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Analysis of Main Factors for CH_4 Emission Factor Development in Manufacturing Industries and Construction Sector

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Abstract: This study has statistically analyzed the effect of boiler type and model year on CH_4 emission factors, focusing on liquefied natural gas (LNG)—the most commonly used fuel in South Korean manufacturing combustion facilities. Samples were collected from the boilers of 39 manufacturing combustion facilities that use LNG fuel. The CH_4 emission factors were developed based on 4 overhead fire-tube boilers, 14 once-through boilers, 14 vertical boilers with vertical water tubes, and 7 other boilers. This resulted in an average value of 0.11 CH_4 kg/TJ, which is considerably lower than the Intergovernmental Panel on Climate Change (IPCC) 1996 guidelines(G/L) emission factor of 5 CH_4 kg/TJ currently used in South Korea. In the Kruskal–Wallis test results, the significance probability was greater than 0.05 for the boiler types and ages, and there was no major difference in the average distributions, according to the boiler type or age. Therefore, according to the results of this study, the differences in the CH_4 emission factors according to the boiler types and ages are not statistically large, and it was determined that there is no major difference even when the emission factors are applied to different fuel types. However, there was a major difference when the developed factors were compared to the CH_4 emission factor proposed by the IPCC. Thus, there is a need to develop manufacturing combustion CH_4 emission factors that reflect national characteristics.

Keywords: manufacturing industries; boiler type; CH_4 emission factor; Kruskal–Wallis test; GHG emission

1. Introduction

The 21st Conference of the Parties (COP) on climate change in 2015 adopted the Paris Agreement regarding a "new climate regime." According to the new climate regime, global stocktaking will be performed at 5-year intervals starting in 2023, and national greenhouse gas (GHG) inventories and greenhouse gas reductions must be reported [1]. To prepare for the global stocktaking, it is important to improve the reliability of greenhouse gas inventories and understand the characteristics of greenhouse gas emission sources [2–4].

In 2016, Korea's total greenhouse gas emissions were 693 million tons CO_2 eq. When compared to OECD (Organization for Economic Cooperation and Development) countries, Korea's GHG emissions were ranked 5th, after the United States, Japan, Germany, and Canada. Therefore, Korea is one of the countries that should work to improve the reliability of its greenhouse gas inventory [5–7].

In 2016, Korea's greenhouse gas emissions in the energy sectors were 604 million tons CO_2 eq, accounting for 87% of Korea's total greenhouse gas emissions. Next, the industrial process was

53 million tons CO_2 eq, agriculture sector 21 million tons CO_2 eq, and waste sector 16.8 million tons CO_2 eq [8].

The total greenhouse gas emissions from fuel combustion by manufacturing and construction industries in South Korea amounted to 184.3 million tons CO_2 equivalent (CO_2 eq) in 2016. This is approximately 30% of the 604 million tons CO_2 eq of greenhouse gases that were emitted by the entire energy sector [9]. Thus, combustion from manufacturing and construction sectors is one of the major sources of greenhouse gas emission in the energy sector. To prepare for the global stocktaking, it is important to improve the reliability of greenhouse gas inventories and understand the emission characteristics of greenhouse gas emission sources [9].

According to the 2006 guidelines (G/L) of the Intergovernmental Panel on Climate Change (IPCC), the non-CO_2 emissions in fuel combustion, such as CH_4 and N_2O, can be affected by factors such as the operating conditions of the fuel and the combustion technology used [10–12].

South Korea uses the values that are proposed in the IPCC 1996 G/L for the manufacturing combustion CH_4 emission factors. However, the IPCC 2006 G/L recommends using country-specific values. Thus, there is a need to develop manufacturing CH_4 emission factors that reflect the characteristics of South Korea.

In South Korean manufacturing fuel combustion, water-tube, once-through, and fire-tube boilers are primarily used. Additionally, hot oil boilers are seldom used. However, the CH_4 emission factors that are currently associated with South Korean manufacturing combustion are related to the fuel type. Thus, there is a need to examine whether the factors are affected by boiler type and model year. This study has statistically analyzed the effect of boiler type and model year on the CH_4 emission factors, focusing on liquefied natural gas (LNG)—the most commonly used fuel in South Korean manufacturing combustion facilities.

