Numerical Investigation on the Effect of Ventilation on the Distribution of Phthalate Esters in the Residential Environment

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Abstract. Semi-volatile organic compounds (SVOCs), such as phthalates and brominated flame retardants, is a kind of emerged pollutants due to wide application in indoor environment. Certain indoor SVOCs have been found to be associated with various adverse health effects, attracting large attentions of researchers. Due to relatively low vapor pressure, SVOCs are easily adsorbed on various surfaces including particles. Therefore, airborne SVOCs are always simultaneously presented in the gas-phase and particle-phase. Ventilation is an important means to improve indoor air quality. Different forms of indoor air distribution will affect the distribution of indoor pollutants and further affect the exposure to the human body. Therefore, in this paper, we selected Di (2-ethylhexyl) phthalate (DEHP) as the target compound and employed computational fluid dynamics (CFD) technique to simulate the emission of DEHP and the concentration distribution with different phases in a modeled room. Euler-Lagarian model is applied to simulate flow field, particle tracks and UDF (user defined function) was implemented to describe the dynamic adsorption of DEHP by the suspended particles. Furthermore, the effect of location of vent and airflow rate on indoor fate of DEHP were discussed and the effect of particle age on indoor fate of DEHP was also investigated.

1 Introduction

With the development of chemical industry, numerous chemical products are used indoors in modern society. Semi-volatile organic compounds (SVOCs) as one of the most important indoor organic contaminants, such as plasticisers, flame retardants and biocides, are widely used in indoor products and building materials [1, 2]. People are exposed to SVOCs via three path-ways: inhalation, ingestion, and dermal absorption [3, 4]. People exposed to certain indoor SVOCs have been found to be associated with various adverse health effects such as endocrine disorders, birth defects, asthma, and cancer [5]. Since people spend about 90% of their lifetime indoors [6], SVOCs can have a great impact on human health. Thus, it is important for improving indoor air quality to study residential exposure to SVOCs.

Aerosol particles are one of the main components in indoor air pollutants. Suspended particles can act as nuclei and carriers for airborne viruses and bacteria, resulting in the spread of diseases. Due to relatively low vapor pressure, SVOCs are easily adsorbed on various surfaces including particles. Therefore, airborne SVOCs are always exist in both the gas-phase and particle-phase [7, 8]. In addition, SVOCs distributed to airborne particles tend to result in higher total concentrations than occur in the gas phase alone [1, 9]. Therefore, these SVOC-containing particles can remain in the air, or settle and resuspend, becoming a source of exposure through inhalation and ingestion of dust [10]. The presence of airborne particles is criticle for SVOC mass transfer in indoor environment and may significantly increase occupant exposure to SVOCs.

Indoor ventilation plays a vital role in indoor air quality, and the diversity of ventilation will lead to different distribution of indoor air distribution, and the form of air distribution will directly affect the spatial non-uniformity of different types of indoor pollutants, thereby affecting the health of indoor personnel. However, most studies are based on the assumption that the air condition of room is well-mixed with particles and SVOCs, thus using the average concentration as an indicator of exposure, which is hardly satisfied in real conditions. The diffusion of gas-phase SVOCs and particles dispersion and deposition are always affected by turbulent flow. Besides, most existing studies assume an instantaneous equilibrium between gas-phase SVOCs and particles in indoor environments. In reality, the instantaneous equilibrium between gas-phase SVOCs and airborne particles maybe not a reasonable assumption. The residence time of particles indoors is often much shorter than the critical time to reach equilibrium between gas-phase SVOCs and airborne particles [1, 11]. Liu et al. [12] assessed the impact of the aerosol dynamics on exposure to the airborne DEHP and it is found that the instantaneous equilibrium assumption may not be reasonable for the less volatile species such as DEHP. Cao et al. [13] proposed the concept of "particle age", which means the time particle takes in the room since it enters the room, to revise the previous equilibrium assumption.

Therefore, the purpose of this study is to analyze the influence of location of vent and ventilation rate condition on the concentration distribution of indoor pollutants SVOCs (gas and particle-phase) through

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numerical simulation. This study will provide further suggestions for improving ventilation methods to reduce human exposure to SVOCs.

2 Numerical method

Di(2-ethylhexyl) phthalate (DEHP) is selected as the target compound. This study built a phthalate transportation and dynamic mass transfer model to obtain the DEHP concentration distribution indoors based on the air flow and particle simulation.

