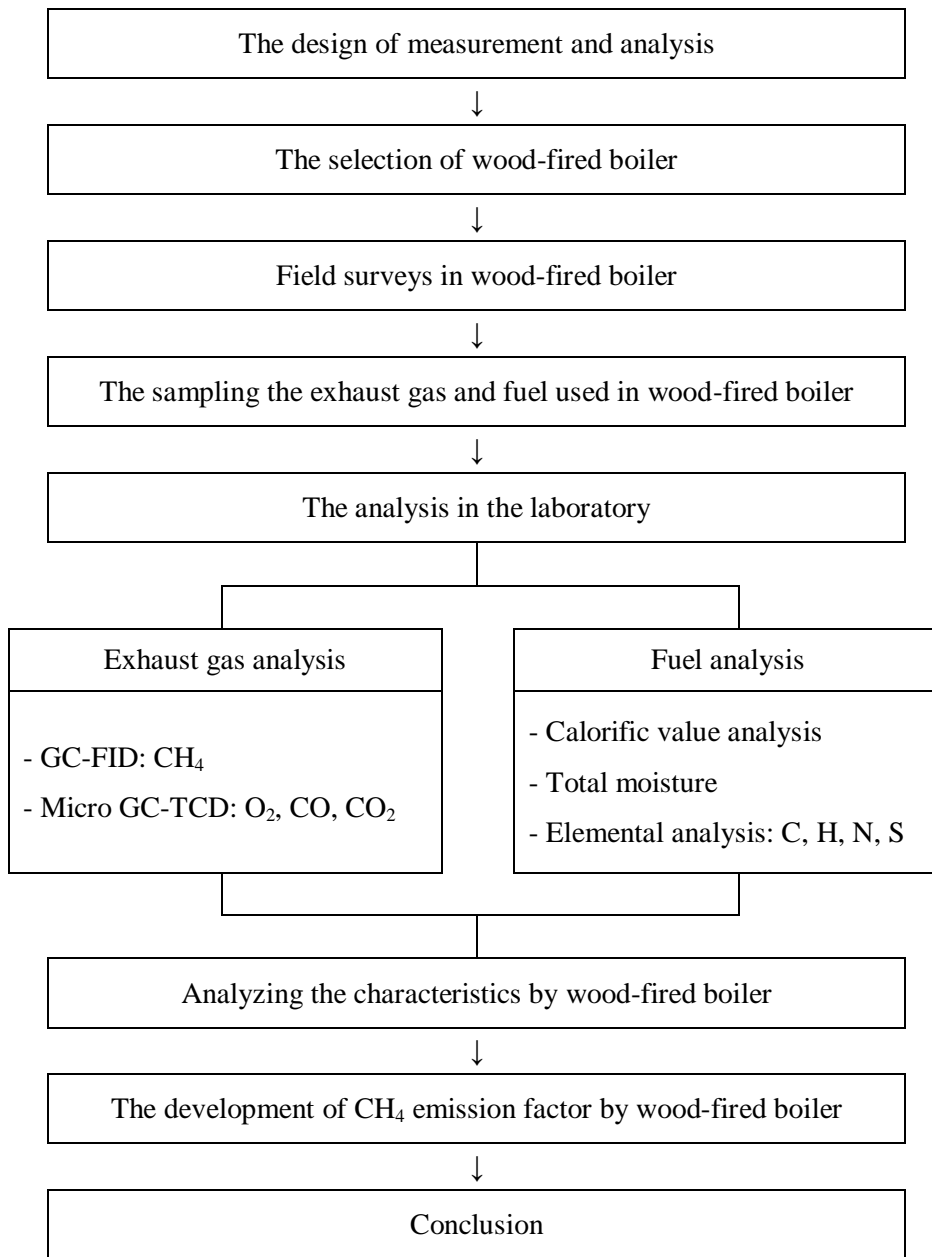
Wood-fired boilers, unlike regular combustion facilities, have incomplete combustion conditions due to intermittent fuel supplies and non-uniformity of fuel.

For these reasons, it is expected that their emission characteristics will be different than other boilers. Non-CO₂, emitted during fuel combustion, is affected by combustion conditions. Also, non-CO₂ emitted by burning firewood is not designated as carbon neutral like CO₂. It is necessary to determine the non-CO₂ emission factor for firewood. The purpose of this study is to identify the emission characteristic and develop the CH₄ emission factor for residential wood-fired boiler.

1.2 Procedure and contents

Wood-fired boilers, burning firewood as their main fuel, are used for residential heating in winter. This study will be developed the CH₄ emission factor and identified characteristics of residential wood-fired boilers. As shown in <Figure 1>, the procedures used are described in more detail.

1. To develop the CH₄ emission factors and identify the emission characteristics of wood-fired boilers, four field surveys of residential wood-fired boilers will be conducted. The surveys will be collected the exhaust gas from the stack over a period of four days.
2. The fuel used in the wood-fired boilers and CH₄ concentration analysis will be later analyzed in the laboratory. The fuels will be analyzed for their calorific value, elements, and total moisture; exhaust gases will be analyzed through gas chromatography.
3. The emission factor of CH₄ will be estimated using the value obtained in the previous steps.
4. The emission factors developed in this study will be compared to the IPCC default values for wood stoves.



<Figure 1> Flow diagram of this study

Chapter 2. Theoretical Background

2.1 Woody biomass

Biomass is fuel developed from organic materials, a sustainable and renewable source of energy used to create electricity or other forms of power (Korea Environment Corporation, 2014).

Woody biomass is comprised of carbohydrates and lignin produced through the photosynthetic process. While this mainly means woody and herbaceous plants, it also includes products derived from them or their waste, such as timber, wood wastes, and paper (Gyeonggi Research Institute, 2009).

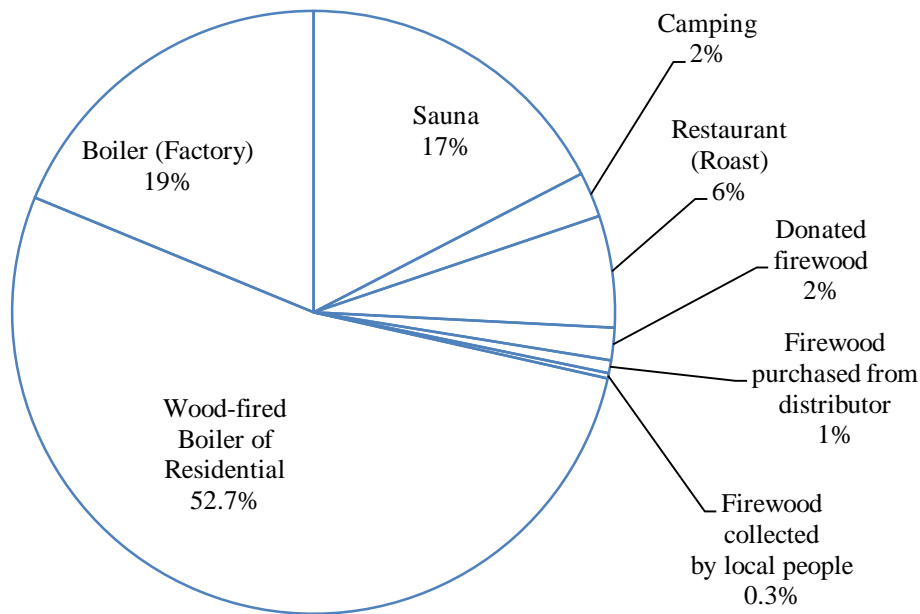
Wood-based biomass accounts for over 90% of total biomass and is one of the most important energy sources in the world (Lee et al., 2013). Woody biomass emits fewer greenhouse gases than fossil fuels. The amount of CO₂ emissions in burning wood is very small, about 1/10 of natural gas and 1/12 of heating oil. The annual amount of woody biomass produced on the Earth is about 170 billion tons, but the amount used by humans is about 6 billion tons, which is about 3.5% of the total. If we develop a technology that can convert unused biomass into available energy, we will have another resource (Ko et al., 2011).