2. Methods

2.1. Characteristics and Population Analysis of Boilers at Manufacturing

To examine the population of manufacturing combustion boilers according to fuel and boiler type, this study used boiler inspection records from 2014 [13]. This data is divided into boiler type and year, and the year classification in this study is based on this statistical data.

These data include the fuel used by the manufacturing boiler, its capacity, the technology type of the combustion facility, and the installation year. 17,688 manufacturing boiler facilities were examined, as shown in Table 1.

The analysis of the manufacturing boilers according to the fuels used showed that 9846 boilers use solid-gas fuel, 6756 use liquid fuel, 46 use solid fuel, and 1041 use other fuels. Therefore, gas fuel is the most common fuel type. If the boilers are classified by capacity, 6718 have a capacity of 3 tons or more, 6630 of 1.5–3 tons, and 4340 of less than 1.5 tons. As for the boiler types, the once-through boilers were the most used with 4440 gas boilers and 3130 liquid boilers. Water-tube boilers were the most used boilers for solid fuels and other fuels with 43 boilers and 391 boilers, respectively.

Table 1. Population of the investigated boilers in manufacturing.

Classification	Fuel type	Facility	< 1.5 tons			< 1.5 ~ 3.0 tons			3.0 tons ≤			Total	
		Capacity / Model years	< 7 years	8-15 years	16 years ≤	< 7 years	8-15 years	16 years ≤	< 7 years	8-15 years	16 years ≤		
Manufacturing Industries	**Solid**	**Overhead fire tube boilers**											46
		Once-through boilers											
		Vertical boilers with vertical water tube						1	18	5	19	43	
		Other			1				1	1		3	
	Liquid	Overhead fire tube boilers	17	36	150	63	129	453	171	344	809	2172	6756
		Once-through boilers	789	571	127	955	470	126	63	20	9	3130	
		Vertical boilers with vertical water tube	1		7	5	5	14	32	28	219	311	
		Other	130	164	299	83	94	208	24	37	104	1143	
	Gas	Overhead fire tube boilers	34	37	50	158	196	165	595	609	618	2462	9845
		Once-through boilers	684	319	92	2150	592	86	417	95	5	4440	
		Vertical boilers with vertical water tube	4	4	6	8	8	10	278	338	545	1201	
		Other	271	242	193	144	161	195	136	147	253	1742	
	Other	Overhead fire tube boilers	10	3	6	17	15	36	99	33	81	300	1041
		Once-through boilers	4	4	3	2	3	1				17	
		Vertical boilers with vertical water tube	5		5	13	4	5	160	95	104	391	
		Other	35	15	22	26	10	19	79	74	53	333	
	Total		1984	1395	961	3624	1687	1319	2073	1826	2819	17,688	
				4340			6630			6718			

2.2. Selection of Objective Facilities

This study developed CH_4 emission factors for combustion facilities that use LNG, which is the most used fuel in the manufacturing industry. Samples were collected from 39 manufacturing combustion facility boilers that use LNG. As shown in Table 2, exhaust gas samples were collected from 4 overhead fire-tube boilers, 14 once-through boilers, 14 vertical boilers with vertical water tubes, and 7 other boilers, and their CH_4 concentrations were analyzed.

Table 2. Sampling status of objective facilities.

Fuel Type	Boiler type	N
LNG	Overhead fire-tube boiler	4
	Once-through boiler	14
	Vertical boiler with vertical water tube	14
	Other	7
	Total	39

2.3. Flue Gas Sampling Method

To determine the CH_4 emission factor from fuel combustion, gas sampling for the exhaust gas discharge, temperature, and water content, is required [14,15].

Because boiler exhaust gas is emitted at a high temperature, the tube for the sample collection must be made of a material that can withstand high temperatures and flow speeds. Thus, in this study, sample collection tubes made of stainless steel (diameter: 7 mm) were manufactured in accordance with the South Korean official test method for air pollution.

