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ABSTRACT

The aim of this article is to present a validated model to study the influence of humidity on formaldehyde sorption of building material and the effect of formaldehyde and moisture buffering capacity of hygroscopic porous material on indoor air environment. New empirical formulas proposed to describe the diffusion and partition coefficients as functions of humidity based on previous experimental data are incorporated in a coupled moisture and pollutants transport simulation model. The numerical model that takes into account the effect of RH (or moisture) in building materials on VOC diffusion can be used to simulate VOC emissions/sorption from building materials. The developed model is implemented in the environment SPARK (Simulation Problem Analysis and Research Kernel) using finite difference technique. The model is then applied to study the hygric and formaldehyde (FOR, a water soluble VOC) behavior of calcium silicate (CS) subjected to different dynamic conditions of RH and formaldehyde. The results obtained in this paper help to emphasize the importance of moisture and formaldehyde buffering capacity as a new key parameter when selecting clean and hygroscopic building materials in building design because they can contribute to dampen indoor RH and formaldehyde variations. In addition, the impact of RH variation is significant and needed to be taken into account in the simulation to correctly predict the indoor formaldehyde concentration. The model developed in this paper can also be used to optimize the hygric performance and IAQ in building.

Keywords: Formaldehyde, VOC, moisture, VOC buffering capacity, moisture buffering capacity, Indoor Air Quality (IAQ), calcium silicate, hygroscopic material

1. Introduction

The means for keeping the indoor relative humidity (RH) and pollutant concentration below a threshold level of interests are necessary and essential to improving building performance in terms of indoor air quality (IAQ), energy performance and durability of building materials. For evaluating the indoor air quality and thermal comfort, concentrations of pollutants such as the volatile organic compounds (VOC), indoor temperature and relative humidity in building are the most important factors.

High moisture levels can damage construction and have an adverse effect on health (Bornehag et al 2001; Bornehag et al 2004). Microbial growth, encouraged by high relative humidity, leads to poor indoor air quality and building deterioration. Indoor moisture levels can be determined from the amount of water vapor added to the house and the amount removed. It has been shown that the use of the moisture buffering capacity of materials is one of passive ways to keep the variation in RH between threshold levels in order to save energy and improve the indoor quality (Olalekan and Simonson 2006, Woloszyn et al 2009, Tran Le et al 2010, Cascione et al 2020, Reuge et al 2020, Vares et al 2020).

Regarding indoor air quality, formaldehyde is the most common and a compound of concern in the indoor environment (Salthammer et al 2010). Most studies focused on the impact of relative humidity (RH) under a constant temperature on VOC emission of building materials which can be characterized by three parameters: the diffusion coefficient ($D_{m,VOC}$), the partition coefficient ($K_{m,VOC}$) and the initial emittable concentration ($C_{m,VOC,0}$). Note that if the temperature is not constant in the experimental tests, the use of absolute humidity, which expresses the amount of water vapor in the wet air of unit volume (1 m^3) and does not change when air is heated or cooled, in stead of relative humidity, is better to analyse the results obtained. According to Bouilly et al (2006), Blondeau et al (2008), the impact of RH on VOC emission can be considered by three kinds of interactions: competition for adsorption at the pore surfaces of the materials, as well as change in the diffusion properties and possible absorption/desorption of VOCs due to capillary condensation of water vapor in the micropores of the materials. Therefore, the VOC behavior of building materials depends on physical properties and environmental condition (temperature, RH, radiation, etc.) under which the material is subjected. Many experimental studies focused on the impact of humidity on formaldehyde emission and showed that the formaldehyde emission rate of building materials which have been identified as VOC emitters, increased with increasing relative

humidity (Fang et al 1999, Zhang et al 2002, Huang et al 2006, Xiong and Zhang 2010, Parthasarathy et al 2011, Frihart et al 2012, Sidheswaran et al 2013). The experimental studies carried out by Liang et al (2016) focusing on a medium-density fiberboard (MDF) showed that mechanism of humidity effect on formaldehyde emission is very complicated and formaldehyde emission from the MDF presented the simultaneous and consistent responses to humidity changes. For the same material, experimental results indicate that initial emittable concentration ($C_{m,0}$) of formaldehyde changes significantly with humidity Huang et al (2015). It should be explained by the hydrolysis of polymers and adsorption competitions between formaldehyde and water molecules in MDF. Note that there is no theoretical correlation between the VOC characteristic (diffusion, sorption and initial emittable concentration) and humidity and there are very limited experimental research focusing on the effect of humidity on the diffusion coefficient and partition coefficient (Huang et al 2006, Xu et al 2009, Xu and Zhang 2011, Liang et al 2016). It is very interesting to underline that clean and hygroscopic building materials could contribute to ameliorate the indoor air quality during the entire service life thanks to their pollutant buffering capacity which could be similar to moisture buffering capacity. This assumption is supported by some studies which showed that there is a similarity between moisture and VOC diffusion through porous media (Salonvaara et al 2006, Xu et al 2009). Compared to the previous experimental study of Xu et al (2009), the similarities between VOC and moisture transport in building materials have been discussed for non-isothermal problems in the framework of Annex 68 (Rode et al 2020). Note that the diffusion and sorption of VOC and water vapor in building materials would be related to physical and chemical properties. Da Silva et al (2016) showed that the adsorption/desorption characteristics are related to material microstructure and polarity of the VOCs. Therefore, the difference in physical properties (size, molar mass, polarity, etc.) would play a role in the similar behavior between VOCs and water vapor which needs to be further investigated. The reduction of indoor VOC through adsorption processes is an important research objective due to its potential to provide improved quality of life for individuals in exposed spaces (Hunter-Sellars et al 2020). Formaldehyde sorption/ desorption process of gypsum boards has been carried out by Matthews et al (1987) and showed that it has a significant storage capacity and influences significantly the formaldehyde concentration. The experimental data and semi-empirical models describing the sorption of organic gases in a simulated indoor residential environment (a 50 m^3 room finished with painted wallboard, carpet and cushion, draperies and furnishings) have been carried out by Singer et al (2004). The results showed that the sorption appears to be a relevant indoor process and can influence gas-phase concentrations.

The study of Gunschera et al (2013) has revealed that the formaldehyde concentration in real indoor air is influenced by multiple parameters and can only be explained accurately when taking into account different parameters such as sorption as well as diffusion.

The literature review showed that there is lack of a comprehensible and validated model which can respond to this need to reduce the tests realized which were time-consuming and expensive. Therefore, in this paper, a new coupled moisture and VOC transport simulation model is developed and presented to study the effect of VOC and moisture buffering capacity of porous material on indoor VOC concentration and RH. It is very important to highlight that the effect of moisture in building material on the diffusion and storage properties of VOC is taken into account in the present model. In this study, formaldehyde (FOR) which is water soluble and a gaseous pollutant produced by both human activity and natural sources, was selected as reference VOCs for this article because it is a typical indoor VOC. Understanding the interaction between indoor building materials and formaldehyde is meaningful to control IAQ in buildings. Therefore, calcium silicate, an inorganic insulation material, which is used in interior thermal insulations systems was chosen as a reference material in this study due to its well-characterized moisture diffusion properties and wide usage as a building insulation material. Besides, calcium silicate is hygroscopic material and clean meaning that it had no VOCs added in the fabrication and very interesting to study its moisture and VOC buffering capacity.

To date, the impact of RH on VOC parameters (D_m, K, $C_{m,0}$) is mainly conducted by experiments under of a set of specific environmental conditions and there is a lack of information concerning the transient effect of RH on VOC sorption of building materials, while such information is essential to understand accurately the impact of building material on IAQ and thermal comfort. Therefore, this study aims to fulfil the gap of knowledge in literature and to investigate the impact of formaldehyde and moisture sorption capacity of hygroscopic material on IAQ and indoor RH under a dynamic condition of humidity and formaldehyde. The paper is organized as follows: First, a coupled moisture, air and VOC transport model taking into account the effect of moisture in building material on VOC diffusion is presented in section 2. Then, the numerical solutions and model validation are detailed in section 3. Next, the impact of RH on formaldehyde and moisture buffering capacity of silicate calcium is discussed in section 4. Finally, the main conclusions are presented in section 5.

2. Numerical model

2.1. Coupled moisture and VOC transport model in porous building materials

A coupled hygric and pollutant simulation model as presented in Figure 1 has been developed to study the similarity between moisture and pollutant behavior of porous building materials. The model includes equations that describe: moisture and VOC diffusion, moisture and VOC sorption, impact of RH and T on moisture and VOC diffusion/sorption, boundary condition between indoor/outdoor air and building envelope surfaces, etc. In this section, the similarity between the moisture and VOC transport and storage coefficients of building materials will be established and explained based on the following accepted assumptions:

- Transport of water vapor in building materials is modelled analogously to the transport of VOC.
- Sorption of water vapor is described by the sorption isotherm curve (due to its multilayer adsorption) while it is modelled by the partition coefficient for VOC because it is generally considered as monolayer adsorption.

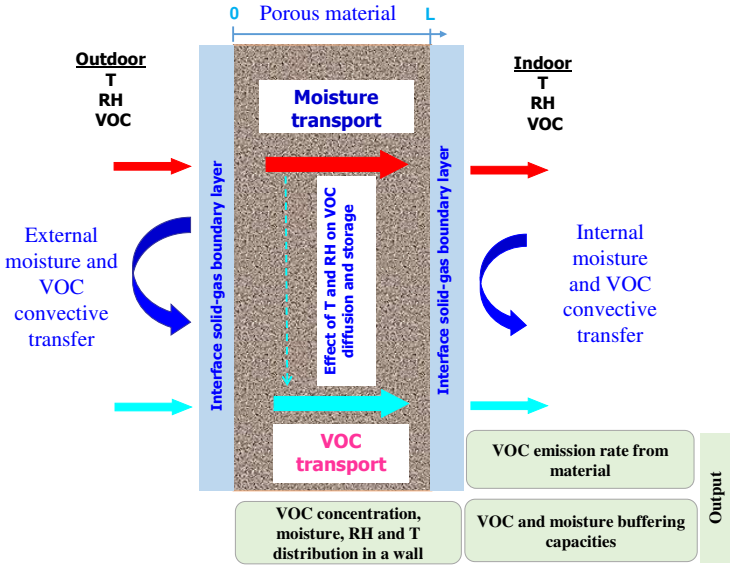


Figure 1: Coupled moisture and VOC transport model in a wall

Note that, the model takes into account the effect of moisture content in building materials on VOC transport and storage if data is available. To establish the similarity between the VOC and moisture diffusion models, only concentration gradient of VOC or moisture is assumed to be driving force in the material and the chemical reactions are neglected in this work.

