



A study of emissions and marker gases from smouldering combustion in *Larix gmelinii* plantations of the Daxing'an Mountains

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Abstract Underground fires are characterized by smouldering combustion with a slow rate of spread rate and without flames. Although smouldering combustion releases large amounts of gaseous pollutants, it is difficult to discover by today's forest fire monitoring technologies. Carbon monoxide (CO), nitrogen oxides (NO_x) and sulfur dioxide (SO₂) were identified as high concentration marker gases of smouldering combustion-easily-be monitored. According to a two-way ANOVA, combustion time had a significant impact on CO and NO_x emissions; smoldering -depth

also had a significant impact on NO_x emissions but not on CO emissions. Gas emission equations were established by multiple linear regression, $C_{CO} = 156.989 - 16.626 t$ and $C_{NO_x} = 3.637 - 0.252 t - 0.039 h$.

Keywords Gas emissions · Marker gases · Influence factors · Smouldering combustion

Introduction

Underground fires in forests are smouldering combustion in humus and peat layers, with frequency of occurrence lower than above-ground forest fires, the damage of which is considerable (Huang and Rein 2017). Researchers have found that particulate emissions from smouldering combustion are much higher than from other forest combustions (Akagi et al. 2011). Suspended in the air for a long time in small sizes, particulate emissions accumulate and lead to large-scale haze (Rein 2013; Black et al. 2016).

During 1997–1998, combustion from smouldering fires in Indonesia led to large-scale hazy conditions in South-east Asia, with millions of people affected with respiratory diseases, millions of hectares of forests destroyed and billions of dollars lost (Tacconi 2003). In 2010, forest fires in Russia resulted in approximately 50 smouldering combustions, and weeks-long air pollution (Cancellieri et al. 2012). In 2015, smouldering combustion caused economic losses of \$ 16 billion in South Sumatra, Indonesia, 1.9% of GDP. Air pollution in Indonesia and surrounding countries was critical, with PM_{2.5}, fine inhalable particles, up more than 1500, causing serious damage to biodiversity and ecosystems (Atwood et al. 2016).

In addition to particulate emissions, smouldering combustion also leads to emission of various gases, with upwards

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of more than 100 types of gases and aerosols being released during the combustion process, such as carbon dioxide (CO₂), carbon monoxide (CO), methane (CH₄), ammonia (NH₃) and alkanes (Stockwell et al. 2014; Hatch et al. 2015), with CO₂ and CO being the highest emissions (Hu et al. 2018a). In addition, because the smouldering combustion is an incomplete process with an oxygen deficit, CO emissions are much higher than from ordinary forest combustion (Rein 2015).

Smouldering combustion spreads slowly without flames, making difficult to discover by standard forest fire monitoring technologies (Huang and Rein 2017). However, the high concentrations of gas emissions might be used as fire monitoring indexes (Hu et al. 2018b) to monitor the occurrence and spread of smouldering combustion.

However, research on gas emissions from smouldering combustion is still in preliminary stages, and there are only a few studies on gas varieties and their concentration that can be used for smouldering combustion monitoring (Hatch et al. 2015; Wilson et al. 2015). The purpose of this study is to explore the marker gases in the smouldering combustion process and to analyze the influence of concentrations.

Materials and method

Study area

The study area is the eastern slope of the Daxing'an Mountains, 15 km south of the Jiagedaqi region in the forest management technology extension station of Jiagedaqi (50°20'–50°23' E, 123°57'–124°0' N). Annual average temperatures range from –1 to 2 °C, with the annual effective accumulative temperature 1800–2000 °C, a frost-free period of 90–120 days, and annual rainfall 450–500 mm. The main tree species are *Quercus mongolica* (Fisch.) Turcz., *Larix gmelinii* (Rupr.) Rupr., *Betula platyphylla* Suks., *B. davurica* Pall. and *Populus davidiana* Dode.

Sample collection

Larix gmelinii plantations were the sample fields selected (Table 1), and 15 soil samples of 50 cm × 50 cm were collected. According to the classification of smouldering (Usup et al. 2004), we divided the sample depth into two groups. The depth in the shallow smouldering furnace was less than 20 cm, and the depth in the deep smouldering furnace was between 20 and 50 cm.