2.1 Air flow model

In ventilated indoor air conditions, airflow is often considered as turbulent. The air phase here is regarded as a continuum and the Eulerian method is used for the numerical analysis. The interaction between fluid phase and particle phase is assumed to be unidirectional, and the effect of the particles on the fluid is negligible. This assumption is feasible because the particle volume fraction is sufficiently small. Compared with other turbulence models, the simulation results of the RNG k-ε model are in good agreement with the experimental data [14]. Therefore, the RNG $k-\varepsilon$ model is selected to predict the incompressible turbulent airflow in a room.

The governing equation can be written as the following general form:

$$\frac{\partial \rho\phi}{\partial t}+div(\rho \boldsymbol{V}\phi-\Gamma_{\phi,eff}\,grad\phi)=S_{\phi} \qquad (1)$$

Where **V** is the velocity vector, t is the time and ρ is the density of indoor air, ϕ is the general variable which can represent solution variables such as velocity and temperature, Γ is the diffusion term coefficient and S_{ϕ} is the source term. For specific equations, such as continuity equations, momentum equations and turbulence equations, ϕ, Γ and S_{ϕ} have specific forms.

2.2 Particle transportation model

The Lagrangian particle tracking method is used to calculate individual trajectories by solving the momentum equation. By equating the particle inertia with external forces, the momentum equation can be expressed as:

$$\frac{d\boldsymbol{u}_p}{dt}=F_D(\boldsymbol{u}-\boldsymbol{u}_p)+\frac{\boldsymbol{g}(\rho_p-\rho)}{\rho_p}+\boldsymbol{F}_a \qquad (2)$$

Where $\boldsymbol{u}$ is the fluid phase velocity, $\boldsymbol{u_p}$ is the particle velocity, ρ is the fluid density, ρ_p is the density of the particle, where F_D is the inverse of relaxation time (s^{-1}) and $\boldsymbol{F_a}$ is an additional force.

2.3 Gas-phase DEHP transport in the air

Since the indoor flowing medium includes air and DEHP, Species Transport Model of the mixture is needed to simulate this situation. When predict the local mass fraction of each species, Y_i, through the solution of a convection-diffusion equation for the i^{th} species. This conservation equation takes the following general form:

$$\frac{\partial}{\partial t}(\rho Y_i)+\nabla\cdot(\rho\boldsymbol{v}Y_i)=-\nabla\cdot\boldsymbol{J}_i+R_i+S_i \qquad (3)$$

Where $\boldsymbol{J_i}$ is the diffusion flux of species i, which arises due to gradients of concentration and temperature. R_i is the net rate of production of species i by chemical reaction and S_i is the rate of creation by addition from the dispersed phase plus any user-defined sources.

Previous studies have shown that the presence of particles influences the gas-phase SVOCs concentration is not appreciable in a large volume room [12], so this study assumes that a long enough time had passed for the room to reach the equilibrium of the gas-phase SVOCs before particle injection, ignoring the adsorption effect of the room wall on the gas-phase SVOCs. This means that the gas-phase concentration field is independent of the particle phase and can be determined in advance.

2.4 Particle-phase DEHP transport in the air

A proper quantitative understanding of the dynamic interaction between gas-phase semi-volatile organic compounds (SVOCs) and airborne particles is important for human exposure assessment and risk evaluation. Eqs. (4) assumes phthalate transfer between the bulk air and the airborne particles [11].

$$V_{\mathrm{p}}\frac{dC_p}{dt}=v_tA_p(C_g-\frac{C_p}{K_{part}}) \qquad (4)$$

where C_p (μg/m^3, mass of SVOCs/volume of particles) is the particle-phase SVOC concentration, v_t (m/s) is the gas/particle mass transfer coefficient, C_g (μg/m^3, mass of SVOCs/volume of air) is the gas-phase SVOC concentration, K_{part} is the gas/particle partition coefficient. V_p (m^3) and A_p (m^2) are the volume and surface area of a single particle.

3 Case study

3.1 Model description

The hypothetic room geometry is 5 m long, 3 m wide, and 3 m hihg, as shown in Fig. 1, which is representative in daily life. The inlet and outlet are both 0.4 m × 0.4 m at the center plane Y = 1.5 m. Both the surfaces of bookcase and desk are the DEHP sources in the room, adjacent to which the gas-phase DEHP concentration equal 1.1 μg/m^3.

Two ventilation modes named side ventilation mode (SVM) and ceiling ventilation mode (CVM) are designed to supply air flow by two inlets located at the side wall and the ceiling wall with particles. Two inlet velocities 0.3 m/s and 1.2 m/s are selected to provide low and high levels of air flow. The particle originated from outdoor air with 100 μg/m^3. The particle diameter is set to 2.5 μm, and its density is 1000 kg/m^3, A monitoring point (X = 5, Y = 1.5, Z = 0.4) at the center of outlet is set up to estimate the time for the steady state of particle concentration field. After simulated for about 15 min with the two inlet velocities, the concentration value at the monitoring point has no longer changed, which could be considered to reach a steady state for both flow and concentration fields to present the final results of the simulation.

