Numerical model for estimating greenhouse gas emissions from pulp and paper industrial wastewater treatment systems in Vietnam

Mô hình số tính toán phát thải khí nhà kính từ hệ thống xử lý nước thải sản xuất giấy tại Việt Nam

Research article

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At present, it is difficult and costly to measure directly greenhouse gas (GHG) emissions from the wastewater treatment system. Application of model will reduce measurement cost and quickly obtain the forecast data set of GHG emissions. This study developed a mathematical model for both steady and dynamic states to calculate GHG (CO₂, CH₄, and N₂O) emissions from wastewater treatment systems for industrial paper processing. These models are constructed based on mass balance equations of species, including substrate balance equations, biomass balance equations for reactors of treatment systems, stoichiometric coefficients of species in biochemical reactions and biological processes. The obtained equations were solved based on algorithm of Runge-Kutta and the model was programmed by MATLAB. Results of applying the model to calculate GHG emissions from the paper industrial wastewater treatment system at Bai Bang and Tan Mai plants are as follows: total GHG emissions and emission factor are 3,070.3 kgCO₂-eq/day, 0.38 kgCO₂-eq/m³, respectively for Bai Bang plant (8,000 m³/day) and 7,413.6 kgCO₂-eq/day, 0.74 kgCO₂-eq/m³, respectively for Tan Mai plant (10,000 m³/day). The research evaluated a number of influencing factors, such as temperature, flow rate of influent, and substrate concentrations, to GHG emissions at the Tan Mai paper plant.

Keywords: emissions, greenhouse gas (GHG), pulp and paper wastewater treatment system

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1. Introduction

Climate change robustly affects the earth environment and human’s life. Climate change causes many problems, such as: exhausted natural resources, harsh climate, abnormal weather, gradually increasing warm earth, gradually destroyed ecosystem. The main cause makes the earth climate change is that increase greenhouse gases (GHG) emissions.

The pulp and paper industry is one of the industries demanding great amount of freshwater and generating consequently huge volume of wastewater [1,8]. The amount of GHG emissions in wastewater treatment depends on the amount of waste water and corresponding chemical oxygen demand (COD) content. Paper and pulp production generates large amounts of wastewater containing high organic content with COD up to 2.94 kg/m$^3$ [10]. The wastewater treatment systems of the paper and pulp industry is also one of the significant sources of GHG emissions in the waste treatment block including indirect emissions from electricity generation for use in the treatment process, production and transportation of fuels and materials, burying of activated sludge generated during the process and directly emissions from the treatment process, such as aerobic treatment tank, anaerobic treatment tank [1,5,10].

Reduction of GHG emissions plays an important role in responding to climate change. A mathematical model for the estimation of GHG emissions will help to reduce the cost of measurement and quickly obtain a set of GHG emission forecast data. It is also a useful tool to estimate the sustainability of a wastewater treatment system [1,2,3,5]. Therefore, the establishment of a reliable model for calculating GHG emissions from wastewater treatment system is necessary.

This study developed a mathematical model for both steady and dynamic states to calculate GHG (CO$_2$, CH$_4$, and N$_2$O) emissions from pulp and paper industrial wastewater treatment systems. These models are constructed using mass balance equations of species, including substrate balance equations, biomass balance equations for reactors of treatment systems, stoichiometric coefficients of species in biochemical reactions and biological processes [4,7,8]. The obtained equations were solved by applying algorithm of Runge-Kutta and coded in MATLAB.

2. Material and methods

2.1. Research subject

Bai Bang paper plant with capacity of 8,000 m$^3$/d uses anaerobic biological treatment technology and anaerobic sludge digestion to treat wastewater. Tan Mai paper plant with capacity of 10,000 m$^3$/d uses anaerobic-aerobic biological treatment technology and anaerobic sludge digestion to treat wastewater.

2.2. Establishing model for calculating GHG emissions from paper and pulp wastewater treatment systems

The model for calculating GHG emissions established for waste paper and pulp wastewater treatment systems is shown in Figure 1.