To remove the moisture in the gas emitted from the boilers, a moisture absorption bottle filled with silica gel was installed in front of the device that collected the emission gas. Therefore, greenhouse gas samples did not contain moisture.

In this study, 10 L Tedlar bags were used to collect the exhaust gas, and the samples were collected according to Environmental Protection Agency(EPA)Method 18 [16]. Figure 1 shows a diagram of the sample collection. Because manufacturing boilers are operated intermittently, oxygen concentrations were monitored in real time, and the samples were collected during boiler operation.

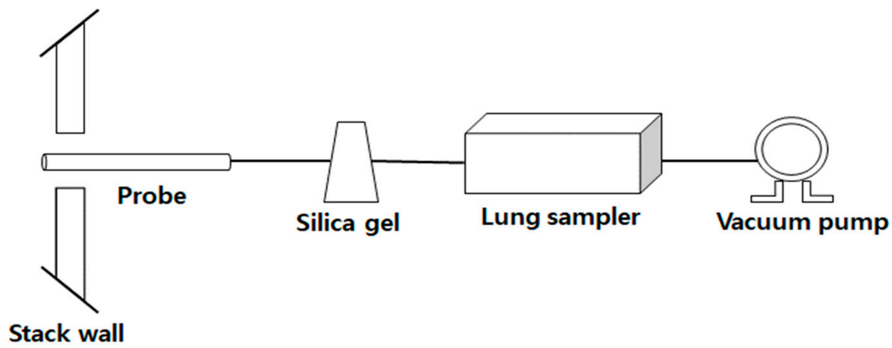

Figure 1. Greenhouse gas sampling using lung sampler.

2.4. Analysis of CH_4 Concentration

A gas chromatography-flame ionization detector (GC-FID) was used to analyze the CH_4 concentrations in the exhaust gas collected from the manufacturing facilities. A Porapack Q 80/100 column was used for the CH_4 concentration analysis, and the carrier gas flow was set to 20 ml/min. The injector temperature was set to 120 °C, the oven temperature at 70 °C, and the detector temperature at 100 °C. High-purity nitrogen (99.999%) was used as the carrier gas. Table 3 shows the GC-FID conditions for the CH_4 analysis.

Table 3. Condition of gas chromatography-flame ionization detector (GC-FID) for CH_4 concentration.

Classifications		GC-FID
Column		Parapack Q 80/100 Mesh
Carrier gas		N_2(99.9999%)
Flow		20 ml/min
Temperature	Oven	70°C
	Injector	120°C
	Detector	100°C

2.5. Development of a CH_4 Emission Factor

Generally, fuel combustion is used to originate the combustion reaction and calculate the theoretical concentration of the fuel gas used for estimating the emission factor. This method is used for estimating the emission factor related to non-CO_2 emissions from a stationary source [17].

Therefore, this study developed the greenhouse gas emission factor, shown in Equation (1), taking into account the experimental amount of exhaust gas from combustion and the theoretical air amounts (that were calculated by considering the measured gas concentrations and fuel characteristics). The net calories for the combusted fuel were collected from the city gas company that served the manufacturing facilities.

$$\mathrm{EF} = C_{CH_4} \times \{G_0 + (m-1) \times A_0\} \times MW / V_m / \ NCV \tag{1}$$

where EF is emission factor (kg CH4/TJ); CCH4 is CH4 concentration in exhaust gas (ppm); G0 is theoretical air volume for each fuel combustion (m^3N/original unit); m is Air ratio = actual air volume/theoretical air volume; MW is molecular weight of CH4 (constant) = 16 (g/mol); Vm is one mole ideal gas volume in standardized condition (constant) = 22.4 (10^{-3} m^3/mol); and NCV is net calorific value for each fuel combustion (MJ/original unit)

However, the air ratio "m" is approximately provided with O_2 concentration in exhaust gas, as shown in the equation below:

$$\mathrm{m} = \frac{21}{21 - C_{O_2}} \tag{2}$$

where CO_2 is O_2 concentration in exhaust gas(%)

2.6. Statistical Analysis Method for Boiler Type and Model Year

Generally, parametric statistical analysis methods assume that the distribution of the population is known; thus, the parametric methods are more efficient than the non-parametric methods that are used when the distribution is unknown. In particular, the statistical analysis method is more reliable when the population has a normal distribution.

