Methodology of methane emission accounting in petrochemical and chemical industries of China

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Abstract. Nowadays, the whole greenhouse effect is becoming more and more serious. It is the common mission of mankind to curb global warming. As the world's second largest greenhouse gas (GHG), the warming potential caused by methane (CH₄) is not negligible, so it is imperative to calculate and reduce its emissions. For the lack of CH₄ accounting methodology in China's petrochemical and chemical industries, this paper proposes a CH₄ accounting methodology based on the enterprise's accounting boundary. Emission sources include CH₄ emissions from fuel combustion, CH₄ emissions from flare gas combustion, CH₄ emissions from escape, CH₄ emissions from anaerobic treatment of sewage, CH₄ emissions from accidental venting. The research results are an important exploration for the accurate measurement of CH₄ emissions from enterprises in China's petrochemical and chemical industries, and also complement the research on China's greenhouse gas accounting methodology.

1. Introduction

With the rapid development of the world economy, the greenhouse effect caused by a large amount of greenhouse gas (GHG) emissions has triggered a series of extreme climate disasters and destroyed the natural ecological balance. Therefore, climate change has become a major global challenge. CO₂ and CH₄ are two important GHG that affect global climate change. Since the industrial revolution, the concentration of CO₂ and CH₄ in the global atmosphere (2011) has increased by 40% and 150% compared with the pre-industrial revolution [1-3]. The warming potential of CH₄ is 28 times stronger than CO₂ [3]. In the past 20 years, the concentration of CH₄ in the atmosphere has increased by 0.8% annually, which is caused by the increase of GHG production and the decrease of its consumption. As the second largest GHG in the world after CO₂ [4], the accurate measurement and accounting of CH₄ is important for the global greenhouse effect.

The problem of CH₄ emissions in China is prominent. So GHG with only consideration of carbon dioxide can't fully represent China's GHG emissions [5]. According to Zhang and Chen [6], in the composition of China's economic sector's GHG emissions in 2007, equivalent carbon dioxide emissions of CH₄ have reached 989.8Mt, which is much higher than the carbon dioxide emissions from fossil fuel combustion in the United Kingdom, Canada, and Germany. Considering the CH₄ emissions is as important as CO₂ in reflecting the history and development trends of China’s GHG emissions. However, the researchers paid more attention on CO₂ in China’s GHG emissions, while ignoring the accounting and reduction of non-CO₂ GHG emissions (such as CH₄). The trading of China's carbon market is limited to CO₂. The GHG emission accounting methods and reporting
guidelines issued by the National Development and Reform Commission for petrochemical and chemical companies are also only for CO$_2$ [7-9]. In the petrochemical and chemical industries, CH$_4$ is both an important raw material and a necessary fuel, which will generate a large amount of CH$_4$ emissions during the process of production. Therefore, it is necessary to calculate the CH$_4$ emissions in the petrochemical and chemical industries [10].

At present, there are some researches on CH$_4$ accounting methodologies in petrochemical and chemical industries. The US Environmental Protection Act regulates the methods used by refinery-related companies to account for greenhouse gases such as CO$_2$, CH$_4$, and N$_2$O. The 2006 IPCC Guidelines for National Greenhouse Gas Inventories use the product as the accounting boundary to account for CO$_2$ and CH$_4$ emissions from the production of typical chemical products. China's guidance document on CH$_4$ accounting methods covers industries such as energy, agriculture and waste, but does not include petrochemical and chemical industries. This research is an important exploration to fill the gaps in CH$_4$ accounting methods in China's petrochemical and chemical industries. It has important reference significance for guiding petrochemical and chemical enterprises to calculate their own CH$_4$ emissions.

2. Framework of accounting methods

2.1. Scope of application
The methodology proposed in this paper provides an accounting method for greenhouse gas methane (CH$_4$) emissions generated by petrochemical and chemical companies based on the enterprise level. It includes accounting boundary, accounting steps, and accounting methods. This methodology can provide a reference for enterprises engaged in petrochemical and chemical production in China to measure their methane emissions.

