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Emission Characteristics of Ammonia at Bituminous Coal Power Plant

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Abstract: This study developed a NH_3 emission factor for bituminous coal power plants in South Korea in order to investigate the NH_3 emission characteristics. The NH_3 concentration analysis results showed that emissions from the selected bituminous coal power plants were in the range of 0.21–0.99 ppm, and that the difference in NH_3 concentration was affected by NOx concentration. The NH_3 emission factor was found to be 0.0029 kg NH_3/ton, which demonstrated that the difference in the values obtained from the research conducted in South Korea was lower than the difference in the emission factor provided by the U.S. EPA, which is currently applied in the statistics of South Korea. NH_3 emissions were compared by using the NH_3 emission factor developed in this study alongside the EPA's NH_3 emission factor that is currently applied in South Korea's statistics; the difference was found to be 206 NH_3 ton/year. This implies that an emission factor that reflects the national characteristics of South Korea needs to be developed. The uncertainty range of the NH_3 emission factor developed in this study was between −6.9% and +10.34% at a 95% confidence level.

Keywords: PM2.5; secondary sources; bituminous coal power plant; uncertainty analysis; ammonia emission

1. Introduction

In 2016, Ultrafine (≤2.5) Particulate Matter (PM2.5) concentration in South Korea was 26 μg/m^3, which was higher than that of Europe, the United States, and Japan. From 1990 to 2015, the average PM2.5 concentration in South Korea was 29 μg/m^3, which was the highest among all the member countries of the Organization for Economic Co-operation and Development (OECD) except Turkey. Moreover, South Korea fared worse than Vietnam, Mongolia, Japan (13 μg/m^3), and Singapore [1].

One of major causes of PM2.5 is the increase in secondary sources of particulate matters (PM), such as NOx, SOx, VOCs, and NH_3 [2–5]. In South Korea, NOx and SOx are controlled by the "Air Pollutant Emission Limit Regulation", with many studies using it for research [6–8]. However, few studies have focused on the emission factor and emission estimation of NH_3 (ammonia).

Among the secondary sources of PM, emission estimation and emission sources of NH_3 are important with respect to air pollution management because emission reduction of NH_3 is closely related to the changes in PM2.5 concentration. A previous study analyzed PM2.5 concentration changes in South Korea based on the reduction in air pollutants (NOx, SOx, NH_3 and PM) using an air quality model (CMAQ) and concluded that a reduction in NH_3 emissions leads to a greater reduction in PM2.5 concentration as compared to any other pollutant [9,10]. Accordingly, there has been an increased focus on research related to NH_3 emission sources and emission estimation [11].

South Korea constructs NH_3 emission inventories using various categories, including energy industry combustion, non-industry combustion, manufacturing industry combustion, production process, off-road mobile sources, waste treatment, agriculture, other area sources and biomass combustion. In the case of energy industry combustion, bituminous coal power plants comprise the majority of power plants [12].

The NH_3 emission factor of bituminous coal power plants is difficult to obtain for South Korea because the U.S. Environmental Protection Agency (EPA) value for the year 1994 is used. Therefore, this study aims to analyze the NH_3 emission of bituminous coal power plants in South Korea and conduct research on the emission characteristics, including the development of an emission factor and an analysis of uncertainty. Furthermore, the differences in NH_3 emissions are examined by using the NH_3 emission factor developed for this study, which reflects the characteristics of South Korea, the EPA's value currently applied in South Korea, and an emission factor value developed previously in South Korea.

2. Method

2.1. Selection of Objective Facilities

This study collected NH_3 samples from three bituminous coal power plants to investigate their NH_3 emission characteristics. Table 1 shows the power generation capacity, fuel consumption, and frequency of sampling conducted at the power plants. Sampling was performed at least three times at each power plant, with ten or more samples collected.

Table 1. Characteristics of the investigated bituminous coal power plant.

