Study on indoor ozone removal by PRM under the influence of typical factors

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Abstract Ozone exposure has been proven to be strongly associated with both morbidity and mortality. In addition, the reaction of ozone with human skin sebum, building materials and indoor other compounds can cause secondary pollution of indoor environment. Therefore, how to effectively control indoor ozone pollution and its induced secondary pollution is an important scientific problem that needs to be solved in the field of indoor air quality. PRM (passive removal materials) can passively remove indoor ozone without energy consumption. According to the mechanism of ozone removal on PRM surface, the effects of material area, reaction probability, transport-limited deposition velocity, outdoor ozone concentration, air change rate (ACH) and indoor reaction rate on ozone removal rate of PRM were analyzed in this study. The results show that the ozone removal rate of PRM is greatly influenced by the reaction probability, the ACH, the surface area and the transport-limited deposition velocity, while the indoor reaction rate and outdoor ozone concentration have no obvious influence. Therefore, in order to remove more indoor ozone by PRM, the position and area of PRM with different reaction probabilities should be chosen reasonably.

Keywords: indoor ozone pollution, passive removal materials, reaction probability, deposition velocity

1 Introduction

As a trace gas in the atmosphere, ozone was identified as one of the six "standard pollutants" by the US Environmental Protection Agency as early as 1970[1], which has a great impact on human health. A large number of epidemiological studies have shown a correlation between ozone exposure and increased short-term mortality[2]-[3]. Ozone, as a strong oxidizing gas, can not only directly damage human health and cause symptoms such as respiratory tract disease and lung function decline[4]-[5], but also react with human surface grease and building materials[6]-[7] to produce formaldehyde, acrylone, acetone[8]-[9] and other pollutants which cause secondary pollution of indoor environment.

Indoor ozone mainly comes from atmospheric ozone which penetrates into the room through ventilation and building enclosure structure. In addition, some indoor electrical appliances such as photocopiers and laser printers also produce ozone during operation[10]. At present, there are two main methods to remove indoor ozone, one is active method to treat fresh air and indoor air with activated carbon filter, the other is using passive removal material (PRM). As a method without energy consuming, recent studies have focused on PRMs. PRMs are materials used on surfaces of buildings such as walls and ceilings, which can react irreversibly with ozone and effectively remove indoor pollutants. Many studies[13]-[14] have proved that PRM does contribute to the reduction of indoor ozone concentration. The influencing factors of its ozone removal capacity should be further studied.

Hoang[10] conducted environmental chamber experiment on 10 common green building materials, quantified ozone removal rate according to deposition velocity and reaction probability, and found degradation and regeneration of ozone removal ability of PRMs. This is consistent with the research result of Rim[16] that PRM's ozone reaction probability decreases with prolonged exposure to ozone. Gall[17] studied the long-term performance of three green building materials in a large-scale environmental chamber and found that perlite-based ceiling, recyclable carpet and recycled drywall have long-term ozone removal ability and long-lasting ozone reaction capabilities. Kunkel[17] measured the ozone decay rate in the environmental chamber with two kinds of materials, and the study showed that the indoor ozone removal rate was less affected by the air exchange rate but more affected by the area and type of materials. Lin[19] studied the influence of physical properties of building materials (specific surface area and total pore volume) on ozone removal ability, and the result showed that the specific surface area of materials had a greater impact than the total pore volume. Gall[20] also studied the influence of the physical properties of porous material (porosity, pore size distribution and material thickness) on ozone reaction. The study found that the ozone deposition rate and reaction probability of material increase with the increase of material thickness, but

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increasing the thickness will not increase the reaction probability under the condition of limited internal transmission within the material. In addition, other scholars have studied the influence of hydrodynamic properties near the material surface on the ozone removal ability of the material[21]-[23].

At present, indoor ozone pollution is becoming more and more serious, and PRM is an effective method to remove indoor ozone. However, it can be seen that the existing research mainly focuses on the influencing factors of PRM ozone removal through the experimental chamber, but there is no systematic theoretical analysis of the impact of different factors on PRM ozone removal, which is also the problem to be solved in this paper. In this study, the effects of PRM surface area, reaction probability, transport-limited deposition velocity, outdoor ozone concentration, air exchange rate(ACH) and indoor reaction rate on the reduction of indoor ozone concentration were analyzed, which may provide guide for the rational application of PRM to remove ozone pollution.