The VOC mass transport within the wall can be described by the one-dimensional diffusion (Yang et al 2001, Zhang 2005) for a dry material with homogeneous diffusivity,:

$$\frac{\partial C_{m,VOC}}{\partial t} = \frac{\partial}{\partial x}\left(D_{m,VOC} \frac{\partial C_{m,VOC}}{\partial x} \right) \quad (1)$$

Where $C_{m,VOC}$ is VOC concentration in the material (kg/m^3), $D_{m,VOC}$ is diffusion coefficient of the VOC in the material (m²/s), x is abscissa (m) and t is time (s). Here, in the developed numerical model, the $D_{m,VOC}$ is a function of relative humidity/moisture in the material while the dependence of $D_{m,VOC}$ on pollutants concentration is neglected as generally accepted under low VOC concentration condition.

There is an equilibrium which exists between the concentration of VOC in a material ($C_{m,VOC}$) and the concentration in air ($C_{a,VOC}$), which is defined by the partition coefficient $K_{m,VOC}$:

$$C_{m,VOC} = K_{m,VOC} . C_{a,VOC} \quad (2)$$

The diffusion coefficient of VOC in the material ($D_{m,VOC}$) can be determined from the VOC diffusion coefficient in the free air (D_{VOC}^{air}) and diffusion resistance factor (μ_{VOC}) of VOC (Xu et al 2009):

$$D_{m,VOC} = \frac{D_{VOC}^{air}}{\mu_{VOC} K_{m,VOC}} \quad (3)$$

Note that the VOC diffusion coefficient in free air (D_{VOC}^{air}) can be calculated if the thermodynamic property of the VOC is known (Stephan and Mayinger, 1988). Concerning the VOC diffusion resistance factor, μ_{VOC}, it has to be determined experimentally using a dual test chamber method (Xu et al, 2009).

At the material-air interface, an instantaneous equilibrium between VOC concentration (kg/m^3) in the air near material surface ($C_{a,VOC,s}$) and the one in the surface layer ($C_{m,VOC,s}$) is assumed:

$$C_{m,VOC,s} = K_{m,VOC} . C_{a,VOC,s} \quad (4)$$

With the following boundary conditions (the external surface x=0 and internal surface x=L):

$$-\left(D_{m,VOC} \frac{\partial C_{m,VOC}}{\partial x} \right)_{x=0,e} = h_{m,VOC,e} \left(C_{a,VOC,e} - C_{a,VOC,s,e} \right) \quad (5)$$

$$-\left(D_{m,VOC}\frac{\partial C_{m,VOC}}{\partial x}\right)_{x=L,i}=h_{m,\mathrm{VOC},i}\left(C_{a,VOC,s,i}-C_{a,\mathrm{VOC},i}\right) \quad (6)$$

Where $C_{a,VOC,i}$ and $C_{a,VOC,e}$ are VOC concentration in the room air and outside (kg/m^3), and $h_{m,VOC,e}$ and $h_{m,VOC,i}$ are convective VOC transfer coefficients (m/s) for the external and internal surfaces, respectively.

Concerning the moisture transport model, the moisture transport within the wall can be described by the one-dimensional diffusion for using moisture content in material as driving force (Philip and De Vries 1957):

$$\frac{\partial\theta}{\partial t}=\frac{\partial}{\partial x}\left(D_{m,\mathrm{wv}}\frac{\partial\theta}{\partial x}\right) \quad (7)$$

Where θ is moisture volumetric content in the material (m^3 of water/m^3 of material), $D_{m,wv}$ is diffusion coefficient of the moisture in the material (m²/s) which is defined by:

$$D_{m,\mathrm{wv}}=\delta_{wv}\frac{P_{v,sat}}{\rho_w}\frac{1}{\partial\theta/\partial RH}=\frac{\delta_{wv}^{air}}{\mu_{wv}}\frac{P_{v,sat}}{\rho_w}\frac{1}{\partial\theta/\partial RH} \quad (8)$$

Where $\partial\theta/\partial RH$ is the slope of the sorption isotherm curve which designates the relationship between the moisture content and the relative humidity at a fixed temperature, δ_{wv} is water vapor permeability of material (kg/(m.s.Pa)) which can be determined in accordance with standard cup-test (such as ASTM standard E96/E96M-05, NF EN 12086), $P_{v,sat}$ saturation pressure of water vapor (Pa), μ_{wv} the vapor diffusion resistance factor and δ_{wv}^{a} is water vapor permeability of still air ((kg/(m.s.Pa)) which can be determined from D_{wv}^{air} (water vapor diffusion coefficient in the free air, m²/s), temperature and the gas constant for water vapor (R_v= 461.5 J/(kg·K)):

$$\delta_{wv}^{a}=\frac{D_{wv}^{air}}{R_vT} \quad (9)$$

By replacing (9) in (8) we have:

$$D_{m,\mathrm{wv}} = \frac{D_{wv}^{air}}{\mu_{wv}\dfrac{\rho_w R_v T}{P_{v,sat}}\dfrac{\partial\theta}{\partial RH}} = \frac{D_{wv}^{air}}{\mu_{wv} K_{m,wv}} \tag{10}$$

As with the VOC, by identifying two equations (3) and (10), the coefficient $K_{m,wv}$ which has been introduced in (10) is the "partition coefficient" for water vapor, which is similar to $K_{m,VOC}$ in (3) for VOC and can be calculated as following:

$$K_{m,wv} = \frac{\rho_w R_v T}{P_{v,sat}}\frac{\partial\theta}{\partial RH} \tag{11}$$

Note that the partition coefficient ($K_{m,wv}$) for water vapor can be calculated by relating gradients of the absorbed moisture content mass by volume of material, to gradients of the humidity of air by volume of the pores at equilibrium condition. Using this definition to calculate $K_{m,wv}$, the same result was obtained (Rode et al 2020).

Concerning the sorption isotherm, in this article, the Guggenheim-Anderson-deBoer (GAB) model (Timmermann 2003) which is extended from Langmuir and BET theories (Langmuir 1918; Brunauer et al 1938) of physical adsorption, is used to describe the sorption curve. Using the GAB model has many advantages such as having a viable theoretical background and giving a good description of the sorption behavior of hygroscopic material (Andrade et al 2010). The GAB model can be written as follows:

$$w = \frac{w_m C_{GAB} K_{GAB} RH}{\left(1 - K_{GAB} RH\right)\left(1 + K_{GAB} C_{GAB} RH - K_{GAB} RH\right)} \tag{12}$$

Where RH is relative humidity, w is the moisture content (kg of water/kg of material), w_m is the monolayer moisture content value, C_{GAB} and K_{GAB} are energy constants of GAB model.

At the material-air interface, an instantaneous equilibrium between water vapor concentration (kg/m^3) in the air near material surface ($C_{a,wv,s}$) and the one in the surface layer ($C_{m,wv,s}$) is assumed. The following boundary conditions applied to water vapor, respectively for the external (x=0) and internal (x=L) surfaces of the wall:

$$-\left(\rho_w D_{m,wv}\frac{\partial\theta}{\partial x}\right)_{x=0,e}=h_{m,\mathrm{wv},e}\left(C_{a,\mathrm{wv},e}-C_{a,wv,s,e}\right) \tag{13}$$

$$-\left(\rho_w D_{m,wv}\frac{\partial\theta}{\partial x}\right)_{x=L,i}=h_{m,\mathrm{wv},i}\left(C_{a,wv,s,i}-C_{a,\mathrm{wv},i}\right) \tag{14}$$

Where $C_{a,wv,i}$ and $C_{a,wv,e}$ are water vapor concentrations in the room air and outside (kg/m^3), and $h_{m,wv,e}$ and $h_{m,wv,i}$ are convective water vapor transfer coefficients (m/s) for the external and internal surfaces.

Still assuming isothermal conditions (turbulent or laminar flows) at the material surface, the convective mass transfer coefficient for VOC and WV ($h_{m,VOC}$, $h_{m,wv}$) can be calculated from the following equations (White 1991, Axley 1991, Blondeau et al 2008):

For turbulent flows

$$Sh=0.037Sc^{\frac{1}{3}}\,\mathrm{Re}^{\frac{4}{5}} \tag{18}$$

For laminar flows

$$Sh=0.644Sc^{\frac{1}{3}}\,\mathrm{Re}^{\frac{1}{2}} \tag{19}$$

$$Sh=\frac{h_m L}{D^{air}} \tag{20}$$

$$Sc=\frac{\upsilon}{D^{air}} \tag{21}$$

where, Sh is Sherwood number, Sc is Schmidt number, h_m is the convective mass transfer coefficient (m/s), L is the characteristic length (m), D^{air} is diffusion coefficient of VOC/water vapor in air (m^2/s), υ is kinematic viscosity of air at 23 °C, which equals 1.544×10^{-5} m^2/s (Cengel and Ghajar 2010). From equations (18) to (21), we can establish the similarity relationship between $h_{m,wv}$ and $h_{m,VOC}$. Note that, because of the same sample and the same air flow field in the airtight chamber as the same test condition for the combined water vapor and VOC study, the same Reynolds number can be considered, and we can have the following formula:

$$\frac{h_{m,\mathrm{wv}}}{h_{m,\mathrm{VOC}}}=\left(\frac{D_{wv}^{air}}{D_{VOC}^{air}}\right)^{2/3} \tag{22}$$

Note that equation (22) permits to determine the convective mass transfer coefficient of VOC, ($h_{m,VOC}$), from the convective mass transfer coefficient of water vapor $h_{m,wv}$ (or inversely) using the diffusion coefficients of VOC and water vapor in the air, respectively.