2.2. Accounting boundary
The accounting boundary of this methodology is defined as the administrative boundary of the enterprise. The accounting entity is an independent legal entity or an independent accounting unit that is treated as a legal entity. It can account for CH$_4$ emissions from all production facilities under its control within the boundaries. Facilities include basic production systems, ancillary production systems, and ancillary production systems directly serving production. And auxiliary production system includes power, power supply, water supply, heating, refrigeration, machine repair, laboratory, instrumentation, warehouse, transportation and so on. The ancillary production system includes the production command management system and the departments and units that serve the production within the plant.

2.3. Emission sources
After many expert discussions, investigation of multiple enterprises and estimation of pilot enterprises, the methodology considers that the categories of emission sources that should be accounted for by the accounting entity include the following five types.
(1) CH$_4$ emissions from fuel combustion. It refers to CH$_4$ emissions from the incomplete combustion of fossil fuels with oxygen in various types of fixed or mobile combustion equipment. The combustion equipment includes boilers, burners, turbines, heaters, incinerators, calciners, kilns, furnaces, ovens, internal combustion engines, etc.
(2) CH$_4$ emissions from flare gas combustion. For safety and other purposes, petrochemical and chemical companies usually concentrate the combustible waste gas generated in each production activity into one or several flare gas systems for combustion before discharge. In addition to CO$_2$ emissions, flare gas combustion may also produce CH$_4$ emissions. And the methodology mainly accounts for CH$_4$ emissions from flare gas systems produced by petrochemical and chemical companies.
(3) CH$_4$ emissions from escape. It mainly refers to the unorganized CH$_4$ emissions from the leakage of equipment in the production business. These devices include valves, flanges, pump wheel seals,
compressor seals, pressure relief valves, sampling interfaces, process drainage, open pipes, casing, tank leakage and other pressure equipment leakage not defined as process emptying. According to the investigation of the current production situation of petrochemical and chemical enterprises, it can be seen that the sealing facilities of enterprises as a whole are good, and the emissions of CH₄ is very small.

(4) CH₄ emissions from anaerobic treatment of sewage. It mainly refers to the CH₄ emissions from the anaerobic treatment of sewage by the accounting entities.

(5) CH₄ emissions from accidental venting. It mainly refers to CH₄ emissions that enterprises have to empty due to accidents.

2.4. Accounting procedures

The workflow of the accounting entity for CH₄ emission accounting includes 4 steps.

(1) The accounting entity must determine the accounting boundary, identify the CH₄ emission facilities it owns, and determine the source of emissions.

(2) The accounting entity should choose the corresponding formulas CH₄ emissions calculation.

(3) The accounting entity needs to obtain data of activity levels and emission factors.

(4) The accounting entity can substitute the collected data into the calculation formulas to obtain the CH₄ emissions.

3. Accounting methods

According to the characteristics of the production process of petrochemical and chemical enterprises, combined with the relevant accounting methods of American and the European Union companies, it is concluded that the total amount of CH₄ emissions from Chinese petrochemical and chemical enterprises is equal to the sum of CH₄ emissions from fuel combustion, CH₄ emissions from flare gas combustion, CH₄ emissions from anaerobic treatment of sewage and CH₄ emissions from accidental venting. The formula is as follows.

\[ E_{CH_4} = (E_{CH_4, \text{combustion}} + E_{CH_4, \text{flare}} + E_{CH_4, \text{sewage}} + E_{CH_4, \text{venting}}) \times GWPC_{CH_4} \]  

where:

\[ E_{CH_4} \] = the total amount of CH₄ emissions from the accounting entity (ton carbon dioxide equivalent, tCO₂eq).

\[ E_{CH_4, \text{combustion}} \] = the CH₄ emissions generated by the accounting entity due to fossil fuel combustion (t).

\[ E_{CH_4, \text{flare}} \] = the CH₄ emissions generated by the accounting entity due to flare gas combustion (t).

\[ E_{CH_4, \text{sewage}} \] = the CH₄ emissions generated by the accounting entity due to anaerobic treatment of sewage (t).

\[ E_{CH_4, \text{venting}} \] = the CH₄ emissions generated by the accounting entity due to accidental venting (t).

\[ GWPC_{CH_4} \] = the global warming potential (GWP) value of CH₄ compared to CO₂. According to the IPCC second assessment report, 1 ton of CH₄ is equivalent to 21 tons of CO₂ in 100 years, so \[ GWPC_{CH_4} = 21 \].