Site	Capacity (MW)	Fuel Type	Sampling
Power Plant A	1020	Bituminous Coal	16
Power Plant B	1050	Bituminous Coal	10
Power Plant C	500	Bituminous Coal	19

2.2. Analysis of Ammonia at Bituminous Coal Power Plant

This study employed the indophenol method presented in the "Odor Analysis Method" and "Standard Methods for the Measurements of Air Pollution" of South Korea to measure the NH_3 emission concentration of bituminous coal power plants [13]. The indophenol method adds phenol-sodium nitroprusside solution and sodium hypochlorite solution to the sample solution for analysis and measures the absorbance of indophenols, which reacts with ammonium ions, to quantify NH_3. To collect NH_3 samples, an ammonia absorbing solution (50 mL 0.5% boric acid solution) was put into two 50 mL capacity flasks, and a mini pump was used to pump in 80 L of emission gas for 20 min at 4 L/min. A moisture absorption bottle containing silica gel was installed in front of the NH_3 sampling device to remove the moisture in the gas emitted from the power plants. Figure 1 shows a schematic diagram of NH_3 sample collection. Furthermore, a spectrophotometer (Shimadzu 17A, Japan) was used to measure the absorbance of the ammonia absorbing solution at a wavelength of 640 nm.

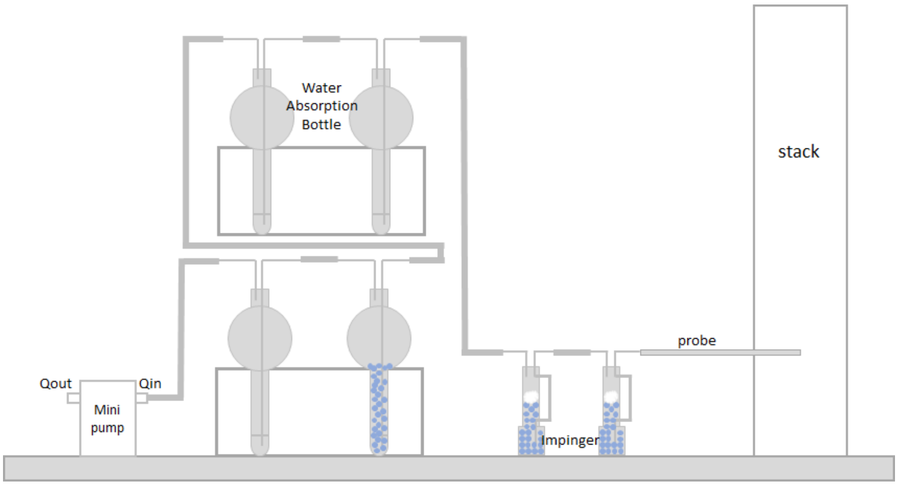

Figure 1. Schematic of the field setup for ammonia sampling at power plant.

2.3. Development of NH_3 Emission Factor

The NH_3 emission factor calculation is shown in Equation (1). CleanSYS data from the three bituminous power plants were used for the flowrate data required in the development of an NH_3 emission factor, and one-day cumulative flowrate data were used for the flowrate. In the case of fuel usage amount, the data were obtained from the power plants.

$$EF_{NH_3} = \left[C_{NH_3} \times \frac{M_w}{V_m} \times Q_{day} \times 10^{-6} \right] / FC_{day} \quad (1)$$

where EF is emission factor (kg NH_3/ton); C_{NH_3} is NH_3 concentration in exhaust gas (ppm); M_w is molecular weight of NH_3 (constant) = 17.031 (g/mol); V_m is one mole ideal gas volume in standardized condition (constant) = 22.4 (10^{-3} m^3/mol); Q_{day} is daily accumulated flow rate (Sm^3/day) (based on dry combustion gas); and FC_{day} is daily fuel consumption (ton/day).

2.4. Uncertainty Analysis by Monte Carlo Simulation

This study used Monte Carlo simulations to estimate the uncertainty of the NH_3 emission factor and performed the analysis in four stages, as shown in Figure 2 [14,15]. First, in the model selection stage, a NH_3 emission factor estimation worksheet was constructed. Second, the probability density functions of input variables needed for the development of the NH_3 emission factor were tested through fitness tests. The level of significance was set to 5% for the hypothesis test, and the probability density functions were calculated through the fitness tests using the data required for NH_3 emission factor development, such as NH_3 emission concentration, emission flowrate, and low calorific value of fuel. Third, when performing the Monte Carlo simulations, random sampling simulations were performed using a "Crystal Ball". Fourth, the uncertainty range of 95% confidence interval was calculated through the simulation results.

