

## Article

# Emission Characteristics of Ammonia at Bituminous Coal Power Plant

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**Abstract:** This study developed a NH<sub>3</sub> emission factor for bituminous coal power plants in South Korea in order to investigate the NH<sub>3</sub> emission characteristics. The NH<sub>3</sub> 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<sub>3</sub> concentration was affected by NO<sub>x</sub> concentration. The NH<sub>3</sub> emission factor was found to be 0.0029 kg NH<sub>3</sub>/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<sub>3</sub> emissions were compared by using the NH<sub>3</sub> emission factor developed in this study alongside the EPA's NH<sub>3</sub> emission factor that is currently applied in South Korea's statistics; the difference was found to be 206 NH<sub>3</sub> 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<sub>3</sub> emission factor developed in this study was between −6.9% and +10.34% at a 95% confidence level.

**Keywords:** PM<sub>2.5</sub>; secondary sources; bituminous coal power plant; uncertainty analysis; ammonia emission

## 1. Introduction

In 2016, Ultrafine (≤2.5) Particulate Matter (PM<sub>2.5</sub>) concentration in South Korea was 26 µg/m<sup>3</sup>, which was higher than that of Europe, the United States, and Japan. From 1990 to 2015, the average PM<sub>2.5</sub> concentration in South Korea was 29 µg/m<sup>3</sup>, 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<sup>3</sup>), and Singapore [1].

One of major causes of PM<sub>2.5</sub> is the increase in secondary sources of particulate matters (PM), such as NO<sub>x</sub>, SO<sub>x</sub>, VOCs, and NH<sub>3</sub> [2–5]. In South Korea, NO<sub>x</sub> and SO<sub>x</sub> 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<sub>3</sub> (ammonia).

Among the secondary sources of PM, emission estimation and emission sources of NH<sub>3</sub> are important with respect to air pollution management because emission reduction of NH<sub>3</sub> is closely related to the changes in PM<sub>2.5</sub> concentration. A previous study analyzed PM<sub>2.5</sub> concentration changes in South Korea based on the reduction in air pollutants (NO<sub>x</sub>, SO<sub>x</sub>, NH<sub>3</sub> and PM) using an air quality model (CMAQ) and concluded that a reduction in NH<sub>3</sub> emissions leads to a greater reduction in PM<sub>2.5</sub> concentration as compared to any other pollutant [9,10]. Accordingly, there has been an increased focus on research related to NH<sub>3</sub> emission sources and emission estimation [11].

South Korea constructs NH<sub>3</sub> 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  $\text{NH}_3$  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  $\text{NH}_3$  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  $\text{NH}_3$  emissions are examined by using the  $\text{NH}_3$  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

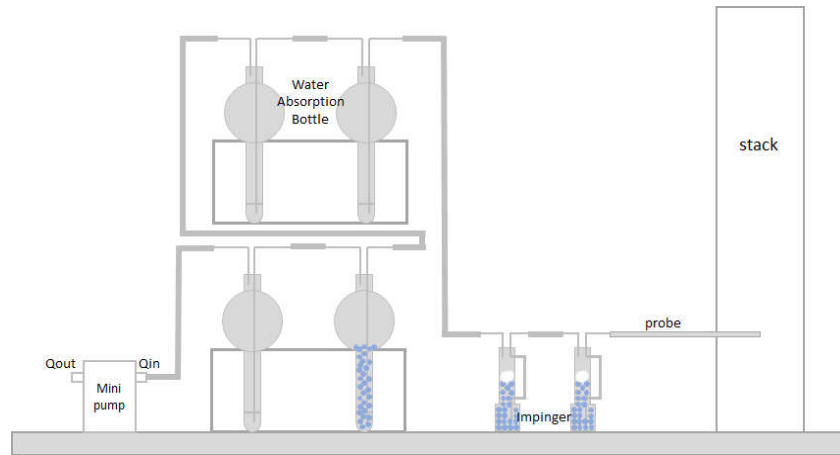
This study collected  $\text{NH}_3$  samples from three bituminous coal power plants to investigate their  $\text{NH}_3$  emission characteristics. Table 1 shows the power generation capacity, fuel consumption, and frequency of sampling conducted at the power plants. Sampling was performed at least three 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  $\text{NH}_3$  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  $\text{NH}_3$ . To collect  $\text{NH}_3$  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  $\text{NH}_3$  sampling device to remove the moisture in the gas emitted from the power plants. Figure 1 shows a schematic diagram of  $\text{NH}_3$  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 $\text{NH}_3$ Emission Factor

