A Study of the Bituminous Coal Oxidation Factor in Large Scale Boilers for Estimating GHG Emissions

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ABSTRACT

Korea-specific GHG emissions should be estimated correctly in order to ensure effective measurement of climate change variables. The use of country-specific data that reflects fuel and technology characteristics is needed for accurate GHG emissions estimation. Oxidation factors are used to convert existing data into equivalent GHG emissions, and changes in these oxidation factors are directly related to changes in emissions. As such, the oxidation factor is one of the most important variables in using country-specific data to determine GHG emissions. In this study, the oxidation factor of bituminous coal in large scale boilers was estimated using 4,527 data points sampled from eight large-scale boilers that had been using bituminous coal for two years. The average oxidation factor was determined to be 0.997, which is lower than the oxidation factor of 1 that is recommended by the IPCC G/L for large scale boilers when estimating national GHG emissions. However, an oxidation factor less than 1 is assumed for fluidized bed boilers, internal combustion engines, and other small-scale boilers. Accordingly, studies on oxidation factor estimation should be continued to allow for accurate estimation of GHG emissions.

Key words: Oxidation factor, Emission factor, Bituminous coal, Combustion efficiency, Residual carbon

1. INTRODUCTION

The Annex 1 countries mandate the reduction of greenhouse gas (GHG) emissions according to the Kyoto Protocol. Among the Organization for Economic Cooperation Development (OECD) member countries, Korea, along with Mexico, is exempted from the requirements of Annex 1 countries (Lim, 2004). However, Korea's GHG emissions rank tenth in the world. In response to mounting pressure for GHG reduction from other international organizations and developed countries, South Korean President, Myung Bak Lee, announced at the G8 Summit held in Japan in August of 2008 that Korea would present GHG reduction goals by the following year.

Since the implementation of the first Comprehensive Strategy for Climate Change Convention in 1998, continuous efforts resulted in the establishment of the third Comprehensive Strategy in 2007 and implementation of the Comprehensive Fundamental Strategy for Climate Change Convention. Additionally, the Korean President announced recently that, rather than viewing climate change as an obstacle, the Korean government will use climate change as an opportunity to develop the Low Carbon Economy and Green Growth Project. In order to establish an effective policy for climate change, proper inventories such as emission sources, emission characteristics, and GHG emission estimation should be made in advance. National GHG emissions are generally estimated using data collected within the country and the Tier 1 method, which applies the default emission factor recommended in the Intergovernmental Panel on Climate Change (IPCC) guidelines, except for some cases (Eom *et al.*, 2007; Jeon *et al.*, 2006; Kim *et al.*, 2004).

In the fuel combustion sector, which is responsible for more than 80% of GHG emissions, GHG emission levels differ markedly according to the characteristics of the boilers and internal combustion engines, such as fuel types used, the quantity of consumed fuel, carbon content, caloric value of the fuel, and so on (IPCC, 2006; EPA, 2002; IPCC, 1996). Therefore, the default emission factor recommended by the IPCC does not accurately reflect Korea's characteristics, and thus cannot be used to determine actual GHG emissions for Korea.

For this reason, the 2006 IPCC guidelines present the Tier 2 and Tier 3 methods, which reflect country-specific and workplace-specific data for estimating emissions. In addition, if country-specific data is secured, it is recommended that Tier 2 and Tier 3 values be

chosen over the default emission factors. When using country-specific data, factors associated with GHG emissions such as caloric value, carbon content, hydrogen content, water content, oxidation factor, and so on are applied. Specifically, the oxidation factor is applied to fuel combustion, which comprises a large proportion of GHG emissions in Korea. Furthermore, changes in the oxidation factor are directly linked to emission changes, so it is a very important variable. Though the 2006 IPCC guidelines suggest a default emission factor, it is recommended that if each country has an oxidation factor, then that specific country's oxidation factor should be used in GHG emissions calculation (IPCC, 2006). Therefore, the aim of this study was to estimate the oxidation factor of large-scale boilers (like those found in a power plant), which are responsible for more than 30% of GHG emissions in fuel the combustion sector in Korea.

