Original Research

Emission Characteristics of Formaldehyde and Particulate Matter in Side-Stream Smoke Emitted from Cigarettes in an Environmental Chamber

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Abstract

Side-stream smoke from cigarettes contains formaldehyde, inhalable particulate matter and other pollutants, which seriously affects human health. The formaldehyde and inhalable particles in side-stream smoke emitted from four common brands of cigarettes in an environmental chamber were monitored by using a hand-held formaldehyde detector and a particulate detector. The concentration of the pollutants was also studied under conditions of closeness, natural ventilation and mechanical ventilation. The results show that the burning rates of four brands of cigarettes range from 2.65 to 3.0 g/h in the enclosed environment chamber and the peak concentrations of formaldehyde emissions range from 1.02 to 1.72 mg/m³. The release process of particulate matter can be divided into release diffusion stage and sedimentation stage. The peak concentrations of PM_{2.5} and PM₁₀ is 0.282 and 3.382 mg/m³, respectively. The sedimentation rates of PM_{2.5} and PM₁₀ are 0.0180 and 0.0289 mg/m³.h⁻¹, respectively. It is found that the removal rate of formaldehyde and particulate matter is positively correlated with the ventilation rate. The fitting correlation coefficients of formaldehyde concentration during the rise and fall phases under closed conditions are greater than 0.968 and 0.979, respectively, and the coefficients of PM_{2.5} and PM₁₀ are greater than 0.904 and 0.929, respectively.

Keywords: formaldehyde, fine particles, side-stream smoke, environmental chamber

Introduction

Formaldehyde and particulate matter are the two most common air pollutants. Indoor formaldehyde mainly comes from decoration paint, furniture board

^{[1].} They can cause a series of diseases such as red cell deformation [2], chronic respiratory diseases, fetal malformation, leukemia and nasopharyngeal cancer. Formaldehyde is classified as a carcinogen by the International Agency for Research on Cancer [3-5]. There are many sources of particulate matter, coal-fired power plants [6], heavy industrial production [7] and the use of fuel vehicles [2], can emit a lot of particulate matter. Particulate pollution can also harm human

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health, and the degree of harm is mainly affected by particle size, concentration and composition [8, 9]. Particulate matter is divided into inhalable particulate matter (particle size $\leq 10\mu$ m, PM₁₀), fine particulate matter (particle size $\leq 2.5\mu$ m, PM_{2.5}) and ultra-fine particulate matter (particle size $\leq 0.1\mu$ m, UFPS, close to the particle size of the virus [10]). Studies have shown that smaller particles do greater harm to human health [11-13].

Cigarette smoke is one of the main pollution sources that affect indoor air quality and harm human health [14, 15], it is mainly divided into mainstream smoke and side-stream smoke. The mainstream smoke refers to the smoke inhaled by smokers through the filter, while the side-stream smoke refers to the smoke released by the cigarette end when the cigarette is burned [16]. The smoke emitted by a cigarette contains more than 4000 toxic substances, including more than 3800 substances harmful to human health, such as formaldehyde, fine particles [17], polycyclic aromatic hydrocarbons (PAHs) [18], heavy metals due to soil and water problems in tobacco growing areas [19, 20]. Many studies on detection of ingredients from cigarette smoke have been carried out. Some scholars determined formaldehyde by optimizing deoxygenant method and evaluated the temperature dependence of formaldehyde distribution in cigarette mass spectrometry. Inhaling formaldehyde from cigarette smoke had a great risk of genotoxicity [21]. Small particles from tobacco products were quite harmful [22]. Formaldehyde in human saliva was used as an indicator of environmental tobacco smoke exposure. Smoking had a significant effect on formaldehyde content in saliva [23].

Obviously, the characteristics of mainstream smoke or the pathological effects of cigarette smoke were focused [24], but it is not enough research in measurement smoke flow. Nicotine, tar content and other parameters on cigarette case are labelled, but the emission characteristics data of harmful components such as formaldehyde and particulate matter in sidestream smoke are missing. It is worth considering whether the different manufacturing process and additives will affect the emission characteristics of smoke pollutants. In addition, how to reduce or remove the pollution of cigarette smoke also needs to be studied.