2.2 Model for a room

In order to model the indoor VOC and humidity in the room, the nodal method has been used, which considers the room as a perfectly mixed zone characterized by a moisture and pollutant concentrations. Nodal method involves equations for moisture/pollutant (VOC) mass balance and equations describing mass transfer through the walls, additional convection between inside wall surfaces and room ambiance. The moisture/VOC level in the room is determined by the moisture/VOC transfer from interior surfaces, moisture/VOC production rate and the gains or losses due to air infiltration, natural and mechanical ventilation, sources due to habitants of room as well as the moisture/VOC buffering capacity of other room elements (such as furniture, bookshelf, woolen carpet, etc.). This yields to the following mass balance equation for water vapor/VOC:

$$V\frac{\partial C_{a,i}}{\partial t} = Q\left(C_{a,o} - C_{a,i}\right) + \sum A.h_{m,i}\left(C_{as,i} - C_{a,i}\right) + G \qquad (23)$$

Where $C_{a,i}$ is the VOC/water vapor concentration at time t (kg/m^3); $C_{a,o}$ is outdoor ventilation air; V is volume space (m^3); A is exposed area of the material (m²), Q is the volume air flow rate into (and out) of the room (m^3/s), G is the generation rate of VOC/water vapor in the room (kg/s).

3. Numerical solutions and validation

The set of equations describing the model has been implemented in **SPARK** which allows to solve efficiently differential equation systems using the finite difference technique with an implicit scheme (Sowell and Haves 2001; Wurtz et al 2006; Mendonça et al 2006; Tran Le et al 2009; Tran Le et al 2016).

Note that the special interests of the developed model in this paper are the dependencies of moisture transport coefficient, pollutant diffusion coefficient, partial coefficient, etc. upon the relative humidity and temperature can be taken into account if the data is available. This makes it possible to consider the effect of T and RH on pollutant and hygrothermal behavior of building materials into the model.

3.1 Moisture and formaldehyde properties of porous materials and case study

Indoor hygrothermal conditions (temperature and humidity) are generally not constant in buildings which are influenced by the outdoor conditions, occupants and HVAC system, etc. Therefore, the understanding of the VOC behavior of building materials in different temperature and humidity conditions is important to predict correctly the IAQ in buildings. In this article, we focus only on the impact of humidity on formaldehyde (FOR) key parameters ($D_{m,FOR}$ and $K_{m,FOR}$) of building materials. The experimental data (Huang et al 2006, Xu et al 2009, Xu and Zhang 2011, Xu 2012) for different materials (Calcium silicate (CS), Medium Density Fiberboard (MDF), Conventional Wallboard (CWB), Green wallboard (GWB)) which were measured at constant temperature (23°C or 25°C) have been gathered, compared together and presented in Figure 2 and Figure 3. Because the temperatures are not the same in the experimental tests (23°C or 25°C), absolute humidity (AH, g/m^3) has been used for the comparison. Figure 2 and Figure 3 showed that the impact of absolute humidity (AH) on the formaldehyde key parameters for different tested materials is very different. Regarding the impact of humidity on $K_{m,FOR}$ for formaldehyde, Figure 2 shows that increased AH results in increased partition coefficient which can be explained by the formaldehyde absorption into the liquid water under the higher humidity condition. The impact is expected to be different for different materials because their sorption capacities are different. Compared to other materials, the partition coefficient of CS is the highest indicating that this one has the largest storage capacity for formaldehyde. In addition, considering the uncertainty of the results, it can be seen that the impact of humidity on the formaldehyde partition coefficient for GWB and CWB is not significant and can be neglected. Concerning the impact of humidity on $D_{m,FOR}$ for formaldehyde, Figure 3 shows that the influence of AH is not always consistent. Considering the uncertainty of the results, the impact of humidity on the diffusion coefficient of formaldehyde for GWB, CWB and MDF is small and can be neglected. Regarding CS, the

decrease of $D_{m,FOR}$ with increased humidity could be explained by the fact that higher relative humidity will increase the water vapor diffusion in the pores where capillary condensation doesn't occur and could be considered as a competitor of formaldehyde diffusion in material (Blondeau et al 2008, Xu and Zhang 2011). In addition, the capillary condensation in which the pores are filled with liquid water due to higher relative humidity will reduce formaldehyde diffusion in the material. Further, it is important to note that some materials presented in the literature are the material systems (i.e., with painted boards, waterproof coating, etc.). Therefore, the data included also the effect of RH on the formaldehyde transport and storage in the paint layer, etc. (Xu, 2012).

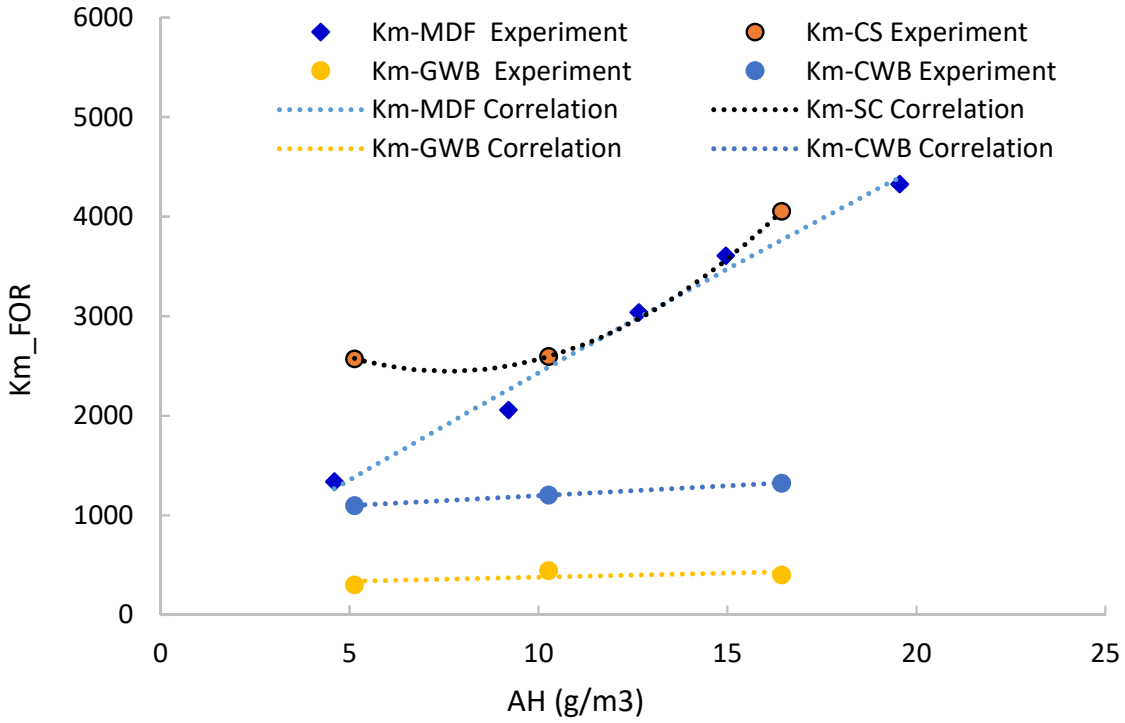


Figure 2: Impact of AH on $K_{m,FOR}$ for formaldehyde (FOR) of different materials based on the experimental results (Huang et al 2006, Xu and Zhang 2011, Xu 2012)

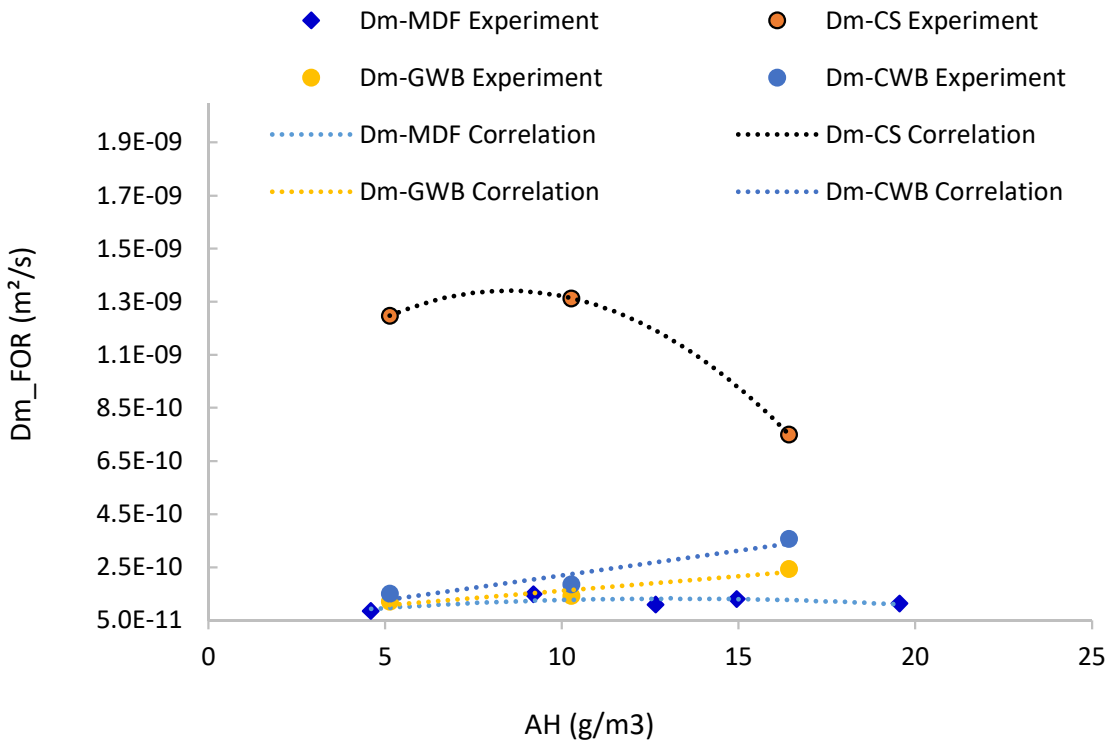


Figure 3: Impact of AH on $D_{m,FOR}$ for formaldehyde (FOR) of different materials based on the experimental results (Huang et al 2006, Xu and Zhang 2011, Xu 2012).