The  $\text{NH}_3$  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  $\text{NH}_3$  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.

$$EF_{\text{NH}_3} = \left[ C_{\text{NH}_3} \times \frac{M_w}{V_m} \times Q_{\text{day}} \times 10^{-6} \right] / FC_{\text{day}} \quad (1)$$

where  $EF$  is emission factor (kg  $\text{NH}_3$ /ton);  $C_{\text{NH}_3}$  is  $\text{NH}_3$  concentration in exhaust gas (ppm);  $M_w$  is molecular weight of  $\text{NH}_3$  (constant) = 17.031 (g/mol);  $V_m$  is one mole ideal gas volume in standardized condition (constant) = 22.4 ( $10^{-3}$  m<sup>3</sup>/mol);  $Q_{\text{day}}$  is daily accumulated flow rate (Sm<sup>3</sup>/day) (based on dry combustion gas); and  $FC_{\text{day}}$  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  $\text{NH}_3$  emission factor and performed the analysis in four stages, as shown in Figure 2 [14,15]. First, in the model selection stage, a  $\text{NH}_3$  emission factor estimation worksheet was constructed. Second, the probability density functions of input variables needed for the development of the  $\text{NH}_3$  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  $\text{NH}_3$  emission factor development, such as  $\text{NH}_3$  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 ( $\pm Z_{\alpha/2}$ ) in the generated emission factors. We can estimate the uncertainty through that range.

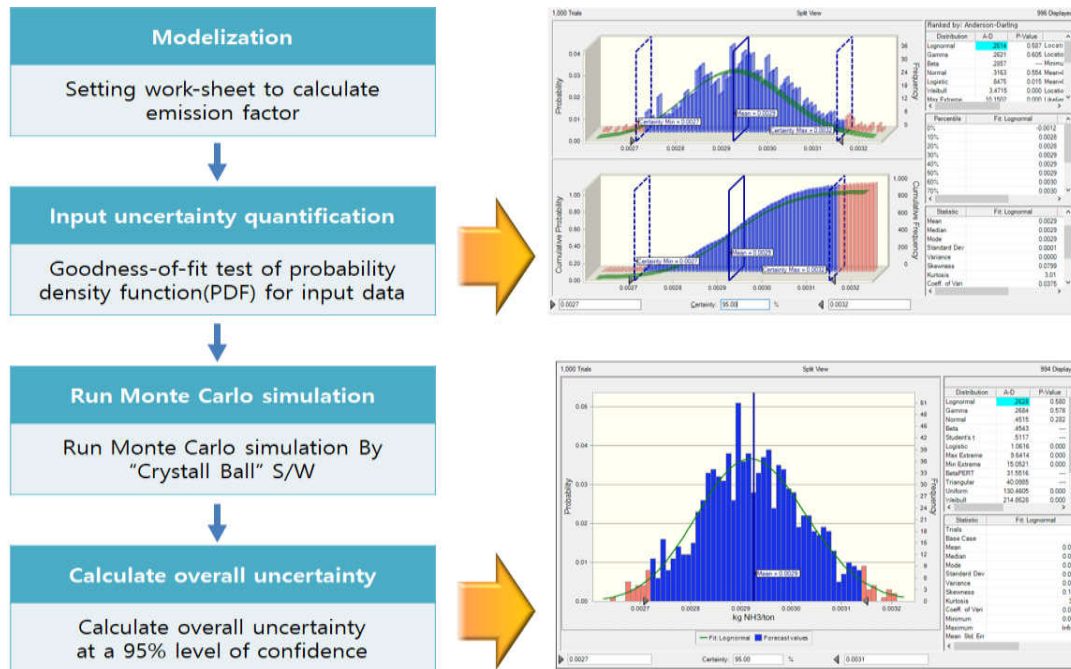


Figure 2. Process of the Monte Carlo Simulation for estimating the uncertainty of the emission factor.