2 Methods

2.1 Calculation model of indoor ozone concentration

The indoor ozone concentrations mainly depend on the outdoor ozone concentrations, ACH, indoor sources of ozone, PRM removal rate, and reactions between ozone and other chemical substances in indoor air. Fadeyi[24] proposed a model equation for estimating indoor ozone concentration using a simplified mass balance equation and steady-state assumptions. Aldred[11] used a well mixed steady-state mass balance equation for ozone in indoor space (as shown in equation (1)) to simulate the influence of activated carbon filtration on indoor ozone concentration:

$$C_{O_3} = \frac{p\lambda_{inf}C_o}{\lambda_{inf}+H_{on}\lambda_{rec}f_{c,O_3}+(H_{on}k^*_{dep,O_3,AC_{on}}+(1-H_{on})k^*_{dep,O_3,AC_{off}})+\sum_j K_jC_j} \quad (1)$$

where λ_{inf} is infiltration air exchange rate, in units of h^{-1}; λ_{rec} is recirculation air exchange rate of HVAC system, in units of of h^{-1}; C_j is the indoor concentration of reactant j, in units of ppb; C_{O_3} is the steady-state concentration of indoor ozone, in units of ppb; C_o is the outdoor ozone concentration, in units of ppb; f_{c,O_3} is ozone removal rate constant of activated carbon filter; H_{on} is average annual fraction of time that the HVAC system operates, in units of %; $k^*_{dep,O_3,AC_{on}}$ is ozone decay rate for integrated background surfaces with HVAC on, in units of h^{-1}; $k^*_{dep,O_3,AC_{off}}$ is ozone decay rate for integrated background surfaces with HVAC off, in units of h^{-1}; K_j is the bimolecular reaction rate constant for ozone and indoor reactant j, in units of ppb^{-1}·h^{-1}; p is the penetration factor for ozone.

In equation (1), the numerator represents the amount of outdoor ozone into the indoor space, and the denominator represents the amount of indoor ozone removed. The bracketed term in the denominator represents the ozone removal of indoor surface with or without HVAC. On this basis, Darlinga[12] used a similar equation (2) to study the impact of PRM on indoor ozone concentration. Compared with equation (1), equation (2) considers the influence of indoor ozone sources on indoor ozone concentration:

$$C_{O_3} = \frac{pC_0+\sum E/\lambda V}{1+\lambda^{-1}[\alpha k_{O_3,surf}+(1-\alpha)k_{O_3,prm}+\sum k_{O_3,j}C_j]} \quad (2)$$

where E is the emission rate of ozone into the space, in units of ppb^{-1}·m^3·h^{-1}; λ *is* air exchange rate, in units of of h^{-1}; V is the volume of the room, in units of m^3. $k_{O_3,surf}$ is the decay rate of ozone in the absence of PRM surfaces, in units of of h^{-1}; $k_{O_3,prm}$ is the decay rate of ozone of PRM surface, in units of of h^{-1}; α is proportion of surface area without PRM coverage; $k_{O_3,j}$ is indoor ozone reaction rate, in units of ppb^{-1}·h^{-1}.

2.2 Ozone deposition velocity on PRM surface

Ozone is a strongly oxidizing gas, which can react irreversibly with chemicals on the surface of materials, thus promoting ozone removal and optimizing indoor air quality. These chemicals include organic compounds, inorganic compounds and organic compounds attached to the surface of the material, which are also called surface reaction sites. Lin[19] pointed out that these surface reaction sites are the reason why PRM can remove ozone. However, with the depletion of reaction sites, the reaction rate between materials and ozone will decrease, which is called the aging phenomenon of materials by Hoang[15]. In addition, the material's ozone removal ability will regenerate without ozone exposure. This may be caused by the deposition of new chemicals on the surface of the material, or the diffusion of chemicals within the material to the surface, and these chemicals become new ozone reaction sites. Meanwhile, ozone reacts with unsaturated organic compounds on the surface of materials to produce by-products, which may affect human health, while the amount of by-products of the reaction between ozone and inorganic compounds can be ignored[25]. The ability of a material's surface to removal ozone can be quantified by the deposition velocity of ozone (v_d), as in equation (3).

$$v_d = \frac{J}{C_f} \quad (3)$$

where v_d is the ozone deposition velocity, in units of m·s^{-1}; J is ozone flow on material surface, in units of of mg·(m^2)$^{-1}$·s^{-1}; C_f is average concentration of ozone, in units of mg·(m^3)$^{-1}$.