Because there is no theoretical correlation between the formaldehyde parameters ($D_{m,FOR}$ and $K_{m,FOR}$) and humidity and there are very limited experimental work for other materials, we suggest the following correlation based on the data shown in Figure 2 and Figure 3:

$$K_{m,\mathrm{FOR}} = 9.6AH^2 - 84.9AH + 1547.8 \tag{24}$$

$$D_{m,\mathrm{FOR}} = -1.18x10^{-12}AH^2 + 3.3x10^{-11}AH + 2.5x10^{-10} \tag{25}$$

Concerning the model validation, because of the significant impact of RH on formaldehyde properties of calcium silicate, this section focuses on the validation of the developed model by comparing the numerical results with experimental ones obtained with CS using the dynamic dual chamber method carried out by Xu and Zhang (2009; 2011). The hygrique properties of calcium silicate measured by Xu and Zhang (2009) are shown in Table 1 and Figure 4 (sorption isotherm curve) and used for the model validation.

ρ (kg/m^3)	μ_{wv}	Sorption isotherm (GAB model parameters, R^2=0.99)
843.38	8.7	w_m=0.002; C_{GAB}=8; K_{GAB}=0.97

Table 1: Hygric properties of calcium silicate for formaldehyde (Xu and Zhang, 2011) and proposed fitting parameters for GAB model.

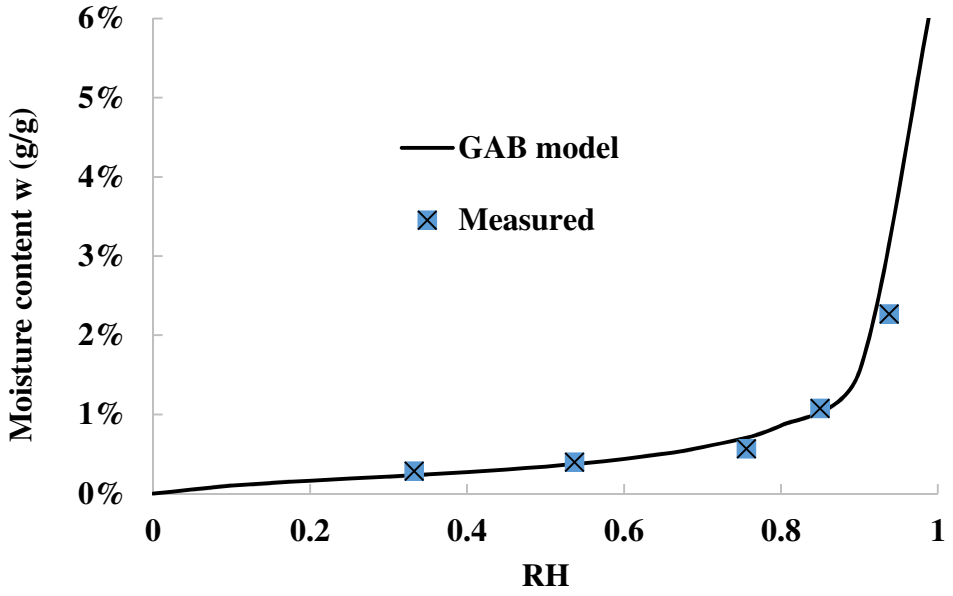


Figure 4: Sorption isotherm of calcium silicate (Xu and Zhang, 2009)

The effect relative humidity on formaldehyde effective diffusion coefficient and partition coefficient of calcium silicate has been measured by Xu and Zhang (2011) under three levels of relative humidity (25%, 50% and 80%) and at constant temperature of 23°C (so AH=5.1, 10.3 and 16.4 g/m^3, respectively) by using a dynamic dual chamber method. Based on the experimental results, we propose two equations which describe the relationship between absolute humidity (AH) and $K_{m,FOR}$ or $D_{m,FOR}$ for calcium silicate as follows:

$$K_{m,\mathrm{FOR}} = 20.56AH^2 - 312.41AH + 3636.23 \tag{26}$$

$$D_{m,\mathrm{FOR}} = -8.52x10^{-12}AH^2 + 1.44x10^{-10}AH + 6.81x10^{-10} \tag{27}$$

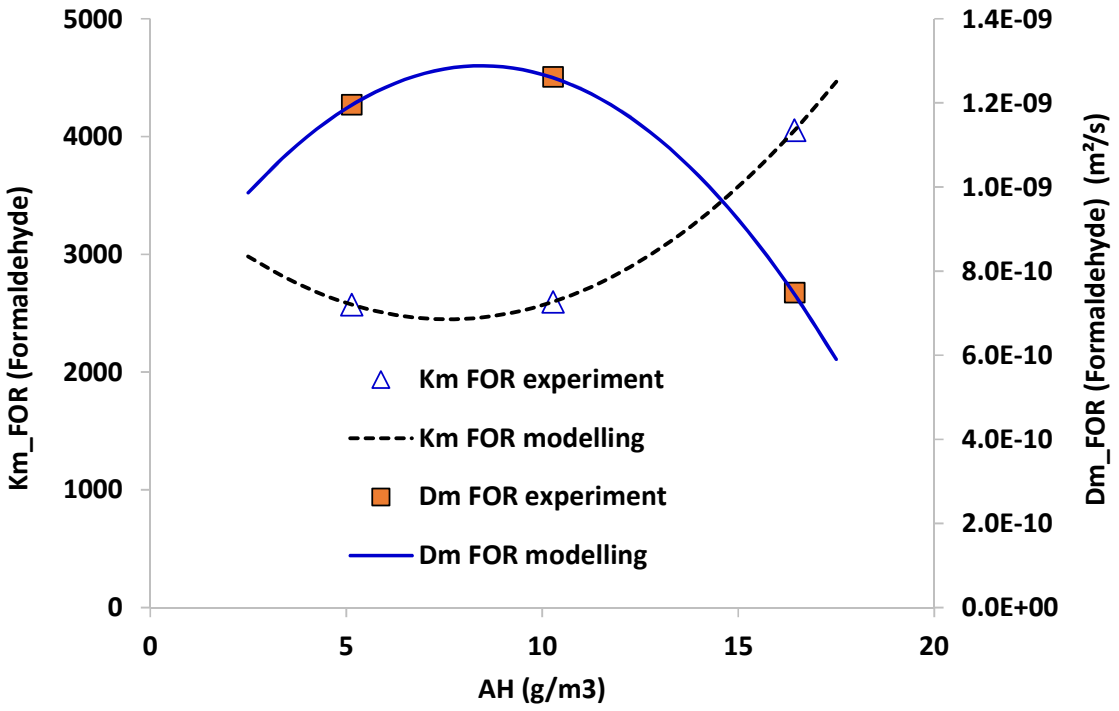


Figure 5: $K_{m,FOR}$ and $D_{m,FOR}$ (formaldehyde) as function of absolute humidity (Xu and Zhang et al, 2009, 2011) of silicate calcium

Figure 5 showed the variations of $K_{m,VOC}$ and $D_{m,FOR}$ of CS as function of absolute humidity (AH). It can be seen that the measurement at AH=16.4 g/m^3 led to a higher partition coefficient and a smaller value of diffusion coefficient for formaldehyde compared to the measurement at AH=10.3 g/m^3. However, between AH=5.1 g/m^3 and AH=10.3 g/m^3, the impact of AH was not significant. More precisely, the measured partition coefficient, $K_{m,FOR}$ is equal to 2574, 2598 and 4057 when the AH=5.1, 10.3 and 16.4 g/m^3, respectively. Note that the impact of AH on $K_{m,FOR}$ is different from one AH range to another because of the non linear nature of the moisture sorption curve (see Figure 4). Concerning the impact of humidity on the effective diffusion coefficients of formaldehyde, Figure 5 showed that it was not significant in the range from AH=5.1 to AH=10.3 (g/m^3), while it is significantly decreased in the range of AH=10.3 to AH=16.4 (g/m^3) ($D_{m,FOR}$ decreases from 1.26×10^{-9} to 7.49×10^{-10} m²/s, respectively).

3.2 Model validation

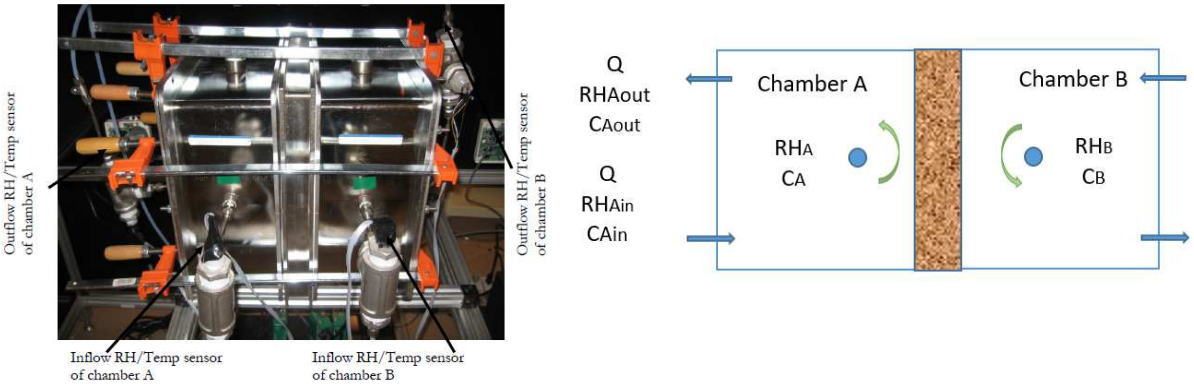
In order to validate the developed model, the two stainless steel chambers which have the dimension of 0.35 m x 0.35 m x 0.15 m each were partitioned by a test specimen (Figure **6**). Each chamber was supplied with inflows under controlled temperature and relative humidity. Both chambers were supplied with the same airflow rate ($Q=6.58\times10^{-2}$ m^3/s) with constant temperature at 23 °C. Concerning the relative humidity of the inflow for both chambers, it was maintained at threshold level by bubbling the liquid water. Regarding formaldehyde, chamber A had a constant formaldehyde injection in the inflow while Chamber B had no formaldehyde injection. The formaldehyde concentration in the inflow of chamber A was set at constant, and the concentration in chambers A and B were continuously monitored until they reached a steady state. Concentrations in the outlets of chamber A and B were monitored as C_{Aout} and C_{Bout}. Under well mixed condition, $C_A=C_{Aout}$ and $C_B=C_{Bout}$ and at steady state, C_{Aout} and C_{Bout} were constant. Formaldehyde concentrations of the outflows of both chambers were measured. Note that, RH and formaldehyde concentration in the inflow of chamber A (C_{Ain} and RH_{Ain}), in chamber B (C_B and RH_B) are used as the input data. The relative humidity and formaldehyde concentration in chamber A (C_A and RH_A) are used for model validation.

