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Experimental Study on Greenhouse Gas Emissions Caused by Spontaneous Coal Combustion

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ABSTRACT: There are large areas of spontaneous coal combustion in northern and northwestern China. The quantification of greenhouse gas (GHG) emissions resulting from spontaneous coal combustion is an important step toward determining proper management practices to reduce such emissions in the future. The present study investigated the GHG emission characteristics of the spontaneous combustion of 10 typical coal types. Furthermore, this study examined the estimation method applied to the GHG emissions caused by spontaneous coal combustion. The experimental results showed that the rates of CH_4 and CO_2 emissions from spontaneous coal combustion resulting from mining activities were greater than the same rates of emissions resulting from mere surface air seepage (by factors of ∼1.8 and ∼1.6, respectively). The emission rate of CH_4 was significantly correlated with the volatile content of coal, while the emission rate of CO_2 was significantly correlated with the moisture, oxygen, and sulfur contents of coal. Three different characteristic phases were observed for the emission of CH_4, with critical temperatures of 200, 400, and 600 °C, respectively. Below 200 °C, CH_4 emissions were very slow; between 200 °C and 450 °C, the emissions increased slowly; and above 450 °C, the emissions increased rapidly. Similarly, the CO_2 emissions were very low at temperatures below 200 °C, slowly grew between 200 °C and 450 °C, and quickly grew after 450 °C. The experimental emission factors of CO_2 and CH_4 caused by spontaneous coal combustion were obtained at different stages of spontaneous combustion. We subsequently determined that the annual GHG emissions from the Wuda coal field fire area of China were ∼956 700 tons of CO_2 equivalents. The results of this research offer a technical basis for quantifying GHG emissions from spontaneous coal combustion.

1. INTRODUCTION

Spontaneous coal combustion events are disasters triggered by both natural and human factors and may occur in underground coal beds, coal gangue dumps, coal piles, and abandoned mines.[1−3] Spontaneous coal combustion has occurred since ancient times. Seriously disastrous spontaneous coal combustion events have more recently occurred in the United States, China, Indonesia, South Africa, Australia, Germany, and Poland.[4−6] Large areas where spontaneous coal combustion may occur exist, particularly in northern and northwestern China. The Chinese government has invested significant manpower and material and financial resources to manage these areas. Spontaneous coal combustion leads to enormous energy waste and releases large amounts of greenhouse gases (GHGs) and hazardous substances,[7] posing severe threats to mine safety, production, local environments, and the health of proximal residents. At the same time, the greenhouse effects caused by spontaneous coal combustion have attracted immense attention from the government and scholars. Typical GHG emissions from spontaneous coal combustion include CO_2 and CH_4.[8,9] Quantifying the regional or national emission rates based on the rates obtained from a macroscopic perspective is key to measuring the GHG contributions caused by large-scale spontaneous coal combustion events. Such quantification is also key to evaluating emissions reductions that may be achieved by the proper management of spontaneous coal combustion. Currently, no adequate data or effective methods exist for evaluating GHG contributions from spontaneous coal combustions. Thus, the effects of spontaneous coal combustion on the environment are highly uncertain.

According to the *United Nations Framework Convention on Climate Change* and the *Kyoto Protocol*, for many years, and via many fields and departments, China has had an international obligation to mitigate GHG emissions. However, no carbon emission factors have yet been determined, with respect to spontaneous coal combustion. Thus, China is in the early stages of quantifying the GHG emissions caused by spontaneous coal combustion. The current methods for quantifying GHG emissions from spontaneous coal combustion are differentiated into two categories: (1) airborne transmission methods and (2) Earth surface measuring methods as applied to the fire area. The former methods are based on satellites or remote sensing data[10,11] and indirectly determine the rough emission flux of CO_2. However, because of the limitations associated with collecting data via remote sensing or satellites, these methods cannot be used to reliably evaluate CO_2 emissions caused by spontaneous coal combustion. The latter methods are based on field Earth surface flux observations and gas dissipation flux monitoring within the soil[12] and directly estimate the GHG emissions from specific fire areas. However, these methods can only be used to measure GHG emissions from partial fire areas, and the estimates are greatly influenced by the monitoring equipment and the geographical conditions of the fire area. China has a wide variety of spontaneous coal combustion areas with complex variations, and field observations of spontaneous coal combustion or observations obtained via remote sensing technology of these areas have been described in some reports. Litschke[13] and Schloemer[14] measured CO_2 emissions from the

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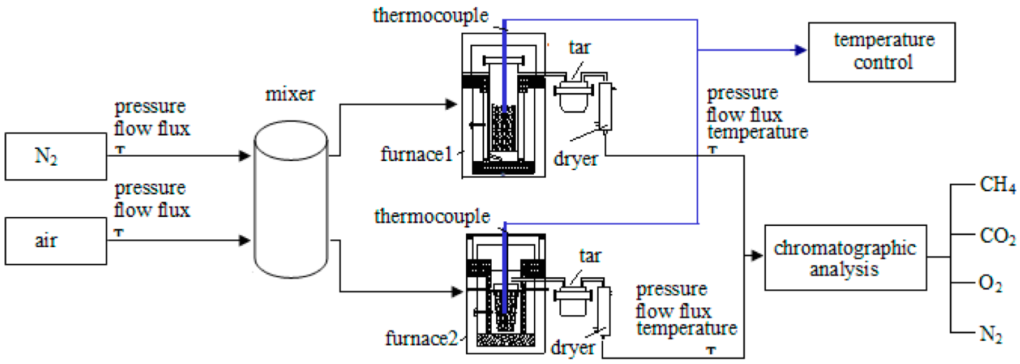

Figure 1. Experimental system.