The overall resistance of the material surface to ozone removal is usually modeled as the sum of transmission resistance and surface reaction resistance, as described in equation (4):

$$\frac{1}{v_d} = r_o = r_t + r_s = \frac{1}{v_t} + \frac{1}{v_s} = \frac{1}{v_t} + \frac{4}{\gamma\langle v\rangle} \quad (4)$$

where r_o is the overall resistance, in units of s·m^{-1}; r_t is the ozone transmission resistance, in units of s·m^{-1}; r_s is the surface reaction resistance, in units of s·m^{-1}; v_t is the transport-limited deposition velocity, in units of m·s^{-1}; v_s is the reaction limited deposition velocity, in units of m·s^{-1}; γ is the reaction probability; $\langle v\rangle$ is the mean Boltzmann velocity of ozone in air (~360 $m \cdot s^{-1}$ at 20°C).

The reaction limited deposition velocity depends on the reaction probability of the material[26], reflecting the influence of the reaction between ozone and the material surface. The reaction probability is the number of reactions in which the molecule collides with the surface divided by the total number of collisions.

The transport-limited deposition velocity depends on the mixing conditions in bulk air and boundary layer fluid mechanics near the material surfaces, which reflects the influence of ozone transport to the material surface. According to Wilson[27], the transport-limited deposition velocity of indoor natural convection is $2.5 m \cdot h^{-1}$, and when air is stirred sufficiently to move loose papers, the transport-limited deposition velocity is $7.2\ m \cdot h^{-1}$. Since that study, Morrison[28] measured the v_t at different locations in the room, in which the v_t near the door and window area is as high as $25.2 m \cdot h^{-1}$, while the v_t near the indoor computer is only $5.2 m \cdot h^{-1}$. The relationship between ozone deposition velocity and ozone decay rate is defined by equation (5):

$$k_{O_3,prm}C_{O_3}V = v_{d,\ prm}C_{O_3}A_{prm} \quad (5)$$

where $v_{d,prm}$ is the ozone deposition velocity, in units of $m \cdot h^{-1}$; A_{prm} is the horizontally-projected surface area of the PRM, in units of m^2; C_{O_3} is the concentration of ozone above the surface calculated in equation (2), in units of ppb.

2.3 Ozone removal rate

Ozone removal rate is defined as the percentage of ozone concentration reduction caused by PRMs:

$$\Omega = 1 - \frac{C_{prm}}{C_{noprm}} \quad (6)$$

where C_{prm} is the ozone concentration with PRM indoor , in units of ppb; C_{noprm} is the ozone concentration without PRM indoor , in units of ppb.

3 Results and discussion

According to the China Statistical Yearbook, the per capita living area of urban residents in China is about $36m^2$. Therefore, the height of the bedroom calculated in this paper is 2.5m, the total surface area is 132 ㎡, and the volume is $90m^3$. According to the study[29], the average ozone deposition velocity of surface without PRM is $0.252 m \cdot h^{-1}$. According to the above analysis of influencing factors of ozone and PRM surface reaction, the surface area, reaction probability, transport-limited deposition velocity, outdoor ozone concentration, air exchange rate, and indoor reaction rate were selected as variables to analyze the efficiency characteristics of PRM in removing ozone pollution.

3.1 Impact of reaction probability

Darling[11] suggested that materials with ozone removal capability should have a higher ozone reaction probability than 1×10^{-5}, so the ozone removal rate of materials with reaction probability higher than 1×10^{-5}[26] is calculated by equation (2) and (6). It is assumed that the material coverage rate is 100%, the transport-limited deposition velocity is taken as $7.2 m \cdot h^{-1}$, the emission rate of ozone from indoor ozone source to air is $4.6\times10^{-3} ppb \cdot m^3 \cdot h^{-1}$[30], the ozone reaction rate is 27.29ppb[31], and the ozone reaction rate is $0.001098 ppb^{-1} \cdot h^{-1}$. The calculation results are shown in Fig 1.