"Crystal Ball" constructs the probability density function of the emission factor as the result of each calculation performed through an iterative process using simulation. It also gives a range of 95% confidence intervals (± $Z_{a/2}$) in the generated emission factors. We can estimate the uncertainty through that range.

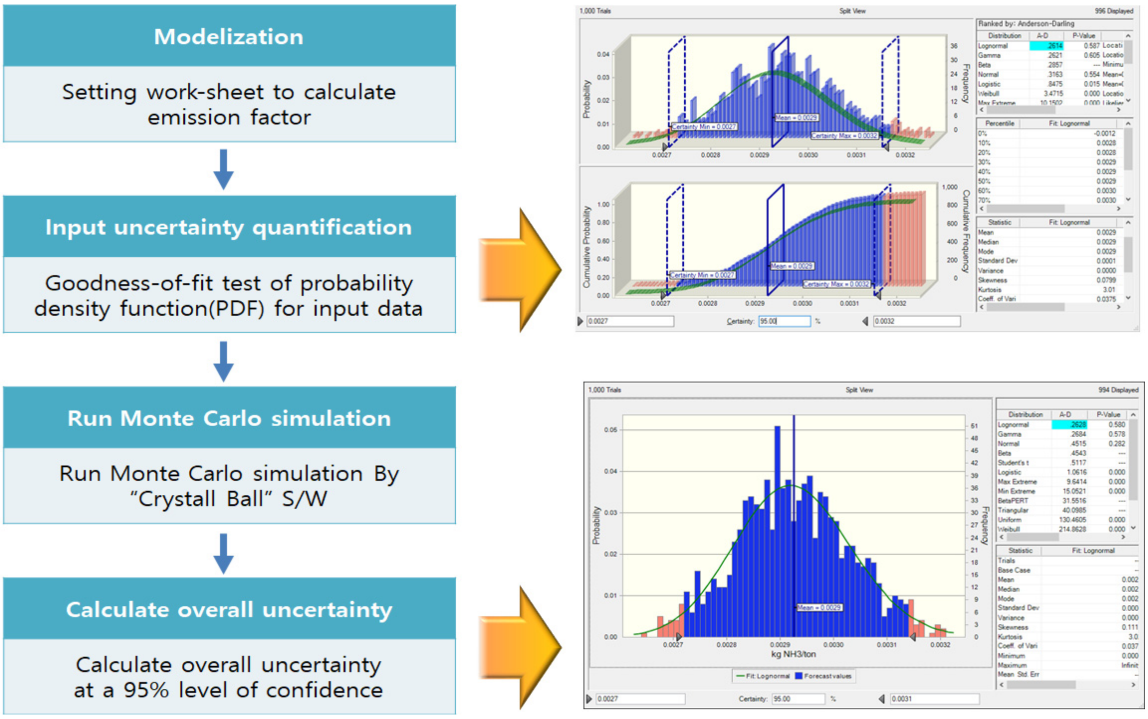

Figure 2. Process of the Monte Carlo Simulation for estimating the uncertainty of the emission factor.

3. Result and Discussion

3.1. Characteristics of NH_3 Emission

Table 2 shows the results of the NH_3 concentration analysis for three bituminous coal power plants. The mean NH_3 concentration of bituminous coal power plant A was 0.21 ppm, with a standard deviation of 0.14 ppm. The NH_3 concentration of bituminous coal power plant C was 0.26 ppm, which showed a similar concentration band as the bituminous coal power plant A. Moreover, it showed a standard deviation of 0.21 ppm, which was higher than that of the bituminous coal power plant A. The NH_3 concentration of bituminous coal power plant B was 0.99 ppm, which was approximately five times higher than that of bituminous coal power plants A and C, and the standard deviation was 0.56 ppm. High NH_3 concentration exhibited by the bituminous coal power plant B was related to NOx concentrations [16]. In the case of coal-fired power plants, NH_3 is injected in the SCR (Selective Catalytic Reduction) in order to reduce NOx concentrations; NH_3 that does not completely react is emitted through the final emission outlet. Therefore, the NH_3 concentration emitted through the final emission outlet will be high in proportion to the amount of NH_3 injected to reduce NOx concentrations. To confirm this, a comparison of the NOx data acquired during the measurement period from the three selected bituminous coal power plants was conducted. The results demonstrated that the NOx concentration of power plant A (20 ppm) and the NOx concentration of Unit No. 6 of power plant C (23 ppm) were higher than that of power plant B (14 ppm). Therefore, it was estimated that the NH_3 concentration of power plant B was high because a large amount of NH_3 was injected to reduce its NOx concentration.