Table 1. Typical Coal Samples and Their Basic Parameters

	Proximate Analysis (%)				Elemental Analysis (%)					
coal type	M_{ad}	A_d	V_d	FC_d	C_{ad}	H_{ad}	O_{ad}	N_{ad}	S_{ad}	calorific value, $Q_{b,ar}$ J/g
brown coal	16	34.8	33.5	31.7	39.1	4.8	24.9	0.83	1.51	15 230.14
jet coal	4.6	18.5	20.5	61	64.4	3.9	12.9	1.29	0.36	24 798.19
noncaking coal	12.8	13.3	29.9	56.8	61.5	6	23.2	0.82	1.19	23 623.27
weakly caking coal	3.3	4.7	28.8	66.5	82.2	0.5	8.8	0.51	0.28	18 112.23
gas coal	2.9	25.8	29.5	44.7	57.1	4.7	12.5	1.28	0.74	23 099.69
fat coal	1.4	12.6	32.5	54.9	73.6	5.7	8.7	1.11	0.34	30 151.41
coking coal	1.1	6.4	24.5	69.1	82.7	6.7	5.5	1.63	0.83	33 561.99
lean coal	0.8	10.4	13.05	76.55	81.3	4.5	4	1.66	0.4	32 110.27
meager coal	1.3	10.7	11.5	77.8	80.8	4.1	4.3	1.61	0.41	31 659.01
blind coal	3.3	13.2	7.8	79	82.2	0.9	4.5	0.54	0.22	30 156.41

No. 3-2 and No. 8 fire areas. Tetzlaff[15] analyzed the GHG levels resulting from the Rujigou and Wuda coal fires at background temperatures of 0−23 °C, using an airborne transmission method. Van Dijk et al.[16] calculated coal fire-related GHG emissions on a provincial scale in China with a CO_2 emission factor determined for complete coal combustion. However, these studies do not accurately and simply estimate the amount of GHGs released by spontaneous coal combustion in China.

Unfortunately, there are no precedents in determining GHG emission factors. In the *National Guideline List of Greenhouse Gas Emission*,[17] released by the Intergovernmental Panel on Climate Change (IPCC), emission factors have been determined only for conditions involving the complete combustion of standard coal and thus are not suitable for estimating GHG emissions that occur during the different combustion stages of spontaneous coal combustion. Therefore, the objectives of the present paper are to experimentally quantify GHG emissions from typical spontaneous coal combustion events and to propose GHG emission quantification factors for the different developmental stages of spontaneous coal combustion (predominantly CO_2 and CH_4 emissions). Our ultimate goal is to obtain essential data and technical support for determining the contributions of large-scale spontaneous coal combustion events to the greenhouse effect and to evaluate the effectiveness of fire-area governance practices on reducing emissions in China.

2. EXPERIMENT

2.1. Equipment. Using a reasonably designed experimental scheme, this paper measures the components and amount of GHG emissions from the spontaneous combustion of typical coals and determines the emission laws for predominant GHGs. These measurements provide the essential data required to calculate GHG emission factors for different stages of spontaneous coal combustion. The self-designed experimental setup in China University of Mining & Technology−Beijing is composed of an experimental platform, a computer monitoring and controlling system, and a gas analysis and acquisition system. The experimental platform includes an air generator, a reactor, a furnace body, a tar-gas separator, and a water-cooling tank. (See Figure 1.) The computer monitoring and controlling system includes a hardware system, which is responsible for data collection and signal processing, and a software system that has process control over an experimental furnace. The measurement and control system contains a high-precision flow meter to manage the air (or oxygen) supply during the experiment. After the coal tar is removed, cooled, and dried, the gas in the air outlet is connected to a high-precision flow sensor in order to collect pressure and temperature data during actual air flow, after which the gas finally enters a chromatographic instrument for gas composition analysis.

2.2. Parameters of the Experiment. For a wide range of applications, to establish quantitative indices for GHG released via spontaneous coal combustion, we selected 10 typical coal samples from northern and northwestern China for our experiments. The values of various indices, such as the basic proximate analysis and calorific value of the coals, are shown in Table 1. Coal samples were sealed on site and were rapidly analyzed. The samples were first sieved to different particle diameters and then enclosed in bottles. The coal samples used in the experiments were a proportional mixture of the following four size categories: 1−3 mm, 3−5 mm, 5−7 mm, and 7−10 mm. The quantity of coal used in each experiment was 200 g. The field survey indicated that, at coal-bed temperatures above 800 °C, spontaneous coal combustion enters a sustained combustion stage.[18] Therefore, the maximum temperature used during the experiments was 800 °C. The heating rate applied during experiments was 2 °C/min.

