

Article

The Characteristics of Gas and Particulate Emissions from Smouldering Combustion in the *Pinus pumila* Forest of Huzhong National Nature Reserve of the Daxing'an Mountains

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Abstract: Smouldering combustion can emit a large amount of CO₂, CO and particulate matter (PM). Moisture content is an important factor of the emission characteristics. As the hot spot of forest smouldering combustion, the gas and particulate emissions of the Huzhong National Nature Reserve with different moisture contents are discussed herein. The emission factors (EF) of CO₂ and CO were 100.71 ± 39.14 g/kg and 11.76 ± 3.89 g/kg, respectively. The EF of PM_{2.5}, PM₄ and PM₁₀ were 87.11 ± 19.47 g/kg, 353.37 ± 159.25 g/kg and 602.59 ± 276.80 g/kg, respectively. PM_{2.5} accounted for 16.59 ± 5.25% of the PM, and PM₄ and PM₁₀ were 54.03 ± 13.46% and 91.00 ± 10.81%, respectively. There was no significant difference in the EF of CO₂ and CO with different moisture contents, nor in the EF of PM_{2.5}, but there was a significant difference in the EF of PM₄ and PM₁₀ with different moisture contents. In addition, the peak of CO₂ and CO appeared at 2~3 h; the peak of PM_{2.5} lagged behind that of PM₄ and PM₁₀. According to the regression analysis, experimental expressions were obtained for the modified combustion efficiency (MCE) and the EF of PM.

Keywords: Huzhong National Nature Reserve; *Pinus pumila*; smouldering combustion; gas emission; particulate emission



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1. Introduction

Forest smouldering combustion mainly occurs in the humus layer and the peat layer, and spreads slowly with no flame. Because of the low degree of external disturbance and strong concealment, the smouldering could last for a long time, even up to several years [1,2]. Smouldering combustion can still spread in the soil layers in winter, like Arctic and boreal areas, and could even survive below −35 °C [3]. Whenever heterogeneous oxidation occurs on the surface of the soil [4], a variety of emissions will be discharged into the environment during smouldering combustion, especially CO₂, CO and the particulate matter (PM) [5–7], and the emission of CO₂ could go up to about 400 t/ha [8]. The PM were dominated by fine particles that were mainly composed of organic constituents [9]. The carbonaceous fraction constituted 50%–70% of the PM and inorganic constituents comprised about 15% of the PM [10]. CH₄, NH₃, SO₂ and other non-methane organic compounds were also detected in smouldering combustion [11–13]. A previous study pointed out that smouldering combustion produced 130% more CO and 670% more hydrocarbons, but 15% less CO₂ and no NO_x [14], while NO and NO₂ were found with low EF values in some other studies [15,16]. In general, CO₂, CO and CH₄ were the most abundant gas types [17,18]. In addition, more than 100 types of gas were released from the smouldering combustion, and the large amounts of terrestrial carbon released in the atmosphere will accelerate climate warming [19–21].

One study found that CO₂ and CO were the largest proportion of gas emissions [22], and they were regarded as the trace gas emissions of the smouldering combustion [18,23,24]. The PM could be suspended in the air for a long time, therefore the PM would continuously

accumulate and lead to large-scale haze [25,26]. Moreover, the PM might contain a large number of harmful components, which could lead to respiratory diseases and acute inflammation [27–29]. The smouldering combustion in Indonesia during 1997–1998 released about 81 to 257 million tons of carbon, which was equivalent to 13%–40% of the mean annual global carbon emissions from fossil fuels, and led to large-scale haze, with more than 2000 mg/m³ TSP detected in Kalimantan and Sumatra [7,30]. The smouldering combustion in Russia in 2010 and in South Sumatra, Indonesia in 2015 led to large-scale of air pollution with PM_{2.5}. The pollutant standards index went up to 1500, which caused serious impacts on the ecological environment and human health [31,32]. There were about 11,000 death reports from nonaccidental causes during the 2010 smouldering combustion in Russia, with the most common cases being related to cardiovascular and respiratory issues [33]. Similarly, in North Carolina during the 2008 smouldering combustion, 2081 respiratory events and 1817 cardiac events were reported [34].

Christian et al. found that the emission factor (EF) of CO₂ could even reach 1703 g/kg from the smouldering combustion of South Sumatra tropical plantations [35]. The EF of CO₂ also rose as high as 1579 g/kg from Malaysian peatlands [36]. However, the EF of CO₂ was only about 420 g/kg from boreal peat [37]. In fact, CO₂ from the smouldering combustion of the tropical area was generally higher than those from the boreal and temperate areas [22,24,25,38]. Apart from that, tropical smouldering combustion emitted more CO, with an average EF of 248 g/kg than those of the boreal and temperate areas, with EF of 179 g/kg [22]. The difference of the carbon content in the soil might be the significant factor for the EF of CO₂ and CO [39–41].

The PM from the smouldering combustion ranged from PM_{0.1} to PM₁, PM_{2.5} and PM₁₀ [42]. The PM of boreal and temperate areas might be higher than that of tropical areas [25,43,44]. The average EF of PM_{2.5} was 19.17 g/kg in boreal and temperate areas, but was 17.3 g/kg in tropical areas [22]. The EF of PM_{2.5} were 16.9 g/kg during flaming combustion and 38.8 g/kg during smouldering combustion [45]. There are several levels of PM (PM₁ to TSP in general), but the research results concerning the EF of PM with different levels were inconsistent. PM_{2.5} was found to be the largest proportion of PM from smouldering combustion in North Carolina, but PM₁₀ was the most emitted PM according to Akagi et al. [15].