The calcium silicate was cut into a 30.5 cm x 30.5 cm x 1.0 cm, and sealed four edge sides by VOC free tape (resulted in a real exposed area of 0.093 m^2) to prevent formaldehyde diffusion through the edges. More information about the testing can be fund in Xu and Zhang (2009, 2011). These tests were used for the model validation:

- Tests for validating the formaldehyde model: formaldehyde concentration has been injected into chamber A while T and RH in both chambers A and B were maintained at 23°C and 50%RH for the first test and 80% RH for the second test. The formaldehyde concentration, RH (C_{Ain} and RH_{Ain}, C_A and RH_A, C_B and RH_B) have been recorded continuously for each test.

- Tests for validating the moisture model: for the first test, the initial relative humidity in both chambers were 25%. Then, the RH of inflow for chamber A was increased to 50% RH while the inflow for chamber B was maintained at a constant relative humidity at 25%. Concerning the second test, the initial relative humidity in both chambers were 50%. Then, the RH of inflow for chamber A was increased to 80% RH while the inflow for chamber B was maintained at a constant RH 50%. Note that the changes of RHs in both chambers were monitored during the testing period.

Concerning the numerical simulation, the specimen has been divided into 25 nodes and the time step is 240s. The initial formaldehyde concentrations C_0 in calcium silicate specimen is 0 ($\mu g/m^3$).



Photograph Schematic

Figure 6: Schematic of dual chamber system (Xu and Zhang 2009; 2011)

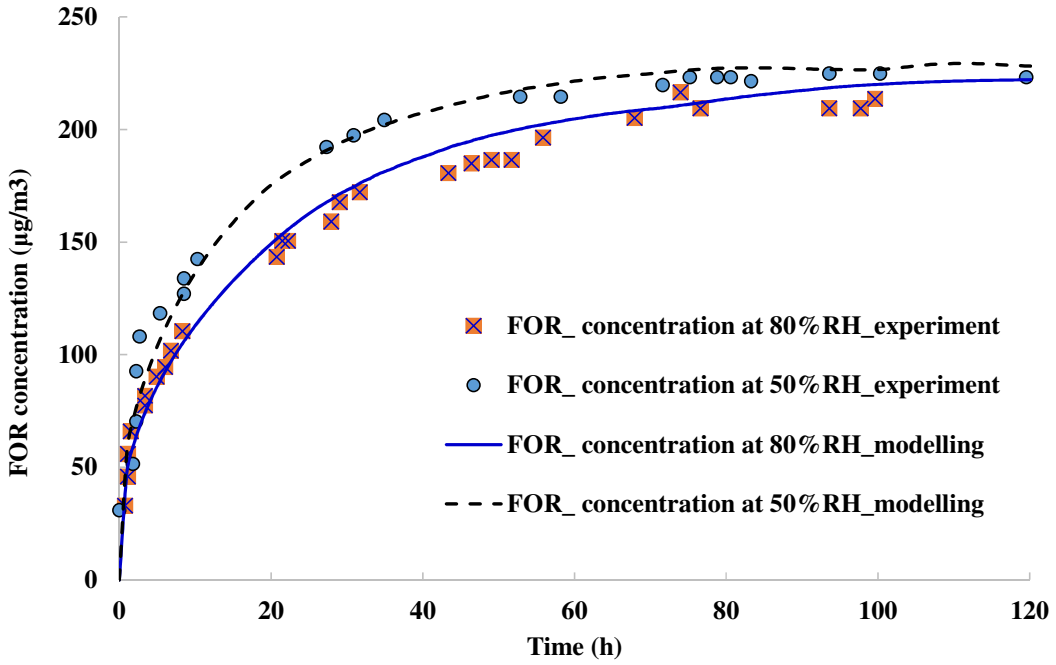


Figure 7: Comparison of experimental and numerical results of indoor formaldehyde (FOR) concentration in the chamber A

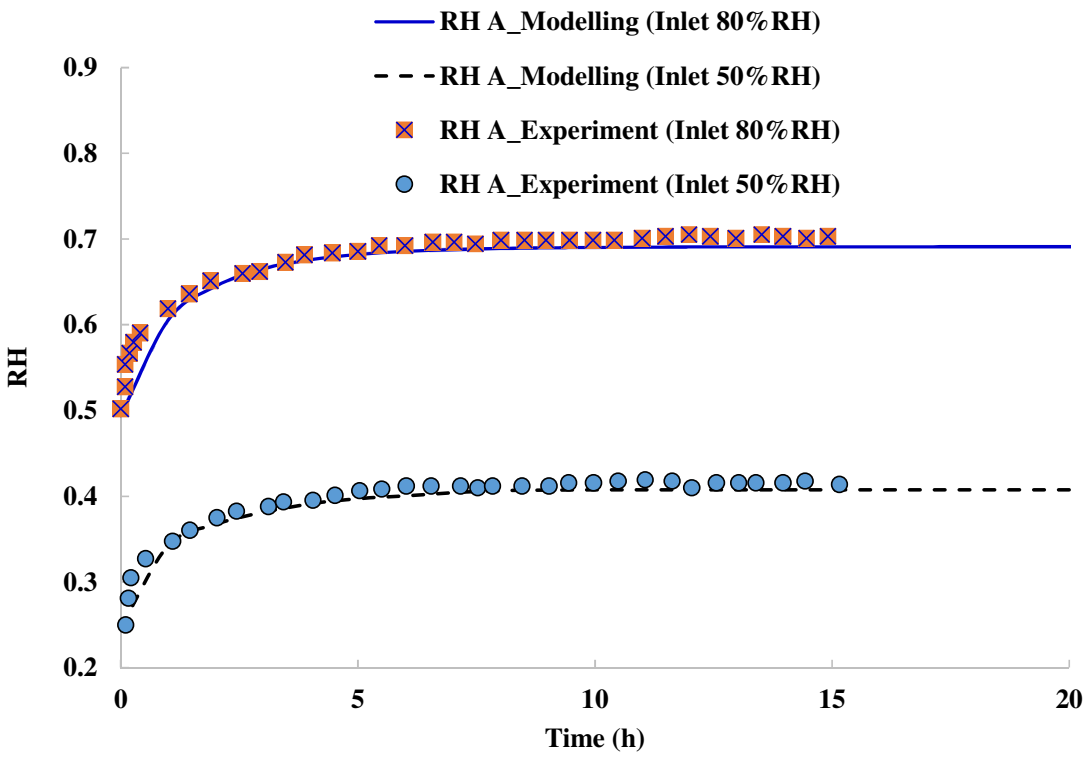


Figure 8: Comparison of results between experimental and numerical results of indoor relative humidity in the chamber A

Figure 7 and Figure 8 compare the simulated formaldehyde concentrations and relative humidity in the chamber A with the experimental data. The results showed a good agreement between the numerical model and experimental results after the few hours for both pollutant and moisture models. The developed model is satisfying to investigate the coupled hygric-pollutant (formaldehyde) behavior of porous building materials. In the next section, the similarity between moisture and formaldehyde buffering capacity and the impact of RH on IAQ and indoor RH will be investigated.

4. Impact of humidity on formaldehyde and moisture buffering capacity of silicate calcium

4.1 Simulation conditions

Regarding the fact that in indoor relative humidity in building may vary in a wider range than the optimal range of relative humidity (e.g. 40-60% RH) and the experimental results showed that the impact of RH on partition coefficient and diffusion coefficient for formaldehyde (see Figure 5) is significant in the range of AH=10.3 g/m^3 and 16.4 g/m^3 (at 23°C, RH= 50% and 80% RH, respectively), a study in depth focusing on the effect of humidity on the formaldehyde behavior of calcium silicate is necessary. In addition, calcium silicate is hygroscopic material and clean meaning that it had no VOCs added in the fabrication, its moisture and formaldehyde buffering capacity is very interesting to study. In order to study the effect of RH on pollutant and moisture inertia of silicate calcium, we consider the same stainless steel chamber (the dimension of 0.35 m x 0.35 m x 0.15 m) as presented above (section dedicated to the experimental validation). The same calcium silicate specimen with a thickness of 1.0 cm and single exposed surface of 30.5 cm x 30.5 cm (other faces are sealed so that no formaldehyde or moisture can pass through) is placed in this airtight chamber in which the temperature and air flow rate are maintained at 23°C and 6.58×10^{-2} (m^3/h), respectively (Figure 9). The initial relative humidity is 50% RH and the initial formaldehyde concentrations C_0 in the calcium silicate sample is 0 (μg/m^3) because it is clean material. The time step is 240s and the CS sample is divided into 25 nodes.