In the process of simulating spontaneous coal combustion, many similarity coefficients were employed; therefore, it was too complex to realize all similarity criteria. Regarding small-scale experiments, compared with the actual fire area in which spontaneous coal combustion occurs, the similarity of gas components and quantity generated by spontaneous coal combustion was superior to determinations via the gas spreading principle. This paper obtained the components and quantities of GHGs released by spontaneous coal combustion while assuming a negligible gas spreading property. The gas component generated by

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spontaneous coal combustion negligibly changes with experimental scale; therefore, the requirements for similar GHG quantities must be satisfied in the simulation processes of smaller-scale experiments. The satisfaction of this condition is dependent on oxygen leaks occurring during spontaneous coal combustion. Generally, there are two types of spontaneous coal combustion, i.e., spontaneous coal combustion involving mining activities (these air leakage patterns are called "Pattern A") and coal-gangue-dump spontaneous combustion, coal-piles spontaneous combustion and unexploited-crop spontaneous combustion, which are simply caused by surface wind leakage (these air leakage patterns are called "Pattern B"). Because of the different oxygen levels in air leaks occurring during the spontaneous coal combustion process, GHG emissions will correspondingly vary. For this reason, the experimental procedure used herein employed two sets of simulation systems: furnace 1 and furnace 2. Furnace 1 was supplied with air via a combination method in which air was supplied from the bottom of the furnace (such that the air first passes through the coal bed) and from the side wall (such that the air first sweeps past the coal bed); these conditions simulate the spontaneous coal combustion process occurring under the conditions of Pattern A. Furnace 2 was supplied with air from the side wall (such that the air first sweeps past the coal bed); this simulates the spontaneous coal combustion process occurring under the conditions of Pattern B. These air leakage patterns are representative of the data in the actual spontaneous-combustion fire area and the characteristics of the container. These patterns satisfy the similarities in air leakage or air leakage rates in the unit coal quantity or unit area.

With regard to determining the quantity of air supplied during experiments, the influence of air supply variations on spontaneous coal combustion should be correlated with their corresponding influence *in situ*. Our previous research results[19] showed the comprehensive and contradictory influence of air supply and quantity and heat loss on spontaneous coal combustion. We observed the ease with which spontaneous coal combustion first increased with increasing air leakage velocity or air supply/quantity (oxidative heat release is the predominant controlling factor in spontaneous coal combustion) and then decreased (heat dissipation is the predominant controlling factor in spontaneous coal combustion). Thus, the maintenance of spontaneous coal combustion requires a more facile oxidation velocity and a very low air leakage velocity. Voracek[20] reported the lowest air leakage velocity required to maintain *in situ*, underground spontaneous coal combustion, for which the oxidation rate was 0.3−0.4 m/min. Taking 0.3 m/min as the minimal air leakage velocity and based on the porosity of mixed coal (∼0.08), the lowest experimental air supply velocity determined in our experiments was 30 mL/min. The experiment in ref 21 measured the air supply velocity for the most facile spontaneous coal combustion obtained for particle sizes ranging from 0.5 mm to 10.5 mm. This air supply velocity value was further used to determine the average air supply velocity value (0.001 m/s) for the easiest spontaneous coal combustion obtained for particle sizes ranging from 1 mm to 9 mm. Therefore, we used 0.001 m/s as the reference air supply velocity for the easiest oxidation occurring during spontaneous combustion. Combining this with the experimental reactor size and entrance diameter of 4 cm, our experiments confirmed that the air supply velocity for the easiest oxidation involved in the spontaneous combustion process was 75 mL/min. Using 75 mL/min as the middle air supply value and 30 mL/min as the lower limit value, and by way of equivalence between the upper and lower limits, we confirmed that the upper limit air supply value in our experiment was 120 mL/min. Considering the influence of air supply on GHG yield, we confirmed the following four air supply states: 30 mL/min, 60 mL/min, 90 mL/min, and 120 mL/min.

2.3. Experimental Method. Data, such as temperature, pressure, flow rate, and oxidation time, were obtained via real-time collection and storage. Gas collection and analysis systems (i.e., gas chromatography) were used to analyze the gaseous products of coal oxidation. For temperature ranges of 30−300 °C, the sampling analysis was performed every 20 °C; within the temperature range of 300−800 °C, the sampling analysis was performed every 30 °C. Based on the production law and the quantities of CO_2 and CH_4 involved in coal oxidation and temperature increases and the accumulated mean values of CH_4 and CO_2 emissions for the four types of air supplies, the level of GHG emissions resulting from spontaneous coal combustion reactions within certain combustion temperatures, we have

$$\bar{x}_k = \frac{\sum_i (x_{ki} + x_{k(i-1)})t_{ki}/2}{\sum_i t_{ki}} \qquad (1)$$

where i is the number of the coal temperature section; k is the air supply number (k = 1, 2, 3, and 4, corresponding to rates of 30, 60, 90, and 120 mL/min, respectively); x is the instantaneous mean GHG emission rate (given in units of g/(t s)); and t is the GHG emission time (in seconds).

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. Influence of Air Leakage Patterns and Coal Quality on GHG Emissions. Figure 2 shows bar charts of accumulated GHG emission rates caused by spontaneous coal combustion, as well as the average values obtained from the accumulation of instantaneous GHG emission rates over time under the influence of different air leakage conditions. The results show that the accumulated GHG emission rates resulting from spontaneous coal combustion that involves mining was significantly higher than those involving surface wind diffusion. The emission rates of CH_4 obtained under Pattern A conditions were, on average, ∼1.8 times greater than the values of those obtained under Pattern B conditions, and Pattern A CO_2 emission rates were ∼1.6 times greater than those obtained via Pattern B. The reason for these differences is that, under Pattern B oxygen supply conditions, the rate at which oxygen permeates inside the coal is slower. Thus, the oxidation rate of coal under Pattern B conditions was slower than that under Pattern A, leading to lower GHG emissions when Pattern B is utilized.