The Daxing'an Mountains are one of the most sensitive regions to global climate change, and the impact of the change in climate could last for a long time [46]. The region is also a hot spot for all kinds of forest fires [47]. The frequency and intensity of forest fires in this region have been on the rise in recent years as a result of global warming [48]. Huzhong National Nature Reserve (HNNR) is located in the core area of the Daxing'an Mountains where the forest vegetation remains in the original state, and as the largest cold temperate coniferous forest ecosystem nature reserve of China, HNNR has great practical and scientific value in the global carbon cycle, biodiversity and environmental protection. HNNR is an important distribution area of *Pinus pumila*, which is commonly used in slope greening and soil and water conservation for its resistance to cold, drought and leanness, and it also serves as an important habitat for rare wild animals and as an economic plant for food and medicine [49]. The smouldering combustion will destroy the physical and chemical composition of the soil, vegetation roots and ecological balance. Moreover, the PM will deposit on the surface of leaves for a long time, and cause a serious impact on the existent environment of *Pinus pumila* [50]. It is difficult for *Pinus pumila* to recover from fires and return to its climax community by natural succession because of its weak seed dispersal ability.

However, studies on the smouldering combustion of *Pinus pumila* are not sufficient. Previous studies on the emissions from smouldering were mostly carried out on laboratory or commercial peat under microcosmic scale, which could hardly reflect the emission characteristics of gas and particles in the actual smouldering process. The EF is an important indicator for the analysis of emissions during the combustion [51], and EF could simplify the vast complexity of natural conditions [37]. Moisture content has been found to be the

most important factor of the ignition and the spread of smouldering combustion [52,53]. Therefore, our research aimed to study the EFs of gas and particulate, and the emission characteristics with different moisture contents in order to determine the actual smouldering situation of *Pinus pumila* in HNNR and provide reference for the smouldering monitoring of *Pinus pumila* in follow-up studies.

2. Methods

2.1. Study Area

HNNR of the Daxing'an Mountains ($122^{\circ}42'14''\sim 123^{\circ}18'05''$ N, $51^{\circ}17'42''\sim 51^{\circ}56'31''$ E) is located in the Daxing'an Mountains of Heilongjiang Province (Figure 1), China, and the northeast slope is between the main vein of the Daxing'an Mountains and the Yilehuli Mountains. HNNR, one of the most typical and intact cold temperate coniferous forest ecosystems in China, is divided into 3 major zones: the core area (54,087 ha), the buffer area (45,493 ha) and the experimental area (67,633 ha). The annual average temperature is -4.3°C and the rainfall is 350~500 mm, with short summers and long winters. The high temperature occurs in spring and autumn, along with low humidity and strong wind that can easily cause forest fires. Forest vegetation features cold and temperate coniferous forest, *Larix gmelina* being the dominant tree species. The main coniferous species are *Larix gmelina*, *Pinus sylvestris* var. *mongolica*, *Picea koraiensis* and *Pinus pumila*. The latter mostly grows in the high-altitude area and constitutes the unique subalpine landscape and the undergrowth shrub of the sparse coniferous forest in the cold temperate zone of the mountains. The main broad-leaf species are *Betula platyphylla*, *Populus davidiana* and *Chosenia arbutifolia* [54,55].

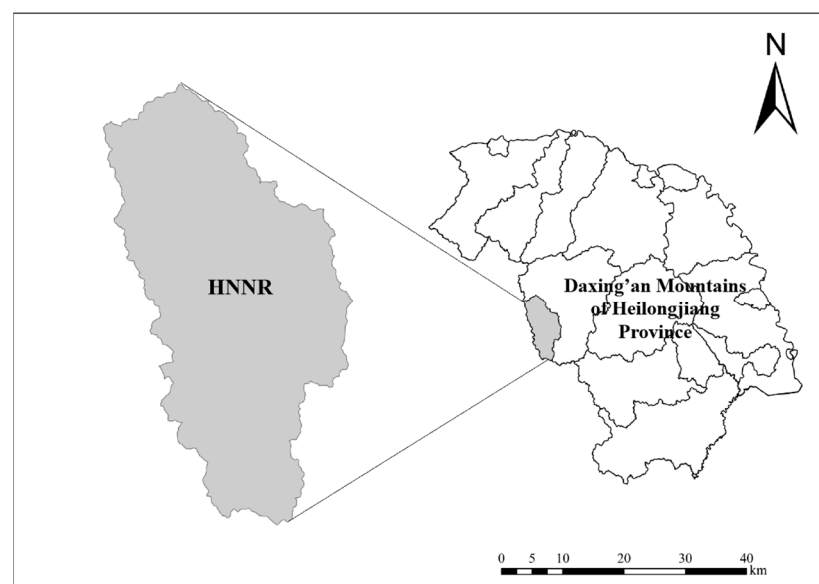


Figure 1. Map of the study area.

2.2. Experimental Method

The *Pinus pumila* forest that had been attacked by the smouldering combustion in HNNR was selected as the study area. Five sample points were randomly selected in the unfired area. We dug quadrats of $0.5\text{ m} \times 0.5\text{ m}$ and took all the subsurface smoldering combustibles in the quadrats to the laboratory (Figure 2a). We mixed the five samples evenly, which were used to represent the soil conditions of the *Pinus pumila*. Based on the results of the field measurement, the samples were placed in a cool and ventilated space to dry naturally (drying them in an oven if necessary), and the MC was measured every 24 h using a rapid moisture meter until the MC reached 2%, 12% and 22%. The soil samples were then sealed for the smouldering experiments. Three duplicate tests were set at each MC gradient and at each point. The samples after smouldering combustion are shown in Figure 2b.