The normality of calculated data could be tested prior to the statistical analysis of the estimated CH_4 emission factor [18]. The Kolmogorov–Smirnov test (K–S test), Shapiro–Wilk test, chi-square test (X^2 test) and quantile-quantile plot (Q-Q plot) are the methods regularly used for the normality test [19–22]. However, when there are less than 30 measured data, it is generally assumed to be non-parametric [23].

To examine how the CH_4 emission factors are affected by the type of LNG boiler used in the manufacturing combustion facilities, this study compared the average distribution of the CH_4 emission factors according to the boiler type and model year. For the statistical analysis, the SPSS 21 program was used. Figure 2 shows the statistical process that was used to analyze the differences in the CH_4 emission factors according to the type and model year.

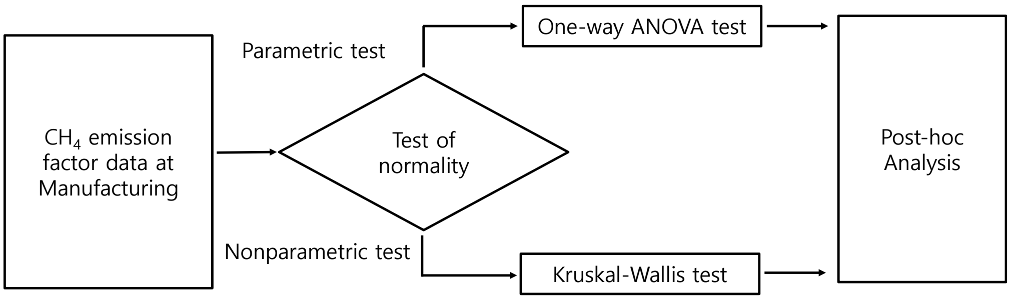

Figure 2. Schematic of statistics analysis.

3. Results and Discussion

3.1. Liquefied Natural Gas (LNG) Boiler CH_4 Emission Factor in Manufacturing

3.1.1. CH_4 Emission Factor According to the LNG Boiler Type

The distribution of developed CH_4 emission factor values according to boilers type calculated by Equation (1) is shown in Figure 3 as a box plot.

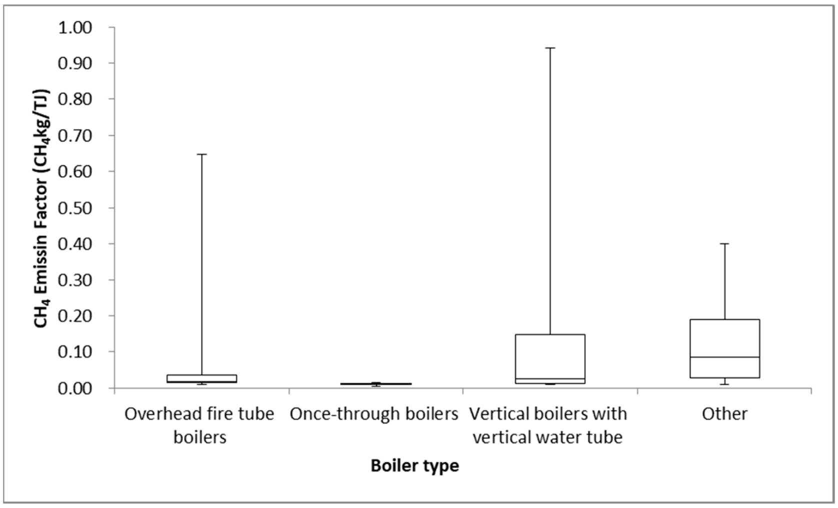

Figure 3. Box plot of developed CH_4 emission factor of boiler type.