Woody biomass is roughly classified into scrap wood and lumber. Waste wood can be classified into forest waste and waste wood. This study, focused on firewood from lumber.

2.2 Status of firewood consumption

Attempts to reduce greenhouse gas emissions and rising oil prices have changed from fossil oil to renewable biofuels in recent years. (Olli Sippula et al., 2009).

Domestic wood-fired boilers are used in rural and forest houses. As of 2013, wood-fired boilers were operated in approximately 45,447 households in Korea (National Institute of Environmental Research, 2016); they consumed an estimated 109,294 tons of firewood. As shown in <Figure 2>, 57,589 tons of firewood were distributed to wood-fired boilers, accounting for 52.7% of total firewood.



<Figure 2> Firewood sales by sector

* Source: Korea Forest Service, The survey of wood utilization, 2013

2.3 Firewood and residential wood-fired boilers

Wood-fired boilers use firewood as their primary fuel; they are used for residential heating in the winter. CO₂ emissions from firewood are 8.3% of those from heating oil and 10% of those from natural gas (Gyeonggi Research Institute, 2009). However, because firewood is biomass, CO₂ emissions from its combustion are “carbon neutral” and are not included in statistics of greenhouse gas emissions (IPCC, 2006). Wood can be used directly as firewood or turned into easy-to-use type such as pellet or wood chip. As shown in <Figure 3>, wood fuels include firewood, pellets, and wood chips (Mirae Energy-code Research Institute, 2014).



<Figure 3> Wood used as energy source

* Source: Mirae Energy-code Research Institute, Safety management system of wood-fired boiler, 2014

The wood-fired boilers are widely used in rural areas, where the goal is to replace fossil fuels such as oil and gas in boilers with firewood. A wood-fired boiler is composed of three parts; input the firewood, a combustion chamber; and a heat exchanger. The wood-fired boilers have advantages; firewood can be easily collected from surroundings and wood-fired boilers have price competitiveness. Also the

installation place is less restricted than pellet boilers in which have pellet storage.

However, wood-fired boilers are less convenient to use and are relatively more vulnerable to fire than other boilers; the user must directly put the fuel into the combustion chamber, unlike oil or gas boilers in which the fuel is automatically supplied through the house (Mirae Energy-code Research Institute, 2014). As shown in <Figure 4>, the wood-fired boilers are four types; general, regenerative, multipurpose, and hybrid type.



General type



Regenerative type



Multipurpose type



Hybrid type

<Figure 4> Type of wood-fired boilers

* Source: Southern Fire Station in Busan, 2014

2.4 Combustion characteristics of firewood

The combustion of wood is an oxidation process; the wood reacts chemically with the oxygen in air. The combustion can be divided into after flame and after glow. The after flame refers to the burning of combustibles that produces flames. And the after glow refers to the phenomenon of continuous burning of carbides remaining after the extinction of flames.

At the beginning of the combustion of wood, the temperature rises and volatile components are generated as the pyrolysis progresses. As wood is heated from room temperature, moisture in the wood evaporates. When the temperature has reached 100 °C, the wood is completely dry as all the moisture in the wood has fully evaporated. When the temperature has reached 150 °C, the color of the surface of wood changes to blackish brown, and when the temperature has reached 200 °C, pyrolysis occurs and flammable gases such as CO, CH₄, C₂H₄, H₂, aldehyde, ketone, and organic acid are generated.

When the temperature has reached 250 to 290 °C, pyrolysis products are increased. And then the flames of the fire are formed. When the temperature has reached 350 to 450 °C, natural ignition occurs.

The combustion of wood is affected by its thermal conductivity, specific gravity, components, water content, and cross-sectional area, as well as air and heating conditions. Heat is generated when wood is burned. The amount of heat generated, when 1 g or 1 kg of wood is completely burned, is calorific value. The maximum calorific value of completely dry wood is about 4,500 cal/g (4,500 kcal/kg) on average. As can be seen in the <Table 1>, pine has the highest calorific value, followed by birch, Douglas-fir, beech, and spruce. Poplar has the lowest calorific value, followed

by maple and alder (Korea Institute of Fire Industry & Technology, 2010).

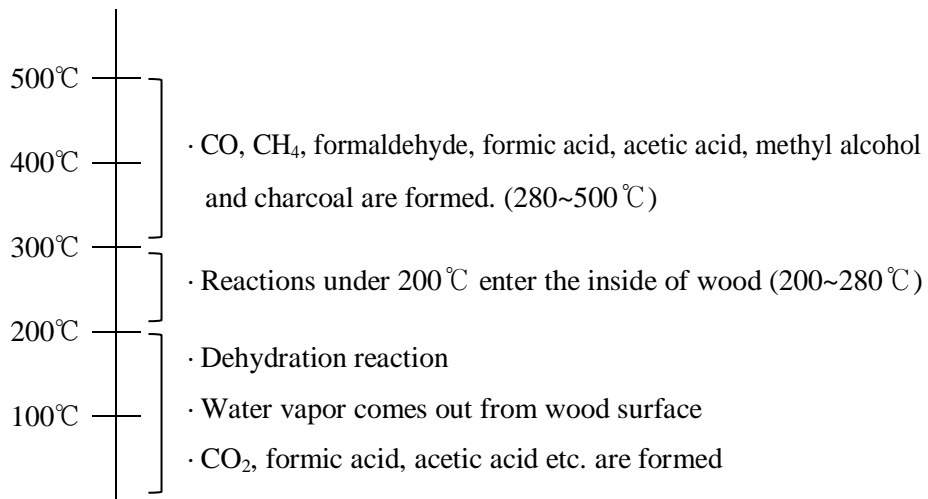
<Table 1> Gross calorific value by wood species

Species	Gross calorific value(kcal/kg)
Douglas-fir	4,580 ~ 5,030
Fir	4,440 ~ 4,650
Pine	4,780 ~ 6,790
Spruce	4,500 ~ 4,700
Alder	4,300 ~ 4,440
Beech	4,500 ~ 4,870
Birch	4,650 ~ 5,190
Maple	4,180 ~ 4,670
Oak	1,390 ~ 5,280
Poplar	4,120 ~ 5,350

* Source: Korea institute of fire industry & technology, 2010

The flaming combustion of wood is possible only when there is an ignition source and a combustible layer of mixed gases has formed around the wood. The pyrolysis of wood produces carbon, tar, and gases; the tar decomposes to release flammable gases, which generate flames.

The combustible layer of mixed gases should produce pyrolysis products faster than the atmospheric diffusion rate; for the consumption to continue, the pyrolysis products should be generated at rates higher than the consumption rate. (Korea Institute of Fire Industry & Technology, 2010).



<Figure 5> Combustion process of wood depending on combustion temperature

* Source: Korea Institute of Fire Industry & Technology, 2010, re-edit

2.5 Literature review

The emissions from combustion of biomass were not included in national totals and the sectoral to avoid double counting. However, the emissions of CH₄ and N₂O are included and estimated in the national totals because their effect is in addition to the stock changes estimated in the AFOLU sector (IPCC, 2006).

Emissions from natural sources account for about 30 percent of total CH₄ emissions, with wetlands the largest natural emission source. Artificial emission sources include the energy industry, ruminants, paddy fields, and landfills; among these, the energy industry and ruminants account for high proportions.

Whereas the causes of CH₄ generation as in paddy fields and landfills have been studied extensively (Kim, 2013; Kim, 2011; Andersen et al, 2010; Jing et al, 2009; Kim et al, 2009), those in the energy industry have not been sufficiently studied.

In the energy industry, CH₄ is known to be a minor component produced by incomplete combustion (Ryu et al, 2011; Korea Energy Agency, 2008; Korhonen, 2001). Previous studies on the development of CH₄ emission factors of wood-fired boilers were reviewed. However, most studies regarding CH₄ emission factors and emission characteristics of wood-pellet boilers have been conducted in Europe, and such studies were mainly conducted on air pollutants. This tendency applies in South Korea as well, where only studies regarding the emission characteristics of air pollutants and black carbon were conducted (Lee et al., 2012; Kim et al. 2014; Park et al., 2015), and studies on CH₄ are insufficient.