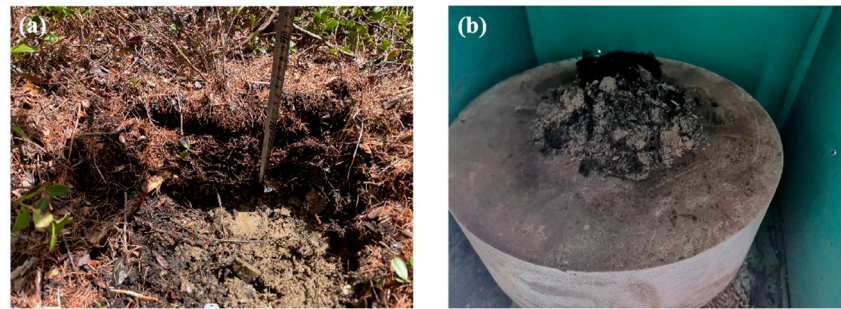


Figure 2. Soil sample before and after the smouldering combustion experiment: (a) before combustion; (b) after combustion.

The smouldering combustion installation is shown in Figure 3. The smouldering furnace was placed into the tank (35 cm in length, 35 cm in width, 90 cm in height) with a monitoring window, which could be opened according to the requirement of measurement. The concentrations of CO₂ and CO were detected using the multifunctional flue gas analyzer (ecom-J2KN), and the concentrations of PM_{2.5}, PM₄, PM₁₀ and TSP were monitored using the particulate monitor (MetOne831). The measurement range of MetOne831 is 0~1000 µg/m³, with a measurement accuracy of 0.1 µg/m³. The sampling time could be up to 6 s/time and was collected 10 times for the mean value. The concentration of the emissions was detected every 30 min according to the results of the preliminary experiment.

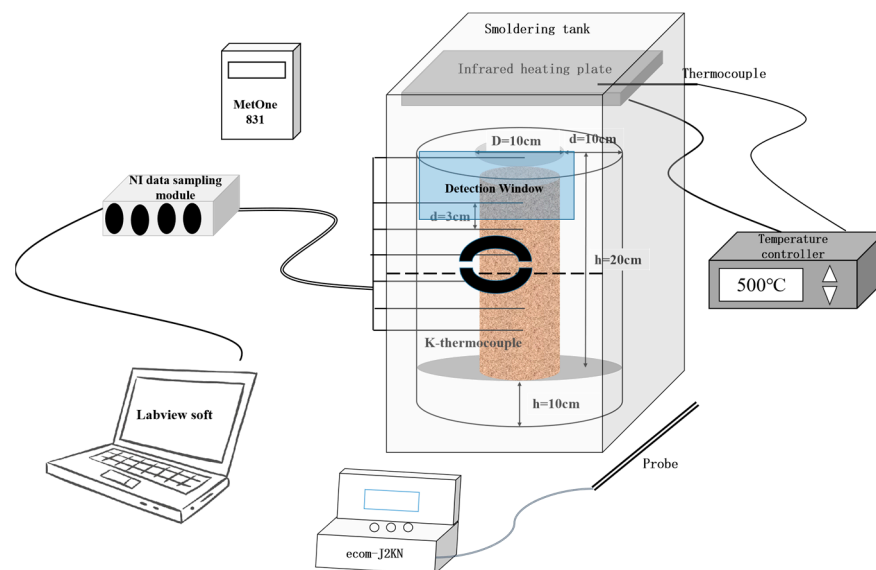


Figure 3. Schematic diagram of the experimental installation.

2.3. Emission Factor

The emission factor (EF) in this paper was calculated as:

$$EF_i = \frac{M_i}{M_{fuel}} \quad (1)$$

where EF_i is the emission factor of the combustion emissions i , g/kg; M_i is the total mass of the combustion emissions i , calculated by the concentration of the emissions i and the volume of the seal box, g; and M_{fuel} is the total mass of the combustion soil in the smouldering furnace, calculated by the soil quality difference in the smouldering furnace before and after smouldering combustion, kg.

2.4. Modified Combustion Efficiency and Emission Ratio

The modified combustion efficiency (MCE) was calculated as:

$$MCE = \frac{C_{CO_2}}{C_{CO_2} + C_{CO}} \quad (2)$$

where MCE is the modified combustion efficiency; C_{CO_2} is the mass concentration of CO_2 , mg/m^3 ; and C_{CO} is the mass concentration of CO , mg/m^3 . Generally, flame combustion has an MCE higher than 0.99, while the smouldering combustion has an MCE between 0.75 and 0.84 [15].

The emission ratio (ER) in this paper was calculated as:

$$ER_{CO/CO_2} = \frac{C_{CO}}{C_{CO_2}} \quad (3)$$

where ER_{CO/CO_2} is the ratio of CO to CO_2 ; C_{CO} is the mass concentration of CO , mg/m^3 ; and C_{CO_2} is the mass concentration of CO_2 , mg/m^3 . ER_{CO/CO_2} can reflect the conversion trend from the smouldering combustion to the flame combustion.

2.5. Statistical Analysis

Statistical analysis was performed by SPSS 19.0. Correlation analysis and nonlinear regression were used to establish the regression equations. Statistical significance was accepted at $* = p < 0.05$, $** = p < 0.01$. The data are shown as mean \pm standard deviation and the confidence interval was calculated at 95% confidence level of normal distribution. The figures were furnished by Origin-Pro 9.1.