Table 2 displays the correlation analyses of the accumulated average GHG emission rates that were above those obtained via the proximate, elemental, and calorific analyses of coal samples. The correlation analyses take into account that the emission of CH_4 had a significant positive correlation with volatiles content and that the emission of CO_2 had a significant positive correlation with water, oxygen, and sulfur contents. The scatter diagram and fitting curve of GHG emission rates are shown in Figure 3, from which it can be seen that the accumulated CH_4 emission rates resulting from spontaneous coal combustion increased as the volatiles content of the coal sample increased. Similarly, the accumulated CO_2 emission rates increased with the water, oxygen, and sulfur contents of the coal samples.

3.2. Influence of Temperature on GHG Emissions. Figures 4 and 5 display the average GHG emission rates arising from the different temperatures of spontaneous coal combustion. Figure 4 shows that, given the different spontaneous combustion temperatures, the emission of CH_4 typically occurs in three stages. Stage one occurs below 200 °C, in which the original CH_4 absorbed by the coal begins to desorb. In this case, CH_4 remains but only in very small amounts. Stage two occurs between ∼200 °C and 600 °C, in which volatile matter, such as CH_4, first begins to dissolve out of the coal. During this stage, other than in blind coal, CH_4 emission first slowly increases and then slowly decreases, with the highest value emerging at ∼400 °C. Stage three occurs above 600 °C, in which semicoke begins pyrolysis and much of the CH_4 dissolves out of the coal. During this stage, other than in blind coal, CH_4 emission tends to initially rapidly increase and, subsequently, slowly increase. Changes observed at temperatures above 700 °C are relatively small.

As shown in Figure 5, during different developmental stages of spontaneous coal combustion, CO_2 emissions display the characteristics described below. Below 200 °C, the emission rate

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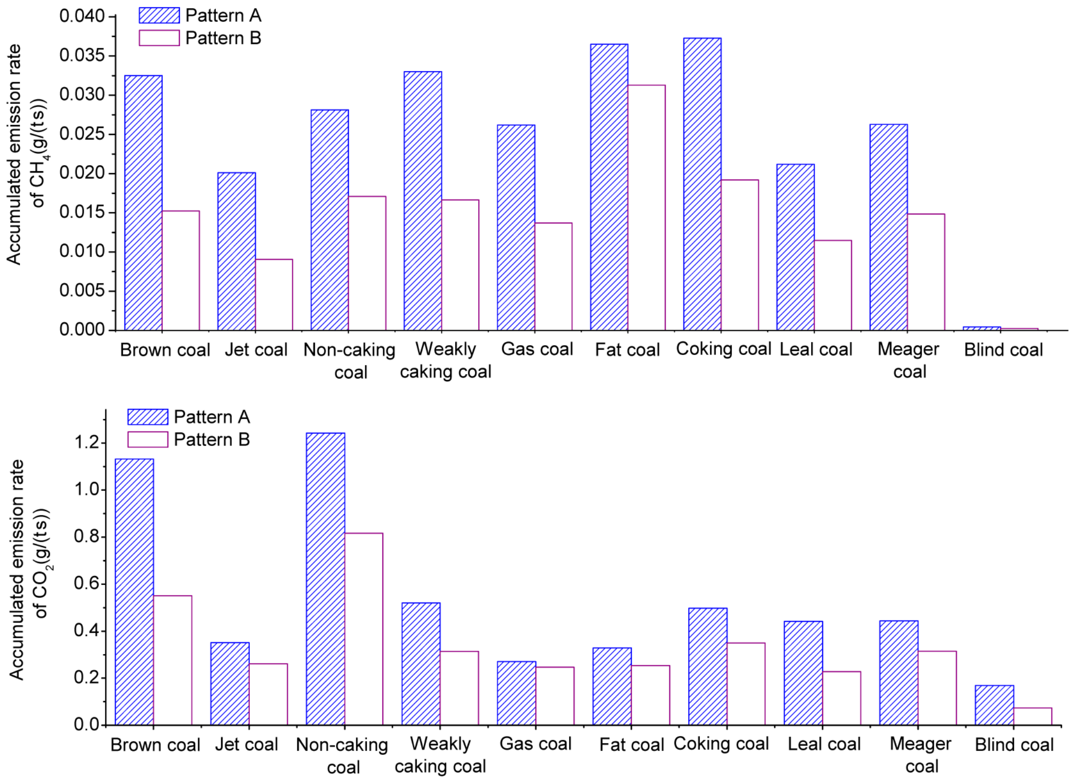

Figure 2. Accumulated GHG emission rates caused by spontaneous coal combustion under conditions of differing leakage patterns.

Table 2. Correlation Analysis of Accumulated GHG Emission Rates Caused by Spontaneous Coal Combustion and Its Relationship to Coal Quality

	Proximate Analysis (%)				Elemental Analysis (%)					
GHG	M_{ad}	A_d	V_d	FC_d	C_{ad}	H_{ad}	O_{ad}	N_{ad}	S_{ad}	calorific value, $Q_{b,ar}$
CH_4										
Pattern A	0.116	−0.0237	0.756[a]	−0.449	−0.201	0.561	0.273	0.241	0.400	−0.213
Pattern B	−0.037	−0.1277	0.684[a]	−0.343	−0.061	0.566	0.134	0.181	0.171	0.014
CO_2										
Pattern A	0.880[b]	0.3241	0.506	−0.503	−0.590	0.366	0.834[b]	−0.248	0.853[b]	−0.532
Pattern B	0.779[b]	0.2116	0.553	−0.465	−0.525	0.480	0.805[b]	−0.160	0.800[b]	−0.436

[a]Significant correlation on the level of 0.05 (two-tailed). [b]Significant correlation on the level of 0.01 (two-tailed).

of CO_2 was very slow; this stage was caused by the resolution of the original adsorption of CO_2 and the oxidation of the coal sample caused by a small amount of physical absorption. From 200 °C to 450 °C, the CO_2 emission rate started to increase slowly. Above 450 °C, the high-temperature reaction was acute, and a large portion of the macromolecular chain fractured, leading to a rapid increase in the emission of CO_2. Above 700 °C, the emission rate was somewhat steady.