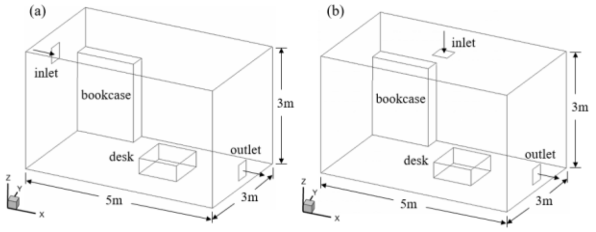

Fig. 1. Schematic of the ventilated room; (a) SVM; (b) CVM.

Three sets of uniform structured grids 132,432, 376,500 and 588,088 are tested for the transient simulation of physical model. The 376,500 grids for the velocity profile and the turbulent intensity along the x-direction of inlet center plane shows a quite similar result to those of the 588,088 grids as shown in Fig. 2. Hence, all the following simulations are based on the 376,500 grids to save the simulation time. All the simulations are carried out by the ANSYS Fluent (version 2020 R2) software.

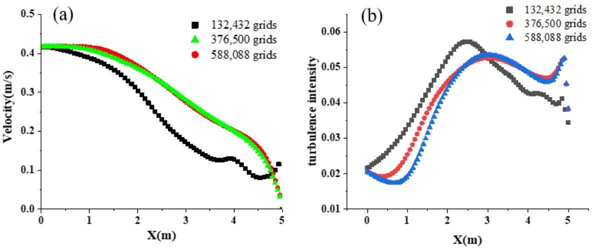

Fig. 2. Grids number independence test; (a) velocity; (b) turbulent intensity.

4 Results and discussion

4.1 DEHP concentration distribution in different ventilation systems

The simulated airflow field and gas-phase DEHP concentration distribution at the center surface are shown in Fig. 3 when the inlet velocity is 0.4 m/s under SVM and CVM conditions. For both cases, the concentration distribution of gas-phase DEHP shows a strong correlation with air flow field. The concentration distribution is intimately coupled to the velocity field, which is consistent with previous study [15].

With SVM (Fig. 3a), the airflow in the upper part of the room is relatively unidirectional and fast, while the airflow in the lower part of the room is well circulated and mixed well. Meanwhile, there exist low concentration in the upper portion of the room due to the constant fresh air flow with certain velocity into the room. In contrast, the gas-phase DEHP is well mixed in the lower part of the room and has a higher concentration near the source location. Similarly, the airflow field and gas-phase DEHP concentration distribution exhibit the same characteristics as the CVM (Fig. 3b). Therefore, it is proved that the concentration field of gas-phase DEHP in room is not uniform, which can be attributed to the airflow pattern in the present air conditioning system.

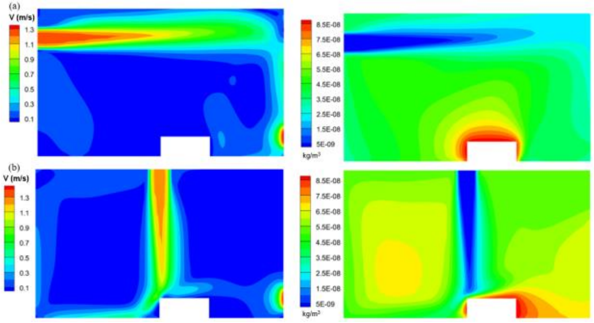

Fig. 3. Simulation result of airflow field and gas-phase DEHP concentration distribution at the center surface when inlet velocity is 1.2 m/s. (a) SVM; (b) CVM.

Fig. 4 shows the particle concentration distribution and particle-phase DEHP concentration distribution at the center surface with SVM and CVM. For both cases, there exist higher particle concentration regions due to the vortex structure induced by the turbulent flow and deposition effect by the gravity. Meanwhile, the higher particle-phase DEHP concentration levels exist in the area with high concentration of particles. However, at the inlet airflow, it takes a period of time for "fresh" particles without DEHP to enter the room from the outside and then into the dynamic action of gas-phase DEHP. Therefore, there is a lower particle-phase concentration level of DEHP at the inlet airflow. As a result, it can be concluded that the influence of particle concentration and particle age, in any kind of airflow pattern, are both important factors affecting the particle-phase DEHP concentration distribution.