3. Results

3.1. Main Gas Emissions in the Smouldering Combustion

3.1.1. EF of CO_2 and CO

In the smouldering combustion of this study, the average EF of CO_2 was 100.71 ± 39.14 g/kg, the EF of CO was 11.76 ± 3.89 g/kg and the EF of CO_2 and CO with different MCs are shown in Table 1. The lowest EF of CO_2 and CO occurred at 12% MC. There were no significant differences in the EF of CO_2 ($p = 0.906$, $p > 0.05$) and the EF of CO ($p = 0.913$, $p > 0.05$) with different MCs.

Table 1. The EF of CO_2 and CO from the smouldering combustion (g/kg).

MC (%)	CO_2	CO
2	107.27 ± 39.83	12.10 ± 3.65
12	91.63 ± 29.36	10.87 ± 1.07
22	103.23 ± 59.01	12.30 ± 6.65

3.1.2. Emission Characteristics of CO_2 and CO

The concentrations of CO_2 and CO with different MCs both showed a rising trend followed by a decline with the ongoing smouldering combustion (Figure 4). With the increasing MC, it took a shorter amount of time for CO_2 and CO to reach the peak concentration. The peak concentration for CO_2 and CO were detected at 3 h with 2% MC, 2.5 h with 12% MC and 2 h with 22% MC.

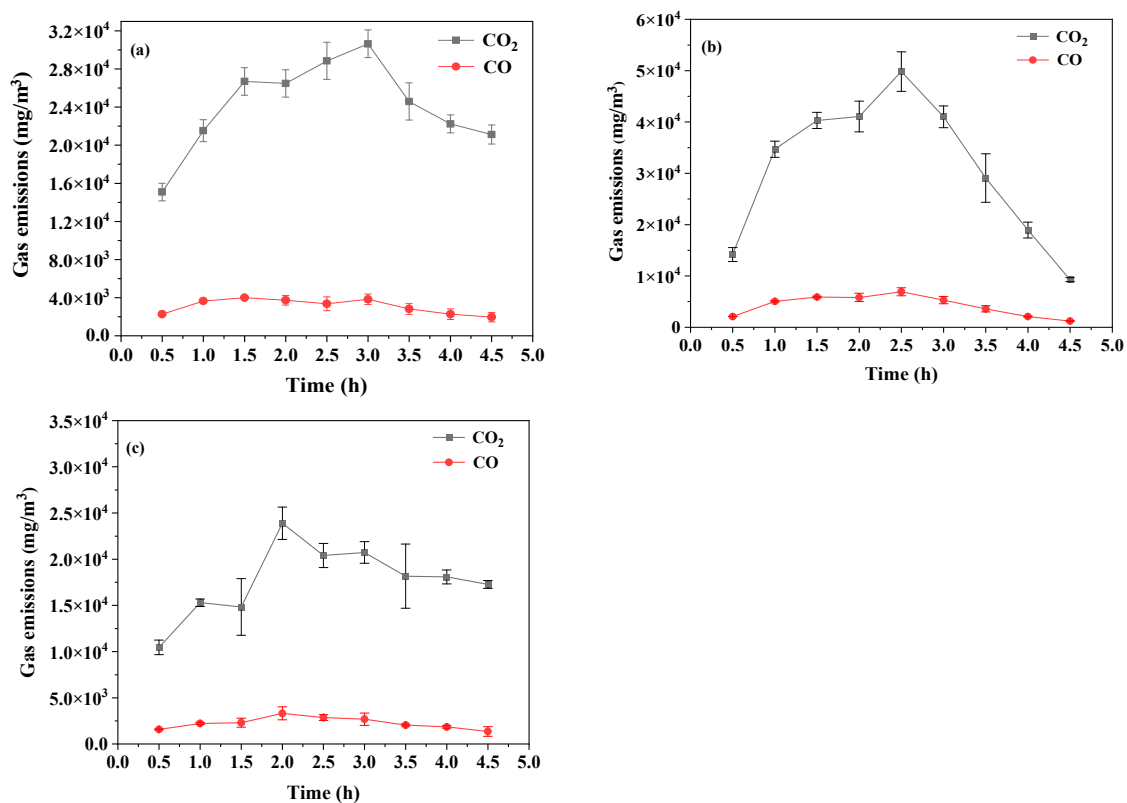


Figure 4. Gas emissions from the smouldering combustion: (a) gas emissions at 2% MC; (b) gas emissions at 12% MC; (c) gas emissions at 22% MC.

When the MC was 2%, the average concentration of CO_2 was $24,138.83 \pm 4720.71 \text{ mg/m}^3$ and the peak concentration was $30,642.86 \text{ mg/m}^3$; the average concentration of CO was $3096.68 \pm 775.79 \text{ mg/m}^3$ and the peak concentration was 3992.17 mg/m^3 . When the MC was 12%, the average concentration of CO_2 was $30,933.35 \pm 13,970.85 \text{ mg/m}^3$ and the peak concentration was $49,834.75 \text{ mg/m}^3$; the average concentration of CO was $4214.74 \pm 2026.51 \text{ mg/m}^3$ and the peak concentration was 6914.33 mg/m^3 . When the MC was 22%, the average concentration of CO_2 was $17,681.63 \pm 3902.97 \text{ mg/m}^3$ and the peak concentration was $23,891.46 \text{ mg/m}^3$; the average concentration of CO was $2249.69 \pm 627.76 \text{ mg/m}^3$ and the peak concentration was 3315.95 mg/m^3 . At 12% MC, the average and peak concentration of CO_2 and CO were the highest, and the variation range of the gas emissions was the largest, while at 22% MC, the average and peak concentration of CO_2 and CO were the lowest, and the variation range of the gas emissions was the smallest.