Table 2. NH_3 concentration of the investigated bituminous coal power plants.

Site	NH_3 Concentration (ppm)	SD(Standard Deviation) (ppm)	Sampling	NOx Concentration (ppm)
Power Plant A	0.21	0.14	16	20
Power Plant B	0.99	0.56	10	14
Power Plant C	0.27	0.21	19	23

The correlation of NH_3 concentration and NOx concentration was examined in detail, using the daily average NH_3 concentration and the daily average NOx concentration of the selected power plants, as shown in Figure 3. The analysis revealed that as the NH_3 concentrations decreased, the NOx concentrations increased, thus exhibiting an inversely proportional relationship. Therefore, as the amount of NH_3 was increased for NOx reduction, NH_3 slip increased, leading to high NH_3 emission concentration.

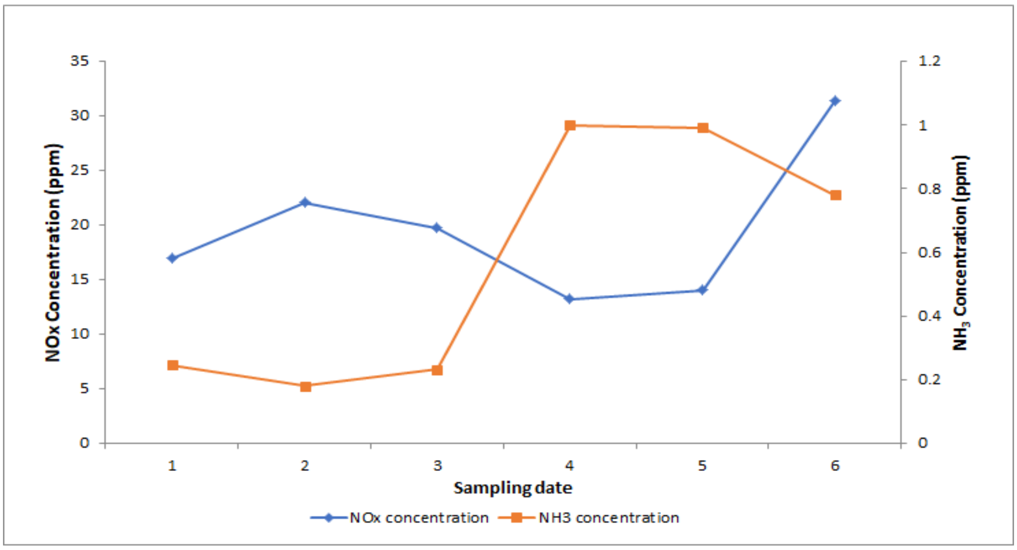

Figure 3. Correlation of NH_3 concentration and NOx concentration.

3.2. *NH_3 Emission Factor and Comparison of NH_3 Emissions*

This study developed the NH_3 emission factor by collecting 45 NH_3 samples from three bituminous coal power plants. The results are shown in Table 3.

The NH_3 emission factor development result was found to be 0.0029 kg NH_3/ton, which is approximately ten times larger than the currently-applied EPA NH_3 emission factor of energy industry combustion that is used in South Korea's national statistics (0.00028 kg NH_3/ton) [17]. This value is about two times lower than the emission factor of 0.0054 kg NH_3/ton for a bituminous coal power plant, which was analyzed in a 2019 South Korean research report [18]. It was also found that this figure is significantly lower than the range of ammonia emission factor (0.07 kg NH_3/ton to 1.17 kg NH_3/ton) for household stoves, which is among the combustion partial ammonia emission factors studied more recently than studies done by the U.S. EPA [19].

Table 3. NH_3 emission factor of the investigated bituminous coal power plant.