### 3. Result and Discussion

#### 3.1. Characteristics of $\text{NH}_3$ Emission

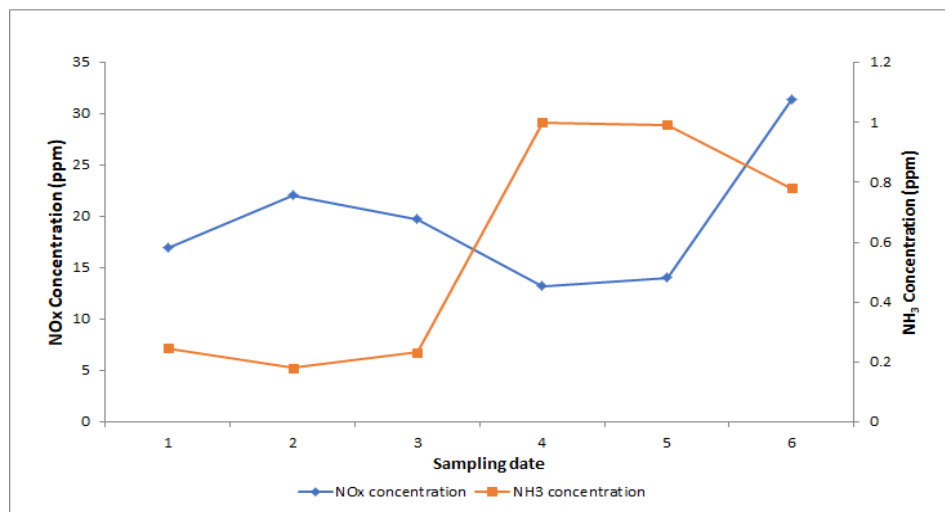
Table 2 shows the results of the  $\text{NH}_3$  concentration analysis for three bituminous coal power plants. The mean  $\text{NH}_3$  concentration of bituminous coal power plant A was 0.21 ppm, with a standard deviation of 0.14 ppm. The  $\text{NH}_3$  concentration of bituminous coal power plant 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  $\text{NH}_3$  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  $\text{NH}_3$  concentration exhibited by the bituminous coal power plant B was related to  $\text{NO}_x$  concentrations [16]. In the case of coal-fired power plants,  $\text{NH}_3$  is injected in the SCR (Selective Catalytic Reduction) in order to reduce  $\text{NO}_x$  concentrations;  $\text{NH}_3$  that does not completely react is emitted through the final emission outlet. Therefore, the  $\text{NH}_3$  concentration emitted through the final emission outlet will be high in proportion to the amount of  $\text{NH}_3$  injected to reduce  $\text{NO}_x$  concentrations. To confirm this, a comparison of the  $\text{NO}_x$  data acquired during the measurement period from the three selected bituminous coal power plants was conducted. The results demonstrated that the  $\text{NO}_x$  concentration of power plant A (20 ppm) and the  $\text{NO}_x$  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  $\text{NH}_3$  concentration of power plant B was high because a large amount of  $\text{NH}_3$  was injected to reduce its  $\text{NO}_x$  concentration.

Table 2.  $\text{NH}_3$  concentration of the investigated bituminous coal power plants.

| Site          | $\text{NH}_3$ Concentration (ppm) | SD(Standard Deviation) (ppm) | Sampling | $\text{NO}_x$ 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  $\text{NH}_3$  concentration and  $\text{NO}_x$  concentration was examined in detail, using the daily average  $\text{NH}_3$  concentration and the daily average  $\text{NO}_x$  concentration of the selected power plants, as shown in Figure 3. The analysis revealed that as the  $\text{NH}_3$  concentrations decreased, the  $\text{NO}_x$  concentrations increased, thus exhibiting an inversely proportional relationship. Therefore, as the amount of  $\text{NH}_3$  was increased for  $\text{NO}_x$  reduction,  $\text{NH}_3$  slip increased, leading to high  $\text{NH}_3$  emission concentration.



**Figure 3.** Correlation of  $\text{NH}_3$  concentration and  $\text{NO}_x$  concentration.

### 3.2. $\text{NH}_3$ Emission Factor and Comparison of $\text{NH}_3$ Emissions

This study developed the  $\text{NH}_3$  emission factor by collecting 45  $\text{NH}_3$  samples from three bituminous coal power plants. The results are shown in Table 3.