![Figure 1. Flow diagram of the pulp and paper wastewater treatment systems](image)

The symbols in the calculation formulas of the model are shown in Table 1.

| Symbol | Definition | Unit |
|--------|------------|------|
| BOD and SS | Biochemical oxygen demand and suspended solid | g/d |
| BOD$_{S,0}$, SS$_{S,0}$ | The amount of BOD and SS is reduced in the primary clarifier | - |
| CO$_{2}$eq | Equivalent carbon dioxide | (kg VSS/kg VSS) |
| $f_d$ | Fraction of biomass as a cell debris | - |
2.2.1. Establishing model for the steady state

a. Calculating in the anaerobic treatment system

The removal of BOD, SS in the primary clarifier can be calculated as follows:

\[ \text{BOD}_{\text{CM}}^\text{LAM}=\text{BOD}_{\text{CM}}\times Q_{\text{inf}}\times S_{\text{inf}} \]
\[ \text{SS}_{\text{CM}}^\text{LAM}=\text{SS}_{\text{CM}}\times Q_{\text{inf}}\times S_{\text{inf}} \]

The formula for the anaerobic digestion is as follows:

\[ \text{V}_{\text{in}}\frac{\text{d}S_{\text{an}}}{\text{dt}}=\frac{Q_{\text{in}}S_{\text{an}}}{S_{\text{inf}}} - (Q_{\text{w}}S_{\text{an}}^\text{w}) + \frac{Q_{\text{in}}}{S_{\text{inf}}} \times \frac{Q_{\text{w}}}{S_{\text{w}}} \times \frac{S_{\text{w}}}{S_{\text{inf}}} \]
\[ \text{V}_{\text{in}}\frac{\text{d}X_{\text{an}}}{\text{dt}}=\frac{Q_{\text{in}}X_{\text{an}}}{X_{\text{inf}}} - (Q_{\text{w}}X_{\text{an}}^\text{w}) + \frac{Q_{\text{in}}}{X_{\text{inf}}} \times \frac{Q_{\text{w}}}{X_{\text{w}}} \times \frac{X_{\text{w}}}{X_{\text{inf}}} \]

Where: \( r_S = \frac{k_{\text{in}}}{k_{\text{in}}+S_{\text{an}}} \times S_{\text{an}} \)
\( r_X = \frac{k_{\text{in}}}{k_{\text{in}}+X_{\text{an}}} \times X_{\text{an}} \)

In the steady-state condition: \( \frac{\text{d}S_{\text{an}}}{\text{dt}} = 0; \frac{\text{d}X_{\text{an}}}{\text{dt}} = 0 \) and using formula:

\[ \frac{1}{\text{HRT}^\text{an}}=\frac{\text{V}_{\text{in}}\text{an}}{\text{V}_{\text{in}}\text{an}} \times \frac{S_{\text{in}}}{S_{\text{inf}}} - k_{\text{d}} \times k_{\text{d}} \times \frac{S_{\text{in}}}{S_{\text{inf}}} \]

The substrate and biomass concentrations inside the anaerobic bioreactor were obtained as:

\[ S_{\text{an}}=\frac{S_{\text{in}}}{S_{\text{inf}}} - \frac{S_{\text{in}}}{S_{\text{inf}}} \times \frac{\text{BOD}_{\text{CM}}\times Q_{\text{inf}}}{Q_{\text{in}}} \]

\[ X_{\text{an}}=\frac{X_{\text{in}}}{X_{\text{inf}}} \times \frac{\text{SS}_{\text{CM}}\times Q_{\text{inf}}}{Q_{\text{in}}} \]

The total mass of solid from the anaerobic bioreactor could be calculated:

\[ \text{P}_{\text{an}}=\frac{Q_{\text{in}}\times\text{Y}_{\text{an}}\times(S_{\text{in}}-S_{\text{an}})}{1+k_{\text{d}}\times\text{SRT}^\text{an}} \]

The oxygen consumption which represents the oxidized amount of BOD can be calculated by the following procedure:

\[ \text{Oxygen consumption (BOD)} = \text{BOD removed from wastewater - BOD in waste sludge} \]

\[ \text{BOD}_{\text{re}}^\text{waste} = \frac{Q_{\text{in}}}{S_{\text{in}}}(S_{\text{in}} - S_{\text{an}}) \times 1.42 \times \text{SS}_{\text{bio}} \]

The amount of decayed biomass:

\[ \text{VSS}_{\text{decay}}^\text{an} = 0.8 \times S_{\text{in}} \times \text{SRT}^\text{an} \times [\frac{S_{\text{in}}-S_{\text{an}}}{1+k_{\text{d}}\times\text{SRT}^\text{an}}] \]