3.2. Particulate Emissions in the Smouldering Combustion

3.2.1. EF of PM

In this study, the average EF of $\text{PM}_{2.5}$, PM_4 and PM_{10} were $87.11 \pm 19.47 \text{ g/kg}$, $87.11 \pm 353.37 \pm 159.25 \text{ g/kg}$ and $602.59 \pm 276.80 \text{ g/kg}$, respectively, as is shown in Table 2. With different MCs, there were no significant differences in the EF of $\text{PM}_{2.5}$ ($p = 0.347$, $p > 0.05$), but there was a significant difference in the EF of PM_4 ($p = 0.011$, $p < 0.05$) and PM_{10} ($p = 0.003$, $p < 0.05$). The lowest EF of $\text{PM}_{2.5}$, PM_4 and PM_{10} all occurred at 12% MC; the EF of $\text{PM}_{2.5}$ at 22% MC was lower than that at 2% MC, but the EF of PM_4 and PM_{10} at 22% were higher than that at 2% MC.

Table 2. The EF of PM from the smouldering combustion (g/kg).

MC (%)	PM _{2.5}	PM ₄	PM ₁₀	TSP
2	100.34 ± 24.05	290.54 ± 29.91	547.29 ± 4.91	549.35 ± 32.80
12	70.64 ± 19.09	219.80 ± 69.65	327.76 ± 44.82	333.96 ± 42.77
22	90.35 ± 6.26	594.76 ± 18.46	932.72 ± 76.93	950.70 ± 90.35

Among all the particulate emissions (TSP) collected in the smouldering experiments, the average EF of PM_{2.5} accounted for $16.59 \pm 5.25\%$, while the proportion for PM₄ and PM₁₀ were $54.03 \pm 13.46\%$ and $91.00 \pm 10.81\%$, respectively. As can be seen in Figure 5, there was a significant difference in the EF proportion of PM_{2.5} ($p = 0.007$, $p < 0.05$), but there were no significant differences in the EF proportion of PM₄ ($p = 0.096$, $p > 0.05$) and PM₁₀ ($p = 0.355$, $p < 0.05$) with different MCs.

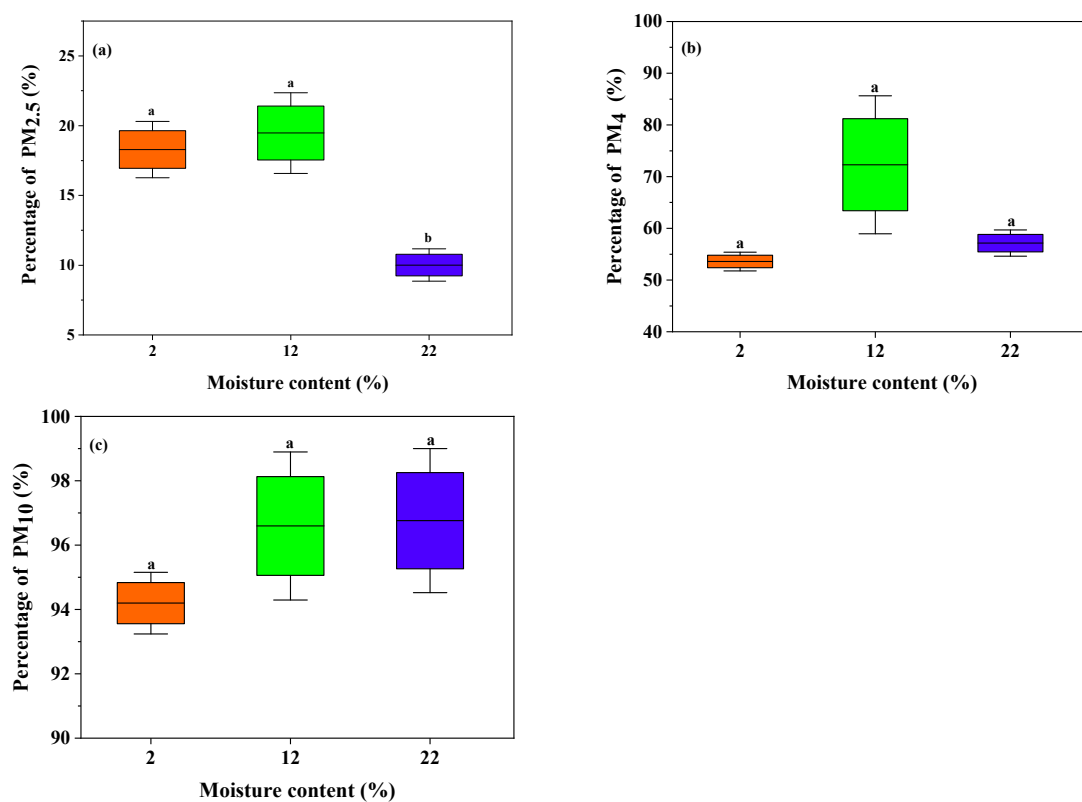


Figure 5. Comparison of the EF proportion of PM with different moisture contents: (a) EF proportion of PM_{2.5}; (b) EF proportion of PM₄; (c) EF proportion of PM₁₀. The letters a and b mean that if there is any same letter in the figure, the difference is not significant.

3.2.2. Relationship between EF and MCE

As a parameter that is commonly used to study the combustion state of biomass, the MCE suggests the flame combustion when it reaches 0.99, and the smouldering combustion when it is 0.75~0.84 [15]. There is a correlation between the MCE and EF of gas emissions according to previous studies [45,56]. Therefore, we researched the relationship between the MCE and EF of the PM.

In the smouldering combustion in this study (Figure 6), when the MCE was 0.75~0.85, the EF of PM_{2.5}, PM₄ and PM₁₀ varied in the same way. The peak values of EF were all detected at MCE 0.81~0.82, and the EF gradually decreased with the increase of MCE.