The  $\text{NH}_3$  emission factor development result was found to be 0.0029 kg  $\text{NH}_3$ /ton, which is approximately ten times larger than the currently-applied EPA  $\text{NH}_3$  emission factor of energy industry combustion that is used in South Korea's national statistics (0.00028 kg  $\text{NH}_3$ /ton) [17]. This value is about two times lower than the emission factor of 0.0054 kg  $\text{NH}_3$ /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  $\text{NH}_3$ /ton to 1.17 kg  $\text{NH}_3$ /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].

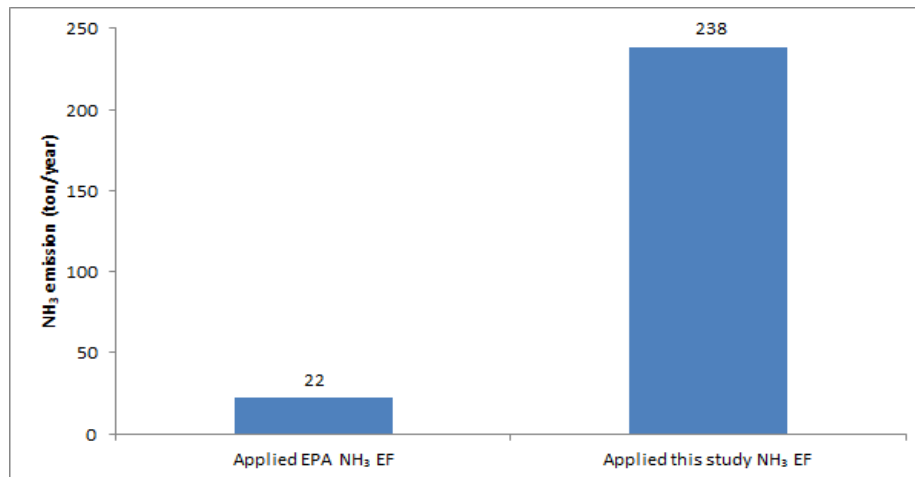
Considering these results, the difference in the emission factor obtained from the research conducted in South Korea is lower than the difference in the emission factor of the EPA, which is currently applied in the statistics of South Korea. Therefore, it is important to develop an  $\text{NH}_3$  emission factor, which reflects the characteristics of South Korea.

**Table 3.**  $\text{NH}_3$  emission factor of the investigated bituminous coal power plant.

| This Study<br>(kg $\text{NH}_3$ /ton) | US EPA(1994)<br>(kg $\text{NH}_3$ /ton) | NIER (2019)<br>(kg $\text{NH}_3$ /ton)[18] |
|---------------------------------------|-----------------------------------------|--------------------------------------------|
| 0.0029                                | 0.00028                                 | 0.0054                                     |

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  $\text{NH}_3$  emissions at the selected bituminous coal power plants. Figure 4 shows the results.

When the emission factor developed in this study was applied, the  $\text{NH}_3$  emission was calculated to be 228  $\text{NH}_3$  ton/year. When compared to the  $\text{NH}_3$  emission estimate (22  $\text{NH}_3$  ton/year), which is calculated by applying the conventional EPA emission factor, the difference was approximately 206  $\text{NH}_3$  ton/year. Therefore, it is necessary to develop a  $\text{NH}_3$  emission factor to improve the confidence level of inventory.



**Figure 4.** The comparison of  $\text{NH}_3$  EF(Emission Factor)for  $\text{NH}_3$  emissions at bituminous coal power plants.

### 3.3. Uncertainty of $\text{NH}_3$ Emission Factor

Monte Carlo simulation was used to estimate the uncertainty of the  $\text{NH}_3$  emission factor of bituminous coal power plants selected for this study. Figure 5 shows the estimation results. The probability density function of the  $\text{NH}_3$  emission factor of bituminous coal power plants developed in this study had lognormal distribution. The mean was 0.0029 kg  $\text{NH}_3$ /ton at a 95% confidence level; the lower 2.5% showed 0.0027 kg  $\text{NH}_3$ /ton and the upper 97.5% showed 0.0031 kg  $\text{NH}_3$ /ton. The uncertainty range of the  $\text{NH}_3$  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  $\text{NH}_3$  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  $\text{NH}_3$  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  $\text{NH}_3$  emission factor is found to be much lower than the basic uncertainty range, that is 50–150% for  $\text{CH}_4$  emission factor. Moreover, the uncertainty is 1000% of the uncertainty of the  $\text{N}_2\text{O}$  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.