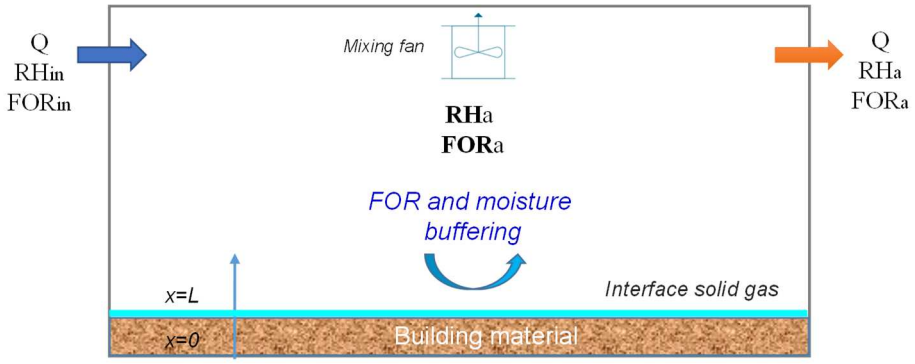


Figure 9: A schematic illustration of a building material placed in an airtight chamber

In this section, two numerical studies have been carried out with two considered models:

- Model with **B**uffering **C**apacity (**BC** model): Simulation taking into account the moisture and formaldehyde sorption capacity.
- Model **Without B**uffering **C**apacity (**Without-BC** model): Simulation neglecting the moisture and formaldehyde sorption capacity.

4.2 Study of the formaldehyde buffering capacity

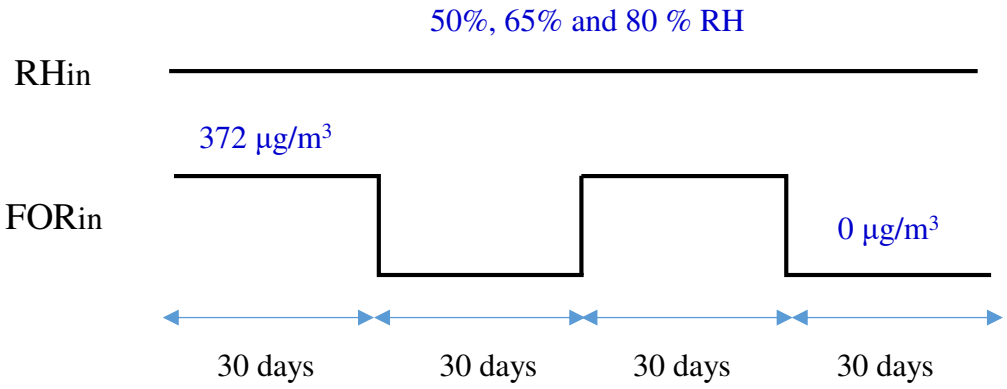


Figure 10: Studied configuration for formaldehyde (FOR) buffering capacity

In order to understand the formaldehyde buffering capacity of silicate calcium, we consider a testing condition which permits to study two distinct adsorption and desorption processes of formaldehyde as shown in Figure 10 : the variation of formaldehyde injected into the airtight chamber has a square-wave form: 30 days of high formaldehyde value FOR_{in}= 372 µg/m^3 (note that this value was chosen because it is the designed value in the dual chamber for the formaldehyde test) followed intermittently by 30 days of low formaldehyde value (0 µg/m^3).

The RH of inflow for chamber was maintained at a constant value and three cases were considered: 50%, 65% and 80% RH. The initial T, RH and formaldehyde in the test chamber and CS board are 23 °C, 50% RH and 0 ($\mu g/m^3$), respectively. To study the impact of RH on formaldehyde buffering capacity of silicate calcium, it is necessary to know the indoor RH in the airtight chamber. Figure 11 shows the indoor RH variation (RH_a) as function of time in which the dotted lines represent the results obtained by **Without-BC** model. Note that, due to the moisture buffering capacity of the calcium silicate board, compared to **Without-BC** model, indoor RH obtained by **BC** model is much more dampened and the equilibrium sate is reached at 3th and 5th days for the RH_{in}=65 and RH_{in}=80% RH cases, respectively. In addition, Figure 11 showed that the moisture buffering capacity has no impact when the equilibrium state is reached and it will be discussed further in the second numerical study.

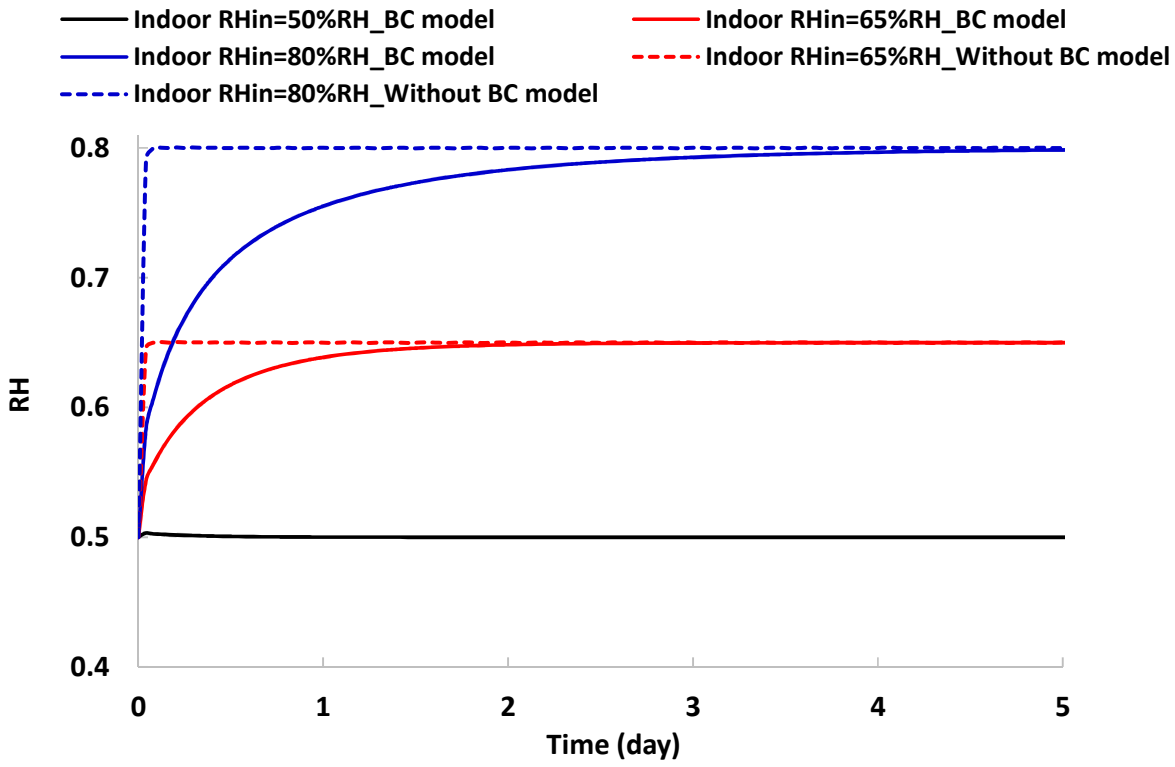


Figure 11: Indoor RH variation in the airtight chamber

The impact of RH on the partition coefficient $K_{m,FOR}$ at the air-material interface (x=L) is shown in Figure 12. It can be seen that in the studied range of RH (50-80% RH), $K_{m,FOR}$

increases significantly with increasing RH. Numerically, at steady state, the $K_{m,FOR}$ are 2586, 3100 and 3997 for the RH_{in}=50% RH, RH_{in}=65% RH and RH_{in}=80 % RH cases, respectively. Figure 13 represents the impact of RH on $D_{m,FOR}$ at the internal surface (x=L) layer of the CS board. It is noted that $D_{m,FOR}$ decreases with increasing RH as explained in Equation 25. At the equilibrium state, the $D_{m,FOR}$ are 1.26×10^{-9}, 1.1×10^{-9} and 7.68×10^{-10} (m²/s) for the RH_{in}=50%, 65% and 80% RH cases, respectively. Note that when RH_{in} increases from 50% to 80%, the diffusion coefficient ($D_{m,FOR}$) decreases 30% and the partition coefficient increases 22.4%.

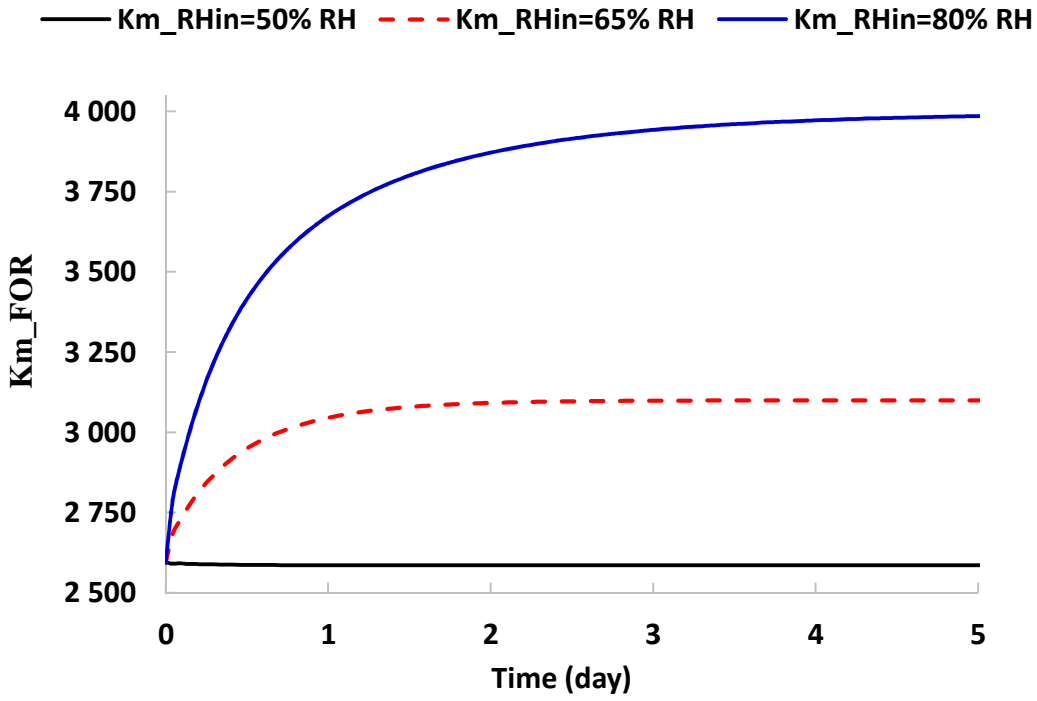


Figure 12: $K_{m,FOR}$ coefficient at the air-material interface (x=L) of the calcium silicate sample