Regression analysis showed that the relationship between the MCE and EF of PM could be fitted by cubic functions (Equations (4)–(6), $p < 0.05$), but the R^2 values were all below 0.80.

$$\ln EF_{PM_{2.5}} = -294.50MCE^3 + 351.31MCE^2 - 69.14 \quad (R^2 = 0.688) \quad (4)$$

$$\ln EF_{PM_4} = -248.68MCE^3 + 397.21MCE^2 - 56.84 \quad (R^2 = 0.655) \quad (5)$$

$$\ln EF_{PM_{10}} = -151.46MCE^3 + 177.82MCE^2 - 29.88 \quad (R^2 = 0.682) \quad (6)$$

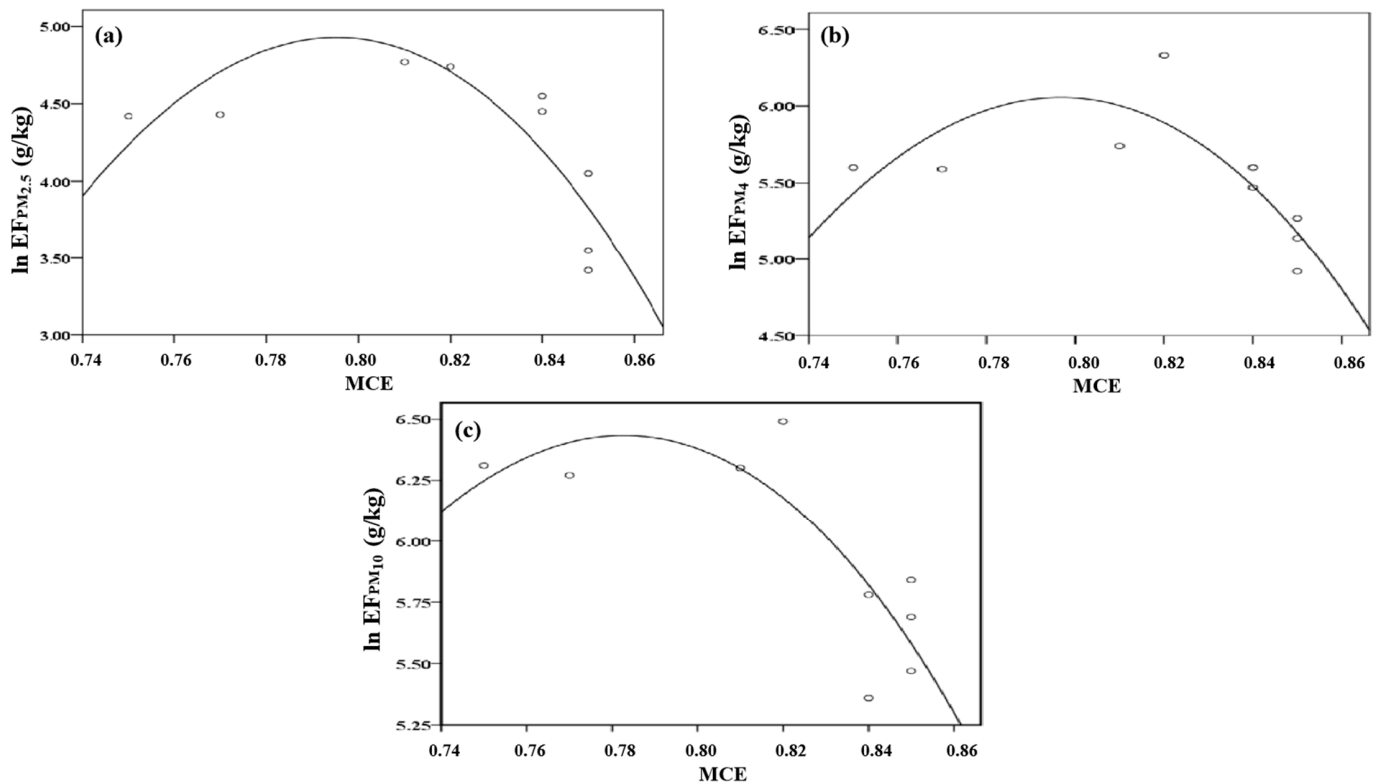


Figure 6. Relationship between EF and MCE: (a) fit between $EF_{PM_{2.5}}$ and MCE; (b) fit between EF_{PM_4} and MCE; (c) fit between $EF_{PM_{10}}$ and MCE.

3.2.3. Emission Characteristics of PM

The variation of particulate emissions with combustion time showed that the particulate emissions first increased and then decreased, and the variation trends of $PM_{2.5}$, PM_4 and PM_{10} were basically the same (Figure 7). The peak concentrations of $PM_{2.5}$, PM_4 and PM_{10} were 1,580,100 mg/m^3 , 6,126,300 mg/m^3 and 8,687,200 mg/m^3 , respectively, at 2% MC; the peak concentrations of $PM_{2.5}$, PM_4 and PM_{10} were 1,585,400 mg/m^3 , 4,882,400 mg/m^3 and 6,621,800 mg/m^3 , respectively, at 12% MC; the peak concentrations of $PM_{2.5}$, PM_4 and PM_{10} were 1,324,200 mg/m^3 , 11,476,900 mg/m^3 and 22,974,400 mg/m^3 , respectively, at 22% MC.

The combustion time for the peak concentration of $PM_{2.5}$ lagged behind that of PM_4 and PM_{10} , with the time lag being 0.5 h at 2% MC and 12% MC, and 1.5 h at 22% MC; the concentration of the particulate emissions decreased significantly after 2.5 h, and although there was a small rise at 4 h, the concentration continued to decrease after 4 h until the smouldering combustion went out.