As shown in <Table 2>, the emission factors compared with the other studies of CH₄ emission factor. The emission factors indicated that were different and high concentrations.

<Table 2> Emission factors of the other studies.

Source	Technology	Configuration	CH ₄ emission factor (kgCH ₄ /TJ)
IPCC ¹⁾	Wood pits		200
	Wood stove in US	Conventional	932
	Wood stove in Asian countries		258 - 2190
	Wood fireplaces		275 - 386
	Agriculture wastes stoves		230 - 4190
Cho et al ²⁾	Biomass fired fluidized bed combustion.	Fuel used of RDF, RPF	1.4
Jung ³⁾	Wood chip fired fluidized bed combustion.	Fuel used of wood chip	0.22
Kim ⁴⁾	Coal briquette stove	Open the air inlet	11.28 ± 0.70
		Close the air inlet	18.14 ± 1.67
Song et al ⁵⁾	Coal briquette boiler		11.77 ± 1.72

The CH₄ emission factors have been separated for different technology types. According to Cho et al (2012) and Jung (2017), these emission factors were developed from fired fluidized bed combustion, however, these are different respectively. It seems to be due to the difference in the characteristics of fuel used (Jung, 2017).

¹⁾ IPCC, 2006, Vol.2; Energy, Chapter 2; Stationary combustion.

²⁾ Changsang Cho, Jaehwan Sa, Kikyo Lim, Taemi Youk, Seungjin Kim, Seulki Lee and Euichan Jeon, 2012, Development of methane and nitrous oxide emission factors for the biomass fired circulating fluidized bed combustion power plant

³⁾ Jaehun Jung, 2017, Development of non-CO₂ emission factor of wood chip fired fluidized bed combustion

⁴⁾ Seungjin Kim, 2013, Development of non-CO₂ emission factor of coal briquette

⁵⁾ Garam Song, 2017, Development of non-CO₂ emission factor of the coal briquette boiler

The CH₄ emission factors of wood stove in Asian countries were in the ranges from 258 to 2190 kgCH₄/TJ. These emission factors were estimated of cook-stoves collected from different Asian countries using wood and charcoal as fuel. It is interpreted that the differences were expected to be due to the technology-specific of CH₄ emission and total moisture content of fuel used.

According to Saryang Kim and Jongsuk Lee (2002), they analyzed composition of exhaust gas from small wood boiler. The CO concentration was obtained more than 3000 ppm (Kim et al, 2002). In general, the CO was formed in incomplete combustion conditions, also CH₄ concentration was too (Kim, 2013; Korea Energy Agency, 2008).

In this study, CH₄ emission factor was developed and the emission characteristics were identified of residential wood-fired boiler.

Chapter 3. Methodology

3.1 Design of measurement and analysis

In this study, CH₄ emission factor was developed and emission characteristics identified for wood-fired boiler. To achieve the aforementioned study objectives, this study conducted four field studies in a household for four days.

The thermal output of wood-fired boiler is 26,050 kcal/h. As shown in <Figure 6>, firewood and exhaust gas were collected during the field study.



The firewood in the storage



The firewood used in wood-fired boiler



The wood-fired boiler



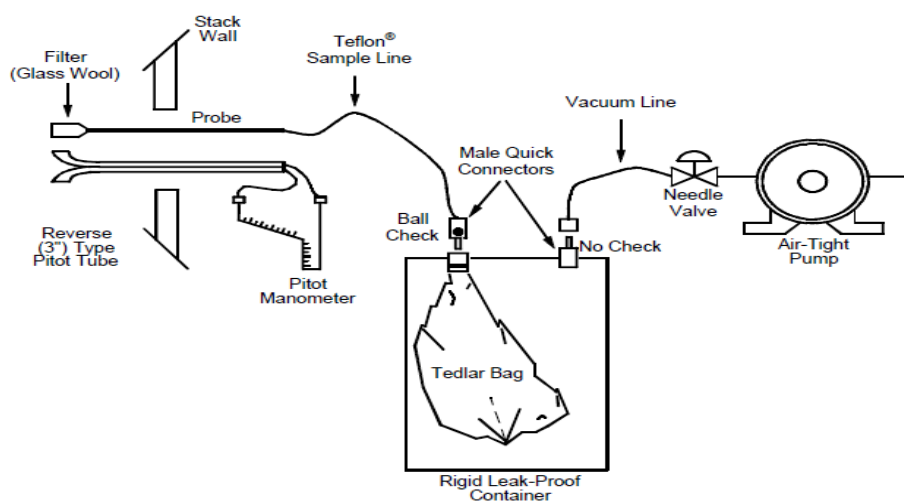
The sampling from the stack

<Figure 6> Wood-fired boiler in the field survey

3.2 Sampling method of the exhaust gas

As shown in <Figure 7>, the exhaust gas from the wood-fired boiler was collected using EPA method 18 (US EPA 2001). This is the most commonly used method for collecting samples of greenhouse gas from combustion facilities.

A 10L tedlar bag (SKC, US) was connected to a lung sampler. The samples were collected by lung sampler, which creates a vacuum inside that uses negative pressure as a pump.



<Figure 7> Schematic diagram of intermittent sampling

The sampling tube and sampling line were stainless steel because the exhaust gases from combustion facilities are large flow and high temperature. The exhaust gases were passed through pretreatment facility in order to remove the moisture (Lee et al., 2009; Jeon et al., 2007). The temperature of exhaust gases was measured using the electronic thermometer of K-type (RS-232 Thermolog, Taiwan).

As shown in <Figure 8>, field surveys were conducted to collect the exhaust gas from the stack. And then, gases collected were analyzed in the Greenhouse Gas Laboratory of Sejong university.

The sampling from the wood-fired boiler was carried out for 60 ~ 70 minutes, and the samples were taken every 5 minutes. Also, the temperature in the furnace was measured using the electronic thermometer of K-type (RS-232 Thermolog, Taiwan) whenever samples were taken.



<Figure 8> Sampling of the exhaust gas

3.3 Analyzing method for the exhaust gas

3.3.1 CH₄ concentration analyzing method

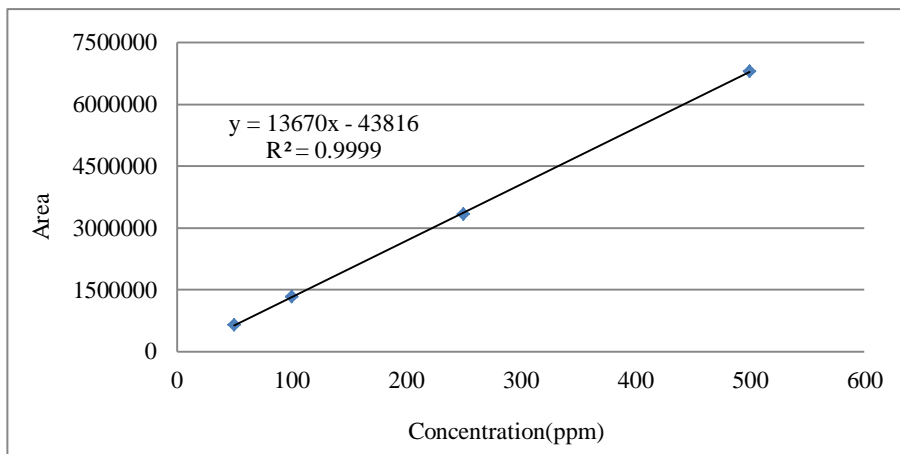
CH₄ concentrations were analyzed by GC-FID (Varian cp-3800). The CH₄ analysis used 3.175 mm outer diameter mesh column with packed Q 80/100 and 1 m stainless steel columns.

The calibration curve was derived from the average value of three repeated analyses using standard gas. The concentration of the sample would be within the calibration curve. The analysis condition for GC is shown in <Table 3>.