3.1. CH₄ emissions from fuel combustion

3.1.1. Accounting formulas

CH₄ emissions of the accounting entity’ s fossil fuel are based on the amount of fossil fuel combustion by each species used within the boundaries, then multiply the corresponding fuel low calorific value and the CH₄ emission factor of the fuel, and add up by layer by layer.

\[ E_{CH_4, \text{combustion}} = \sum_k FA_k \times NCV_k \times EF_k \times 10^{-3} \]  

Where:

\[ FA_k \] = the consumption of fossil fuel k in the production of the accounting entity (t CH₄).

\[ NCV_k \] = the low calorific value of fuel k (GJ/t).

\[ EF_k \] = the emission factors for CH₄ of fuel k (kg/GJ).
3.1.2. Monitoring and acquisition of activity data
The amount of fossil fuel consumed: The level data of fossil fuel combustion activities of different varieties shall be determined according to the original record of energy consumption or the statistical account of the accounting entity. It is required to indicate fossil fuels that are sent to various types of combustion equipment for combustion as fuel, including energy sources that are self-produced and recycled by the accounting entity.

Low calorific value of fossil fuels: The determination of fuel low calorific value should follow GB/T 213 coal calorific value determination method, GB/T 384 petroleum products calorific value determination method, GB/T 22723 natural gas energy determination and other relevant standards. The coal shall be tested at the time of each batch of fuel entering the plant or once a month at least, with the weighted average of fuel entering the plant or monthly consumption as the low calorific value of the fuel variety. Oil products can be tested at the time of each batch of fuel entering the plant or quarterly, and the arithmetic mean value is taken as the low calorific value of oil products. Gas fuel such as natural gas can be tested at the time of each batch of fuel entering the factory or every half year, and the arithmetic mean value is taken as the low calorific value.

Table 1. Default values of characteristic parameters of common fossil fuels.

| Types of fossil fuels | Low calorific value | Low calorific value unit | Emission factor for CH$_4$ ($\text{kg/GJ}$) |
|-----------------------|---------------------|--------------------------|--------------------------------------------|
|                       |                     |                          | Default emission factors | Lower limit | Upper limit |
| Solid                 |                     |                          |                            |             |             |
| Anthracite            | 24.515              | GJ/t                     | $1 \times 10^{-3}$         | 0.3         | 3           |
| Bituminous coal       | 19.570              | GJ/t                     | $1 \times 10^{-3}$         | 0.3         | 3           |
| Lignite               | 14.449              | GJ/t                     | $1 \times 10^{-3}$         | 0.3         | 3           |
| Coke                  | 28.446              | GJ/t                     | $1 \times 10^{-3}$         | 0.3         | 3           |
| Liquid                |                     |                          |                            |             |             |
| Crude                 | 42.620              | GJ/t                     | $3 \times 10^{-3}$         | 1.0         | 10          |
| Fuel oil              | 40.190              | GJ/t                     | $3 \times 10^{-3}$         | 1.0         | 10          |
| Gas                   |                     |                          |                            |             |             |
| Refinery Gas          | 46.050              | GJ/t                     | $1 \times 10^{-3}$         | 0.3         | 3           |
| Liquefied petroleum gas | 47.310          | GJ/t                     | $1 \times 10^{-3}$         | 0.3         | 3           |
| Natural gas           | 389.310             | GJ/10000Nm$^3$           | $1 \times 10^{-3}$         | 0.3         | 3           |
| Coke oven gas         | 173.854             | GJ/10000Nm$^3$           | $1 \times 10^{-3}$         | 0.3         | 3           |
| Other Gases           | 52.340              | GJ/10000Nm$^3$           | $1 \times 10^{-3}$         | 0.3         | 3           |