2. THEORETICAL REVIEW OF THE OXIDATION FACTOR

2.1 CO_2 Emission and the Oxidation Factor of a Combustion Facility (IPCC)

In the process of combustion, Carbon (C) in fuel is oxidized and emitted as carbon dioxide. However, a very small amount of carbon is not oxidized due to fuel characteristics and combustion technology, and this unburned carbon is emitted in the combustion byproducts such as ash, dust, and soot. Thus, fuel characteristics and combustion technology should be considered when developing country- or technology-specific emission factors.

Table 1 shows the average carbon oxidation factor by fuel type presented in the IPCC guidelines. When comparing the 1996 and 2006 IPCC guidelines, one of the most notable points is the carbon oxidation factors for fuels. Oxidation factors for fuels ranged from 0.91 to 0.995 in the 1996 report; however, in the 2006 guidelines, all oxidation factors for fuels in stationary sources were assumed to be 1. This is because generally combustion facilities, especially those with large-scale boilers (e.g., power plants), are designed and managed to maximize the combustion efficiency of carbon (the main energy source in fuel). Therefore the oxidation factor is negligible in these situations.

However, there are remarkable differences in the oxidation rates of solid fuels, especially coal, because the characteristics of coal and the corresponding combustion technologies vary significantly. For this reason, the use of country-specific oxidation factors, which are calculated based on the amount of unburned carbon present in ash, is recommended when estimating GHG

Table 1. The oxidation factors recommended in the IPCC guidelines.

Type of fuel	Default carbon oxidation factor	
	IPCC 1996	IPCC 2006
Coal	0.98[1]	1
Natural gas	0.995	1
Oil and oil products	0.99	1
Peat for electricity	0.99[2]	1

NOTE: [1]The default carbon oxidation factor of coal, 0.98, is a world average value. If the type of coal is significantly different, the lowest value, 0.91 can be used.
[2]If peat is used for home use (non-electricity generating), the value can be lower than 0.99.

emissions for coal using Tier 2 and 3 methods.

2.2 Oxidation Factor Case Studies

In the National GHG Inventory Report (NIR), there are some instances in which the country-specific oxidation factors were applied in the calculation of national CO_2 emissions. Since the IPCC recommends using country-specific oxidation factors, the Annex I countries have tried to secure country-specific oxidation rate data.

In 2004, Japan conducted experiments to determine the oxidation rate of different fuels under actual combustion conditions. The oxidation factor was measured based on the concentration of unburned carbon in soot. The carbon in the LNG for gas power plants is considered to be oxidized completely (oxidation factor is 1). In the liquid fuel (oil) power plants, the un-oxidized carbon rate is under 0.5% (oxidation factor is over 0.995). However, due to strict Japanese regulations for combustion and emission gases, the oxidation factor of liquid fuel is regarded as 1. Between 1990 and 2003, the unburned carbon concentration in ash (fly and bottom ash) was measured to determine the oxidation factor for coal (solid fuel). As a result, the oxidation factor was estimated to be 0.996 (Japan, 2007).

Australia estimated oxidation factors for 46 power plants. Specifically, the unburned carbon concentration in ash was measured for all power plants (27) that use two kinds of coal (black coal and brown coal). As a result, the oxidation rate was measured with a minimum of 0.998 and maximum of 1, and these oxidation factors were used to estimate GHG emissions for each plant (Australia, 2005).