Sample processing

The soil samples were placed into a smouldering furnace 1.5 m long × 0.5 m wide × 0.5 m deep (Fig. 1). A far-infrared heating plate (700 °C) was used as the ignition device; a k-type thermocouple and data acquisition module were used to record the surface soil temperatures during the smouldering combustion process. The concentration of gas emissions was detected by an emission analytical instrument (ecom-J2KN) every two hours.

Statistical analysis

Statistical analysis was performed by SPSS 19.0, and two-way ANOVA used to assess the influence factors of gas emissions. Least significant difference (LSD) was used in the multiple comparisons and correlation analysis and multiple linear regression used to establish the regression equations. Statistical significance was accepted at * = $P < 0.05$, ** = $P < 0.01$. Figures were furnished by OriginPro 9.1.

Results

Smouldering combustion process

The process was carried out in the combustion furnace and the soil surface temperature measured every two hours.

Table 1 Sample plot information

NO	Tree species composition	Altitude (m)	Longitude and latitude	Diameter at breast height (cm)	Age (a)	Canopy density
1	8 <i>Larix gmelinii</i> 1 <i>Betula platyphylla</i> 1 <i>Quercus mongolica</i>	566.0	124.04°E 50.34°N	22.4	22	0.7
2	<i>Larix gmelinii</i>	406.3	124.09°E 50.318°N	22.5	27	0.5
3	9 <i>Larix gmelinii</i> 1 <i>Betula platyphylla</i>	379.7	124.11°E 50.30°N	21.2	26	0.8
4	<i>Larix gmelinii</i>	407.2	124.08°E 50.30°N	20.6	28	0.8
5	8 <i>Larix gmelinii</i> 1 <i>Betula platyphylla</i> 1 <i>Quercus mongolica</i>	553.8	124.02°E 50.35°N	14.8	16	0.7