To explore the above problems, four brands of cigarettes in the market were selected, and their production process, tar content and other parameters are different. In the closed conditions, the emissions of formaldehyde and particulate matter were measured. At the same time, the effect of ventilation on particles from cigarettes was also investigated.

Materials and Methods

Test Devices

The test was performed in an environmental chamber with the volume of 1m³, as shown in Fig. 1. As a cigarette placement point, a cylinder with a height of 0.6 m (including the thickness of the circular tray) was erected on the bottom of the chamber. A monitoring equipment placement place was set on the wall with a height of 0.6 m (including the thickness of the bracket) opposite to the cigarette placement point. The distance from the center of the round tray of the burning point to the center of the placing board of the monitoring equipment was 0.8 m. In addition, two vents of with length and width of 130 mm and 130 mm were installed on both sides of the wall of the vent and two baffles for closures were set on both sides of the vents.

In this study, four common brands of cigarettes on the market were tested. The basic parameters of the cigarettes are shown in Table 1.

A PC-3A(S) type dust detector with a sensitivity of 0.001 mg/m³ was used to measure $PM_{2.5}$ and PM_{10} concentrations. The detection ranges for $PM_{2.5}$ and PM_{10} are 0~10 mg/m³ and 0~20 mg/m³, respectively. A HTV-M type detector was used to measure the concentration of formaldehyde. It has a detection

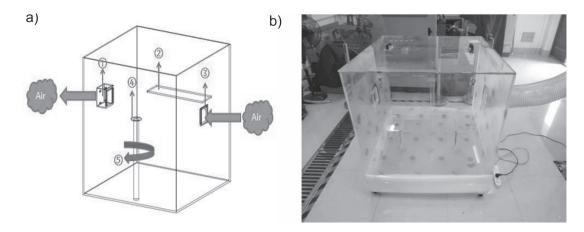


Fig. 1. Tested environmental chamber a) 3D drawing: ① Fan vent, ② Instrument, ③ Vent, ④ Cigarette tray, ⑤ Cylinde, b) Physical picture.

Cigarette brands	Cigarette types	Tar/mg	Nicotine content in smoke /mg	Flue gas - carbon monoxide /mg
А	Flue-cured	11	1.0	13
В	Flue-cured	11	1.0	11
С	Flue-cured	8	0.8	10
D	Blended	8	0.7	9

Table 1. Basic parameters of the tested cigarettes.

sensitivity of 0.001 mg/m³ and a detection range of $0\sim12.3$ mg /m³. A blower was used for air exchange in the chamber. The blower has an air flow of 1.15 m³/min at the rated speed of 2000 r/min.

Experimental Procedure

During the test process, the indoor temperature was set to $16\pm2^{\circ}$ C, and relative humidity to $35\pm5\%$. The two vents were opened under natural ventilation condition. The air flow of the blower was $1.15 \text{ m}^3/\text{min}$ under mechanical ventilated condition. The tested cigarette was put on the cylinder with a height of 0.6 m [25]. According to the average social distance [26], the distance from the detectors to the cigarette in the horizontal direction was 0.8 m. After the cigarette was lit, the detectors started to record the emission data. The monitoring time was from 200 to 600 minutes under closed conditions. The monitoring time was 192 minutes under nature and mechanical ventilation conditions.

Characteristic Parameters

During cigarette combustion, according to the law of conservation of mass, Equation (1) is established, which is often used to analyze the distribution and migration of indoor and outdoor particulate matter [27, 28].

$$\frac{dC_{IN}}{dt} = P\alpha C_{OUT} + \frac{Er}{V} - (\alpha + \lambda_{SINK})C_{IN}$$
(1)

where C_{IN} is the concentration of particulate matter in the environmental chamber (mg/m³), *P* is the penetration coefficient, α is air exchange rate (1/h), C_{OUT} is the concentration of particulate matter outside the environment chamber (mg/m³), *Er* is the quantitative release rate (mg/h), *V* is the volume in the environment chamber (m³) and λ_{SINK} is the deposition rate of particulate matter (mg/m³.h⁻¹).