The amount of CO₂ and CH₄ production by anaerobic bioreactor could be obtained as follows:

\[ \text{CO}_2^\text{an} = Y_{\text{CO}_2} \times \text{BOD}_{\text{re}} + Y_{\text{CO}_2,\text{decay}} \times \text{VSS}_{\text{decay}} \]

\[ \text{CH}_4^\text{an} = Y_{\text{CH}_4} \times \text{BOD}_{\text{re}} + Y_{\text{CH}_4,\text{decay}} \times \text{VSS}_{\text{decay}} \]

The total amount of GHG by anaerobic bioreactor can be calculated as follows:

\[ \Sigma \text{CO}_2^\text{an} + \text{CO}_2^\text{burn} \text{GHG} + 23 \times (\text{CH}_4^\text{an} \times \text{CH}_4,\text{dissolve} + \text{CH}_4,\text{leak}) \]

b. Calculating in the aerobic treatment system:

The substrate and biomass concentration inside the aerobic bioreactor were obtained as:

\[ S = \frac{k_{\text{an}}}{k_{\text{an}}+S_{\text{RT}X}} \times \frac{X_{\text{in}}}{X_{\text{inf}}} \]

\[ S_{\text{an}}=\frac{S_{\text{in}}}{S_{\text{inf}}} - \frac{S_{\text{in}}}{S_{\text{inf}}} \times \frac{S_{\text{in}}}{S_{\text{inf}}} \times \frac{\text{BOD}_{\text{CM}}\times Q_{\text{inf}}}{Q_{\text{in}}} \]

Total suspended solids in the system:

\[ X_{\text{TSS}}=X+X_{\text{nh}} \]

\[ X_{\text{nh}}=\frac{Q_{\text{in}}}{S_{\text{inf}}} \times \frac{Q_{\text{inf}}}{Q_{\text{in}}} \times \frac{X_{\text{nh}}}{X_{\text{inf}}} \times \frac{S_{\text{inf}}}{S_{\text{in}}} \]
\[
X_{\text{SST}} = \text{VSS} \times (1 - \frac{b_{\text{COD}}}{c_{\text{COD}}}) \quad \text{(2.21)}
\]

Total solid is generated during reaction:
\[
P_{\text{SS,bio}} = \frac{X_{\text{V}}}{\text{SRT}} + f_d \times k_d \times X \times V \times \frac{X_{\text{SST}}}{\text{SRT}} \quad \text{(2.22)}
\]

The amount of oxygen is consumed in the aerobic bioreactor:
\[
\text{BOD}_o = Q_i (S^a - S) - r_{\text{oxy,decay}} (P_{\text{SS}} - Q^a \times X_{\text{SST}}) \quad \text{(2.23)}
\]

The amount of carbon dioxide due to BOD removal can be obtained:
\[
\text{CO}_2 \times \text{BOD}_o = Y_{\text{CO}_2} \times \text{BOD}_o \quad \text{(2.24)}
\]

The amount of decayed biomass:
\[
\text{VSS}_{\text{decay}} = 0.85 \times V \times k_d \times X \quad \text{(2.25)}
\]

The amount of CO₂ emission from biomass decay:
\[
\text{CO}_2 \times \text{VSS}_{\text{decay}} = Y_{\text{CO}_2} \times \text{VSS}_{\text{decay}} \quad \text{(2.26)}
\]

\[
\text{BOD}_{\text{ox,decay}} = r_{\text{oxy,decay}} \times \text{VSS}_{\text{decay}} \quad \text{(2.27)}
\]

The total amount of needed oxygen is equal to the amount of oxygen consumed for BOD oxidation and endogenous degradation:
\[
\text{BOD}_{\text{ox,total}} = \text{BOD}_o + \text{BOD}_{\text{ox,decay}} \quad \text{(2.28)}
\]

The total amount of CO₂ by aerobic bioreactor can be calculated as follows:
\[
\text{CO}_2 \text{ aerobic tank} = \text{CO}_2 \times \text{BOD}_o + \text{CO}_2 \times \text{VSS}_{\text{decay}} \quad \text{(2.29)}
\]

c. Calculating in the anaerobic digester:

The total sludge that enters the anaerobic digester is estimated from the sources identified in Figure 1 as follows:
\[
p_{\text{SS}} = \text{SS}_{\text{SST}} + P_{\text{SS}} + P_{\text{SS,bio}} \quad \text{(2.30)}
\]

Total mass production in the anaerobic digester can be calculated as follows:
\[
p_{\text{SS,bio}} = p_{\text{SS}} \times \text{BOD}_{\text{SST}} + p_{\text{SS}} \times \text{BOD}_{\text{SST}} \quad \text{(2.31)}
\]

\[
p_{\text{SS,bio}} = \frac{p_{\text{SS}} \times \text{BOD}_{\text{SST}} + p_{\text{SS}} \times \text{BOD}_{\text{SST}}}{1 + p_{\text{SS}} \times \text{SRT}_{\text{ph}} / \text{SRT}_{\text{ph}}} \quad \text{(2.32)}
\]

\[
p_{\text{SS,ST}} = f_d \times k_d \times \text{SRT}_{\text{ph}} \times \frac{Q^p \times \text{VSS}_{\text{ph}} - \text{SS}_{\text{ph}}}{1 + k_d \times \text{SRT}_{\text{ph}}} \quad \text{(2.33)}
\]

Therefore, total CO₂ and CH₄ production by anaerobic digester could be obtained as follows:
\[
\text{CO}_2 = Y_{\text{CO}_2} \times \text{BOD}_{\text{SST}} + Y_{\text{CO}_2} \times \text{VSS}_{\text{ph}} \quad \text{(2.34)}
\]

\[
\text{CH}_4 = Y_{\text{CH}_4} \times \text{BOD}_{\text{SST}} + Y_{\text{CH}_4} \times \text{VSS}_{\text{ph}} \quad \text{(2.35)}
\]

2.2.2 Establishing model for the dynamic condition

In the dynamic condition, the amount of accumulation changes over time, that is, \( \frac{dx}{dt} \) and \( \frac{dy}{dt} \) is not equal to zero. With the assuming that \( X_{\text{in}} = 0, X_e = 0 \), we have:
\[
\frac{dx}{dt} = \frac{X_{\text{in}}}{\text{HRT}} - \frac{s}{\text{HRT}} + \frac{s^a}{\text{HRT} \times \text{tan}} + \frac{\text{san}}{\text{HRT} \times \text{tan}} \quad \text{(2.36)}
\]

\[
\frac{dV}{dt} = \frac{s}{\text{HRT}} - \frac{\text{VSS}}{\text{HRT} + c_{\text{S}} + s_{\text{a}}} \quad \text{(2.37)}
\]

\[
\frac{ds}{dt} = \frac{s}{\text{HRT}} - \frac{\text{VSS}}{\text{HRT} + c_{\text{S}} + s_{\text{a}}} \quad \text{(2.38)}
\]

The present study used the Runge-Kutta fourth order method to solve Equations 2.36 to 2.37 using MATLAB. Applying the RungeKutta method solves the mass balance equations in the state condition, which could determine the values of \( S^a, X^a \), and \( X \) at each point in the survey range, using the equations of the emission computation in the stable steady model to calculate the amount of the GHG emission at that time.

2.3 Input parameters for the model

Numerical model in steady state was tested by using measured data of pulp and paper wastewater treatment plant in Montreal, Quebec, Canada. Results of numerical simulation of model were compared with data from the published research [1] as shown in figure 2.

\[
\frac{dX}{dt} = \frac{Q_{\text{in}} X_{\text{SST}}}{V} - \left( \frac{Y_{\text{K}_S + S} X_{\text{in}}}{K_S + S} \times d \right) \quad \text{(2.39)}
\]
The wastewater treatment system of Bai Bang paper plant uses anaerobic biological treatment technology including some tanks: primary sedimentation, UASB, secondary sedimentation, and anaerobic digestion. The wastewater treatment system of Tan Mai paper plant uses anaerobic biodegradation technology combined with activated sludge technology including some tanks: agglutina, primary sedimentation, UASB, aerotank, secondary sedimentation and anaerobic digestion. Data on influent and Kinetic parameters of two plants for the model are shown in Table 2 and 3.