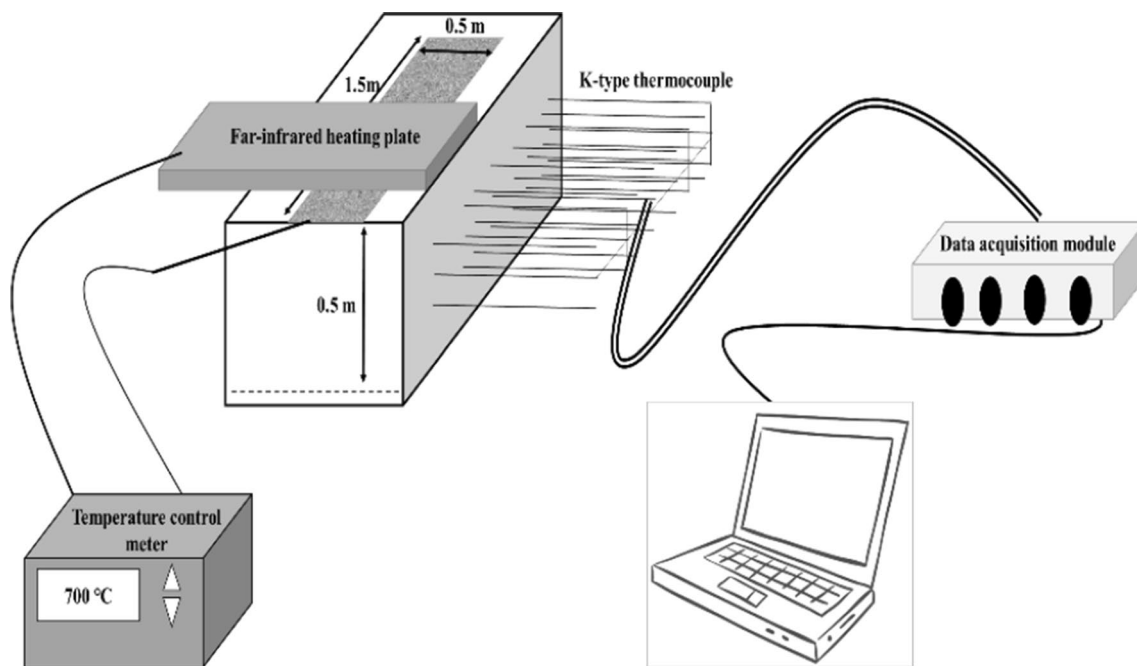


Fig. 1 Schematic diagram of the smouldering combustion experiment

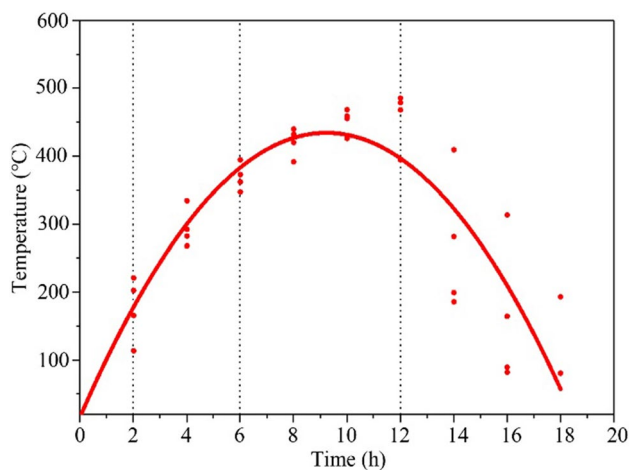


Fig. 2 Surface temperature of the soil in the smouldering combustion process

The results showed that the variations in temperature and combustion kinetics at different times were inconsistent (Fig. 2) (Hu et al. 2018a) and the process was divided into four stages: (1) Ignition Stage (0–2 h): soil temperature rose rapidly to more than 100 °C; (2) Combustion Stage (2–6 h): soil temperature rose gradually to about 400 °C; (3) Steady Stage (6–12 h): soil temperatures stayed around 400 °C to 500 °C; (4) Extinguishing Stage (after 12 h): soil temperatures decreased slowly by degrees, and no CO, NO_x and SO₂ could be detected after 12 h.

Gas emissions detection

According to the depth of the combustion, smouldering combustion could be classified as shallow (< 20 cm) and deep (> 20 cm) (Usup et al. 2004). In this study, the soil surface temperature ranged from 81.0 °C to 486.0 °C, and the average combustion temperature was 312.2 ± 19.6 °C. Carbon monoxide, NO_x and SO₂ were detected during the smouldering but surface temperatures had no significant influence on emissions ($P > 0.05$).

High concentrations of CO were detected and declined gradually as the combustion time increased; the rate of decline in the steady stage was higher than that in the combustion stage. The release of NO_x was mainly concentrated in the ignition and combustion stages but could also be detected in the steady stage. Small quantities of SO₂ could be detected in the combustion and steady stages (Fig. 3).

The highest emission of CO was 187.5 mg/m³ after 2 h, gradually dropping and by 16 h could not be detected in some experiments. The average CO concentration was 66.2 ± 8.8 mg/m³.

The emission characteristics of NO_x in various smouldering furnaces were different. Concentrations in the deep smouldering furnace ranged from zero to 3.4 mg/m³, with an average concentration of 0.9 ± 1.1 mg/m³. Levels consistently decreased and could not be detected after 6 h.

NO_x concentrations in the shallow smouldering furnace ranged from zero to 4.7 mg/m³, with an average of