Ozone removal rate increases with the increase of reaction probability. It can be seen from the fitting curve that when the reaction probability increases to a certain extent, the ozone removal rate tends to be infinitely stable to a certain value. This indicates that the removal rate of ozone cannot be continuously improved by simply increasing the reaction probability of materials.

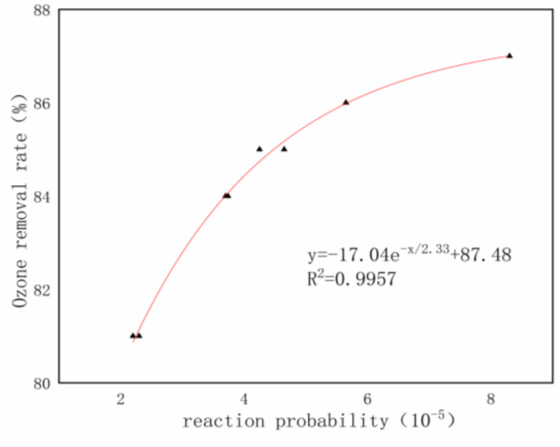

Fig. 1. Trend of ozone removal rate with reaction probability

3.2 Surface area of PRM

In order to analyze the effect of PRM surface area on ozone removal rate, the proportion of PRM covered area changed from 10% to 100%. Three materials with ozone removal capability were selected, and their reaction probability were 2.20×10^{-5} 、 4.25×10^{-5} 、 8.30×10^{-5} respectively. As shown in Fig 2, the ozone removal rate of the material increases with the increase of PRM area proportion, which is consistent with the results of Kunkel[13] experiments. Moreover, the higher the reaction probability, the smaller the PRM coverage required for the same ozone removal rate. Meanwhile, similar to the effect of reaction probability, the increment of ozone removal rate gradually decreases with the further increase of PRM area, and the ozone removal rate infinitely approaches a fixed value. Therefore, the ozone removal rate can be maximized by properly arranging the coverage area of different PRMs.

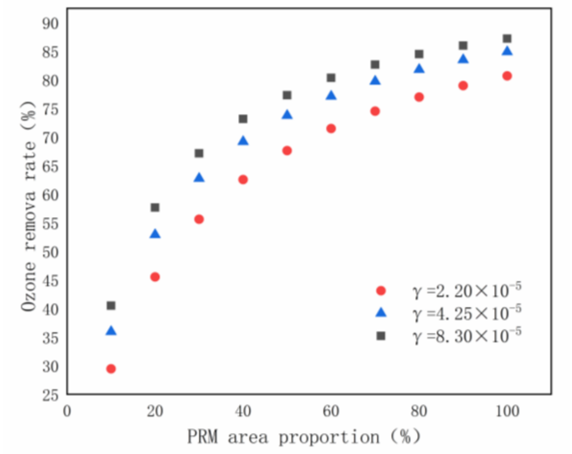

Fig. 2. Trend diagram of ozone removal rate changing with PRM area proportion

3.3 Impact of transport-limited deposition velocity

The transport-limited deposition velocity depends mainly on the fluid mechanics near the material surface, so different wind velocity at different locations indoor will result in different transport-limited deposition velocity. According to the transport-limited deposition velocity measured by Morrison[28] at different positions indoor, its variation range was set from 5m·h^{-1} to 25m·h^{-1}. It can be seen from Fig 3 that the ozone removal rate increases with the increase of the transport-limited deposition velocity. When the transport-limited deposition velocity increase from 5m·h^{-1} to 15m·h^{-1}, the ozone removal rates of the three materials increase by 10%, 11% and 11% respectively, while the transport-limited deposition velocity increase from 15m·h^{-1} to 25m·h^{-1}, and the ozone removal rates of the three materials only increase by 2%. This indicates that increasing the transport-limited deposition velocity alone will not continuously improve the indoor ozone removal rate. The transport-limited deposition velocity of PRM surface is related to the position of PRM indoor, and the deposition velocity near the door and window is much higher than that at other locations. Therefore, in order to remove more ozone, the indoor location of PRM with different reaction probability should be planned reasonably.