<Table 3> Analysis condition of GC for CH₄

Classification		Analysis condition
Column		Porapak Q 80/100 Mesh
Carrier gas		N ₂ (99.999%)
Flow	N ₂	25 mL/min
	H ₂	30 mL/min
	Air	300 mL/min
Temperature	Oven	70 °C
	Injector	120 °C
	Detector	250 °C

To draw the calibration curves, concentrations of CH₄ were set as 50, 100, 250 and 500 ppm, in standard conditions. When a high concentration of sample was analyzed, it was diluted. As shown in <Figure 9>, the result indicates excellent linearity.



<Figure 9> Calibration curve for CH₄

3.3.2 CO, O₂, and CO₂ concentration analyzing method

The exhaust gases were sent to an on-line micro-GC (Inficon 3000A, Switzerland) for quantification and identification of CO, O₂, and CO₂. The micro-GC was equipped with a capillary column which is more detailed in <Table 4>.

<Table 4> Analysis condition of micro-GC

Classification	Analysis condition	
	Channel A	Channel B
Channel A, column	Molsieve, 10 m × 0.32 mm × 30 μm	
Channel B, column	PLOTU, 8 m × 0.32 mm × 30 μm	
Oven and GC setting	Channel A	Channel B
Sample inlet	100 °C	100 °C
Injector	100 °C	80 °C
Column	80 °C, 25 psi	70 °C, 25 psi
Run time	3 min	3 min

* Source: Inficon 3000A, Switzerland, re-edit

3.4 Analyzing method of fuel

3.4.1 Calorific value analyzing method

In this study, the calorific value was analyzed using a calorimeter (IKA-C2000, Germany) in the laboratory. The quantification value of standard sample (Benzoic acid C 723, IKA) was measured using an electronic scale (Mettler Toledo-AB204S, Switzerland) with 0.0001 g sensitivity, as shown in <Figure 10>.

The temperature of the cooling water was set at 25 °C using a water temperature controller (IKA-KV600, Germany) to analyze calorific value. The pure water was used as cooling water.



<Figure 10> Calorimeter and Electronic scale

3.4.2 Element analyzing method

The samples were analyzed using an automatic element analyzer (Thermo Finnigan-Flash EA 1112, USA) for carbon and hydrogen, as shown in <Figure 11>. For this analysis, a 2-m column (Para QX) was used. The flow rate of carrier gas, oxygen, and reference gas were set each at 140, 240, and 100 mL/min respectively. The temperatures of the furnace and TCD oven were set at 900 °C and 70 °C respectively.



<Figure 11> Automatic Elemental Analyzer

3.5 Quality assurance and quality control

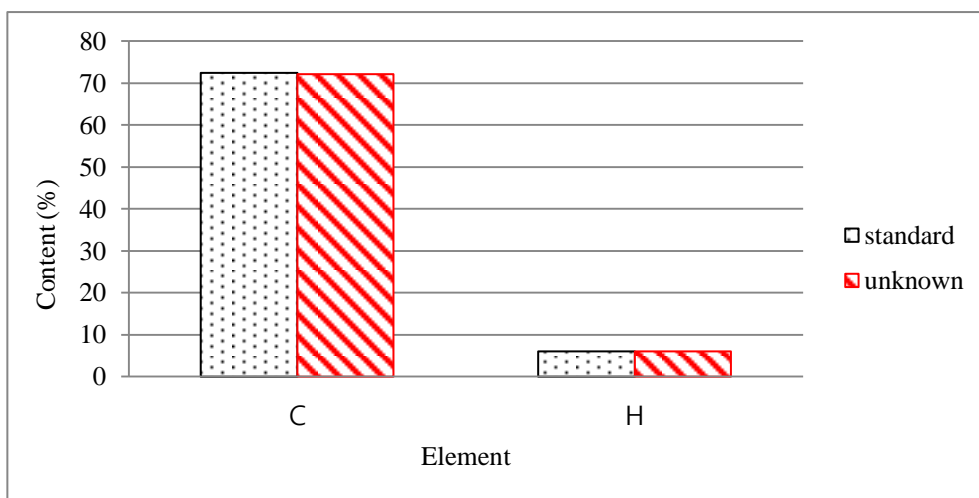
The repeatability test of calorific value was conducted with benzoic acid (IKA, caloric, 6323 ± 4.57 cal/g). The gross calorific value of benzoic acid was repeatedly analyzed five times. The relative standard deviation (RSD), shown in <Table 5>, was 0.13%. This result indicates excellent repeatability.

<Table 5> Repeatability test of calorific value analysis using standard sample

Sample	Mass of standard (g)	Gross calorific value (cal/g)
1	0.4884	6544
2	0.5012	6598
3	0.4792	6535
4	0.4823	6555
5	0.4983	6531
Mean		6541
SD		8.36
RSD (%)		0.13

3.5.1 Repeatability test of elemental analysis for carbon and hydrogen

The repeatability test for element analysis was implemented using BBOT (2,3-bisthiophene). The repeatability test of elemental analysis was evaluated by analyzing between standard and unknown. The analyzing standard was first step; BBOT was instilled into the element analyzer with the entering element contents and the information of element contents was analyzed. And the analyzing unknown was second step; BBOT used at first step was analyzed without entering element contents. These procedures were done repeatedly. The difference of absolute value was 0.32% carbon and 0.02% for hydrogen respectively. This result, shown in <Figure 12>, indicated excellent repetition.



<Figure 12> Repetition test of elemental analysis for carbon and hydrogen

3.5.2 Repeatability test for CH₄ concentration

Standard gas with a concentration of CH₄ was analyzed 10 times to confirm repeatability. The results are shown in <Table 6>. For CH₄, the average value was evaluated as 250.10 ppm, the standard deviation as 0.32 ppm, and the relative standard deviation as 0.13%.

<Table 6> Results of reproducibility test using standard gas

Number of analysis	CH ₄ concentration(ppm)
1	249.99
2	250.34
3	250.27
4	250.27
5	250.36
6	249.23
7	250.16
8	250.02
9	250.38
10	250.02
Mean	250.10
SD	0.32
RSD (%)	0.13

3.5.3 Repeatability test for CO, CO₂, and O₂ concentration

In order to evaluate the relative standard deviation (RSD), the standard deviation of CO, CO₂ and O₂ was measured three times. The result of the repeatability test is shown in <Table 7>.

<Table 7> Results of reproducibility test for CO, CO₂ and O₂

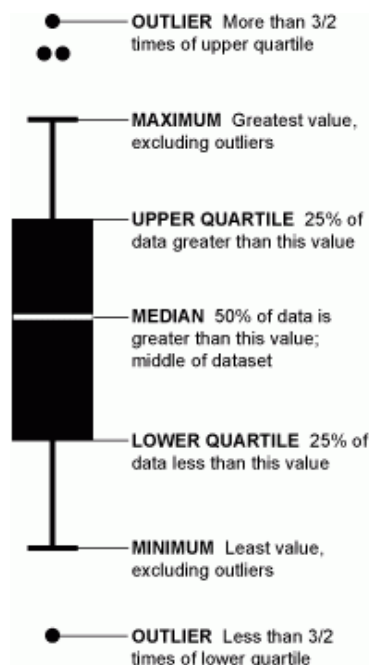
Number of analysis	CO(%)	CO ₂ (%)	O ₂ (%)
1	0.05	0.10	20.99
2	0.05	0.12	20.98
3	0.07	0.14	21.03
4	0.05	0.14	21.01
5	0.04	0.12	21.00
6	0.04	0.10	21.09
7	0.06	0.08	21.05
Mean	0.05	0.11	21.02
SD	0.01	0.02	0.04

3.5.4 Method of Interquartile Range (IQR)

The interquartile range (IQR) is a measure of where the “middle fifty” is in a data set. As shown in equation (1), the IQR is the first quartile subtracted from third quartile.

$$IQR = Q_3 - Q_1 \quad (1)$$

IQR is a way to find the abnormal values through a box plot diagram, which is also called a whisker plot. The box plot diagram typically uses a value such as the measure of dispersion and median. The purpose of the diagram is to identify outliers and discard them from the data series before making any further calculations. The search for the outliers using the IQR is shown in <Figure 13>.