Because the environmental chamber with adhesive strips at the cracks has good sealing performance, the particle penetration is ignored. Therefore, Equation (1) can be rewritten as:

$$\lambda_{SINK} = \frac{1}{t} \ln \frac{C_{MAX} - C_0}{C_t - C_0} - \alpha \tag{2}$$

...where C_0 is the background concentration in the (mg/m³) and C_t is the concentration in the chamber at *t* time (mg/m³).

Assuming that the bottom concentration in the environmental chamber is zero, Equations (1) and (2) can be further deduced:

$$C_{IN} = \frac{Er(1 - e^{-kt})}{Vk} (0 \le t \le T)$$
(3)

$$C_{IN} = C_{MAX} \left(e^{-k(t-T)} \right) \left(T > t \right)$$
(4)

$$k = \alpha + \lambda_{SINK} \tag{5}$$

where k is the particle removal rate (mg/h). T is the burning time of cigarette (h) and C_{MAX} is the maximum concentration (mg/m³).

The emission rate of particulate matter and formaldehyde can be given by:

$$Er = \frac{C_{MAX}\alpha V}{1 - e^{-\alpha T}} \tag{6}$$

The burning rate of the cigarettes is written as:

$$Br = \frac{M}{T}$$
(7)

where M is the weight of the tested cigarettes (g).

The particulate matter and formaldehyde emission factors are presented as:

$$Ef = \frac{Er}{Br}$$
(8)

where Ef is the emission factor (mg/g).

Results and Discussion

Emission Characteristics of Formaldehyde and Particulate Matter under Closed Conditions

Table 2 shows the combustion characteristics of four kinds of cigarettes under closed conditions. It is

Cigarette brands	Cigarette burning rate (g/h)	Pre-combustion mass/g	Post-combustion mass/g	Combustion quality/g	Burning time/min
А	3.000	0.64	0.09	0.55	11
В	2.800	0.62	0.06	0.56	12
С	2.782	0.63	0.12	0.51	11
D	2.650	0.66	0.13	0.53	12

Table 2. Combustion characteristics of four kinds of cigarettes under closed condition.

found that the burning rate of cigarettes range from 2.650 to 3.000 g/h. The burning rate of each cigarette is related to its own characteristics. Combined with the basic characteristic data of cigarettes in Table 1, the burning rate of cigarettes is positively correlated with the amount of carbon monoxide in the smoke. When the burning rate of the cigarette becomes fast, the cut tobacco burns incompletely. As a result, the carbon monoxide content in the smoke increases.

The curves of formaldehyde concentration in the side-stream smoke of four kinds of cigarettes are shown in Fig. 2. Table 3 shows the peak concentration, emission rates and emission factors of formaldehyde for the four kinds of cigarettes under closed conditions. The peak concentration of formaldehyde ranges

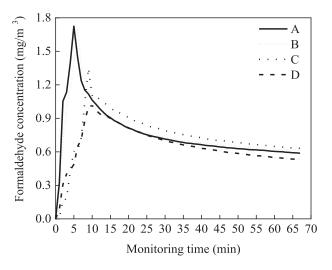


Fig. 2. Formaldehyde concentrations of four kinds of cigarettes under closed condition.

from 1.022 to 1.724 mg/m³. After about 15 minutes, formaldehyde concentration for cigarette A reaches its peak. However, for cigarette C and D, the peaks of formaldehyde concentration occur after 30 minutes. The order of formaldehyde emission factors of four kinds of cigarettes is A, B, C and D. During the combustion stage, the formaldehyde concentration rises to a peak value rapidly. However, after the cigarette is burned out, the concentration of the formaldehyde decreases slowly due to gas leaks.

