Formaldehyde emission behavior of building materials: On-site measurements and modeling approach to predict indoor air pollution

Delphine Bourdin [a,b], Pierre Mocho [c,*], Valérie Desauziers [a], Hervé Plaisance [a]

[a] Pôle RIME C2MA, Ecole des Mines d'Alès, Hélioparc, 2 Avenue Pierre Angot, 64053 Pau Cedex 9, France
[b] NOBATEK, 67 Rue de Mirambeau, 64600 Anglet, France
[c] Laboratoire Thermique Energétique et Procédés, Université de Pau et des Pays de l'Adour, BP 7511, 64075 Pau, France

HIGHLIGHTS

- New method to measure gas phase concentration of formaldehyde at material surface.
- New method to quantify the mass transfer of formaldehyde at material surface.
- Assessment of a mass balance model to predict indoor air quality.
- On-site measurements were required to predict their real impact on indoor pollution.

ARTICLE INFO

ABSTRACT

The purpose of this paper was to investigate formaldehyde emission behavior of building materials from on-site measurements of air phase concentration at material surface used as input data of a box model to estimate the indoor air pollution of a newly built classroom. The relevance of this approach was explored using CFD modeling. In this box model, the contribution of building materials to indoor air pollution was estimated with two parameters: the convective mass transfer coefficient in the material/air boundary layer and the on-site measurements of gas phase concentration at material surfaces. An experimental method based on an emission test chamber was developed to quantify this convective mass transfer coefficient. The on-site measurement of gas phase concentration at material surface was measured by coupling a home-made sampler to SPME. First results had shown an accurate estimation of indoor formaldehyde concentration in this classroom by using a simple box model.

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1. Introduction

Exposure to indoor air pollutants was one of the primary environmental health stressors, since people spend 80–90% of their time within enclosed living spaces [1]. Among indoor pollutants, volatile organic compounds (VOCs) were of environmental interest because they could be responsible for health hazards and/or malodorous atmospheres [2]. Regarding the large number of indoor sources such as building materials, furniture, burning of petroleum products, smoking, electrical appliances, use of cleaning products and other household chemicals, they were found to be present at higher concentrations in indoor air than outdoors [3]. To limit their levels, one strategy consisted on control of their major emission sources, especially building materials [4,5]. In France, as in several European countries, new products were evaluated through a time-consuming procedure involving a 28-days emission test within an environmental chamber or an emission cell [6]. These
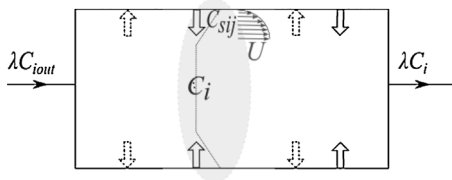
Fig. 1. Physical concept of the one-box model.

test methods were based on a dynamic sampling mode to transfer the VOCs emitted from the material to an active sampling tube to concentrate the compounds [7,8]. Thus, clean air supply, air flow meters and control of temperature and humidity were needed. These protocols, covered by ISO standards [9,10], were suitable for laboratory testing for material labeling but were not able to evaluate the material behavior in real indoor environments in the aim of determining their impact on indoor air quality. For this purpose, static sampling methods [11–14] have been recently investigated to obtain simpler and faster on-site sampling. One of these methods consisted of coupling a standard FLEC® emission cell in static mode with solid-phase microextraction (SPME) for rapid sampling and simple thermal desorption, directly performed in a GC injector and allowed a multi-pollutant analysis [15,16]. Assuming that equilibrium was reached inside the emission cell, the headspace concentration could be considered as the gas phase concentration at the material surface and quantified with a SPME fiber [17]. The ability to measure in situ the surface concentration of building materials offered opportunities to predict the indoor air quality (IAQ) and personal exposure level by modeling approaches. Two major types of computer simulation techniques for modeling IAQ were developed: mass-balance models and computational fluid dynamics (CFD) techniques [18]. Mass-balance models were used to simulate average indoor air pollutant concentration as a function of outdoor concentration, building characteristics (volume, air exchange rate...) and indoor sources/sinks. They were widely used due to the simplicity of the mathematics involved [19–21]. CFD models concerned a microscopic view of IAQ by examining the detailed flow fields and pollutant concentration distributions within a room. Some significant difficulties with CFD concerned the conception, the meshing of the geometry and the setup of the model. Moreover, calculation step was time consuming [22–24]. The developed model was inspired from box models, which were the oldest and most widely used to study the indoor environments [19]. A CFD model was also implemented to discuss the relevance of the box model in the studied indoor environment.