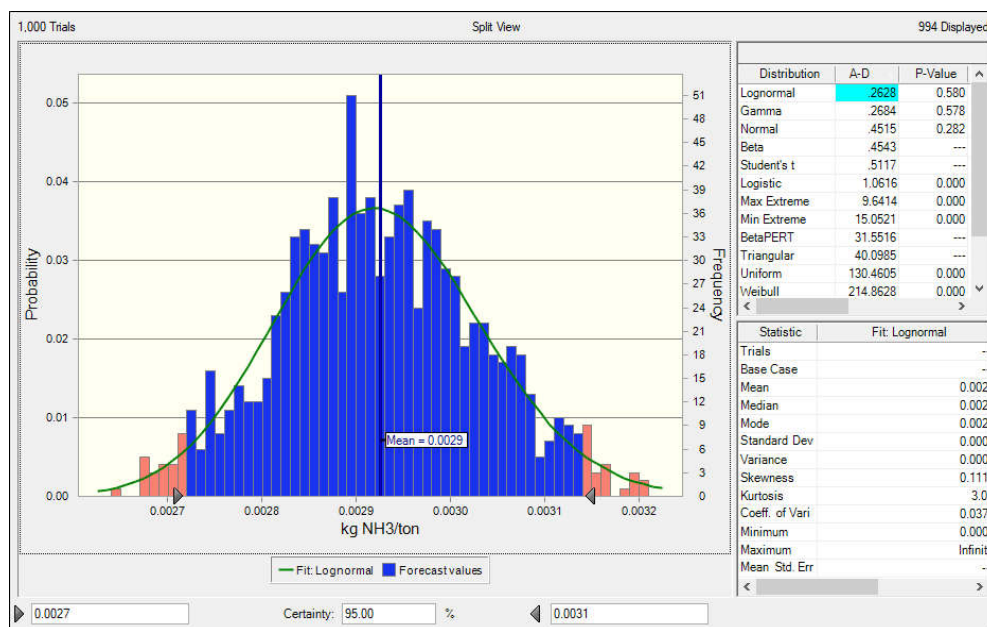


Figure 5. Process of the Monte Carlo Simulation for estimating the uncertainty of the emission factor.

#### 4. Conclusions

This study developed a NH<sub>3</sub> emission factor for bituminous coal power plants in South Korea to investigate the NH<sub>3</sub> emission characteristics. Furthermore, three different emission factors were used to compare the NH<sub>3</sub> 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<sub>3</sub> emission characteristics, including NH<sub>3</sub> emission factor development.

The NH<sub>3</sub> 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<sub>3</sub> concentration was affected by NO<sub>x</sub> concentration. The NH<sub>3</sub> emission factor was found to be 0.0029 kg NH<sub>3</sub>/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<sub>3</sub> emissions were compared by using the NH<sub>3</sub> emission factor developed in this study alongside that of the EPA's NH<sub>3</sub> emission factor that is currently applied in South Korea's statistics, the difference was found to be 206 NH<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> 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<sub>4</sub> and N<sub>2</sub>O. If the uncertainty ranges are provided for air pollutants, like those of greenhouse gas, quantitative evaluation will be feasible.

This study investigated the NH<sub>3</sub> emission factor and emission characteristics for only three bituminous coal power plants. In the future, if a NH<sub>3</sub> emission factor is developed for a larger number of facilities by considering the seasonal effects, the confidence level of NH<sub>3</sub> inventory in South Korea will significantly improve.

**Author Contributions:** All authors contributed to the research presented in this work. Their contributions are presented below. “Conceptualization, E.-c.J.; Methodology and writing- original draft preparation, S.K. and Analysis, S.D.; All authors have read and agreed to the published version of the manuscript.

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