This Study (kgNH_3/ton)	US EPA(1994) (kgNH_3/ton)	NIER (2019) (kgNH_3/ton) [18]
0.0029	0.00028	0.0054

Considering these results, the difference in the emission factor obtained from the research conducted in South Korea is lower than the difference in the emission factor of the EPA, which is currently applied in the statistics of South Korea. Therefore, it is important to develop an NH_3 emission factor, which reflects the characteristics of South Korea.

The emission factor that was developed in this study and the EPA's emission factor, which is currently applied in the statistics of South Korea, were used to compare the difference in NH_3 emissions at the selected bituminous coal power plants. Figure 4 shows the results.

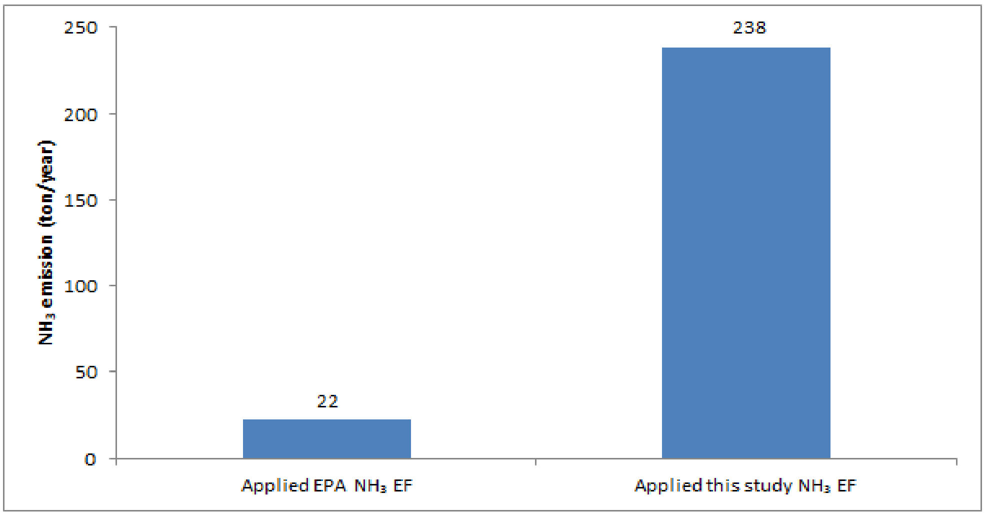

Figure 4. The comparison of NH_3 EF(Emission Factor)for NH_3 emissions at bituminous coal power plants.

When the emission factor developed in this study was applied, the NH_3 emission was calculated to be 228 NH_3 ton/year. When compared to the NH_3 emission estimate (22 NH_3 ton/year), which is calculated by applying the conventional EPA emission factor, the difference was approximately 206 NH_3 ton/year. Therefore, it is necessary to develop a NH_3 emission factor to improve the confidence level of inventory.

3.3. Uncertainty of NH_3 Emission Factor

Monte Carlo simulation was used to estimate the uncertainty of the NH_3 emission factor of bituminous coal power plants selected for this study. Figure 5 shows the estimation results. The probability density function of the NH_3 emission factor of bituminous coal power plants developed in this study had lognormal distribution. The mean was 0.0029 kg NH_3/ton at a 95% confidence level; the lower 2.5% showed 0.0027 kg NH_3/ton and the upper 97.5% showed 0.0031 kg NH_3/ton. The uncertainty range of the NH_3 emission factor was estimated using these values from −6.9% to +10.34% at 95% confidence level. At present, the values and range are not available for NH_3 uncertainty. Therefore, comparison with relevant cases is difficult. However, in the case of greenhouse gas, uncertainty range and values are available.

When the uncertainty of the NH_3 emission factor of bituminous coal power plants from this study is compared with the greenhouse gas uncertainty range provided by the Intergovernmental Panel on Climate Change (IPCC), the NH_3 emission factor is found to be much lower than the basic uncertainty range, that is 50–150% for CH_4 emission factor. Moreover, the uncertainty is 1000% of the uncertainty of the N_2O in the stationary combustion sector of energy provided in the 2006 IPCC guidelines, but is larger than the uncertainty (−1.0 to +1.04) of the carbon emission factor of bituminous coal [20]. In South Korea, the uncertainties in air pollutants are expressed in ranks and evaluated by experts. If the uncertainty range of air pollutants is provided, as in the case of greenhouse gases, it is possible to evaluate them quantitatively.