2. Theory/calculation

2.1. One-box model

The simple one-box model (or single zone model) described the change of a pollutant's indoor concentration in a well-mixed room as a differential equation, in which production processes add to the concentration with time and loss processes subtract from the concentration with time. Variants of the model relied on the definition of production and loss processes. Many studies had focused on the chemical reactivity of indoor pollutants, based on data from tropospheric chemistry [25–28] or on the sorption of VOCs on building materials [29]. In our approach, building materials were considered both as VOC sources and sinks from indoor air. If VOC concentration at material surface was higher than indoor air concentration then the material was considered as a VOC emission source (Fig. 1). Inversely, if surface concentration was lower than indoor air concentration then the material was a VOC sink. Therefore, the behavior of the building material was a function of the

value of indoor air concentration, which depended itself on outdoor air exchange rate (defined by outdoor airflow to room volume ratio). The originality of this work was the quantification of real contribution of materials on IAQ by on-site measurements of both gas phase concentration at material surface and indoor air concentration. The mass balance for a controlled volume could be expressed by the following differential equation (Fig. 1):

$$\frac{\partial C_i}{\partial t} = \sum_{j=1}^{m} Q_{ij} + \lambda C_{iout} - \lambda C_i \tag{1}$$

where C_i was the average indoor air concentration of the pollutant i ($\mu g\,m^{-3}$), Q_{ij} the contribution of the material j to the IAQ (source or sink of pollutant i) ($\mu g\,m^{-3}\,s^{-1}$), λ the outdoor air exchange rate (s^{-1}), C_{iout} the average outdoor air concentration of pollutant i ($\mu g\,m^{-3}$), t the time and m the total number of materials within the room. In Fig. 1, C_{sij} was the gas phase concentration of the pollutant i at the material j surface.

At the material/air interface, VOC mass transfer could be expressed as:

$$Q_{ij} = h_{ij} \frac{A_j}{V}(C_{sij} - C_i) \tag{2}$$

where h_{ij} was the convective mass transfer coefficient of pollutant i through the boundary layer over the material j, A_j the surface area of the material j and V the volume of the room.

Substituting (2) into (1), we obtained:

$$\frac{\partial C_i}{\partial t} = \sum_{j=1}^{m} h_{ij} \frac{A_j}{V} C_{sij} + \lambda C_{iout} - \left(\sum_{j=1}^{m} h_{ij} \frac{A_j}{V} + \lambda \right) C_i \tag{3}$$

Assuming h_{ij}, C_{sij}, λ, A_j, V and C_{iout} were constant, Eq. (3) could be integrated analytically to give:

$$C_i(t) = \frac{\sum_{j=1}^{m} h_{ij}(A_j/V)C_{sij} + \lambda C_{iout}}{\sum_{j=1}^{m} h_{ij}(A_j/V) + \lambda} \left(1 - e^{-\left(\sum_{j=1}^{m} h_{ij}(A_j/V) + \lambda \right) t} \right)$$
$$+ C_{i0} e^{-\left(\sum_{j=1}^{m} h_{ij}(A_j/V) + \lambda \right) t} \tag{4}$$

where C_{i0} was the indoor air concentration of pollutant i at $t = 0$ (generally, $C_{i0} = C_{iout}$).

At steady state, Eq. (3) became:

$$C_i = \frac{\sum_{m}^{j=1} h_{ij}(A_j/V)C_{sij} + \lambda C_{iout}}{\sum_{m}^{j=1} h_{ij}(A_j/V) + \lambda} \tag{5}$$

The h_{ij} coefficient was determined by two ways: one based on empirical equations and the other on experimental protocol using a chamber test, as detailed on Material and Methods section.

2.2. Determination of the convective mass transfer coefficient from empirical equations (h_{ij})

In laminar flow over a flat surface, the convective mass transfer coefficient (h_{ij}) could be deduced from relationship among Sherwood number (Sh), Schmidt number (Sc) and Reynolds number (Re) according to the following equation [30]:

$$Sh = 0.664 Sc^{1/3} Re^{1/2} \tag{6}$$

where $Sh = h_{ij}L_c/D_{iair}$, $Sc = \nu/D_{iair}$, $Re = UL_c/\nu$, ν was the kinematic viscosity of the air ($m^2\,s^{-1}$), U the mean air flow velocity over the material ($m\,s^{-1}$), L_c the characteristic length of the material/fluid system (m) and D_{iair} the molecular diffusion coefficient of the compound i in the air ($m^2\,s^{-1}$). This coefficient could be estimated through two main methods: the Fuller, Schettler and Giddings (FSG) method and the Wilke and Lee (WL) method [31].