Source:  
$^a$ Low calorific values from China Energy Statistics Yearbook 2012 and Study on China's Greenhouse Gas Inventory in 2005.  
$^b$ Emission factor for CH$_4$ from IPCC Guidelines for National Greenhouse Gas Inventories 2006.
The accounting entity without the measured conditions can also refer to Table 1 for the default values of the low calorific value of some common fossil fuels.

3.1.3. Emission factors for CH$_4$ of fossil fuel

The determination of emission factors for CH$_4$ of fossil fuels should be in accordance with relevant standards or by qualified third-party verification agencies. Accounting entities without measured conditions can also refer to Table 1 for the default values of Emission factors for CH$_4$ of some common fossil fuels.

3.2. CH$_4$ emissions from flare gas combustion

The CH$_4$ emissions from flare gas combustion can be calculated indirectly from the CO$_2$ emissions from flare gas combustion. Therefore, the CO$_2$ emissions from flare gas combustion are calculated first.

Flare gas combustion in petrochemical and chemical enterprises can be divided into two types: flare gas combustion under normal working conditions and flare gas combustion caused by accidents. The data monitoring basis of the two types of flare gas is different, so it should be separately accounted.

\[ E_{CO_2,flare} = E_{CO_2,normal,flare} + E_{CO_2,accident,flare} \]  

(3) where:

- $E_{CO_2,flare}$ = the CO$_2$ emissions from flare combustion (t CO$_2$).
- $E_{CO_2,normal,flare}$ = the CO$_2$ emissions from flare gas combustion under normal working conditions (t CO$_2$).
- $E_{CO_2,accident,flare}$ = the CO$_2$ emissions from flare gas combustion caused by accidents (t CO$_2$).

3.2.1. The CO2 emissions from flare gas combustion under normal working conditions

1) Accounting formulas

\[ E_{CO_2,normal,flare} = \sum_{i} \left[ Q_{normal,flare} \times \left( CC_{non,CO_2} \times OF \times \frac{44}{12} + V_{CO_2} \times 19.7 \right) \right] \]  

(4) where:

- $i$ = flare gas system serial numbers.
- $Q_{normal,flare}$ = the flare gas flow of the flare gas system $i$ under normal working conditions (10000Nm$^3$).
- $CC_{non,CO_2}$ = the total carbon content of other carbon-containing compounds other than CO$_2$ in the flare gas (t/10000 Nm$^3$). Please refer to formula (5) for its calculation method.
- $OF$ = the carbon oxidation rate of the flare gas system $i$. If there is no measured data, the default value is 0.98.
- $V_{CO_2}$ = the volume concentration of CO$_2$ in the flare gas (%).
- 19.7 = the density of CO$_2$ under standard conditions.

2) Monitoring and acquisition of activity data

For the flare gas system under normal working conditions, the flare gas flow during the reporting period can be obtained according to the flow monitoring system, engineering calculation or similar estimation method.

The $V_{CO_2}$ in formula (3) should be obtained from the gas component analyser or the flare gas source. The $CC_{non,CO_2}$ in formula (3) should be calculated according to the volume concentration of each gas component and the number of carbon atoms in the chemical formula of the component. The formula is as follows.

\[ CC_{non,CO_2} = \sum_{n} \left( \frac{12 \times V_{n} \times CN_{n} \times 10}{22.4} \right) \]  

(5) where:

- $n$ = the various gas components of the flare gas, except for CO$_2$.
- $V_{n}$ = the volume concentration of the number $n$ carbon-containing compound (including carbon monoxide) other than CO$_2$ in the flare gas (%).
- $CN_{n}$ = the number of carbon atoms in the chemical formula of the number $n$ carbon-containing compound (including carbon monoxide) in the flare gas.
3.2.2. The CO2 emissions from flare gas combustion caused by accidents