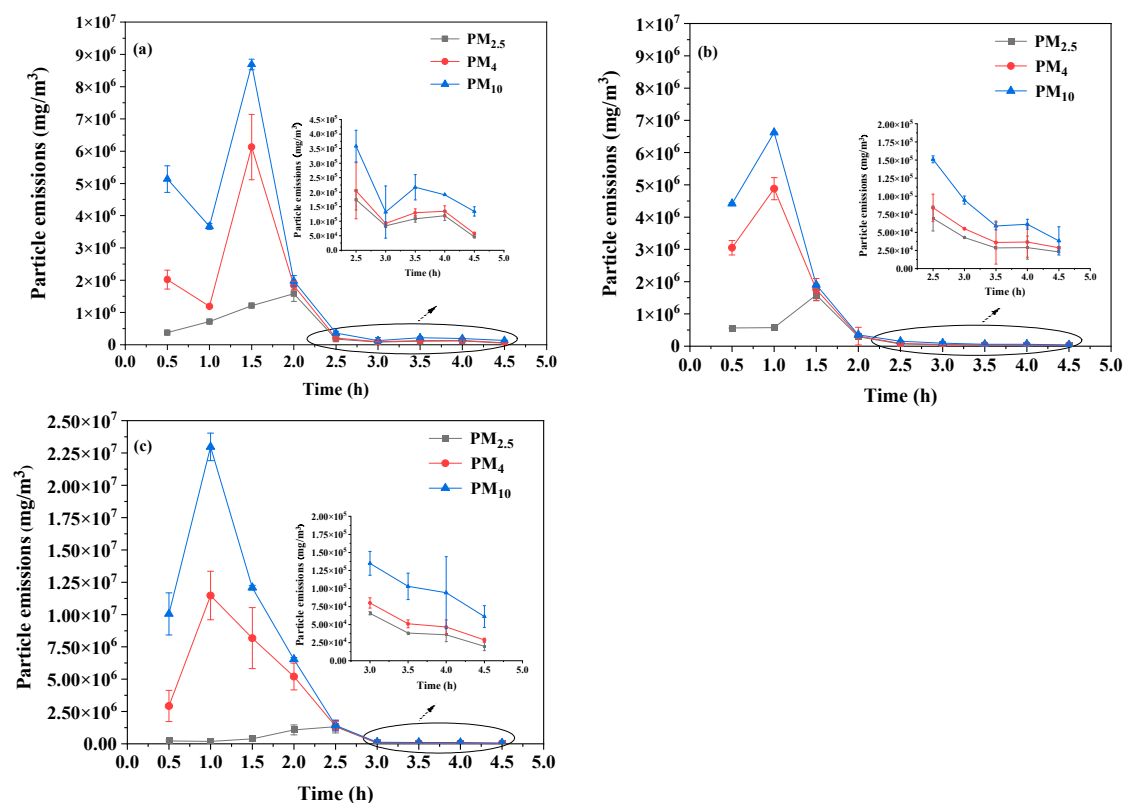


Figure 7. Particle emissions from the smouldering combustion: (a) particle emissions at 2% MC; (b) particle emissions at 12% MC; (c) particle emissions at 22% MC.

4. Discussion

4.1. EF of the Emissions and Variation Trend

Hu et al. [57] found that the EF of CO₂ ranged from 3056~3320 g/kg to 3042~3452 g/kg, and the EF of CO ranged from 201~432 g/kg to 171~227 g/kg from the flame combustion in the *Pinus pumila*-*Larix gmelinii* forests and *Pinus pumila* forests of the Huzhong region in the Daxing'an Mountains. Chang et al. [58] found that the EF of CO₂ was 1393~3328 g/kg and the EF of CO was 75~195 g/kg from the flame combustion of different forest types in the Daxing'an Mountains. The studies on the main forest types in the Daxing'an Mountains showed that, in flame combustion, the average EF of CO₂ of branches (leaves) was 1509.25 (1496.59) g/kg and that of CO was 181.51 (180.17) g/kg [59]. All the EF of CO₂ and CO in the studies above were higher than our results. This is consistent with the report that flaming combustion emitted more gas emissions than smouldering combustion [60]. However, the ER_{CO/CO2} values in the studies above were lower than those in our study, which resulted from the lack of oxygen and incomplete combustion, and was consistent with the reports of Bonsang et al. [61].

Reisen et al. [45] found that the EF of PM_{2.5} was 38.8 g/kg from the smouldering combustion. Hu et al. [6] pointed out that the average EF of PM_{2.5} was 23.12 ± 1.19 g/kg at the stable phase of smouldering. Hu et al. [60] reported that the peak EF of PM_{2.5} was about 20 g/kg. All the results were lower than the average EF of PM_{2.5} in this study (87.11 ± 19.47 g/kg). The higher EF of PM_{2.5} mainly related to the large carbon content of the soil samples from HNNR. In addition, the soil samples of this study were from the area with no experience of smouldering combustion in recent years, so the emissions were higher than the samples that have experienced fires [25,62,63].

Consistent with previous studies [25,64], the R values for the regression of MCE and EF were lower (below 0.80) and the curves cannot adequately describe the process of PM emissions. Considering the complex mechanism of smouldering combustion, temperature, particle size, porosity and other factors will also affect combustion. Moreover, the

lower range of MCE during the smouldering combustion would also affect the regression results [25]. Further studies on the EF of PM remain necessary since little research has been conducted into particulate emissions during the smouldering combustion.