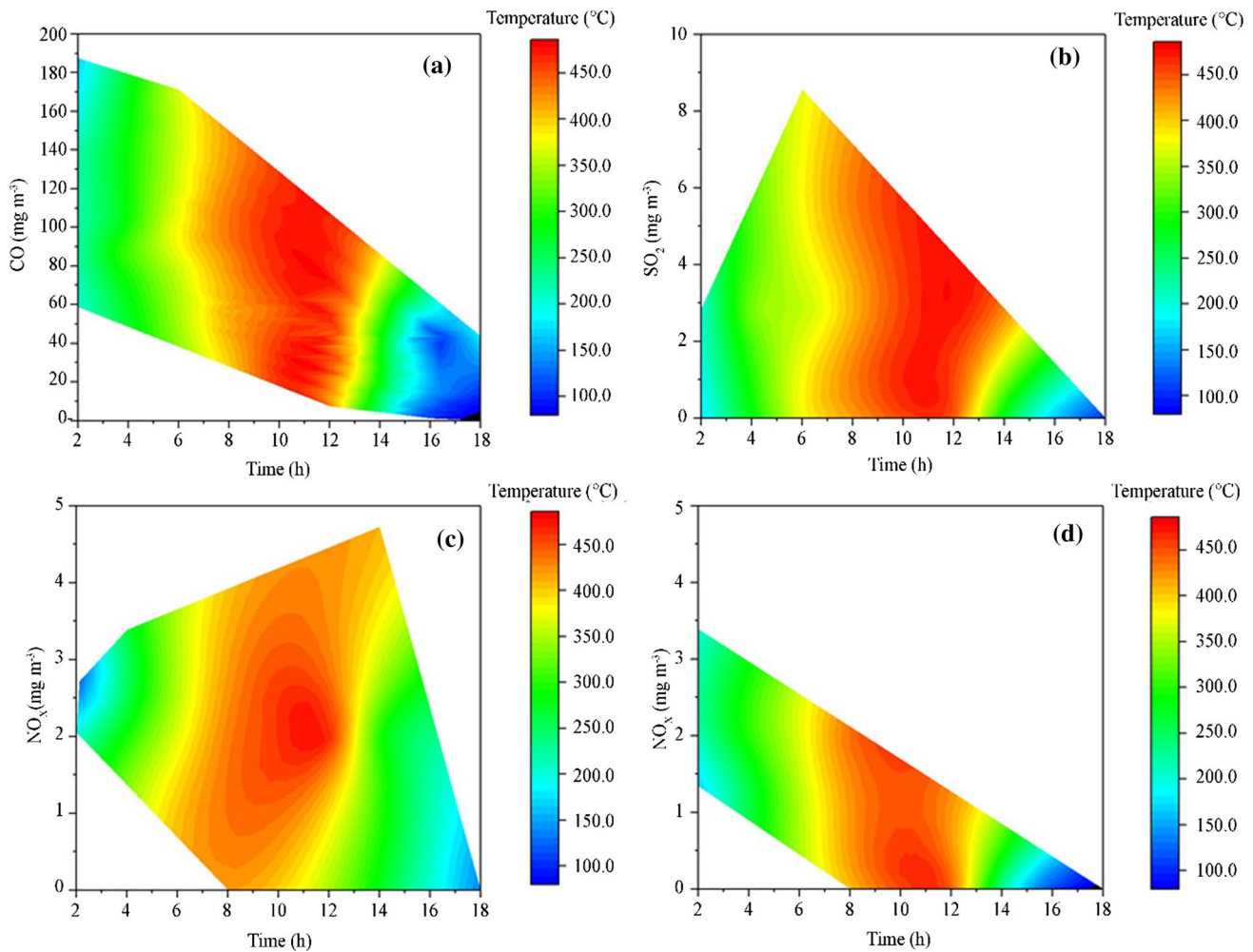


Fig. 3 Gas emissions of the smoldering combustion process: **a** CO emissions; **b** SO₂ emission; **c** NO_x emission of the shallow smoldering furnace; **d** NO_x emission of the deep smoldering furnace

$2.0 \pm 1.1 \text{ mg/m}^3$. Unlike the deep smoldering furnace, the concentration continued upward after 6 h until 12 h.

SO₂ concentrations ranged from zero to 8.6 mg/m^3 , with the average being $0.8 \pm 1.8 \text{ mg/m}^3$. A peak was detected at 6 h and then rapidly decreased. However, the data samples of SO₂ were not abundant in this study and further research is required.

Effect of combustion time and smoldering furnace depth on gas emissions

This study analyzed the effect of combustion time and smoldering furnace depth on CO and NO_x, with SO₂ not further studied for a lack of data.