3.3. Emission Factors. The survey results obtained for the existing spontaneous-combustion fire area demonstrate[18] that, from the start of oxidation to conditions sustaining combustion, spontaneous coal combustion can be divided into the following five stages: (1) a slow oxidation stage, (2) a stage of spontaneous combustion development, (3) a combustion-center formation stage, (4) a developing combustion-system developmental period, and (5) a stage of sustained combustion. The stages correspond to (1) temperatures below 100 °C, (2) temperatures between 100 °C and 250 °C, (3) temperatures between 250 °C and 400 °C, (4) temperatures between 400 °C and 600 °C, and (5) temperatures above 600 °C, respectively. Moreover, each stage has its own typical geologic features. To distinguish and correctly differentiate GHG emissions caused by spontaneous coal combustion during different developmental stages, the experimental CH_4 and CO_2 results and the fire area survey results were used to classify spontaneous coal combustion into four stages, based on coal temperatures: (i) <200 °C, (ii) 200−400 °C, (iii) 400−600 °C, and (iv) >600 °C. Generally, with regard to carbon emission factors,[22] if the data are from random and representative samples, a distribution can be established directly via typical statistical analyses. We adopted a basic statistical approach to average the GHG emissions caused by spontaneous coal combustion during different combustion stages under the Pattern A and Pattern B conditions. A 95% confidence interval was used to estimate the uncertainty in GHG emissions caused by spontaneous coal combustion, the results of which are shown in Table 3. Under normal conditions, the emission factors of GHGs emitted in the actual fire area can be used to select the mean value for quantification directly. However, regarding certain spontaneous coal combustion events involving greater relative changes in coal quality, the emission factors can be obtained with relatively greater accuracy. For example, the CO_2 and CH_4 emission factors during the spontaneous coal combustion of coal samples with high

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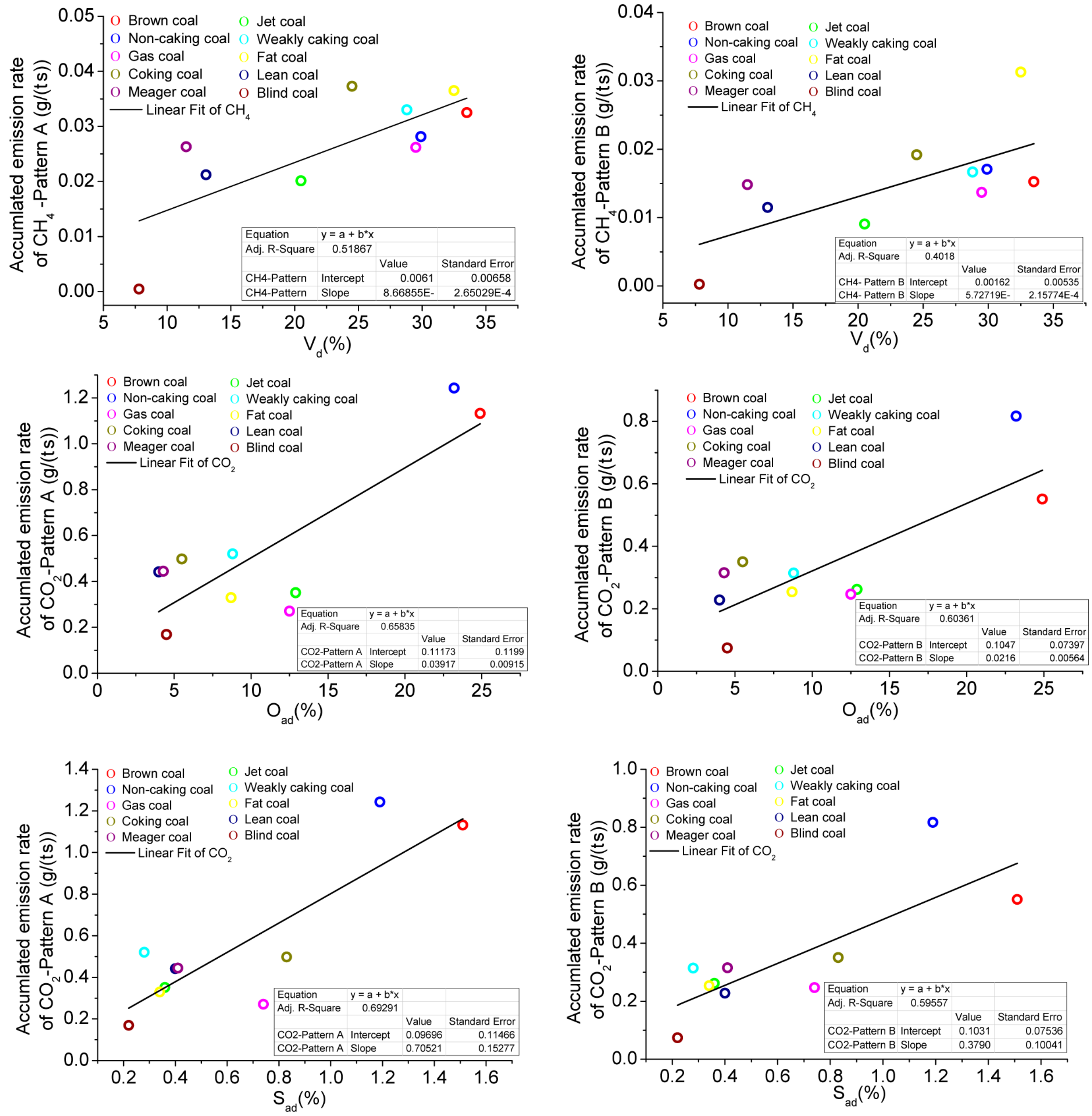

Figure 3. Scatter and fitting curves of accumulated GHG emission rates caused by spontaneous coal combustion.

volatility and sulfur contents can be identified between the mean value and the upper limit of the 95% confidence interval.