The overhead fire tube boilers were analyzed with a maximum value of 0.65 CH_4kg / TJ and a minimum value of 0.011 CH_4kg/TJ, and median with 0.015 CH_4kg/TJ. Once-through boilers were analyzed with a maximum value of 0.015 CH_4kg / TJ and a minimum value of 0.005 CH_4kg/TJ, and median with 0.011 CH_4kg/TJ. Vertical boilers with vertical water tube were analyzed with a maximum value of 0.942 CH_4kg / TJ and a minimum value of 0.011 CH_4kg/TJ, and median with 0.027 CH_4kg/TJ. Other boilers were analyzed with a maximum value of 0.400 CH_4kg / TJ and a minimum value of 0.010 CH_4kg/TJ, and median with 0.085 CH_4kg/TJ.

As shown in Table 4, this study developed CH_4 emission factors for each boiler type to understand the characteristics that affect the CH_4 emission factors. The average CH_4 emission factor obtained for manufacturing combustion facilities that use LNG boilers was 0.11 CH_4 kg/TJ, considerably lower than the IPCC 1996 G/L emission factor of 5 CH_4 kg/TJ that is currently used in South Korea. Although according to the difference from the IPCC 2006 G/L, CH_4 emission factor of 1 CH_4 kg/TJ is smaller, the value obtained experimentally is still lower [17,24]. Therefore, it is believed that there is a need to apply an emission factor that reflects national characteristics.

Table 4. CH_4 emission factor of LNG boiler type.

Fuel Type	Boiler Type	This Study (CH_4kg/TJ)	N	IPCC 1996 CH_4 Emission Factor (CH_4kg/TJ)	IPCC 2006 CH_4 Emission Factor (CH_4kg/TJ)
	Overhead fire-tube boiler	0.11	4		
	Once-through boiler	0.01	14		
LNG	Vertical boiler with vertical water tube	0.18	14	5	1
	Other	0.13	7		
	Total	Mean: 0.11	Total: 39		

When the CH_4 emission factors were examined according to the LNG boiler type, the highest value was found for the vertical boiler with vertical water tubes, which emitted 0.18 CH_4 kg/TJ, followed by other boilers (0.13 CH_4 kg/TJ), overhead fire-tube boilers (0.11 CH_4 kg/TJ), and once-through boilers (0.01 CH_4 kg/TJ).

3.1.2. CH_4 Emission Factor According to the LNG Boilers Model Years

The distribution of developed CH_4 emission factor values according to boilers model years in Vertical boilers with vertical water tube calculated by Equation (1) is shown in Figure 4 as a box plot.