The results of the analysis by two-way ANOVA showed that combustion time had a significant impact on the concentration of CO ($P < 0.01$), while the smoldering furnace depth had no significant impact ($P > 0.05$) (Table 2). As the

Table 2 The effect of combustion time and smoldering furnace depth on CO emissions by two-way ANOVA

Influence factor	<i>df</i>	<i>F</i>	<i>Sig</i>
Combustion time	8	5.773	0.001
Smoldering furnace depth	1	0.415	0.527
Combustion time*Smoldering furnace depth	6	0.376	0.886

result of difference analysis, the concentration of CO had no significant difference between ignition stage (0–2 h), combustion stage (2–6 h), and the early time of the steady stage (6–8 h) (Fig. 4).

The combustion time and depth of the smoldering furnace had a significant impact on the concentration of NO_x ($P < 0.01$) based on the results of two-way ANOVA which was different from CO factors (Table 3). This was

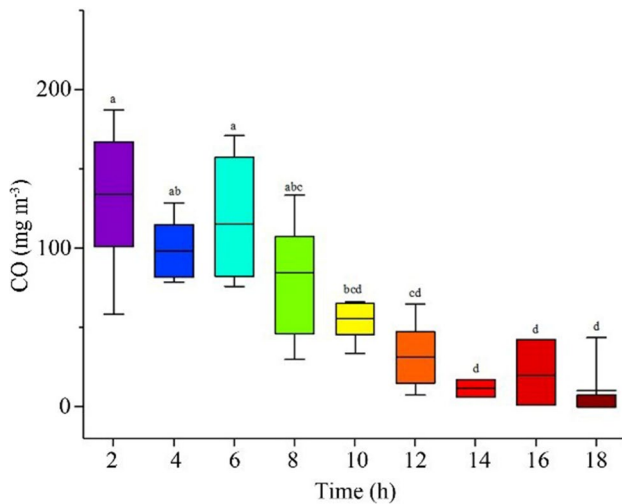


Fig. 4 Effect of combustion time on CO emissions by multiple comparison (no significant difference between values that have at least one identical letter; there is a significant difference between values that have no identical letter)

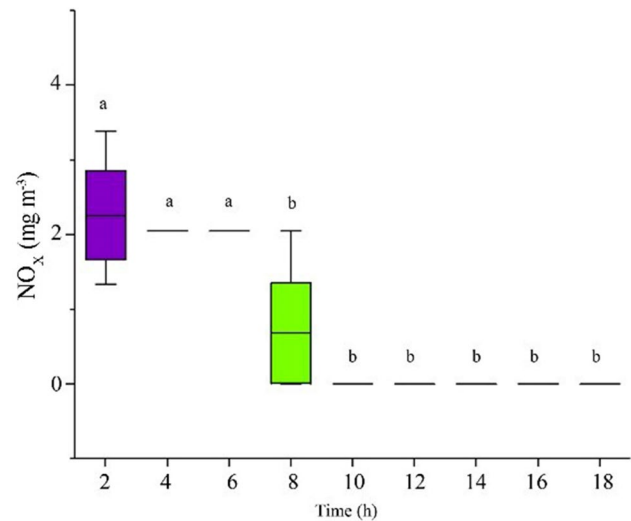


Fig. 5 Effect of combustion time on NO_x emissions in the deep smouldering furnace by multiple comparison; (no significant difference between values that have at least one identical letter; there is a significant difference between values that have no identical letter)

Table 3 Effect of combustion time and smouldering furnace depth on NO_x emissions by two-way ANOVA

Influence factor	df	F	Sig
Combustion time	8	6.870	0.000
Smouldering furnace depth	1	34.771	0.000
Combustion time*Smouldering furnace depth	8	4.153	0.002

consistent with the smouldering experiments, i.e., the NO_x emissions in the deep smouldering furnace and shallow smouldering furnace was not the same (Fig. 3).