4. APPLICATION

4.1. Methodology. Spontaneous coal combustion includes spontaneous combustion via coal field fires, coal gangues, and coal storage piles, of which the most universal are coal field fires in northern and northwestern China. Therefore, the mathematical model for GHG emissions caused by spontaneous coal combustion is based on coal field fires. With regard to the other types of spontaneous coal combustion, the model can be modified according to their own characteristics. With regard to coal field fire areas, particularly large areas, the development of the fire area is a dynamic process. However, the areas of coal field fires change within a wide time scale (for example, years or months) and are easy to obtain using conventional techniques. Therefore, based on the area of the coal field fire and the abundance of resources in different combustion stages, the amount of raw coal contributing to spontaneous combustion in relevant fire areas can be determined as follows:

$$M_{ij} = \Delta S_{ij} H_{ij} \tag{2}$$

where i is the number of fire areas, j is the number of the spontaneous combustion stage, M_{ij} is the amount of raw coal t of fire area i in combustion stage j per unit time t, ΔS_{ij} is the change in area of fire area i in combustion stage j in the period of time of Δt (m^2), and H_{ij} is the abundance of resources (i.e., the raw amount of coal per unit area contributing to combustion, t/m^2).

The amount of GHGs emitted is determined by the emission factors multiplied by the amount of coal undergoing spontaneous combustion. Thus, based on eq 2, the quantity of GHG emissions in the coal-field fire area is

$$C_m = \sum \sum M_{ij} K_{ij,m} \qquad (i = 1, 2, ..., k;\ j = 1, 2, ..., t) \tag{3}$$

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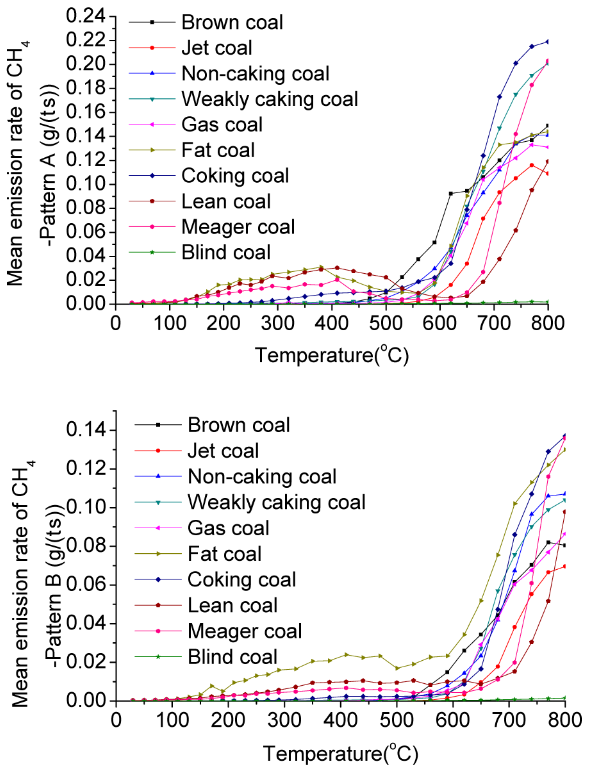

Figure 4. Mean emission rate of CH_4 under different coal temperatures.

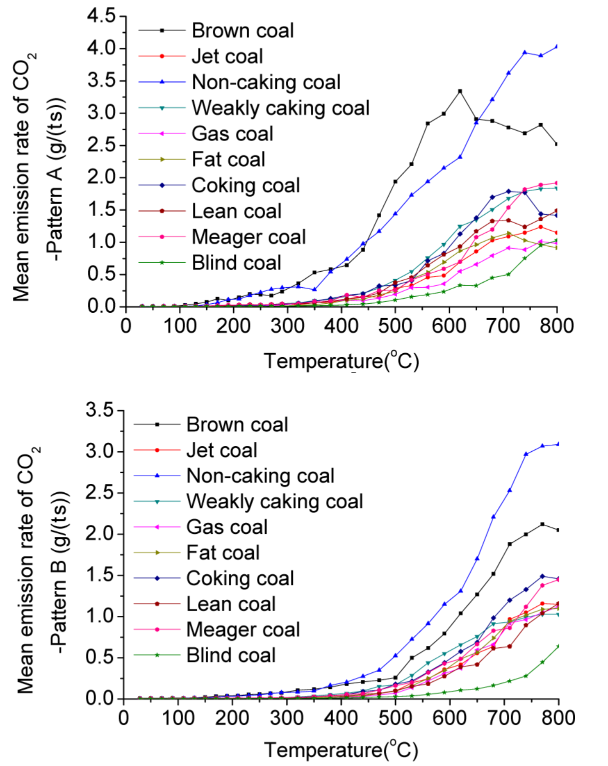

Figure 5. Mean CO_2 emission rates as a function of coal temperature.

where C_m refers to m-type GHG emissions (g/s) and $K_{i,j,m}$ is the emission factor for the m-type GHG emissions in combustion stage j of fire area i (g/(t s)). The emission factor, K_{ij}, can be determined using the accumulated emissions obtained by simulating a specific coal bed, as demonstrated in Table 3.