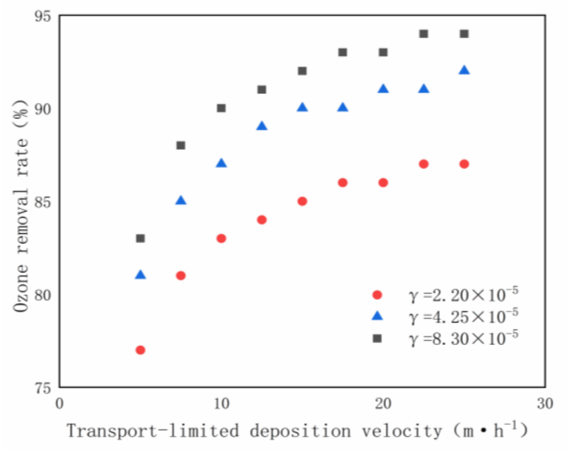

Fig. 3. Trend diagram of ozone removal rate changing with transport-limited deposition velocity

3.4 Impact of outdoor ozone concentration

According to national environmental monitoring data, the maximum environmental ozone concentration in China is about 150 ppb. In order to analyze the influence of outdoor ozone concentration on ozone removal rate, the range of outdoor ozone concentration variation was set from 80 ppb to 150 ppb. It can be seen from Fig 4 that outdoor ozone concentration has little effect on ozone removal rate. On the one hand, the ozone removal rate is defined as the ratio of indoor ozone concentration with PRM and indoor ozone concentration without PRM, as shown in equation (7):

$$\Omega = 1 - \frac{\mathrm{C_{prm}}}{\mathrm{C_{noprm}}} = 1 - \frac{1+\lambda^{-1}[k_{O_3,prm}+\sum k_{O_3,j}C_j]}{1+\lambda^{-1}[k_{O_3,surf}+\sum k_{O_3,j}C_j]} \qquad (7)$$

According to equation (7), outdoor ozone concentration has no effect on PRM ozone removal rate. On the other hand, because of the mechanism of ozone removal by PRM, the initial ozone reaction sites on the material surface will not change with the increase of outdoor ozone concentration without changing the material type, surface area and air flow conditions. As with the study of material ozone removal rate under ultra-high ozone concentration (1000-1200ppb) by Popendiecks[32], the results also show that even higher ozone concentration has no effect on material ozone removal rate. However, over a period of time, the ozone reaction probability of the material decreases because the high ozone concentration consumes the surface reaction sites of the material very quickly.

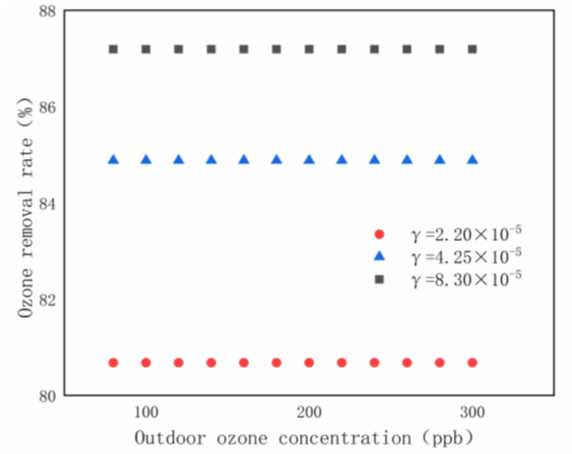

Fig. 4. Trend diagram of ozone removal rate changing with outdoor ozone concentration

3.5 Impact of ACH

In order to analyze the influence of the ACH on ozone removal rate, the ACH was taken as a variable, and the results are as shown in Fig 5. Indoor ozone concentration mainly depend on the outdoor, so the increase of ACH will lead to an increase in the indoor ozone concentration. According to the previous discussion about the influence of outdoor ozone concentration on the ozone removal rate of PRM, the effect of increasing indoor ozone concentration on ozone removal rate is not obvious. However, it can be seen from in Fig 5 that the material ozone removal rate decreases with the increase of ACH. This is because the increase in the ACH shortens the contact time between ozone and PRM, which affects the ozone removal rate on PRM. When the ACH increases from 0.5h^{-1} to 5h^{-1}, the ozone removal rates of the three materials decrease by 43%, 37% and 33% respectively. Moreover, the effect is greater on material with low reaction probability.