1) Accounting formulas

At present, there is no specific monitoring record for the combustion of flare gas caused by accidents in Chinese petrochemical and chemical industry enterprises. So, it is very difficult to obtain flare gas flow data. It is recommended to estimate the amount of accident flare combustion based on the average gas flow rate of the accident facility leading to the flare gas systems and the duration of the accidents. And then they can estimate the CO$_2$ emissions from flare gas combustion caused by the accidents.

$$E_{CO_2, \text{accident flare}} = \sum_j \left[ GF_{\text{accident}, j} \times T_{\text{accident}, j} \times CC_{(\text{non CO2})j} \times OF \times \frac{44}{12} + V_{(CO2)j} \times 19.7 \right]$$  \hspace{1cm} (6)

where:

- $j =$ the accident $j$.
- $GF_{\text{accident}, j} =$ average flare air velocity at the accident $j$ during the reporting period (10000Nm$^3$/h).
- $T_{\text{accident}, j} =$ the duration of the accident $j$ during the reporting period (h).
- $CC_{(\text{non CO2})j} =$ the total carbon content of other carbon-containing compounds other than CO$_2$ in the accident $j$ flare gas (t/10000 Nm$^3$). Please refer to formula (5) for its calculation method.
- $OF =$ the carbon oxidation rate of the accident $j$ flare gas. If there is no measured data, the default value is 0.98.
- $V_{(CO2)j} =$ the volume concentration of CO$_2$ in the flare gas of the accident $j$ (%).

2) Monitoring and acquisition of activity data

$GF_{\text{accident}, j}$ and $T_{\text{accident}, j}$ should refer to the accident investigation report for value. The accident flare gas component of the petroleum refining system is calculated as C$_5$. And the accident flare gas component of the petrochemical system is calculated as C$_3$.

3.2.3. CH4 emissions from flare gas combustion

The accounting entity can further calculate the CH$_4$ emissions from the flare combustion based on the calculated CO$_2$ emissions from the flare combustion.

1) Calculation formula

$$E_{CH_4, \text{flare}} = \left( E_{CO_2, \text{flare}} \times \frac{EF_h}{EF} \right) + \left( E_{CO_2, \text{flare}} \times \frac{0.02}{0.98} \times \frac{16}{44} \times f_{CH_4} \right)$$  \hspace{1cm} (7)

where:

- $EF_h =$ 2.844kg/GJ. It refers to the default CH4 emission factor for flare gas. And this value is based on the Code of Federal Regulations (CFR).
- $EF =$56.87kg/GJ. It refers to the default CO$_2$ emission factor for flare gas. And this value is based on the Code of Federal Regulations (CFR).
- $f_{CH_4} =$ the weight fraction of carbon in the flare gas before combustion. Its default value is 0.4
- 0.02/0.98 = correction factor for flare gas combustion efficiency.
- 16/44 = molecular weight correction factor ratio of CH$_4$ to CO$_2$.

3.3. CH$_4$ Emissions from anaerobic treatment of sewage

1) Calculation formulas

The calculation formula for the CH$_4$ emissions caused by the accounting entity to treat its own or external sewage by anaerobic process is shown in formula (8).

$$E_{CH_4, \text{sewage}} = (TOW - S) \times EF_{CH_4, \text{sewage}} \times 10^{-3}$$  \hspace{1cm} (8)

where:

- $E_{CH_4, \text{sewage}} =$ the CH$_4$ emissions generated of anaerobic treatment of sewage (t).
- $TOW =$ the total amount of degradable organic matter in sewage (kg) . It uses chemical oxygen demand (COD) as a measurement index.
- $S =$ the total amount of organic matter that is disposed of by sludge (kg). It uses chemical oxygen demand (COD) as a measurement index.
- $EF_{CH_4, \text{sewage}} =$ the CH$_4$ emission factors for anaerobic treatment of sewage (kg CH$_4$/kg COD).
If the accounting entity has the COD statistics removed by the sewage treatment system, it can be directly used as the TOW value. If there is no COD statistic removed by the sewage treatment system, the following formula can be used to estimate.