The predominant GHGs produced by spontaneous coal combustion are CH_4 and CO_2. The greenhouse effect of CH_4 is 21 times greater than that of CO_2.[16] Thus, the CO_2 equivalents emitted by spontaneous coal combustion is

$$C_{\mathrm{GHG}} = 21\sum\sum M_{ij}K_{ij,\mathrm{CH_4}} + \sum\sum M_{ij}K_{ij,\mathrm{CO_2}} \quad (4)$$

With regard to gangue spontaneous combustion, we refer to the IPCC method for measuring the difference between GHG emissions from different coals.[17] We modify eq 4 by taking the ratio f between the net calorific value of the gangue and the net calorific value of the corresponding raw coal. Thus, the CO_2 equivalents caused by the spontaneous combustion of gangue is

$$C_{\mathrm{GHG}} = f(21\sum\sum M_{ij}K_{ij,\mathrm{CH_4}} + \sum\sum M_{ij}K_{ij,\mathrm{CO_2}}) \quad (5)$$

If the mass of the coal (gangue) pile during different combustion stages can be directly determined, the mass value can be directly represented as M_{ij}.

4.2. Annual GHG Emissions from the Wuda Coal Field Fire Area in China. The Wuda coal field is located in Wuhai City in Inner Mongolia, China, on the north end of Helan Mountain, east of Ulanbuh Desert, and 10 km from the Yellow River in the east. Its geographic coordinates are between 39°28′21.15″N and 39°34′6.01″N, 106°36′21.83″E, and 106°39′15.53″E. The Wuda coal field has a dry climate, experiences large temperature changes between day and night and has adequate sunlight. The coal field covers an area of 27.6 km^2. Shaft building was initiated in the Wuda mine area in 1958. This area, which contains the Suhaitu, Huangbaici, and Wuhushan coalmines, has become one of the most important coal enterprises in Inner Mongolia. In 1961, the spontaneous combustion of coal with particularly high sulfur contents occurred in the No. 9 and No. 10 coalbeds of the Suhaitu coalmine. From the 1970s to the 1990s, because of its high number of small coal mines, spontaneous combustions occurred in many coal beds within the Wuda coal field. In July 2002, the number of different fire areas in Wuda reached 26, comprising a total area of 3 076 000 m^2.[23] After many years of government oversight, this area has been significantly reduced. Based on surveys in 2001, 2006, and 2008, and on the application of a gradual self-adaption threshold value method[24] to remote sensing data obtained on March 27, 2013 (collected at night by the ASTER imager and Terra satellite), the spontaneous combustion area of the Wuda coal field was determined to be ~1 231 000 m^2.[25]

According to the 2008 survey, the combusting coal beds of the Wuda coal field predominantly included Nos. 1, 2, 4, 6, 7, 9, 10, and 12.[18] The average thickness of the coal beds that participated in combustion was 7.06 m.[25] Based on mining records, the abandoned coal rate was determined to be ~0.6; thus, the average thickness of the combusting coal beds areas was estimated to be 7.06 × 0.6 = 4.236 m. The average density of coal in the Wuda coal field was 1.53 t/m^3,[26] so the total amount of coal within the Wuda coal field fire areas marked by the satellite was 1 231 000 × 4.236 × 1.53 = 5 215 000 tons.

The Shenhua Group explored a typical Wuda coal field fire area (No. 18). The surface area of spontaneous combustion (in the No. 18 fire area) was ~174 748 m^2. The actual projected area of spontaneous combustion was confirmed to be 20 600 m^2.[27] Therefore, the projected area of coal beds on the surface that were participating in combustion accounted for ~12% of the total marked fire area. At 18 locations in the southern, middle, and northern sections, temperatures were recorded in the

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Table 3. GHG Emission Factors Caused by Spontaneous Coal Combustion during Different Combustion Stages

	CO_2 (g/(t s))				CH_4 (g/(t s))			
emission factor	mean value	lower	upper	standard deviation	mean value	lower	upper	standard deviation
				<200 °C				
Pattern A	0.014263	0.008500	0.022376	0.012478	0.000989	0.000062	0.001929	0.001509
Pattern B	0.008206	0.006187	0.010933	0.004177	0.000406	0.000044	0.000876	0.000676
				200−400 °C				
Pattern A	0.127233	0.034156	0.267184	0.220010	0.006146	0.001096	0.011970	0.009126
Pattern B	0.025322	0.013544	0.041629	0.023164	0.002556	0.000216	0.005875	0.004654
				400−600 °C				
Pattern A	0.555238	0.273733	0.974278	0.568106	0.009371	0.005551	0.013559	0.007022
Pattern B	0.210990	0.123727	0.308782	0.164346	0.004812	0.001738	0.009092	0.006476
				≥600 °C				
Pattern A	1.506458	1.024472	2.114004	0.887533	0.085777	0.060633	0.107708	0.039530
Pattern B	0.980497	0.691468	1.330751	0.552235	0.045193	0.031844	0.056829	0.021701