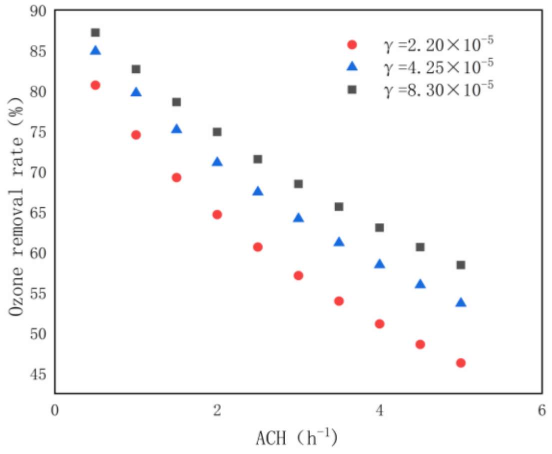

Fig. 5. Trend chart of ozone removal rate changing with air change rate

3.6 Impact of the indoor reaction rate

Ozone can react with many compounds indoor. In order to analyze the influence of indoor ozone reaction on the ozone removal rate of PRM, the indoor reaction rate was taken as a variable, ranging from 0.000549$ppb^{-1}\cdot h^{-1}$-0.00549$ppb^{-1}\cdot h^{-1}$. As can be seen from Fig 6, the ozone removal rate of PRM decreases slightly with the increase of indoor reaction rate due to more ozone removed by gas phase reaction. However, when the indoor reaction rate increases by an order of magnitude, the ozone removal rates of the three materials decrease by 2.3%, 1.8% and 1.5% respectively, which indicates that the indoor reaction rate had no significant effect on the ozone removal rate of PRM.

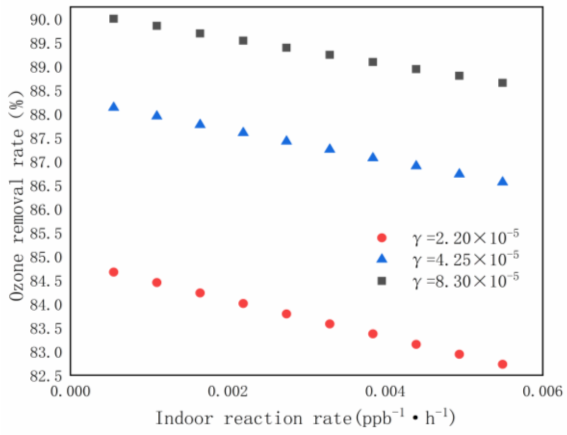

Fig. 6. Trend chart of ozone removal rate changing with indoor reaction rate

4 Conclusions

Based on theoretical methods, this paper systematically analyzed the effect of PRM material area, reaction probability, transport-limited deposition velocity, outdoor ozone concentration, ACH and indoor gas phase reaction rate on the ozone removal rate of PRM, and the following conclusions were drawn:

(1)The ozone removal rate increases with the increase of the reaction probability, but with the further increase of the reaction probability, the change of ozone removal rate tend to be stable.

(2)The ozone removal rate increases with the increase of PRM coverage area, but with the continuous increase of PRM coverage area, the increase of ozone removal rate decreases. Increasing PRM coverage area alone can not effectively improve indoor ozone removal rate.

(3)The ozone removal rate increases with the increase of the transport-limited deposition velocity, but increasing the transport-limited deposition velocity alone cannot continuously improve the indoor ozone removal rate.

(4) Outdoor ozone concentration has little effect on the ozone removal rate of PRM, but the higher the ozone concentration, the faster the depletion of the reaction sites on the PRM surface.

(5)The ozone removal rate decreases with the increase of ACH, and the effect on materials with low reaction probability is more significant.

(6)The ozone removal rate decreases with the increase of the reaction rate in the chamber, but the effect is not obvious.

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