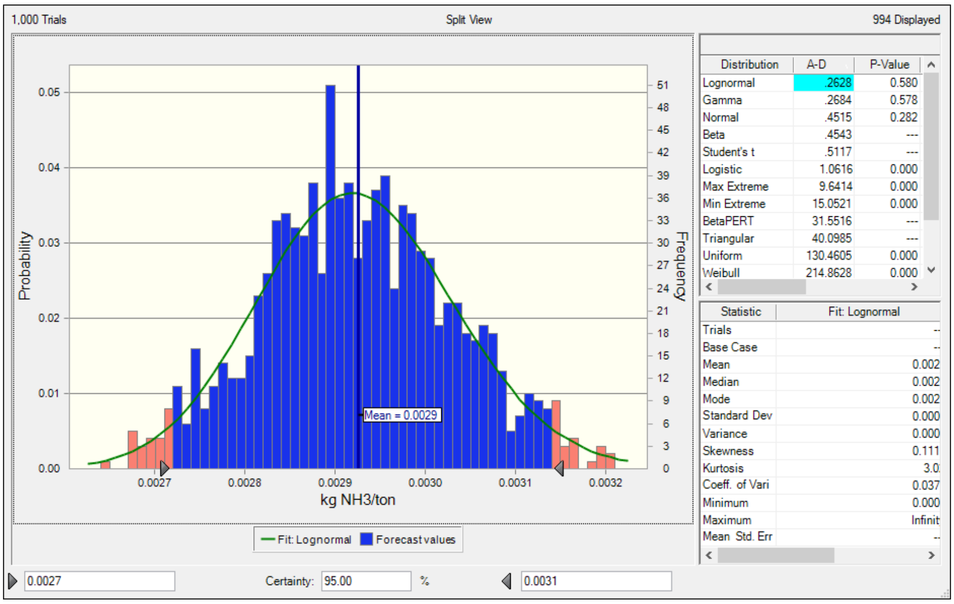

Figure 5. Process of the Monte Carlo Simulation for estimating the uncertainty of the emission factor.

4. Conclusions

This study developed a NH_3 emission factor for bituminous coal power plants in South Korea to investigate the NH_3 emission characteristics. Furthermore, three different emission factors were used to compare the NH_3 emissions, namely the EPA value, which is currently applied in South Korea, a previously developed emission factor value in South Korea, and the emission factor value developed in this study, which reflects the characteristics of South Korea. Three bituminous coal power plants were selected to compare the NH_3 emission characteristics, including NH_3 emission factor development.

The NH_3 concentration analysis results showed that emissions from the selected bituminous coal power plants were in the range of 0.21–0.99 ppm, and that the difference in NH_3 concentration was affected by NOx concentration. The NH_3 emission factor was found to be 0.0029 kg NH_3/ton, which demonstrated that the difference in the values obtained from the research conducted in South Korea was lower than the difference in the emission factor from the EPA, which is currently applied in the statistics of South Korea. Furthermore, when NH_3 emissions were compared by using the NH_3 emission factor developed in this study alongside that of the EPA's NH_3 emission factor that is currently applied in South Korea's statistics, the difference was found to be 206 NH_3 ton/year. This implies that an emission factor needs to be developed which reflects the national characteristics of South Korea.

The uncertainty range of the NH_3 emission factor developed in this study was between −6.9% and +10.34% at a 95% confidence level. At present, numerical values of uncertainty are not available for air pollutants, thus making their comparison difficult. When compared with the uncertainty of greenhouse gas, the NH_3 emission factor's uncertainty was higher than that of the carbon emission factor of bituminous coal and lower than that of the emission factors of CH_4 and N_2O. If the uncertainty ranges are provided for air pollutants, like those of greenhouse gas, quantitative evaluation will be feasible.

This study investigated the NH_3 emission factor and emission characteristics for only three bituminous coal power plants. In the future, if a NH_3 emission factor is developed for a larger number of facilities by considering the seasonal effects, the confidence level of NH_3 inventory in South Korea will significantly improve.

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