3. RESEARCH METHODS

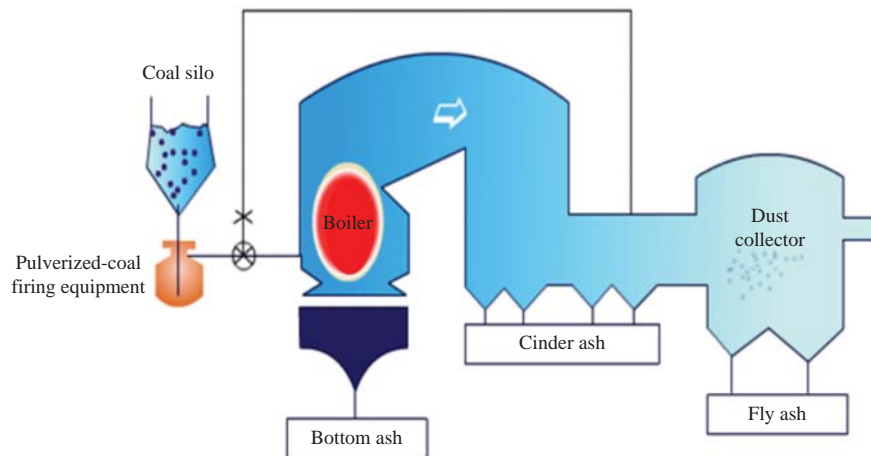Fig. 1 shows the generative process of ash in a coal fired power plant. Ash is a byproduct of pulverized

Fig. 1. Ash generation process in a coal-fired power plant.

Table 2. Operating information of large-scale combustion facilities (boilers) in this study.

Boiler No.	Fuel type	Combustion type	Capacity (kg/H)	Steam condition (kg/cm^3)	Efficiency (%)
Boiler 1-4	Bituminous coal	Pulverized-coal firing	1,720,000	255 kg/cm^3, 541°C	88.95
Boiler 5-8	Bituminous coal	Pulverized-coal firing	1,610,000	255 kg/cm^3, 541°C	90.37

coal combustion in the power plant and is classified into fly ash, bottom ash, and cinder ash according to its site. Fly ash, which consists of micro particles, comprises about 75-80% of all generated ash. Bottom ash is a massive or large-diameter particle that drops into the bottom of the boiler. Bottom accounts for about 20% of the overall ash generation. Cinder ash is collected in the hopper located under the cutter or air preheater and makes up around 5% of the total coal generated. Cinder ash and fly ash are handled and collected in a single line, as shown in the ash-handling diagram in Fig. 1. Therefore, fly ash and cinder ash are generically referred to as fly ash (Lee and Lee, 1998).

3.1 Unburned Carbon Analysis

Fly ash and bottom ash generated in large-scale combustion facilities (Table 2) were collected three times daily. They were pulverized to less than 74 μm using sticks, then an unburned carbon sample in a plate was heated to 800 ± 10°C for more than 30 min. Standard samples were made in 50-mm-diameter balanced weighing bottles after cooling to room temperature (KS E 3716, 1993).

For the loss on ignition method, a 1-2 g sample was put into a 15 mL melting pot for precise weighing to 0.1 mg. The sample was subsequently ignited in an 800°C electric furnace for 15 minutes, and its weight was measured after cooling in a desiccator. Finally,

Table 3. Repeatability test of unburned carbon content measurement in random samples.

Frequency of analysis	Unburned carbon content (%)	
	Sample 1	Sample 2
1	4.397	8.896
2	4.417	8.891
3	4.521	8.916
4	4.453	8.932
5	4.447	8.84
Average	4.447	8.895
Standard deviation	0.047	0.035
Maximum value	4.521	8.932
Minimum value	4.397	8.840
Error (%)	0.124	0.092

weight loss was measured when the sample reached a constant weight (KS E 3716, 1993).

$$C = \frac{m_1}{m_2} \times 100$$

C : weight loss on ignition (%)
m_1: sample weight (g)
m_2: weight lost after reaching a constant weight (g)