The peak particulate emissions were detected in the early smouldering combustion period (before 2.5 h); the peak CO₂ and CO were also detected in 2~3 h. Gas and particulate emissions both indicated a decreasing trend after 3 h. According to the analyses, the content of O₂ in the soil was relatively higher at the beginning phase of smouldering, and the major reactions, including a pyrolytic reaction and a redox reaction, produced a mass of heat and released plenty of gas and particulates including coke simultaneously [65]. The results are also consistent with the finding that PM emissions were mainly in the ignition and spread stage at different MCs [60]. The heat then diffused outwards continuously with the spread of the smouldering combustion, the combustibles decreased gradually, and the reaction rates of the pyrolytic reaction and the redox reaction went down [66,67], as did the gas and particulate emissions.

4.2. Composition of Particulate Emissions in the Smouldering Combustion

Hu et al. [6] found that PM₁₀ accounted for up to 99% and PM_{2.5} accounted for 87% of particulate emissions from the smouldering, and their proportions were close. However, the proportion of PM_{2.5} was much lower than that of PM₁₀ in this study. The soil types, smouldering temperature and detection methods could all affect the composition of the particle emissions. There should be an in-depth discussion on the factors that influence the composition of particulate emissions in subsequent research [22,68].

Although the proportion of PM_{2.5} was lower in this study, the large amounts of PM₁₀ could still lead to respiratory diseases, conjunctivitis and dermatitis [69]. Uttajug et al. [70] also reported that the concentration of PM₁₀ during forest fire or vegetation burning has a significant influence on the occurrence of respiratory diseases. In addition, PM₁₀ suspended in the air will deposit on the surface of plant leaves through adsorption or stagnation and affect the surface morphology and physiological parameters of plants, resulting in damage to the cuticle and diminution of the photosynthetic rate and the stomatal numbers in leaves [48,71,72].

4.3. Effect of MC on Gas and Particulate Emissions

Preheating, drying, pyrolysis and oxidation were the main process of the smouldering combustion [60], and CO₂ and CO were mainly emitted by a char oxidation process [6]. Although the gas emissions decreased with the increase of MC in the research of Huang et al. [1,73], there were no significant differences in the EFs of CO₂ and CO in this study. The main reason was the lower MC of the soil samples, while the MC was up to 160% in the previous studies [60], resulting in a very serious heat consumption that would significantly affect the gas emissions [74].

The EF of PM₄ and PM₁₀ at 22% MC were about twice as much as those at 2% MC. The main reason is that the specific heat capacity of water is large, therefore the increase of MC will weaken the heat accumulation during combustion, and the evaporation process will increase the heat loss, leading to the intensification of incomplete oxidation and particulate emissions [75]. Additionally, water molecules could intensify the carbonization of carbonaceous organic material through an aromatization reaction and increase the concentration of particulate emissions [76].

Different from PM₄ and PM₁₀, the MC had no significant effect on the EF of PM_{2.5}. The main reason was the fine particle aggregation of the water droplets and PM_{2.5} particles by the inertial collision and mixing reaction. Then the ongoing smouldering led to the decrease of MC and the particle aggregation also gradually decreased. The peak concentration of PM_{2.5} lagged behind PM₄ and PM₁₀, and the hysteresis was more obvious at 22% MC. There was a significant difference in the proportion of PM_{2.5} emission factors at different MCs; the proportion of PM_{2.5} emission factors went down significantly at 22% MC. All the results could confirm the inference above.

Compared with other MCs, the characteristics of gas and particulate emissions at 12% MC are worthy of attention. The EF of CO₂ and CO were the lowest at 12% MC (Table 1), and the smouldering combustion was evident with an average MCE of 0.81, lower than that at 2% and 22% MC. The EF of PM (PM_{2.5}, PM₄, PM₁₀, TSP) at 12% MC were all lower than those at other MCs (Table 2). In terms of the composition of PM, the proportions of PM_{2.5} and PM₄ at 12% MC were higher than those at other MCs, but the proportion of PM₁₀ was close to those at other MCs. It can be inferred that particulates with smaller sizes are more likely to be released at 12% MC. Combined with the discussion above on MC and PM, it could be concluded that being affected by the multiple reactions of evaporation, incomplete oxidation, carbonization and aggregation, 12% MC is a turning point in this study on the characteristics of gas and particulate emissions. Based on 12% MC, follow-up studies should be conducted to refine the MC gradient and to further study the effect of refined MC on the emissions.

5. Conclusions

During the smouldering combustion, the average EF of CO₂ was 100.71 ± 39.14 g/kg and the average EF of CO was 11.76 ± 3.89 g/kg. The average EF of PM_{2.5}, PM₄ and PM₁₀ was 87.11 ± 19.47 g/kg, 353.37 ± 159.25 g/kg and 602.59 ± 276.80 g/kg, respectively; the proportion of PM₁₀ was more than 90% and PM_{2.5} was less than 20%. The MC had no significant effect on the EF of CO₂ and CO; MC also had no significant effect on the EF of PM_{2.5}, but had a significant effect on the EF of PM₄ and PM₁₀. The peak concentrations of CO₂ and CO were detected at 2~3 h; the peak of the particulates was detected before 2.5 h and the peak of PM_{2.5} lagged behind that of PM₄ and PM₁₀.

Due to the limitations of the experimental conditions, CH₄ and other organic gases were not examined in this study. As one of the trace emissions in the smouldering combustion, CH₄ should be considered in future studies. Soil conditions could also affect the emissions and samples from more areas should be studied in order to accomplish a more comprehensive analysis of smouldering combustion in the Daxing'an Mountains.

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Abbreviations

HNMR	Huzhong National Nature Reserve
MC	Moisture content
EF	Emission factor
MCE	Modified combustion efficiency
ER	Emission ratio

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