Fig. 3 presents the particulate matter concentration of four kinds of cigarettes. Table 4 shows the statistical characteristics of particulate matter of four kinds of cigarettes. It can be seen that the peak concentration of PM_{25} and PM_{10} are in the range of 0.272 to 0.282 mg/m³ and 3.294 to 3.455 mg/m³. In addition, the PM₂₅ concentration trend is basically the same. However, the PM25 concentrations of cigarette A and D are significantly higher than those of cigarette B and C after about 15 minutes, and the highest concentration difference between cigarette B and cigarette D is 0.564 mg/m³. Obviously, the peak concentration of PM225 of cigarette D is the lowest, while the peak concentration of PM₁₀ of cigarette D is the highest. It is also found that the concentrations of particulate matter of the four kinds of cigarettes increase rapidly during combustion stage. However, the concentrations of particulate matter remain at high level for a long time after the cigarette is burned out. The reason is that the environmental chamber has a good sealing performance. In addition, the concentrations of PM₁₀ of the four kinds of cigarettes are much higher than those of PM₂₅. It can be seen that the emission rates of PM_{2.5} and PM₁₀ of the four kinds of cigarettes range from 1.506 to 1.684 mg/h and from 18.834 to 20.190 mg/h.

Table 3. Formaldehyde emission characteristics of four kinds of cigarettes under closed condition.

Cigarette brands	Peak concentration of formaldehyde (mg/m ³)	Rate of formaldehyde emission (mg/h)	Formaldehyde emission factor (mg/g)
А	1.724	10.292	3.431
В	1.699	9.373	3.347
С	1.349	8.053	2.895
D	1.022	5.638	2.128

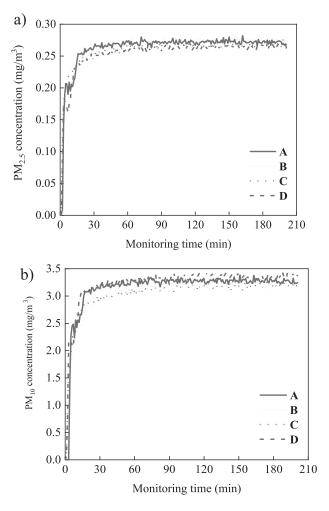


Fig. 3. $PM_{2.5}$ and PM_{10} concentrations of four kinds of cigarettes under closed conditions: a) $PM_{2.5}$, b) PM_{10} .

To study the deposition of particulate matter, a longer test for cigarette A was conducted under closed conditions, and the duration of the experiment was extended to 10 h. The emission curve of particulate matter is shown in Fig. 4. It can be seen that the concentration of $PM_{2.5}$ decreases more slowly than that of PM_{10} . According to Equation (2), the deposition rates of $PM_{2.5}$ and PM_{10} are 0.018 and 0.0289 mg/m³.h⁻¹. This is consistent with the conclusion of the reference [29].

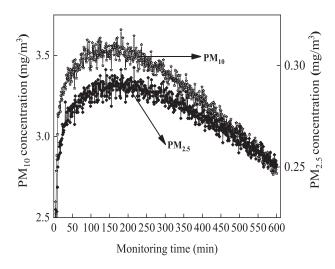


Fig. 4. $PM_{2.5}$ and PM_{10} concentrations from Cigarette A under closed conditions for a long time.

Curve Fitting of Formaldehyde and Particulate Matter under Closed Conditions

It can be seen from Fig. 2 that the change process of formaldehyde concentration in the chamber is divided into the release stage and the escape stage. A little amount of gas escapes during the release stage. According to Equations (3) and (4), a staged curve fitting was carried out, and the fitting formula for the rising segment is given by:

$$y = a(1 - \exp(-bx)) \tag{9}$$

The fitting formula for the descending phase is written as:

$$y = A_1 \exp(-x/t_1) + y_0$$
 (10)

The fitting results of formaldehyde concentrations are shown in Fig. 5. The fitting correlation coefficients range from 0.96817 to 0.98017 during the release stage and from 0.97947 to 0.99883 during the escape stage, indicating that the deduced formula is reliable. It can be seen that the fitting result of escape stage is better than that of release stage. The main reason is that the

Table 4. Statistical characteristics of particulate matter of four kinds of cigarette particles under closed condition.