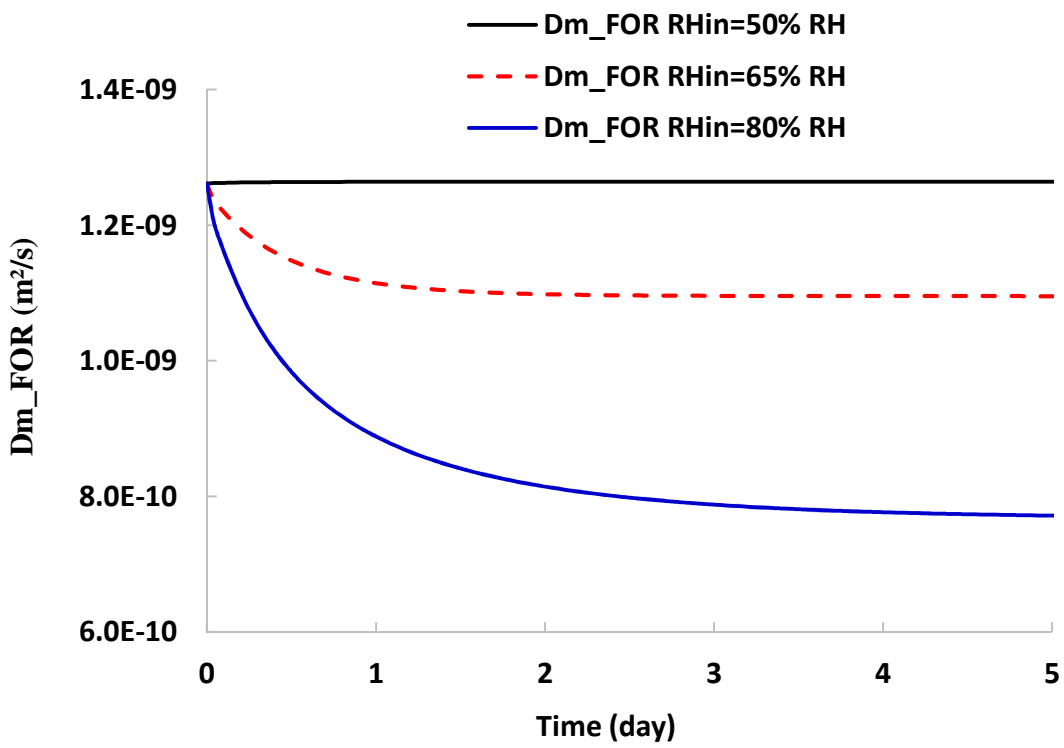


Figure 13: Variation of $D_{m,FOR}$ at the internal surface (x=L) layer of the CS board

According Yang (2001) and Zhang et al (2005), for a porous material, there are three mass transfer regions which may be associated with the VOC sorption: VOCs transferred from the air to the material-air interface, VOCs in both the air phase and material phase are exchanged which can be represented by $K_{m,VOC}$, and finally the VOCs diffuses into the material which is defined by $D_{m,VOC}$ coefficient. In this study, this is a complex nature of the sorption process because $K_{m,FOR}$ and $D_{m,FOR}$ vary inversely with increasing RH, more precisely, $K_{m,FOR}$ increases while $D_{m,FOR}$ decreases continuously in the material until reach the equilibrium state.

Figure 14 compares the indoor formaldehyde concentration in the airtight chamber obtained by **BC** model for different values of RHin (50%, 65% and 80% RH) and the one obtained by **Without-BC** model. When the formaldehyde sorption capacity of the CS board is neglected (**Without-BC** model) the indoor formaldehyde concentration increases suddenly to the equilibrium state compared to 20th day for **BC** model because the formaldehyde in the

chamber will be gradually adsorbed by CS board until an equilibrium is reached. Figure 14 shows clearly the impact of RH on indoor formaldehyde concentration; increasing RH results in decreased indoor formaldehyde concentration (because the partition coefficient $K_{m,FOR}$ increases) and increased formaldehyde buffering capacity of the CS board. Regarding desorption period from 30th day of simulation, the indoor formaldehyde concentration obtained by **BC** model decreases gradually to the equilibrium state (C=0) due to the release of formaldehyde in material which has been adsorbed in the CS board during the adsorption period. In this period of the desorption process, the formaldehyde concentration in the airtight chamber is higher with higher RH_{in} because of higher partition coefficient $K_{m,FOR}$ (which is constant in this studied period, see Figure 12). The simulation results confirm that the surface sorption represented by partition coefficient $K_{m,FOR}$ is the dominant mechanism affecting the formaldehyde concentration in the test chamber.

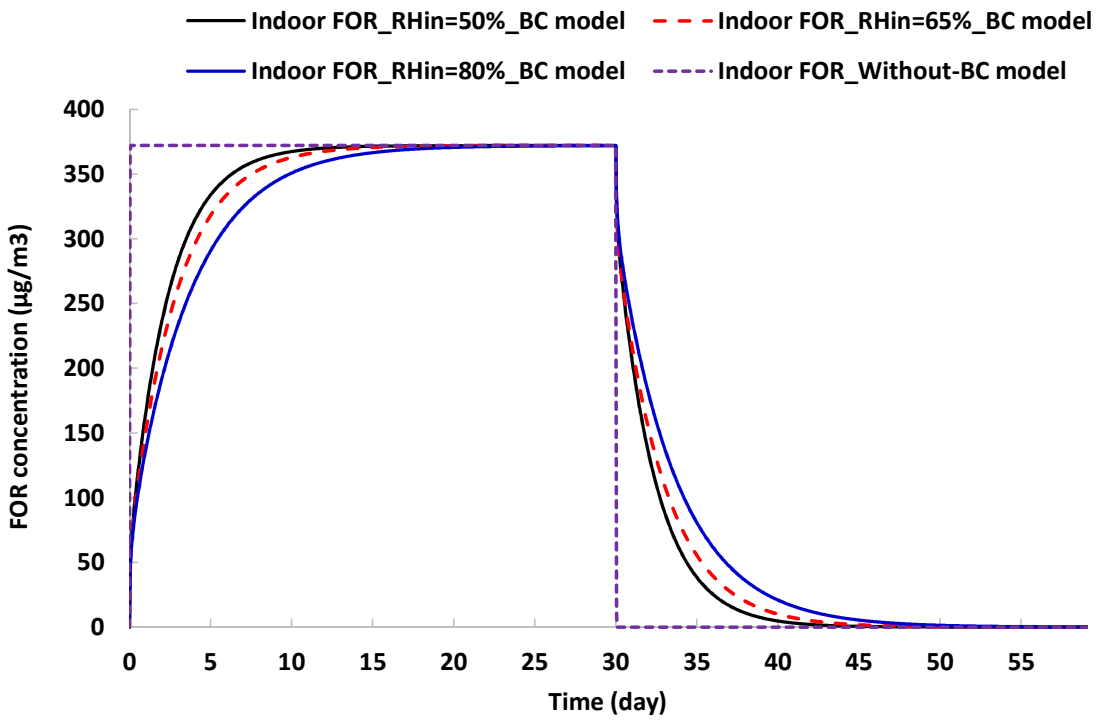


Figure 14: Effect of RH and formaldehyde (FOR) buffering capacity on indoor formaldehyde concentration

As shown in Figure 14, the impact of RH on formaldehyde buffering capacity of the CS board depends on the exposure time. From IAQ analysis and design point of view, it is very interesting to define a parameter called "*peak reduced factor-PRF_{VOC}*" which is calculated from the VOC concentration in the test chamber with and without VOC buffering capacity (C_0 corresponds to the case without VOC buffering capacity calculated using **Without-BC** model):

$$PRF_{VOC} = \frac{C_0 - C}{C_0} \tag{22}$$

The PRF_{VOC} value which varies between 0% (no buffering impact, e.g. $C=C_0$) and 100% allows to quantify the VOC buffering capacity of building materials. Figure 15 shows that PRF_{FOR} value (for formaldehyde) decreases as function of the exposure time. Numerically, after one hour of exposure, the PRF_{FOR} values is about 85% and will decrease to 15%, 21% and 28% for RH_{in} =50%, 65% and 80% RH, respectively, after 4 days of exposure. The results suggest that a combination between formaldehyde buffering capacity of building material and ventilation system is necessary to ensure a good IAQ in long-term pollutant source.