3.2 QA/QC of the Unburned Carbon Analysis

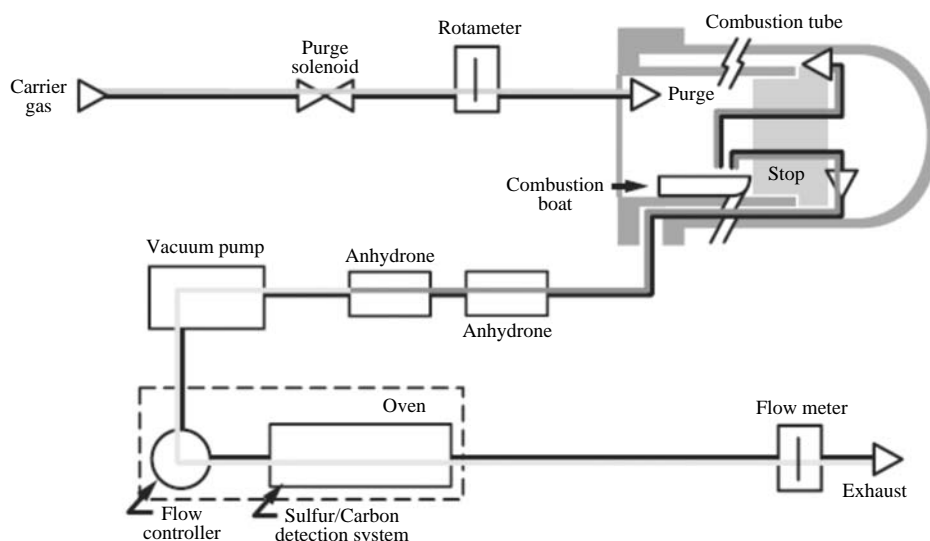To determine the repeatability of the ignition loss method, two types of identical samples were analyzed

Fig. 2. Flow diagram illustrating the operation of an elemental analyzer.

Table 4. Allowable intra- and inter-lab error ranges for unburned components[1].

Test item	Allowable intra-lab error (Measurement)	Allowable inter-lab error (Reported value)
Coal fly ash Elemental analysis	0.40	0.80

[1]The Korean Industrial Standards (1993), KS E 3716: Methods for Analysis of Coal Ash and Coke Ash.

Table 5. Unburned carbon content by loss on ignition and instrumental analysis.

Analytic method	Unburned carbon content (%)	
	Sample 1	Sample 2
Loss on ignition analysis	4.447	8.895
Instrumental analysis	4.320	8.710
Error	0.127	0.185

Table 6. Error rate between unburned carbon concentration values determined by loss on ignition and instrumental analysis.
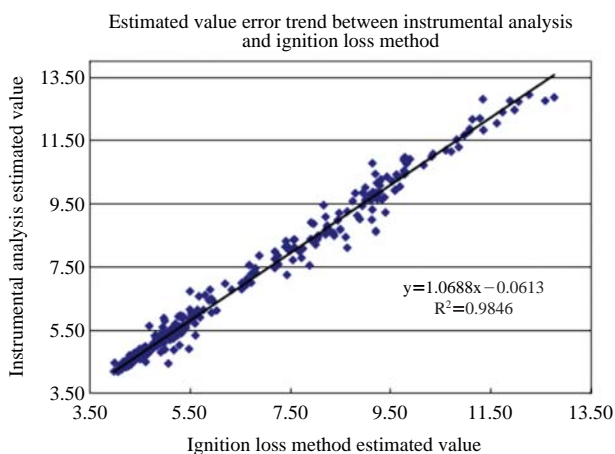
Unburned carbon concentration (%)	Error rate (%)
4	0.20
5	0.26
6	0.33
7	0.39
8	0.46
9	0.59
10	0.65
12	0.72

five times each. As shown in Table 3, the unburned carbon in 'sample 1' was 4.521-4.397% and with a standard deviation of 0.0047. The unburned carbon content in 'sample 2' ranged from 8.840% to 8.932% with a standard deviation of 0.035.