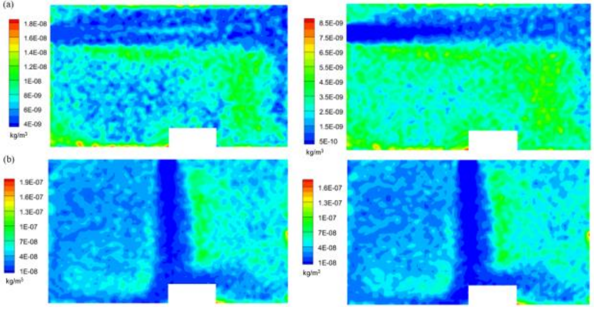

Fig. 4. Simulation result of particle concentration distribution and particle-phase DEHP concentration distribution at the center surface when inlet velocity is 1.2 m/s. (a) SVM; (b) CVM.

4.2 DEHP concentration distribution in different ventilation rate

As shown in Fig. 5a and b, the location of the sources and the indoor airflow field are still the decisive factors for the concentration distribution of gas-phase DEHP. However, with the decrease of inlet velocity and lower turbulence intensity, particle deposition exhibits the most obvious effect (Fig. 5c). The particle-phase DEHP has a high concentration near the ground due to the massive deposition of particles (Fig. 5d). In this case, the exposure of children to DEHP will be greatly increased, and the particle resuspension caused by the movement of indoor personnel will further increase the risk of human exposure.

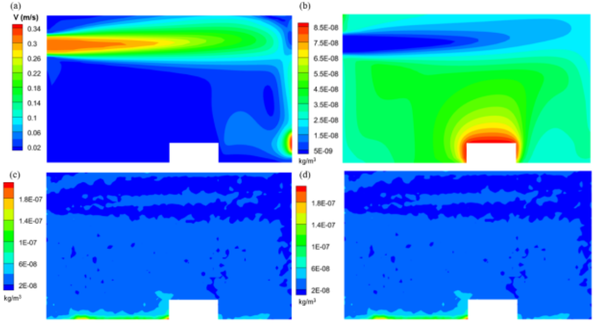

Fig. 5. Simulation result at the center surface when inlet velocity is 0.3 m/s with the SVM. (a) airflow field; (b) gas-phase DEHP concentration distribution; (c) particle concentration distribution; (d) particle-phase DEHP concentration distribution.

In order to explore the effect of particle age on the dynamic interaction between particles and gas phase DEHP, 50 particles were randomly sampled at the outlet for statistics. Fig.6 shows that the concentration of DEHP adsorbed on a single particle C_{pp} (kg/m^3) is closely related to the particle age. The previous results shows that the in the region close to the inlet, the particles mostly have smaller particle age and therefore lower particle-phase DEHP concentration. Regions farther from the inlet, most notably particles deposited on the ground, have a greater proportion of "old" particles and therefore higher concentration of particle-phase DEHP. Therefore, it proved that particle age is also a key factor affecting the particle-phase concentration.

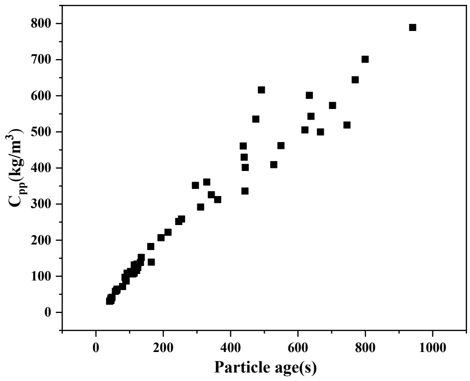

Fig. 6. DEHP concentration on individual particle with different particle age at the outlet surface.

5 Conclusion

1. Results of the life-size house modeling work suggest that gas-phase DEHP concentration distribution shows a strong correlation with air flow field. In addition, the location of DEHP source also significantly affects the concentration distribution.
2. The effect of particle dynamics on particle-phase DEHP concentration distribution is still evident throughout the calculation time. Particle dynamics is influenced by airflow field, inlet velocities, particle size and other factors. For example, the deposition effect is most pronounced when the inlet velocity is small, and we know that the deposition effect is closely related to the particle size. In contrast, the distribution of gas-phase SVOCs has no apparent influence on particle-phase concentration distribution.
3. The concentration of DEHP adsorbed on a single particle is closely related to the particle age. It proves that these "old" particles, which stay indoors longer, pose a greater threat to human exposure.
4. Previous studies have shown that airborne DEHP is dominated by the particle phase [12], which is closely related to particle dynamics. Therefore, further studies on the dynamic interaction between indoor particle dynamics and gas-phase DEHP are needed to reduce residents' exposure to airborne DEHP.

This study was financially supported by National Natural Science Foundation of China (Grant No. 51878553).

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