The kinetic parameters were selected from calculations in research of aerobic aeration tank and UASB reactor, and then these parameters were calibrated according to the BOD effluent of model and BOD effluent of Bai Bang and Tan Mai wastewater treatment systems with absolutely error of 4.68% and 11.11%, respectively.

### 3. Results and discussion

#### 3.1 Results in steady state

Results of calculating GHG emissions from Bai Bang and Tan Mai pulp and paper wastewater treatment plants are shown in Figure 3 and Table 4.

![Figure 3. The GHG emissions at Bai Bang and Tan Mai paper plants](image)

**Figure 3. The GHG emissions at Bai Bang and Tan Mai paper plants**

| Parameter | Bai Bang paper plant | Tan Mai paper plant |
|-----------|----------------------|---------------------|
| $f_d$     | 0.1                  | -                   |
| $k$       | 2                    | -                   |
| $k_d$     | 0.2                  | -                   |
| $K_s$     | 60                   | -                   |
| $Y$       | 0.6                  | -                   |
| $K_{an}$  | 3.15                 | 3.5                 |
| $k_{an}$  | 0.029                | 0.03                |
| $K_{an}$  | 460                  | 379.473             |
| $Y_{an}$  | 0.08                 | 0.08                |

The GHG emissions depend not only on the capacity of the treatment plant, but also on the applied treatment technology, and the concentration of organic inputs. With the combined technology of aerobic and anaerobic treatment, the GHG emission from Tan Mai paper is 0.74 kgCO$_2$eq/m$^3$ influent, nearly two times higher than that from Bai Bang plant.

![Figure 4. The GHG emissions from degisters in Bai Bang and Tan Mai paper plant](image)

**Figure 4. The GHG emissions from degisters in Bai Bang and Tan Mai paper plant**

The amount of GHG emission from the sludge degister at Tan Mai paper plant is nearly 10 times higher than the GHG emission from the Bai Bang paper plant. The solid content in the input wastewater of the Tan Mai paper plant is much higher than that of the Bai Bang paper plant (3,022mg/l compared to 400mg/l).

#### 3.3. Some factors affecting the greenhouse gas emissions

The assessment some of factors affecting GHG emissions was made at the Tan Mai plant. Results are as follows:

a. According to solid retention time (SRT) in the anaerobic bioreactor:

Considering the change in GHG emissions by SRT varies from 20 to 35 days in anaerobic bioreactor, the results are shown in Figure 5.

![Figure 5. Total GHG emissions in wastewater treatment system and treatment efficiency in anaerobic tank](image)

**Figure 5. Total GHG emissions in wastewater treatment system and treatment efficiency in anaerobic tank**

Increasing SRT results in a gradual increasing efficiency of the process, but also increasing GHG emissions. Initially, SRT increased from 20 to 23 days, GHG emissions from anaerobic tanks increased by 317,1 kg CO$_2$eq/d; SRT increased from 29 to 32 days, GHG
emissions from anaerobic tanks slowly increased by 134.9 kg CO$_2$eq/d. Therefore, to ensure both treatment efficiency and reduction of GHG emission, the optimal SRT of 30 days was chosen.

b. According to solid retention time (SRT) in the aerobic bioreactor:

Considering the change in GHG emissions by SRT varies from 4 to 14 days in aerobic bioreactor, the results are shown Figure 6.

![Figure 6. Total GHG emissions in wastewater treatment system and treatment efficiency in aerobic tank](image)

The amount of GHG emissions in the aerobic treatment tank increases as the solid retention time in the aerobic tank increases and vice versa. As SRT changed from 4 to 14 days, the GHG emissions from aerobic tanks increased from 675.28 to 1,236 kgCO$_2$eq/d, and the processing efficiency increased by 12.21%. As the SRT is higher, the GHG emissions increase more slowly, the efficiency increases slowly, combined with the chart shows that the value of SRT = 10 days is the optimal value to ensure the efficiency of wastewater treatment and reduction GHG emissions.

c. According to the temperature in the anaerobic bioreactor:

Considering the change in GHG emissions by temperature varies from 28 to 40°C in anaerobic bioreactor, the results are shown Figure 7.

![Figure 7. Effect of temperature on GHG emissions](image)

As temperatures increased from 28°C to 40°C, GHG emissions of the whole system increased negligibly from 29,386 to 30,107 kgCO$_2$eq/d, but the treatment efficiency increased robustly from 68.7 to 98.16%. As the temperature of the anaerobic treatment tank increased, processing efficiency increased with more sludge generated resulting in increased GHG emissions of the whole system.