Cigarette brands		centration /m ³)	The ratio of $PM_{2.5}$ to PM_{10} peak	Emission rate (mg/h)		Emission factor (mg/g)	
	PM _{2.5}	PM ₁₀	concentration (%)	PM _{2.5}	PM ₁₀	PM _{2.5}	PM ₁₀
А	0.282	3.382	8.338	1.684	20.190	0.561	6.730
В	0.281	3.347	8.396	1.506	18.834	0.538	6.726
С	0.276	3.294	8.379	1.648	19.665	0.591	7.056
D	0.272	3.455	7.873	1.501	19.060	0.566	7.193

fluctuation in the temperature and humidity affects the diffusion of formaldehyde gas.

Compared with gas, particles are more difficult to escape from the gap in the environmental chamber. The change process of particle concentration in the chamber is divided into the release stage and the deposition stage. During the release stage, particles from cigarettes are endowed with initial kinetic energy by combustion and the deposition effect can be ignored. After a certain time, energy of motion of particles is not enough to

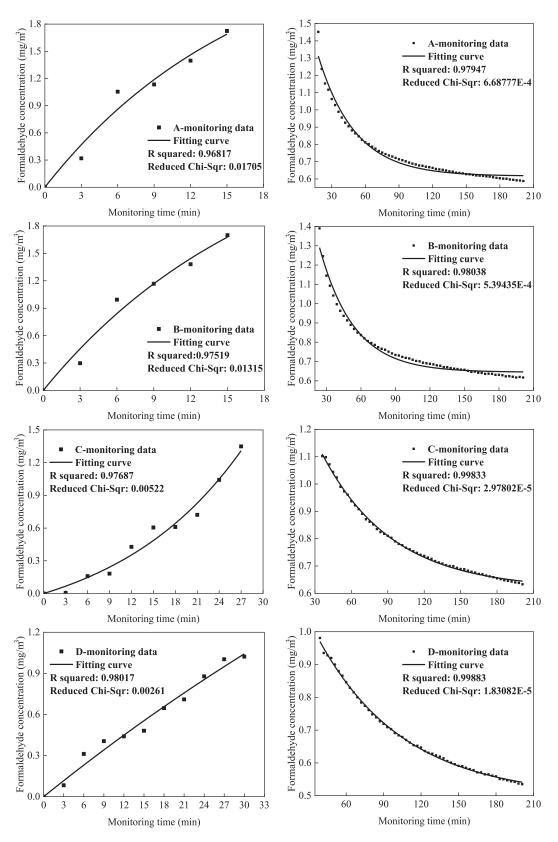


Fig. 5. Fitting of tested formaldehyde concentrations.

support their continued movement due to energy lose in the collision. Many particles begin to fall due to gravity action during the second stage.

The concentrations of particulate matter keep near the peak value for a long time. Therefore, it is judged to belong to the release stage. For this stage, the fitting formula is:

$$y = a(1 - \exp(-bx)) \tag{11}$$

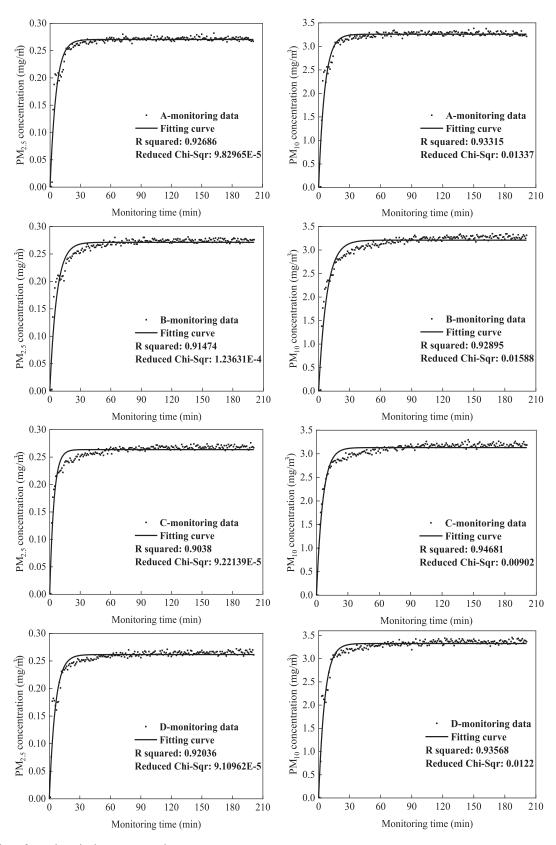


Fig. 6. Fitting of tested particulate concentrations.