\[
TOW = W \times (COD_{in} - COD_{out})
\]

where:
- \(W\) = the amount of sewage from anaerobic treatment (m³/year).
- \(COD_{in}\) = the average COD concentration of sewage entering the anaerobic treatment system (kg COD/m³).
- \(COD_{out}\) = the average COD concentration of sewage from the outlet of an anaerobic treatment system (kg COD/m³).

\[
EF_{CH_4, sewage} = B_0 \times MCF
\]

where:
- \(B_0\) = the maximum production capacity of CH\(_4\) in sewage anaerobic treatment systems (kg CH\(_4\)/kg COD).
- \(MCF\) = the CH\(_4\) correction factors. It refers to the extent to which different treatment systems or discharge pathways achieve maximum methane production capacity and also reflects the anaerobic extent of the treatment system.

2) Monitoring and acquisition of activity data
The amount of sewage from the anaerobic treatment of the accounting entity, the amount of COD removed by the anaerobic treatment system, and the amount of COD removed by sludge should be determined according to the original record of the accounting entity or the statistical account. And the amount of COD removed by sludge should be assumed to be zero if the accounting entity does not have statistics.

The COD concentration in the sewage should be the average value of the accounting entity. The measurement method needs to meet the standard monitoring method for chemical oxygen demand in water quality monitoring of the Ministry of Environmental Protection of China. Water samples were collected at least once every 2 hours, and mixed samples were taken 24 hours for determination.

(3) Monitoring and acquisition of emission factors
For the maximum production capacity of CH\(_4\) in sewage anaerobic treatment system, the default value is 0.25 kg CH\(_4\)/kg COD. The accounting entity should be updated according to official data issued by the competent authority in the future. For MCF, the qualified accounting entity can carry out the actual measurement or entrust qualified professional institutions to carry out the inspection. The accounting entity without a specific MCF value can refer to Table 2 for the default value.

**Table 2.** MCF default values for various sewage treatment systems.

| Treatment and discharge pathways or system types | MCF  | Range | Remarks |
|-------------------------------------------------|------|-------|---------|
| Ocean, river or lake discharge                   | 0.1  | 0 - 0.2| High concentration of organic sewage entering the river may produce anaerobic reactions |
| Aerobic treatment facility                       | 0    | 0 - 0.1| Must be well managed |
| Aerobic treatment facility                       | 0.3  | 0.2 – 0.4| Incomplete management, overload |
| Sludge anaerobic digestion tank                   | 0.8  | 0.8 – 1.0| CH\(_4\) recovery not considered |

3.4. \(CH_4\) emissions from accidental venting
Due to process modification and maintenance, the companies need to vent the remaining CH\(_4\) gas stored in process pipelines and equipment with the consideration of safety production. The calculation of these emissions should be estimated based on the company's venting history and the corresponding monitoring records.

4. Conclusion
China has already issued accounting and reporting requirements for greenhouse gas emissions in 24 industries. But China has not yet issued corresponding accounting methodologies for CH\(_4\) emissions in
the petrochemical and chemical industries. It is necessary to calculate CH$_4$ because of its important position in the petrochemical and chemical industries. The methodology proposed in this paper is aimed at the five major sources of CH$_4$ in petrochemical and chemical industry enterprises: CH$_4$ emissions from fuel combustion, CH$_4$ emissions from flare gas combustion, CH$_4$ emissions from escape, CH$_4$ emissions from anaerobic treatment of sewage, CH$_4$ emissions from accidental venting. The conclusion of the study can be mentioned as support for the introduction of China's greenhouse gas accounting policy. The default emission factors in this methodology are mainly provided by the 2006 IPCC National Greenhouse Gas Inventory. Due to the huge differences in production processes, it is easy to lead to deviations in the calculation. The future research will focus on accurate measurement of CH$_4$ emission factors to strengthen this methodology.

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