At temperatures higher than 34°C, the amount of biogas and the treatment efficiency increase negligibly, so the temperature range of 31-34°C is the best for both anaerobic wastewater treatment and biogas recovery process.

3.4. Investigating the greenhouse gas emissions in dynamic condition

Firstly, the model will be run with the stable input parameters for the system until it reaches the equilibrium state. Initial conditions of the system are given in Tables 2 and 3. The BOD contents of the anaerobic and aerobic tank at the time of initiation were assumed to be $S_{an}=160$ mg/l and $S=30$ mg/l.

* The results of the model prediction of the effects of temperature, flow rate and loading of influent organic matter on the GHG emissions from wastewater treatment systems are shown in Figures 8 to 13.

* The effect of temperature changes on GHG emissions

![Figure 8. CO$_2$ variability in aerobic tanks according to temperature](image)

![Figure 9. CO$_2$ variability in anaerobic tanks according to time](image)

![Figure 10. CO$_2$ and CH$_4$ variability in anaerobic digester according to time](image)

The predicted results showed that the temperature has a significant effect on GHG emissions from wastewater treatment in aerobic bioreactors. When the temperature increases, GHG emissions also increase and vice versa. At the same time as the temperature rises, the BOD treatment efficiency in the aerobic system increases, CO$_2$ emissions increase. Within the surveyed temperature range, the model also showed a large difference between the highest and the lowest GHG emission yield of 1,317.2 kgCO$_2$/d at 27.9°C and of 1,021.7 kgCO$_2$/d at 18.1°C.
The amount of GHG generated by the sludge treatment is relatively balanced at 3,600 kgCH₄/d and 6,600 kg CO₂/d when the temperature is 18.1°C and 27.9°C, respectively.

* The effect of influent BOD concentration changes on greenhouse gas emissions

GHG emissions in the treatment tanks tend to increase as the input flow of the treatment system increases and vice versa. Flow rate fluctuated from 9,300 m³/d to 10,600 m³/d, the amount of GHG increased from 986.6 kgCO₂/d to 1,245.6 kgCO₂/d in aerobic tanks and from 1,881.5 kgCO₂/d to 2,222.7 kg CO₂/d in anaerobic tank. Increasing the flow will be detrimental to controlling the GHG emissions. The large flow output discharge will increase the GHG output generated during the day and reduce efficiency of the waste treatment.

The influent BOD concentration is a factor influencing on microbial growth. Therefore, BOD affect treatment efficiency. When the influent substrate concentrations varied from 1,600 mg/l to 2,000 mg/l, the amount of GHG increased from 984.4 kgCO₂/d to 1,213.2 kgCO₂/d in aerobic tank, and 882.5 to 1,292 kgCH₄/d in the anaerobic tank, respectively.

4. Conclusion

This study established a model based on mass balance equations of species including substrate balance equations, biomass balance equations for reactors of treatment systems, stoichiometric coefficients of species in biochemical reactions and biological processes. The model is programmed by MATLAB.

The steady-state model using the data set from Tan Mai pulp and paper wastewater treatment plant with flow rate of 10,000 m³/day showed that the calculated total on-site GHG emissions from the wastewater treatment process is significantly high (0.74 kgCO₂eq/m²). This value was twice as much as the value in Bai Bang plant (0.38 kgCO₂eq/m²) due to different flow rates and wastewater components between these two plants.

Based on the evaluation of operational factors in the Tan Mai wastewater treatment system, it showed that the optimum SRT in the anaerobic and aerobic treatment tanks is 30 and 10 days, respectively; the optimum temperature in the range of 31 - 34°C will help to reduce GHG emission while still ensuring the efficiency of treatment. Recovering and utilization of biogas will reduce GHG emissions by about 4.5 times according to CO₂ equivalent.

The dynamic model showed that the temperature of the aerobic bioreactor, the flow rate of waste sludge, and the influent concentration of organic matter are directly proportional to the GHG emissions during the wastewater treatment process. Therefore, it is necessary to check and control these parameters in order to minimize GHG emissions. Using the model in calculating GHG emissions is recommended to reduce measurement cost and quickly obtain the forecast data set of GHG emissions.

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