As a comparative study with the loss on ignition method, the fly ash and bottom ash samples were also analyzed with an elemental analyzer (LECO SC-144DR Sulfur/Carbon Determinator, USA) to evaluate the precision of the unburned carbon concentration. The infrared elemental analyzer measured the concentration of CO_2 emitted from the unburned carbon that remained in the ash via a carbon IR cell (Fig. 2). Subsequently, the unburned carbon content was calculated using the input equation (calibration line, sample weight, moisture content and so on) determined by analyzing standard samples containing a known amount of carbon at 1,350°C. A total of 0.001 mg of sample was measured accurately using a precision balance (Metter Toledo-MX5, Switzerland) and then injected. Intra- and inter-lab error for the proportion of unburned components comprised of coal fly ash from a domestic boiler are recommended to be at or below 0.40% and

0.80%, respectively (Table 4).

Table 5 shows unburned carbon content determined via instrumental analysis in comparison to the loss on ignition method. The unburned carbon content of 'sample 1' was 4.320% by instrumental analysis and 4.447 % by the loss on ignition method. As for 'sample 2', the unburned carbon content measured by the instrument and the loss on ignition method was 8.710% and 8.895%, respectively. The error was 0.124% for 'sam-

Table 7. Average oxidation factor in this study.

Number of measurements	Coal consumption (ton)	Ash		Unburned carbon content (wt%)		Unburned carbon quantity (ton)			Oxidation factor
		Generation rate (wt%)	Generation quantity (ton)	Fly ash	Bottom ash	Fly ash	Bottom ash	Total	
4,527	3,730	11.17	418.40	3.88	2.55	11.25	1.04	12.29	0.997

Estimated value error trend between instrumental analysis and ignition loss method

$y = 1.0688x - 0.0613$
$R^2 = 0.9846$

Fig. 3. Trend in estimated value error between the loss on ignition method and instrumental analysis.

ple 1' and 0.092% for 'sample 2'; both were below the recommended error thresholds. As shown in Fig. 3 and Table 6, which shows a trend in the occurrence of error in these measurements, a total of 272 measurements were compared. When the measurement value was lower than 7%, the error between the values was below the acceptable intra-lab error shown in Table 4.

4. RESULTS AND DISCUSSION

In this study, fly ash and bottom ash from large scale bituminous coal boilers were sampled for two years in order to measure the unburned carbon content and determine the corresponding oxidation factor. According to a total of 4,527 analyses, the unburned carbon content of fly ash and bottom ash were 3.85% and 2.55%, respectively. As shown in Table 7, because the unburned carbon content was lower than 4%, the error between the values determined via instrumental analysis and the loss on ignition method is considered to be much lower (Tables 4, 6). We used the measurements of unburned carbon content in ash to estimate the oxidation factor according the total ash (fly ash and bottom ash) generated and coal consumption. The average oxidation factor for the bituminous coal boiler based

Table 8. Oxidation factor determined in this study.

	Oxidation factor
Number of measurements	4527
Average	0.997
Maximum	1.000
Minimum	0.978
Standard deviation	0.002

on 4,527 measurements was about 0.997, with a maximum of 1, a minimum of 0.978, and a 0.002 standard deviation.

Table 9 lists oxidation factors in the EU, Japan, Australia, and Finland, including the IPCC G/L. The 1996 IPCC G/L indicated that the oxidation factor of coal, 0.98, could be altered to 0.91 depending on the type of coal and stationary combustion facility. The 2006 IPCC G/L assumed the oxidation factor of all fuels available in a stationary combustion facility to be 1. However, they recommend measuring and applying country-specific oxidation factors given the fact that there are different kinds of fuel.