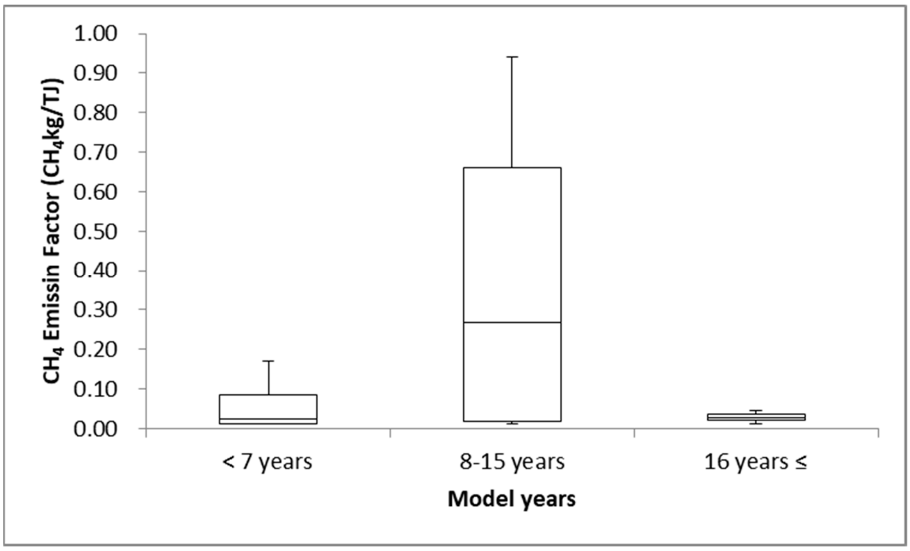

Figure 4. Box plot of developed CH_4 emission factor of boilers model year

The Vertical boilers with vertical water tube of less than 7 years were analyzed with a maximum value of 0.169 CH_4kg/TJ and a minimum value of 0.012 CH_4kg/TJ, and median with 0.025 CH_4kg/TJ. Vertical boilers with vertical water tube of 8–15 years were analyzed with a maximum value of 0.942 CH_4kg/TJ and a minimum value of 0.011 CH_4kg/TJ, and median with 0.269 CH_4kg/TJ. Vertical boilers with vertical water tube of more than 16 years were analyzed with a maximum value of 0.045 CH_4kg / TJ and a minimum value of 0.013 CH_4kg/TJ, and median with 0.028 CH_4kg/TJ.

CH_4 emission factors were measured for the water-tube boilers of manufacturing facilities and were compared according to the LNG boiler age. The results are shown in Table 5.

Table 5. CH_4 emission factor of LNG boiler model years.

Fuel Type	Boiler Type	Model Years	This Study (CH_4kg/TJ)	N	IPCC 1996 CH_4 Emission Factor (CH_4kg/TJ)	IPCC 2006 CH_4 Emission Factor (CH_4kg/TJ)
LNG	Vertical boiler with vertical water tube	<7 years	0.06	5	5	1
		8–15 years	0.37	6		
		16 years ≤	0.03	3		
		Total	Mean: 0.18	Total: 14		

CH_4 emission factors were developed for manufacturing combustion facility water-tube boilers according to boiler age. The highest factor was 0.37 CH_4 kg/TJ for vertical boilers with vertical water tube of 8–15 years, followed by 0.06 CH_4 kg/TJ for vertical boilers with vertical water tube of less than 7 years, and 0.03 CH_4 kg/TJ for vertical boilers with vertical water tube of more than 16 years. This was considerably lower than the LNG emission factor of the IPCC G/L.

3.2. Kruskal–Wallis Test of the CH_4 Emission Factor for Boiler Type and Model Year

3.2.1. Kruskal–Wallis Test of CH_4 Emission Factor for LNG Boiler Type

Generally, normal distribution data must be conducted to compare the differences in the data sets using one-way analysis of variance (ANOVA) [25]. For the data of three or more groups that do not show normal distribution the Kruskal–Wallis test can be used [26,27]. Because there were less than 30 CH_4 emission factor data for each boiler type, a non-parametric distribution was assumed, and the non-parametric Kruskal–Wallis test was selected for comparing the average distributions, determining whether the CH_4 emission factor was affected by the boiler type. The results are shown in Table 6.

Table 6. The result of Kruskal–Wallis test by CH_4 emission factor for LNG boilers type.

Hypothesis Test	Null Hypothesis	Test	Signigicant Level	Decision
CH_4 emission factor for LNG boilers type	The distribution of CH_4 emission factor is the same across categories of LNG boiler type	Independent-samples Kruskal–Wallis Test	0.93	Retain the null hypoyhesis

In the Kruskal–Wallis test results, the significance was greater than 0.05, and the test supported the null hypothesis of "there is no difference in CH_4 emission factors according to the manufacturing boiler types (once-through, hot oil, water-tube, and other)". As such, it was determined that the type of manufacturing combustion facility LNG boiler does not affect the CH_4 emission factors.

3.2.2. Kruskal–Wallis Test of CH_4 Emission Factor for LNG Boiler Model Year in Manufacturing

Because there were fewer than 30 CH_4 emission factor data for each boiler age, a non-parametric distribution was assumed, and the non-parametric Kruskal–Wallis test was selected as the method for comparing the average distribution of the data. The results are shown in Table 7.

Table 7. The result of Kruskal–Wallis test by CH_4 emission factor for LNG boiler model year.

Hypothesis Test	Null Hypothesis	Test	Signigicant Level	Decision
CH_4 emission factor for LNG boiler model year	The distribution of CH_4 emission factor is the same across categories of LNG boiler model year	Independent-samples Kruskal–Wallis Test	0.94	Retain the null hypoyhesis

The Kruskal–Wallis test was used to compare the average distributions and find out whether the CH_4 emission factors were affected by the boiler age.