<Figure 13> Box and whisker plot

* Source: Student learning centre, 2013, Box & Whisker plots, <http://www.flinders.edu.au/SLC>

3.6 Method of development of CH₄ emission factor

In this study, the CH₄ emission factor was developed using measured CH₄ concentrations, calculated combustion exhaust emissions, and theoretical air. In equation (2), EF is the emission factor of CH₄ (kg/TJ), C_{CH_4} (ppm) is the CH₄ concentration, G_{od} (Sm³/kg) is the theoretical dry exhaust emissions of the combusted fuel, and A_0 (Sm³/kg) is the theoretical air of the combusted fuel.

As in equation (3), the O₂ in the exhaust gas was used for m , the excess air ratio. MW is the molecular weight of CH₄, which is 16 g/mol, V_m is the volume of 1 mol of the ideal gas in the standard state, which is $22.4 \times 10^{-3} \text{ m}^3/\text{mol}$.

$$EF_{CH_4} = C_{CH_4} \times \{ G_{od} + (m - 1) \times A_0 \} \times (MW/V_m)/NCV \quad (2)$$

$$m = 21/(21 - C_{O_2}) \quad (3)$$

In order to develop the CH₄ emission factor of wood-fired boilers, we need the Net Caloric Value (NCV), measured in MJ/fuel. The calculation of NCV is shown in equation (4).

$$NCV = GCV - [6 \times (\text{Moisture} (\%) + 9 \times \text{Hydrogen} (\%))] \quad (4)$$

The emission factor was calculated using elemental analysis of fuel, the calorific value analysis result, and the CH₄ concentration measurement results from the wood-fired boilers. As shown in <Table 8>, it shows the worksheet used to calculate the CH₄ emission factor.

In the first step, the hydrogen and carbon content were analyzed using the element analyzer and the total moisture content was analyzed. Using the hydrogen content and the total moisture content, the gross calorific value of firewood was converted into net calorific value. In the second step, CH₄ concentration was analyzed using GC-FID (Varian cp-3800), and the exhaust gas was analyzed using micro-GC. In the third step, the theoretical oxygen demand, the theoretical air, and the excess air ratio were calculated by using the oxygen concentration of the exhaust gas. In the fourth step, the theoretical dry exhaust gas amount was calculated using the parameters of the third step. Finally, in the fifth step, the emission factor of CH₄ was developed using the value obtained in the previous four steps.

<Table 8> Work-sheet for developing the CH₄ emission factors

Step 1 (Fuel data)						
Item	Elementary analysis					Net calorific value
	Carbon	Hydrogen	Oxygen	Nitrogen	Surfer	
Sub-Item	A	B	C	D	E	F
Unit	%(wt)	%(wt)	%(wt)	%(wt)	%(wt)	MJ/kg
Step 2 (Exhaust data)						
Item	Concentration(CH ₄)			Exhaust oxygen concentration		
Sub-Item	G			H		
Unit	ppm			%(vol)		
Step 3 (Parameter)						
Item	Theoretical oxygen(O ₀)	Theoretical air(A ₀)	Excess air ratio(m)			
Sub-Item	I	J	K			
Unit	m ³ /kg	m ³ /kg	-			
Calculation	$1.867 \times A + 5.6 \times (B - C/8) + 0.7 \times E$	$I/0.21$	$21/(21 - H)$			
Step 4 (Amount of theoretical dried combustion gas)						
Item	Amount of theoretical dried combustion gas(G _{0d})					
Sub-Item	L					
Unit	m ³ /kg					
Calculation	$(1 - 0.21) \times J + 1.867 \times A + 0.7 \times E + 0.8 \times D$					
Step 5 (Emission Factor)						
Item	CH ₄ Emission factor					
Sub-Item	M					
Unit	kgNon-CO ₂ /TJ					
Calculation	$[(L + (K - I) \times J) \times G \times (16/22.4)] \div F$					

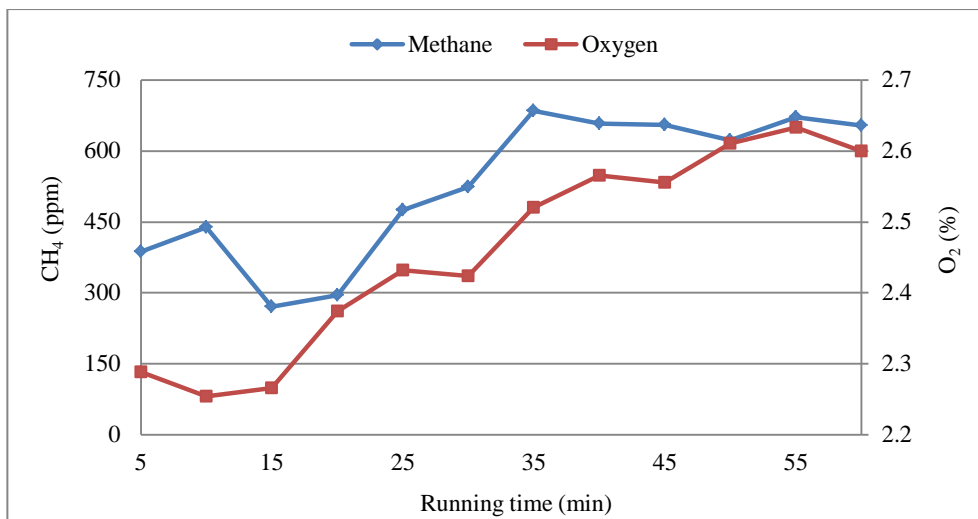
Chapter 4. Emission characteristics of exhaust gas

4.1 Analyzing emission characteristic

4.1.1 CH₄ concentration with fan turned off

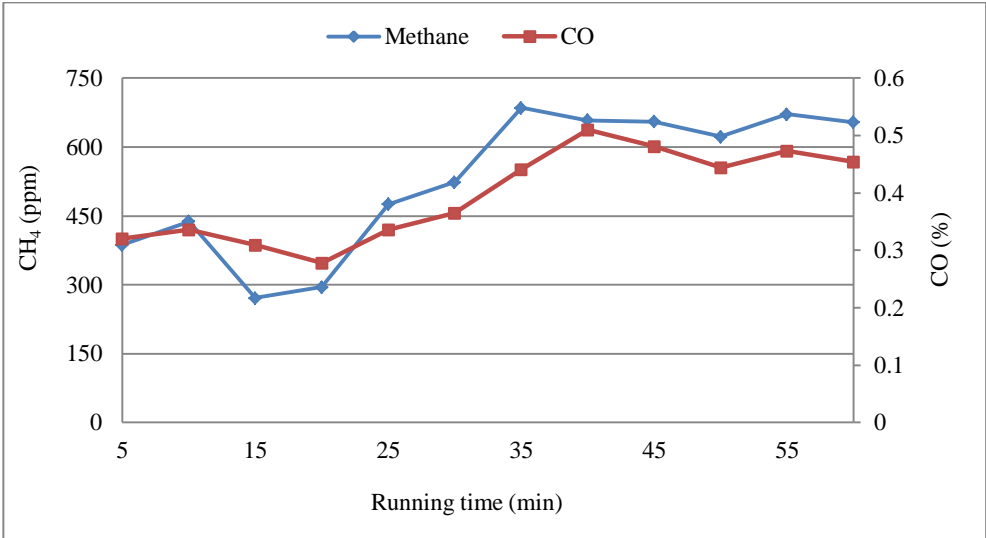
An experiment was carried out for 60 minutes to measure the concentration of CH₄ in exhaust gas with fan turned off and the samples were taken every 5 minutes. The concentrations of CH₄ were relatively high, ranging from 271.13 ppm to 676.91 ppm. Changes in the concentrations of CH₄ and exhaust gases with fan turned off are shown in <Figures 14 ~ 17>.

As shown in <Figure 14>, the emission characteristic of CH₄ and O₂ were different at the beginning of operation. However, the emission characteristic of O₂ concentration was similar to CH₄ concentration after 20 min.