Through further analysis, this study examined the effect of combustion time on NO_x emissions when the smouldering furnace depth was the same. Combustion time had no effect on NO_x emissions in the shallow smouldering furnace ($P > 0.05$), but did affect emissions in the deep smouldering furnace ($P < 0.05$); high concentrations of NO_x occurred at 2 h, 4 h and 6 h, with a significant difference between 2 and 6 h and the other combustion times ($P < 0.05$, Fig. 5). The main reason for the significant difference was the sharply decreased concentration of NO_x after 6 h which could not be detected after 8 h.

This study evaluated the effect of smouldering furnace depth on NO_x emissions when combustion times were the same. As the results show (Table 4), depth had no significant effect on the NO_x emissions at 2 h, 4 h, 6 h, 8 h and 18 h ($P > 0.05$); depth had a significant effect on NO_x emissions at 10 h, 12 h, 14 h, 16 h ($P < 0.05$). This was because NO_x could still be detected after 10 h of the shallow smouldering furnace but the concentration tended

to zero and could not be detected after 10 h of the deep smouldering furnace.

Regression analysis of the CO and NO_x emissions

Concentrations of CO had high negative correlation with combustion time as shown by multivariate linear regression, and with the increase of combustion time, concentration of CO gradually decreased. However, there was no correlation between the concentration of CO and the smouldering furnace depth (Table 5). The regression equation for the concentration of CO was $C_{CO} = 156.989 - 16.626 t$ ($R^2 = 0.606$), where t is the combustion time.

The concentration of NO_x was highly negatively correlated with both combustion time and smouldering furnace depth (Table 5) and the regression equation for the concentration of NO_x was $C_{NOx} = 3.637 - 0.252 t - 0.039 h$ ($R^2 = 0.458$), where t is the combustion time; h is the smouldering furnace depth.

Discussion

Combustion times and smouldering temperatures

This study divided the smouldering combustion process into the ignition stage, combustion stage, steady stage, and extinguishing stage, according to temperature variations. This is consistent with the conclusion of Hu et al. (2018a). In the ignition stage, oxygen in the soil taking part in the reaction was an important factor for accelerated combustion

Table 4 ANOVA for the effect of smoldering furnace depth on NO_x emissions

Influence factor	df	F	Sig
Smoldering furnace depth within combustion time (2 h)	1	0.09	0.767
Smoldering furnace depth within combustion time (4 h)	1	0.09	0.771
Smoldering furnace depth within combustion time (6 h)	1	0.09	0.767
Smoldering furnace depth within combustion time (8 h)	1	0.37	0.548
Smoldering furnace depth within combustion time (10 h)	1	5.85	0.021*
Smoldering furnace depth within combustion time (12 h)	1	5.85	0.021*
Smoldering furnace depth within combustion time (14 h)	1	12.98	0.001**
Smoldering furnace depth within combustion time (16 h)	1	5.85	0.021*
Smoldering furnace depth within combustion time (18 h)	1	0.37	0.548

* = $P < 0.05$, there is a significant difference; ** = $P < 0.01$, there is a highly significant difference

Table 5 Significance test of regression equations (P value)

Gas emission	Combustion time	Smoldering furnace depth	Constant
CO	0.000	0.491	0.000
NO _x	0.000	0.000	0.000

(Ohlemiller 1985) so temperatures rose rapidly in a short time. In the combustion stage, heat energy diffused into the surrounding areas and sustained the combustion without the ignition source (Hu et al. 2018a). In the steady stage, the burning areas extended to the maximum, combustion heat accumulated continually, and the smoldering combustion process reached the peak temperature (Rein et al. 2009). In the extinguishing stage, the smoldering combustion did not continue because of loss of heat energy, reduced combustibles, and shortage of oxygen, and the combustion temperatures fell until the smoldering was extinguished.

Marker gases of the smoldering combustion

There are considerable CO₂ and CO gases in the combustion for the oxidation of carbon particles and shortage of oxygen (Rein et al. 2009). High concentrations of CO were continuously detected during the process, above the ambient air quality standards of China and the background value of CO in the general forest environment (Table 6) (Su et al. 2013). CO emissions were also the predominant emissions in this study, consistent with the results of Hu et al. (2018a). Nitrogen and sulfur in the semi-decomposed and humus layers were also released as NO_x and SO₂ and the concentrations were much higher than the ambient air quality standards of China and the background value of NO_x and SO₂ in the general forest environment (Su et al. 2013).