In the Kruskal–Wallis test results, the significance probability was greater than 0.05, and the test supported the null hypothesis: "There is no difference in the CH_4 emission factors according to the manufacturing boiler age (7 years or less, 8–15 years, 16 years or more)". Therefore, it was determined that the age of the manufacturing combustion facility LNG boiler does not affect the CH_4 emission factors.

4. Conclusions

South Korea uses the basic values that are proposed in the IPCC 1996 G/L for its manufacturing fuel combustion CH_4 emission factors. However, the IPCC 2006 G/L recommends using country-specific values

The boilers that are used in South Korean manufacturing combustion mainly include water-tube boilers, once-through boilers, and fire-tube boilers, although hot oil boilers are also used. According to the IPCC G/L, non-CO_2 emission factors are significantly affected by the boiler combustion type. However, the CH_4 emission factors are currently used for fuel combustion in the manufacturing industry of South Korea according to the fuel type. Therefore, there is a need to examine whether the factors are affected by the boiler type and boiler age.

This study has used statistical methods to analyze the effect of boiler type and age on the CH_4 emission factors, focusing on LNG, which is the most-used type of fuel in South Korean manufacturing combustion facilities.

Samples were collected from the boilers of 39 manufacturing combustion facilities that use LNG fuel. The CH_4 emission factors were developed based on 4 overhead fire-tube boilers, 14 once-through boilers, 14 vertical boiler with vertical water tubes, and 7 other boilers.

This resulted in an average value of 0.11 CH_4 kg/TJ, which is considerably lower than the IPCC 1996 G/L emission factor of 5 CH_4 kg/TJ currently used in South Korea. Compared with the CH_4 emission factor of the IPCC 2006 G/L, which is 1 CH_4 kg/TJ, the difference of the developed factor is smaller than with the IPCC 1996 G/L one. However, the developed value is still lower. Therefore, we believe that there is a need to develop and apply emission factors that reflect national characteristics.

When the CH_4 emission factors were developed according to the LNG boiler type, the highest factor (0.18 CH_4 kg/TJ) was found for Vertical boilers with vertical water tube, followed by other boilers (0.13 CH_4 kg/TJ), Overhead fire tube boilers (0.11 CH_4 kg/TJ), and Once-through boilers (0.01 CH_4 kg/TJ).

However, when CH_4 emission factors were developed for Vertical boilers with vertical water tube according to their age, the highest factor found was 0.37 CH_4 kg/TJ for Vertical boilers with vertical water tube of 8–15 years, followed by 0.06 CH_4 kg/TJ for those of 7 years or less, and 0.03 CH_4 kg/TJ for Vertical boilers with vertical water tube of 16 years or more. These are considerably lower than the LNG emission factor proposed by the IPCC.

The average distributions of the CH_4 emission factors were compared to analyze whether the factors are affected by the type or age of the LNG boilers of the manufacturing combustion facilities.

Because there were fewer than 30 data for each boiler type and age, a non-parametric distribution was assumed, and the non-parametric Kruskal–Wallis test was selected as the method for comparing the average distribution. In the Kruskal–Wallis test results, the significance probability was greater than 0.05 for the boiler types and ages, and there was no major difference in the average distributions, according to the boiler type or age.

Therefore, according to the results of this study, the differences in the CH_4 emission factors according to the boiler types and ages are not statistically large, and it was determined that there is no major difference even when the emission factors are applied to LNG fuel. However, there was a major difference when the developed factors were compared to the CH_4 emission factor proposed by the IPCC. Thus, there is a need to develop manufacturing combustion CH_4 emission factors that reflect national characteristics. In this study, since it was conducted only on LNG fuel, it is necessary to identify the difference according to the use of other fuels in the future. Furthermore, in the future, the manufacturing combustion greenhouse gas inventories can be improved by examining the uncertainty in the emission factors and performing similar studies on N_2O to determine whether its emission factor is also affected.

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