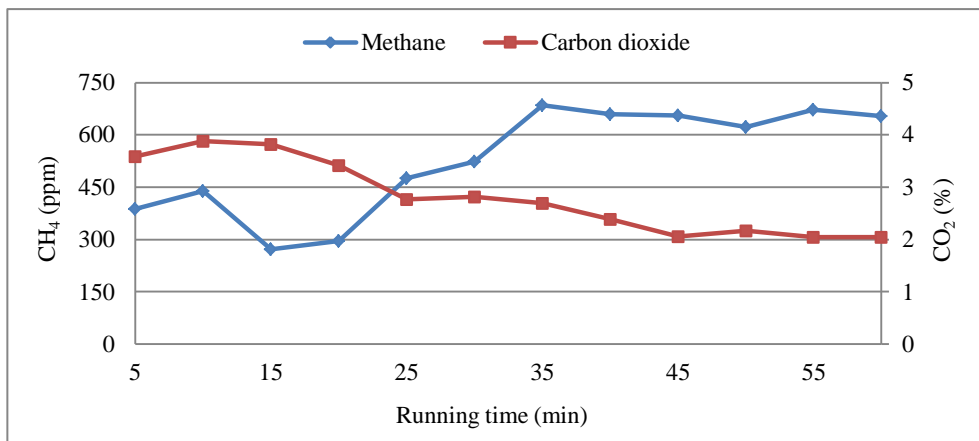
<Figure 14> CH₄ and O₂ concentrations with fan turned off

As shown in <Figure 15>, the emission characteristic of CO concentration was similar to CH₄ concentration when boiler was operated. And the CO and CH₄ concentrations were increased after 20 min. It can be seen that the higher the concentration of CO in the exhaust gases, the higher the concentration of CH₄.



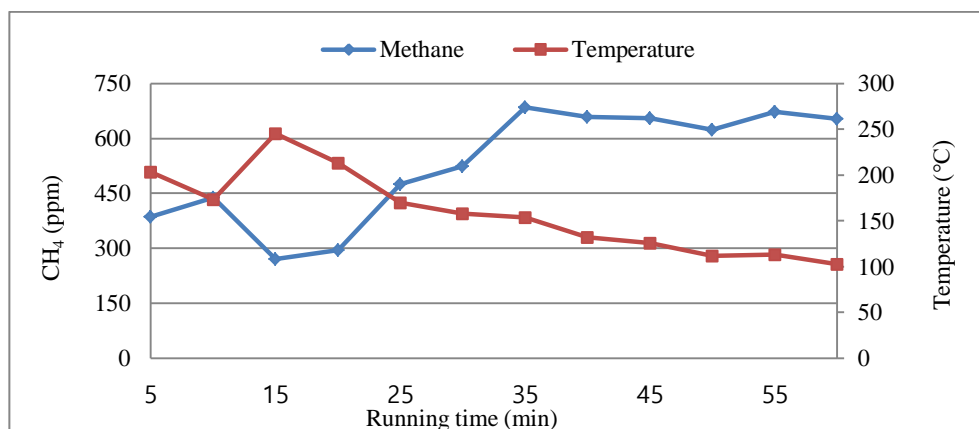
<Figure 15> CH₄ and CO concentration with fan turned off

As shown in <Figure 16>, the emission characteristic of CH₄ and CO₂ concentrations were different after 20 min. The CO₂ concentration was continuously decreased after 20 min. On the other hand, CH₄ concentration was increased after 20 min.



<Figure 16> CH₄ and CO₂ concentration with fan turned off

As shown in <Figure 17>, the temperature and CH₄ concentration were showing different trends while the boiler was operated. It can be seen that the lower the temperature, the higher the concentration of CH₄. This is considered that the emission characteristics of CH₄ were formed by incomplete combustion with insufficient air and a low combustion temperature (Kim et al., 2013; Korea Energy Agency, 2008).



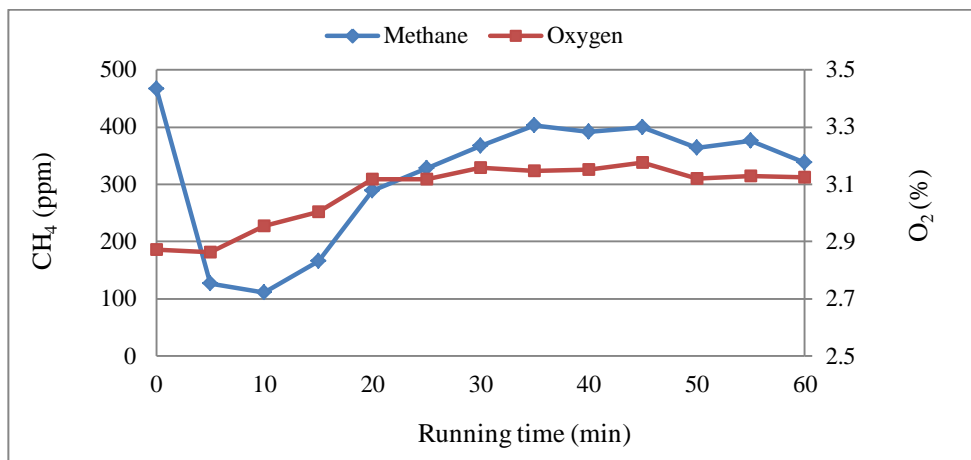
<Figure 17> CH₄ concentration and Temperature with fan turned off

4.1.2 CH₄ concentration with fan turned on

An experiment was carried out for 60 minutes to measure the concentration of CH₄ in exhaust gas with fan turned on, and the samples were taken every 5 minutes.

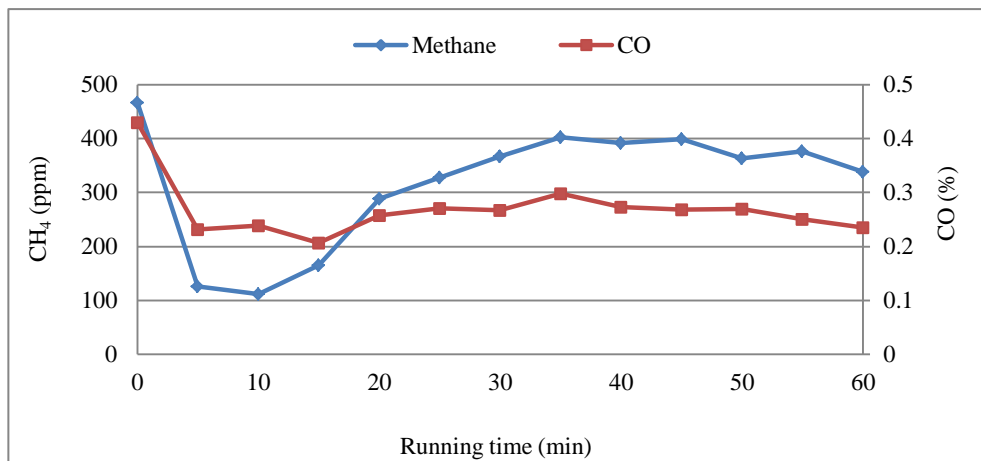
The concentrations of CH₄ were ranging from 111.28 ppm to 466.49 ppm. The CH₄ concentration with fan turned on was relatively lower than fan turned off. Changes in the concentrations of CH₄ and exhaust gases are shown in <Figures 18 ~ 21>.

As shown in <Figure 18>, the emission characteristic of O₂ concentration was similar to CH₄ concentration after 20 min. There's no difference between fan turned off and turned on. However, O₂ concentration with fan turned on was slightly changed in comparison with fan turned off.