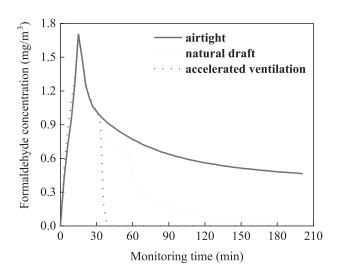


Fig. 7. Formaldehyde concentrations under different ventilation modes.

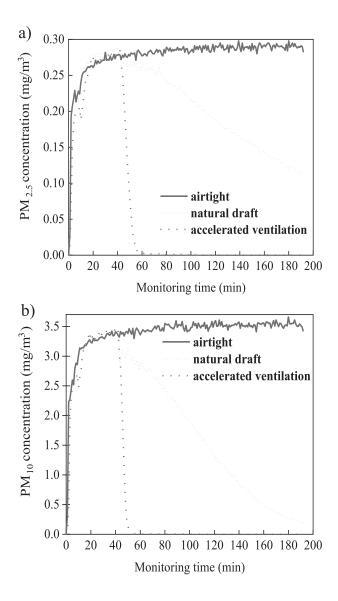


Fig. 8. Concentrations of $PM_{2.5}$ and PM_{10} under different ventilation modes: a) $PM_{2.5}$, b) PM_{10} .

The fitting results of particle concentrations are shown in Fig. 6. It can be seen that the fitting correlation coefficients of $PM_{2.5}$ and PM_{10} range from 0.9038 to 0.92686 and ranged from 0.92895 to 0.94681 during the release stage, respectively. Obviously, the performance of linear regression of PM_{10} is better than that of $PM_{2.5}$, because gravity has little effect on small particles while other external forces have important effect on them. In addition, the fitting result of particulate matter is worse than that of formaldehyde, especially during the rise period. This reason is that the burning of cigarettes causes drastic changes in the temperature and humidity of the surrounding environment affecting the diffusion and condensation of particulate matter [30-32].

Pollutant Characteristics under Ventilation Conditions

The concentrations of formaldehyde and particulate matter from A cigarette under ventilation conditions are shown in Figs 7 and 8. It is found that the concentrations of formaldehyde and particulate matter drop rapidly under natural and mechanical ventilation conditions. This proves that ventilation plays a significant role in improving the air quality in the chamber [33]. It is also found that the concentration of $PM_{2.5}$ decreases more slowly than that of PM_{10} under ventilated conditions. This reason is that $PM_{2.5}$ as fine particles are affected by air drag force, adhesion force and levitation force [34, 35].

Conclusions

When four common brands of cigarettes are burned in a closed environment chamber, they release formaldehyde with a peak concentration of $1.022\sim1.724 \text{ mg/m}^3$, PM_{2.5} with a peak concentration of $0.265\sim0.271 \text{ mg/m}^3$ and PM₁₀ with a peak concentration of $3.183\sim3.354 \text{ mg/m}^3$.

After the cigarettes are burned out, the particle concentrations under closed conditions still keep at high level for a long time, and the particle deposition rate is very small. The removal rate of particulate matter with large particle size is higher than that of particulate matter with small particle size under ventilation conditions.

The fitting of formaldehyde under closed conditions is more reliable. The fitting correlation coefficients of formaldehyde range from 0.96817 to 0.98017 in the release stage and from 0.97947 to 0.99883 in the escape stage, while the fitting correlation coefficients of $PM_{2.5}$ and PM_{10} range from 0.9038 to 0.92686 and ranged from 0.92895 to 0.94681 in the release stage, respectively.

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Conflict of Interest

The authors declare no conflict of interest.

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