In Japan, the oxidation factor for coal was measured using the same method used in this study; based on the unburned carbon content contained in ash. As a result, the estimated unburned carbon concentration in ash was 0.996. Accordingly, the oxidation factor of coal was set at 1. As for Australia, the plant-specific oxidation factor was calculated for a total of 27 coal power plants using brown and black coals. The oxidation factors ranged from 1 to 0.988, and the average oxidation factor was 0.9932 and 0.9943 for brown coal and black coal, respectively. Further, the average oxidation factor of coal was found to be 0.9937 for all power plants in Australia. In contrast, Finland measured an oxidation factor for a bituminous coal power plant, and the value was 0.99. Finland uses IPCC recommendations for other coals. In addition, according to the National Report published by Canada, the US, and others, Finland considers carbon in fuel to be completely oxidized and thus uses the oxidation factor recommended by the IPCC G/L for stationary combustion facilities (Canada, 2007; US, 2007).

This study determined the oxidation factor in eight large-scale combustion facilities (boiler) in a bitumin-

Table 9. Comparison of the results of this study with those of other studies.

Stationary combustion facility	Carbon oxidation factor					
	IPCC, 1996	IPCC, 2006	EU[1], 2004	Japan[2], 2007	Australia[2], 2007	Finland[2], 2007
Coal (Bituminous coal)	0.98[3]	1[4]	1	1[5]	PS[6]	0.99[7]

[1]EEA, 2004, [2]National Report, [3]An oxidation factor as low as 0.91 could be used based on coal quality, [4]Apply an oxidation factor of 1 to coal including bituminous coal (measurable according to coal quality), [5]Coal (measured as 0.996, regarded as 1), [6]Measured the oxidation factor of each power plant (mean value=0.9934), [7]Bituminous coal (applied IPCC oxidation factor to the other coals).

ous coal power plant over two years. The average oxidation factor was estimated to be 0.997 based on a total of 4,527 unburned carbon content measurements. This result is quite similar to the oxidation factor measurement in Japan (0.996).

5. CONCLUSIONS

Korea, one of the top ten GHG emitters, is exempt from the obligation to reduce GHG emissions imposed on the Annex 1 countries set forth by the Kyoto Protocol. However, Korea has recently been pressured by other international organizations and developed countries to submit to the GHG reduction obligation. Since President Lee announced the Low Carbon Economy and Green Growth plans, the government has regarded climate change as a potential opportunity for growth rather than a crisis. Lee's administration is preparing a positive climate change countermeasure. The first step to implementing effective action toward controlling climate change is to correctly estimate domestic GHG emissions and build databases that reflect GHG emission characteristics.

In particular, to the use of country-specific data that reflect domestic characteristics is recommended in order to more accurately estimate GHG emissions. Important country-specific data parameters include the caloric value of fuels, carbon content, hydrogen content, moisture content, oxidation factors, and others. Among them, the oxidation factor is utilized in calculating GHG emissions from the fuel combustion sector, which produces the highest GHG emissions. The oxidation factor is very important because it is directly linked to emission changes. For this reason, the aim of this study was to estimate the oxidation factor in large scale combustion facilities in Korea since power plant boilers account for more than 30% of the GHG emissions in the fuel combustion sector.

Fly ash and bottom ash were sampled from a large-scale bituminous coal boiler for two years, and then unburned carbon content was measured in order to the estimate oxidation factor. Instrumental analysis was used to measure unburned carbon content, and a total of 4,527 data points were used to determine the oxidation factor. The average oxidation factor was 0.997, which is lower than the oxidation factor of 1 recommended in the 2006 IPCC guidelines. In particular, large-scale boilers oxidize as much CO_2 as possible to maximize the energy contained in the fuel. Therefore, for large-scale coal boilers, applying the oxidation factor (1) recommended by the 2006 IPCC to calculate national CO_2 emissions.

However, for fluidized combustion facilities, small scale power plants, and domestic combustion boilers, the use of specific oxidation factors, which are estimated by more accurate methods, is recommended. In addition, when estimating Tier 3-based plant-specific CO_2 emissions for an individual workplace, applying an oxidation factor measured for a combustion facility in similar workplaces is appropriate. In conclusion, studies focused on determining the oxidation factor for coal combustion facilities other than large-scale boilers and individual workplaces should continue.

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