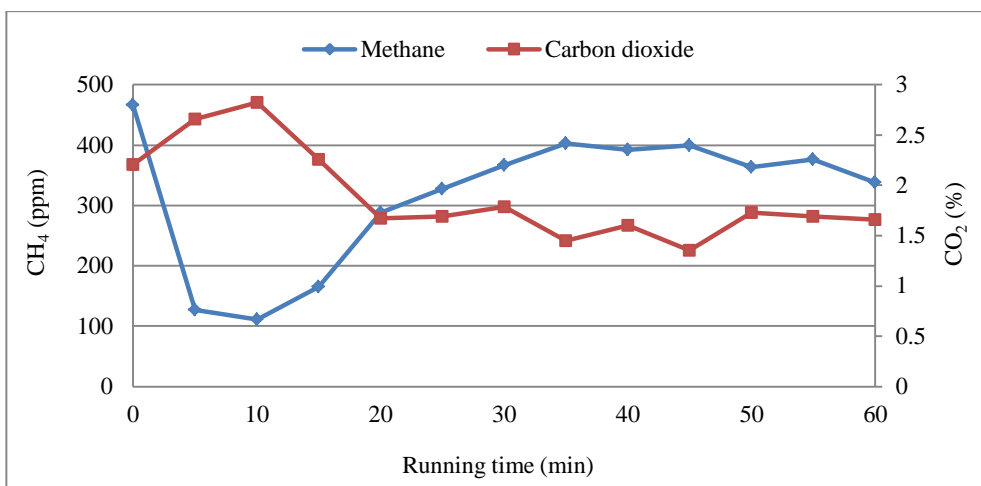
<Figure 18> CH₄ and O₂ concentration with fan turned on

As shown in <Figure 19>, the emission characteristic of CH₄ concentration was proportional to CO. The change of CO concentration was similar to CH₄ concentration after 20 min. And also, the CO concentration with fan turned on was relatively lower than fan turned off.



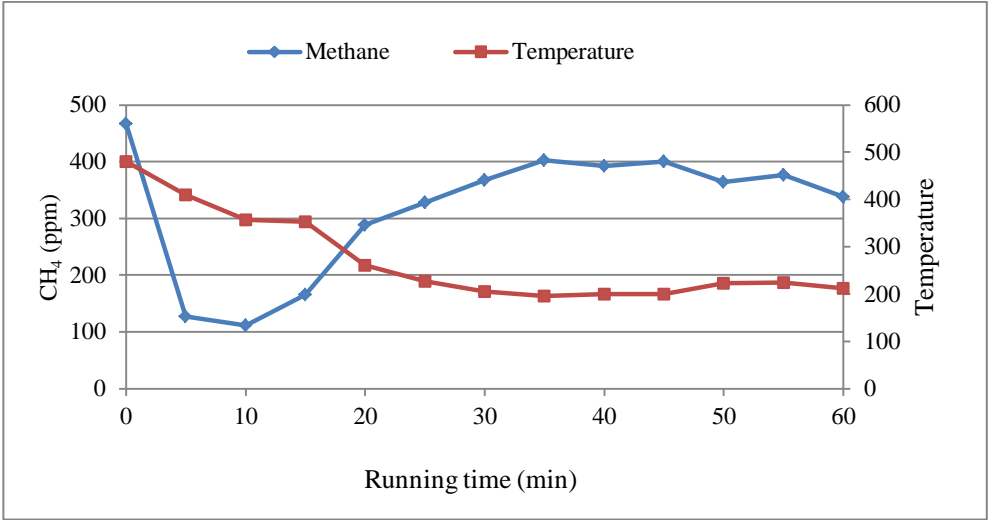
<Figure 19> CH₄ and CO concentration with fan turned on

As had been seen with fan turned on, CH₄ concentration was proportional to O₂ and CO. However, as shown in <Figure 20>, CH₄ concentration was in inverse proportion to CO₂ when boiler was operated.



<Figure 20> CH₄ and CO₂ concentration with fan turned on

As show in <Figure 21>, the temperature and CH₄ concentration were different when boiler was operated. And the temperature with fan turned on was high in comparison with fan turned off.



<Figure 21> CH₄ concentration and Temperature with fan turned on

The Changes with fan turned off were no different than with fan turned on. CH₄ concentration was proportional to O₂ and CO but it was in inverse proportion to CO₂ and temperature.

4.2 Analyzing correlations between exhaust gases

The correlations between CH₄ concentration and the O₂, CO, and CO₂ in exhaust gases were examined using SPSS. Before the correlation analysis, normality tests were performed to understand the distribution of the results.

If the significance probability is greater than 0.05 in the results of normality tests, the data can be judged to follow a normal distribution. According to the results for normality tests of CH₄, O₂, CO, and CO₂ concentrations among exhaust gases, all the significance probabilities of the test values were lower than 0.05, indicating that the test values did not follow a normal distribution.

Therefore, nonparametric Spearman's rho correlation analyses were performed to investigate the correlations between CH₄ concentrations and O₂, CO, and CO₂ among exhaust gases and temperatures in the furnace. The results are shown in <Table 9>.

The significance probability (2-tailed) of the correlation between CH₄ and CO concentrations among the exhaust gases is less than 0.01, indicating that the correlation between them is statistically significant. The correlation coefficient of CH₄ and CO concentrations among the exhaust gases was 0.756, indicating a clear positive linear relationship. Therefore, it can be seen that the higher the concentration of CO in the exhaust gases, the higher the concentration of CH₄.

CH₄ was formed in an environment of incomplete combustion at a low combustion temperature (Kim et al, 2013; Korea Energy Agency, 2008); CO concentration in exhaust gases was also associated with incomplete combustion. It is concluded that the higher the CO concentration was, the more CH₄ was emitted through incomplete combustion. According to the results of analysis of the correlation between CH₄ concentration and in-furnace temperatures, the significance probability (2-tailed) was

lower than 0.05, indicating that CH₄ concentration and in-furnace temperatures were correlated with each other. The correlation coefficient was -0.303, indicating the negative linear relationships. Therefore, it can be seen that the higher temperature in the furnace, the lower the concentration of CH₄. Because CH₄ concentration tends to decrease steadily as combustion temperature increases (Kim et al., 2013; Ryu et al., 2011; Korehonen., 2001), it is concluded that CH₄ concentration is lower, when the temperature is higher.

<Table 9> Result of correlation analysis

Classification		CH ₄	O ₂	CO	CO ₂	Temperature	
Spearman's rho	CH ₄	Correlation Coefficient	1.00	-.107	.756**	.114	-.303*
		Sig. (2-tailed)	.	.420	.000	.389	.020
		N	59	59	59	59	59

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

Chapter 5. Development of CH₄ emission factors

5.1 Fuel analysis results

The emissions from fuel combustion are dependent on fuel characteristics such as the type of fuel, water content, and calorific value. The amount of emissions from same type of wood fuels may vary with the aforementioned characteristics, particularly water content (Korea Rural Economic Institute, 2005).

In this study, field surveys were conducted four times. The results of analysis of the wood fuel used in this study are shown in <Table 10>. To estimate the net calorific value from the total calorific value of a fuel, the hydrogen content and the total water content of the fuel must be known. The average of calorific value as dry basis was 4,421 kcal/kg. And the average of net calorific value as received basis was 3,494 kcal/kg.

<Table 10> Fuel analysis of wood-fired boiler

No.	Configuration	Total moisture (%)	Element content as dry basis (%)		Gross calorific value as dry basis (kcal/kg)	Net calorific value as received basis (kcal/kg)
			C	H		
1	Fan off	16.21	51.73	5.78	4,417	3,342
2		11.68	50.01	5.99	4,448	3,573
3	Fan on	11.28	49.15	5.76	4,313	3,483
4		12.11	49.73	5.82	4,469	3,579
Mean		12.82	50.16	5.84	4,412	3,494
SD		2.29	1.11	0.10	69	111
RSD (%)		17.83	2.21	1.79	2	3

5.2 Estimation of the CH₄ emission factor for residential wood-fired boiler

5.2.1 Result of CH₄ concentration

In order to identify the emission characteristic, the experimental condition was dependent upon the operation of fan. The results of the analysis of CH₄ concentrations are shown in <Table 11>.

The CH₄ concentrations with fan turned off were relatively high ranging from 271.13 ppm to 676.91 ppm. The average CH₄ emission concentration was 521.02 ppm.

The CH₄ concentrations with fan turned on were relatively low ranging from 111.28 ppm to 466.49 ppm. The average CH₄ emission concentration was 280.19 ppm.