Table 4. GHG Emissions Caused by Spontaneous Combustion in the Wuda Coal Field Fire Area in 2013

combustion stage	raw coal amount ($\times 10^4$ t)	emission factor of CO_2 (g/(t s))	emission factor of CH_4 (g/(t s))	emission of CO_2 ($\times 10^4$ t/a)	emission of CH_4 ($\times 10^4$ t/a)	total GHG ($\times 10^4$ t/a CO_2 equivalent)
<200 °C	58.8	0.014263	0.000989	26.45	1.83	
200−400 °C	3.8	0.127233	0.006146	15.25	0.74	
total	62.6			41.70	2.57	95.67

combustion center of fire areas. Among these, only one point measured within the northern section of the fire area (231 °C), while all other measured points were between 60 and 180 °C (i.e., the proportion of fire areas below 200 °C was 17:18 = 94%, and that between 200 °C and 400 °C was 1:18 = 6%). Therefore, based on the exploration results obtained from the No. 18 fire area of the Wuda coal field, the total amount of raw coal participating in spontaneous combustion in the Wuda coal field was 5 215 000 × 12% = 626 000 tons. The amount of raw coal participating in spontaneous combustion occurring below 200 °C was 626 000 × 94% = 588 000 tons, and the amount of raw coal participating in spontaneous combustion within the temperature range of 200−400 °C was 626 000 × 6% = 38 000 tons.

The six fire subareas first formed in 1978 and are located in the goaf of the Suhaitu coal mine. From 1985 to 1995, small-scale coal mines grew rapidly. In this time period, 10 coal field fire subareas occurred in the Wuda coal field, because of mining in the small-scale coalmines, and eight additional fire subareas developed as a result of national mining coal mines located in Wuda. The fires were combined through the airways of the small-scale mines and, consequently, the Wuda coal field became a large-scale coal field fire area under the combined goafs within the small-scale coalmines and national, large-scale coal mines. Overall, the Wuda coal field fire area was in a middle, shallow zone of these coal measures. The coal-related spontaneous combustion fire involved middle metamorphic grade bituminous coal, such as fat, coking, and gas coals, with a volatile content of ∼30%. The sulfur content of the eight spontaneous combustion coalbeds was 0.4%−0.93%, except within the Nos. 9 and 10 coalbeds (which were ∼3%).[23] Thus, based on the experimental GHG results listed in Table 3, we adopted the GHG emission factors of Pattern A to calculate the entire GHG quantity involved in the Wuda coal field fire. It is noteworthy that, in the spontaneous combustion process, only a small portion of the area reached the high-temperature stage (>600 °C), and changes in the high-temperature proportion (generally called the coal fire center) were very small because of the limited oxygen supply, as has been observed with remote-sensing impact analysis (see ref 24 for further details). Therefore, by neglecting the impact of the areas reaching temperatures above 600 °C, we can further confirm the GHG emission factors in the Wuda fire area, which are shown in Table 4. Meanwhile, by eqs 3 and 4, we determined the annual GHG emissions in the Wuda coal field fire area, as listed in Table 4.

In June 2012, a research team from the Chinese Academy of Sciences (CAS) built experimental areas in Wuda.[28] Independently developed data-acquisition equipment was applied to the fracture environment of the coal field fire area. By constructing observation towers around the fractures, the research team synchronously collected environmental parameters, which included wind direction, wind speed, temperature, air pressure, and CO_2 concentrations at different geographic positions and altitudes. Based on the traditional aerodynamics method, the CO_2 emission fluxes in the spontaneous coal combustion fractures were calculated. The results showed that, within the experimental area, the observed CO_2 emission flux over 3 h was between 4 mg/(m^2 s) and 14 mg/(m^2 s).[28] When the annual CO_2 emissions determined herein were converted to the unit-marked fire area, the emission flux was ∼11 mg/(m^2 s). This value was close to the observed CAS value.

5. CONCLUSION

In the present study, we designed a new experiment to evaluate the greenhouse gas (GHG) emissions caused by spontaneous coal combustion. We investigated the characteristics of GHG emissions caused by spontaneous coal combustion and found that CH_4 and CO_2 emission rates resulting from spontaneous coal combustion associated with mining activity were higher than those resulting from mere surface air seepage. The main factors determining the influence of coal quality on GHG emission rates were obtained and the CO_2 and CH_4 emission factors were calculated for different stages of spontaneous combustion. Based on the coal fire area and its coal resource abundance, the obtained

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emission factors were used to evaluate the annual GHG emissions from the Wuda fire area, and the results have been compared with data obtained by monitoring the fire area.

Importantly, the quantification of the GHG emissions caused by spontaneous coal combustion remains a challenging problem worldwide. No effective method exists to estimate these GHG emission quantities. The estimation method put forward in this paper further requires additional investigation to determine its applicability and usefulness.

First, a confirmation of the different combustion stages that occur in spontaneous coal combustion areas is of great importance to estimating the associated GHG emissions. In the future, it will be easier to calculate the areas of the different stages by inverting the coal temperature by means of remote sensing and infrared technology. Second, the estimations do not exclude the carbon absorbed by the stratum and transformed at high temperatures and do not exclude the additional carbon that cannot be released to the atmosphere because of other factors. GHG emissions resulting from spontaneous coal combustion are influenced by geology, geography, and climate. The major controlling factors include the degree of fractures development, vegetation on the ground, the heat conductivity of surrounding rock, combustion depth, and seasonal variations. Thus, additional studies are necessary to determine the effects of the geological complexity of spontaneous coal combustion, coal occurrence, rock structure, and heat conductivity. Future studies in this laboratory will examine the influences of the above factors to propose a more accurate method for calculating GHG emissions caused by spontaneous coal combustion.

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Notes

The authors declare no competing financial interest.

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