CO, NO_x and SO₂ can be detected by a handheld device or online monitoring instrument, and therefore easier monitored in the field. Compared to other gas emissions during the smoldering combustion such as methane (CH₄),

Table 6 Concentration of marker gases in different combustion stages (mg/m³)

Combustion stage	CO	NO _x	SO ₂
Ignition stage	134.4 ± 54.2	2.3 ± 0.8	0.0
Combustion stage	97.1 ± 26.2	2.5 ± 1.1	2.3 ± 1.3
Steady stage	56.8 ± 17.7	1.0 ± 0.6 (<20 cm)	0.9 ± 1.6
Background value	0.4	5.1 × 10 ⁻³	3.9 × 10 ⁻³
Standard concentration of China	10.0	0.3	0.5

Standard concentration of China as the <ambient air quality standards of China> (GB3095-2012)

particulate matter (PM), and other organic gases (VOCs), the detection of CO, NO_x and SO₂ is relatively simple. These gases can therefore be used as marker gases to detect the occurrence of smoldering combustion. As noted earlier, smoldering combustion is difficult to monitor because it is flameless (Huang and Rein 2017), and thus marker gases could help to solve the difficulty in locating and monitoring smoldering combustion.

Carbon monoxide may be regarded as the key marker gas for large emissions, NO_x as the periodical marker gas for different emission trends in different smoldering depths. Although there was only a little SO₂ released during the smoldering combustion process, SO₂ emissions were much higher than general forest emissions. Therefore, SO₂ could be regarded as a secondary marker gas.

Influence factors of gas emissions

Heat energy spreads continuously into the deeper soil layer as the combustion time increases, so this layer also burns (Svensen et al. 2003). Soil carbon is continuously converted into CO gas and released due to a lack of oxygen. Nitrogen in the semi-decomposed and humus layers is also be transformed into NO_x and released.

The concentration of CO in the ignition stage was the highest in this study. Because smoldering combustion mainly

focused on the soil surface layer which is rich in humus and is soft and high in organic carbon, the rate of combustion increases rapidly (Hu et al. 2018a). Although combustion spreads downward with the increase in temperature, the concentration of CO in the combustion and steady stages was lower than in the ignition stage because of the reduced carbon and oxygen contents.

As noted previously, both combustion time and smoldering furnace depth had a significant effect on NO_x emissions. NO_x could only be continuously detected in the shallow smoldering furnace and was undetected after 6 h in the deep smoldering furnace. The main reason for this is that, as the smoldering combustion spreads downward, humus and total nitrogen (TN) contents decline (Knops and Bradley 2009; Yao et al. 2020), so the concentration of NO_x is extremely low after 6 h in the deep smoldering furnace. In addition, as the smoldering combustion proceeds, a large mass of combustion particles are released and leads to plugging of soil pores (Hu et al. 2018b). Gas emissions of the deep smoldering furnace are unable to spread to the surface smoothly, and NO_x was not detected after 10 h.

Conclusions

Gas emissions from smoldering combustion were investigated in a smoldering furnace. The main conclusions are as follows:

High concentrations of CO were continuously released during the smoldering combustion process. Combustion time had a significant impact on CO emissions with a peak detected in the early stage of the process. In order to improve the efficiency of monitoring smoldering combustion, CO may be considered as a key marker gas as it was highly concentrated and easily monitored.

Both combustion time and furnace depth had a significant impact on NO_x emissions. NO_x were continuously detected during the entire combustion process in the shallow smoldering furnace, and only detected during the ignition and combustion stages in the deep smoldering furnace.

The concentration of SO₂ was higher than the ambient air quality standards of China and the background value of SO₂ in forest. But SO₂ emissions were difficult to analyse in depth in this study because the sample was inadequate. Further research will be carried out in the future.

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