The CH₄ concentration with fan turned on was lower than the CH₄ concentration with fan turned off. It is concluded that CH₄ concentration, appears to be low when the fan was turned on because of the amount of combustion air being added into the furnace.

<Table 11> CH₄ concentration analysis of wood-fired boiler

No.	Number of samples	Configuration	CH ₄ (ppm)	Mean (ppm)
1	14	Fan off	547.56	521.02
2	13		494.47	
3	12	Fan on	254.10	280.19
4	15		306.28	

5.2.2 Estimation of the CH₄ emission factor

In this study, the CH₄ emission factors of the wood-fired boiler were developed in case of fan turned on or off. The average CH₄ emission factor was 88.31 kg CH₄/TJ with fan turned on.

The average CH₄ emission factor was 171.98 kg CH₄/TJ with the fan turned off. The CH₄ emission factor with fan turned off is about 1.9 times higher than with fan turned on.

These differences were assumed to be caused by the difference in the amount of combustion air. CH₄ concentration was formed by incomplete combustion at insufficient air and low combustion temperature (Kim, 2013; Korea Energy Agency, 2008). The CH₄ emission factors when the wood-fired boiler fan is turn on or off are shown in <Table 12>.

<Table 12> Result of CH₄ emission factors by condition

Number of operation	CH ₄ emission factor (kg CH ₄ /TJ)	
	Fan on	Fan off
1 st operating	78.97	186.08
2 nd operating	97.65	157.88
Mean	88.31	171.98
SD	13.21	19.94

The emission factors developed in this study were compared with the IPCC default values for the residential sector. However, emission factor of wood-fired boiler was not found in the residential sector of the 2006 IPCC guidelines. Those the emission factors developed in this study were compared with emission factors of similar facilities of burning the firewood.

The CH₄ emission factors were shown in <Table 13>. The emission factors developed in this study were lower than the other emission factors. Comparison between wood stove in Asian countries and wood fired-boiler with fan turned off show that CH₄ emission factor developed in this study was 1.5 times lower than wood stove in Asian countries. Also, the emission factor with fan turned on was 2.9 times lower than wood stove in Asian countries. These differences were expected to be due to the technology-specific of CH₄ emission and total moisture content of fuel used.

<Table 13> Comparison of CH₄ emission factors

Source	Technology	Configuration	CH ₄ emission factor (kgCH ₄ /TJ)
This study	Wood-fired boiler	Fan on	88.31
		Fan off	171.98
IPCC	Wood pits		200
	Wood stove in US	Conventional	932
	Wood stove in Asian countries		258 - 2190
	Wood fireplaces		275 - 386
	Agriculture wastes stoves		230 - 4190

Chapter 6. Conclusion

6.1 Summary

In this study, emission characteristics were identified and CH₄ emission factors were developed for wood-fired boiler. In order to do that, field surveys were conducted to collect the exhaust gases from the wood-fired boiler. And the fuel used in the wood-fired boiler was analyzed in the laboratory.

In the fuel analysis, the gross calorific value as dry basis was 4,421 kcal/kg, carbon content was 50.16%, and hydrogen content was 5.84%. Using the hydrogen content, the total moisture content and the total calorific value of firewood were converted into net calorific value as received basis.

The CH₄ concentration was from 271.13 ppm to 676.91 ppm with fan turned off, and from 111.28 ppm to 466.49 ppm with fan turned on. CH₄ emission factors were 171.98 kg CH₄/TJ with fan turned off, and 88.31 kg CH₄/TJ with fan turned on.

The difference between the CH₄ concentrations and emission factors found in this study depends on the average furnace temperature and the amount of combustion air added. The emission factors developed in this study were compared to the IPCC default values for wood stoves. The difference found between the CH₄ emission factors of this study and those of the IPCC are probably caused by combustion technology and the total moisture content of the fuel used.

6.2 Limitations and implications

To identify the emission characteristic and develop the CH₄ emission factors for residential wood-fired boiler, this study conducted for field surveys in the same house. For that reason, this study has limitation on the representativeness of samples. In order to ensure reliability of the emission factors, a variety of field surveys should be conducted for residential wood-fired boiler.

However, this study investigated the variation of CH₄ concentration and the correlations between CH₄ concentrations and affecting factors (exhaust gas and temperatures in the furnace) while wood-fired boiler was operated. Therefore, it is expected that this study will help to understand CH₄ emission characteristics which have not been sufficiently studied for residential wood-fired boiler.

This study developed the CH₄ emission factors for residential wood-fired boiler. It is expected that the result of this study can increase the reliability of national greenhouse gas inventory.

Regarding future research, studies on the development of emission factors and inventories for wood-fired boilers should be continued not only for CH₄ but also for N₂O. And also, those studies should be conducted for other facilities that used firewood as fuel.

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국문초록

가정용 화목보일러의 CH₄ 배출계수 및 배출특성 개발

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화목보일러는 전기나 기름 연료 대신 장작을 이용하여 겨울철 난방용으로 사용된다. 화목보일러는 2013년 기준 약 45,447대가 가동 중이며, 가정뿐만 아니라 상업시설에서도 난방을 목적으로 화목난로 및 보일러를 사용하고 있는 추세이다. 그러나 화목보일러의 Non-CO₂ 배출계수는 개발된 사례가 미흡한 실정이다.

따라서, 본 연구에서는 가정용 화목보일러를 대상으로 현장조사를 통해 CH₄ 배출특성을 확인하고 CH₄ 배출계수를 개발하고자 하였다. 현장조사는 가정에 설치된 화목보일러의 배기가스를 채취하여 배기가스 중 CH₄, CO, O₂, CO₂ 농도 등을 분석하고, 대상 화목보일러에서 사용하는 장작을 실험실에서 분쇄한 뒤 발열량, 원소 함량, 그리고 수분함량 등을 분석하였다.

가정용 화목보일러의 CH₄ 배출특성을 확인한 결과, 화목보일러의 송풍기를 가동하지 않았을 때의 CH₄ 농도는 송풍기를 가동하지 않았을 때보다 비교적 높게 배출되었다. 또한, CH₄ 농도와 배기가스 중 O₂, CO, CO₂, 노내온도의 상관관

계를 알아보기 위해 Spearman rho 상관분석을 실시하였다. 그 결과, 배출가스 중 CO 농도가 높으면 높을수록 CH₄ 농도도 높아지며, 노내온도가 높으면 높을수록 CH₄ 농도는 낮아진다는 것을 알 수 있었다.

화목보일러의 CH₄ 배출계수는 보일러의 송풍기를 가동했을 때와 가동하지 않았을 때로 나누어 각각 산정하였다. 화목보일러의 송풍기를 가동했을 때의 CH₄ 배출계수는 88.31 kgCH₄/TJ으로 산정되었으며, 화목보일러의 송풍기를 가동하지 않았을 때의 CH₄ 배출계수는 171.98 kgCH₄/TJ으로 산정되었다. 화목보일러의 송풍기를 가동하지 않을 경우, 보일러내의 연소 공기량이 부족하게 되므로 일반적으로 연소 공기량이 부족하거나 연소온도가 낮은 불완전연소에 의해 생성되는 CH₄의 배출농도가 상대적으로 높게 배출된 결과로 판단된다.

개발된 가정용 화목보일러의 CH₄ 배출계수는 2006 IPCC 가이드라인에서 제시하고 있는 화목난로의 기본배출계수와 비교하였다. 본 연구에서 개발된 배출계수 중 송풍기를 가동하지 않았을 때와는 약 2.9배, 송풍기를 가동했을 때에는 약 1.5배 정도 낮은 것으로 나타났다. 이는 화목보일러의 경우 화목난로와는 달리 연소온도와 사용된 연료의 수분함량이 다르기 때문에 차이가 나는 것으로 해석할 수 있다. 우리나라의 온실가스 인벤토리 신뢰도를 높이기 위해서는, 화목보일러뿐만 아니라 화목난로 등과 같이, 다양한 연소시설에서의 Non-CO₂ 배출계수 개발과 관련된 연구가 수행되어야 할 것이다.

주요어 : Greenhouse gas, Wood-fired boiler, CH₄ emission factor