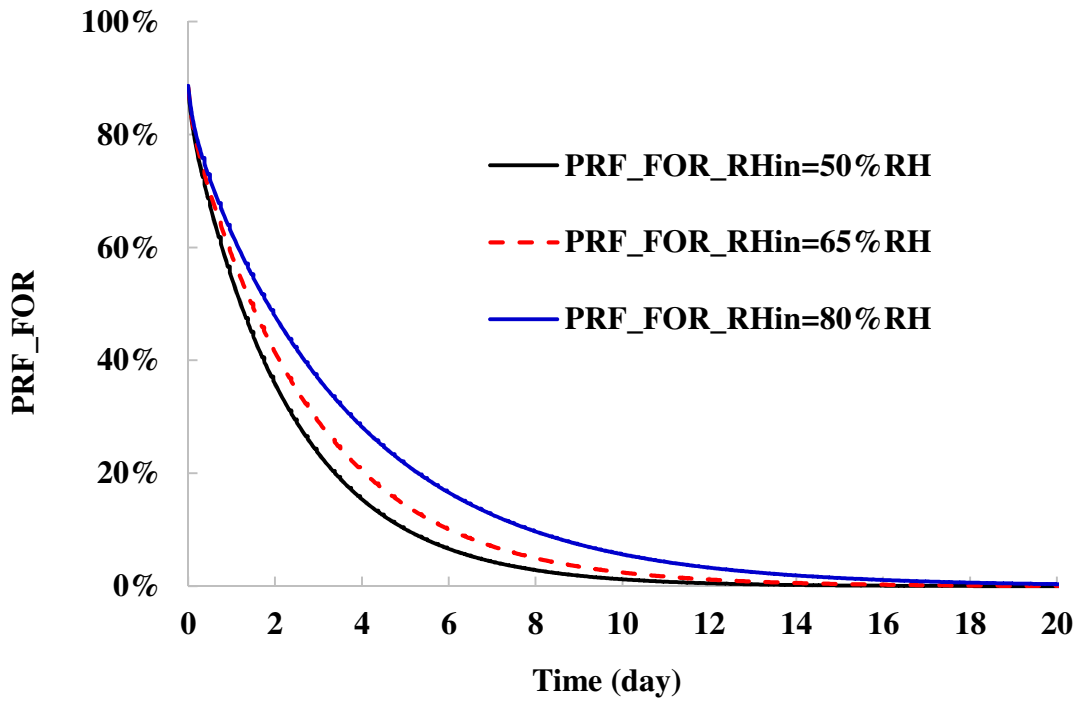


Figure 15: Effect of RH on PRF_{FOR} during adsorption period

In this section, formaldehyde buffering capacity of the CS board has been investigated to show that hygroscopic materials can moderate formaldehyde variation in the test chamber thanks to its formaldehyde sorption capacity. Therefore, regarding the health effect, formaldehyde buffering capacity can reduce the cause eye, nose and throat irritation related to short-term exposure to formaldehyde (Yang et al 2001, Lang et al, 2008). It is very important to underline that the formaldehyde buffering capacity of the CS board has no impact on the formaldehyde concentration in the airtight chamber at the equilibrium state as shown in Figure 15 (in which the PRF_{FOR} value is equal to 0). Considering now the variation of the formaldehyde concentration in the internal surface (x=L) layer of the CS board (Figure 16), when the one reaches the saturation value which depends also on the material thickness related to the formaldehyde diffusion, the CS board does not contribute to dampening the formaldehyde in the test chamber. Note that, the impact of RH on formaldehyde concentration in the CS board is significant and increased RH results in increased formaldehyde concentration in the CS board. At the equilibrium state, the formaldehyde concentrations in the surface (x=L) are 962, 1153 and 1486 mg/m^3 for RH_{in}=50, 65 and 80% cases, respectively. Therefore, the concept of formaldehyde buffering capacity which is important to consider in the building design, is only applied for the transient state and depends on the material thickness and humidity.

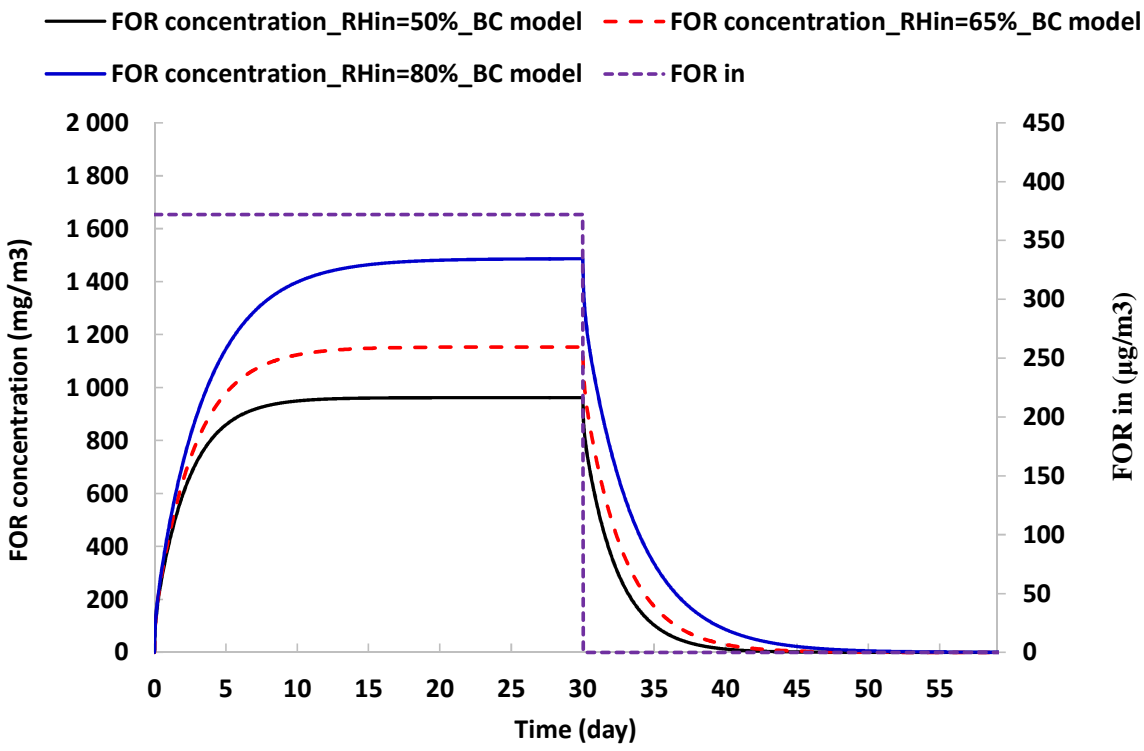


Figure 16: Formaldehyde (FOR) concentration in the surface (x=L) of the CS board

4.3 Study of the impact of RH variation on moisture and formaldehyde buffering capacity

In the section above, the formaldehyde buffering capacity has been studied with a constant inlet relative humidity (RH_{in}=constant). In reality, the humidity in buildings can have daily variation due to many factors such as ventilation, internal moisture sources, etc. Therefore, to study the impact of RH daily variation on formaldehyde and moisture buffering capacity of silicate calcium, we considered the boundary conditions as follows (see Figure 17):

- The formaldehyde injected into the airtight chamber was maintained at a constant value (FOR_{in}= 372 µg/m^3)
- The RH of inflow for chamber has a square-wave form: 12h of high RH value (RH_{in}= 80%) followed intermittently by 12 h of low RH value (50%). The time scheme has been chosen based on the recommendation of ISO 24353.

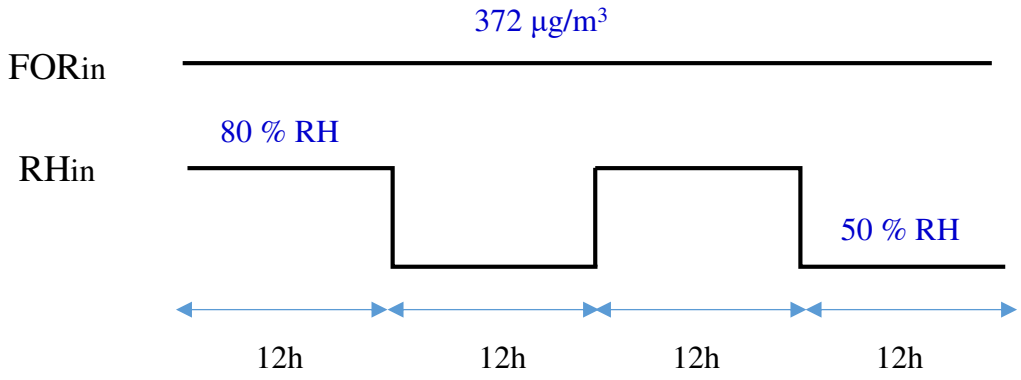


Figure 17: Studied configuration for RH_{in} and FOR_{in}

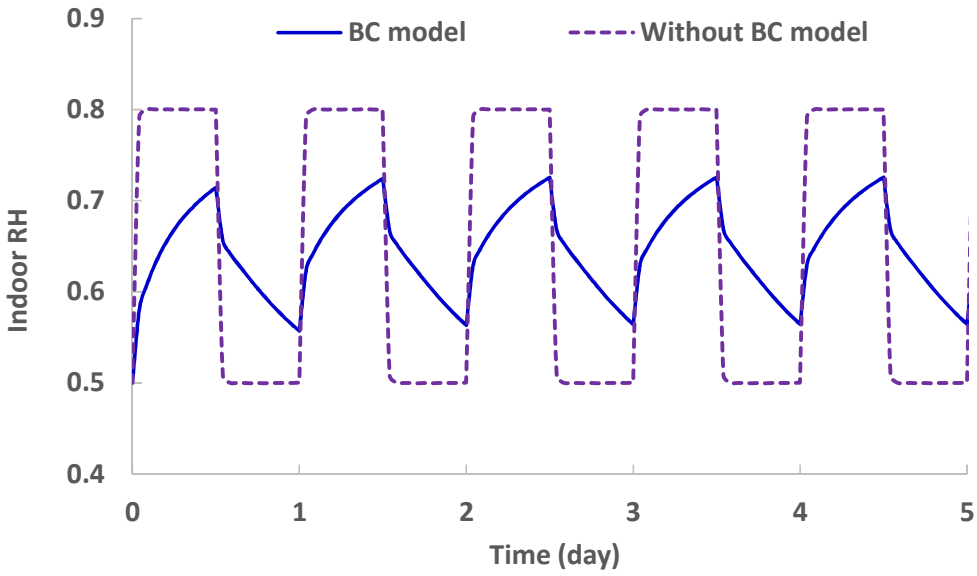


Figure 18: Impact of moisture buffering capacity of calcium silicate on indoor RH

Figure 18 represents the variation of the indoor relative humidity in the airtight chamber. The results showed that the hygroscopic material allows to dampen the indoor relative humidity variation thanks to its moisture sorption capacity. More clearly, the CS sample can adsorb/desorb water vapor from the indoor air when the indoor RH increases/decreases. Numerically, the maximum indoor RH values decrease from 80% to 72.6% RH (a difference of 7.4% RH and PRF_{RH}=9%) for **Without-BC** and **BC** models, respectively. For indoor humidity, we define a parameter called "*amplitude reduced factor-RF_{RH}*" which is calculated from the amplitude of indoor relative humidity variation with and without moisture buffering capacity (A_0 corresponds to the case without moisture capacity):

$$ARF_{RH} = \frac{A_0 - A}{A_0} \tag{23}$$

The ARF_{RH} value allows to quantify the hygric buffering capacity of building materials. In addition, an ARF_{RH} value of 53.7% is obtained showing that taking moisture buffering capacity of calcium silicate into account can reduce the indoor RH variation amplitude by 53.7%.

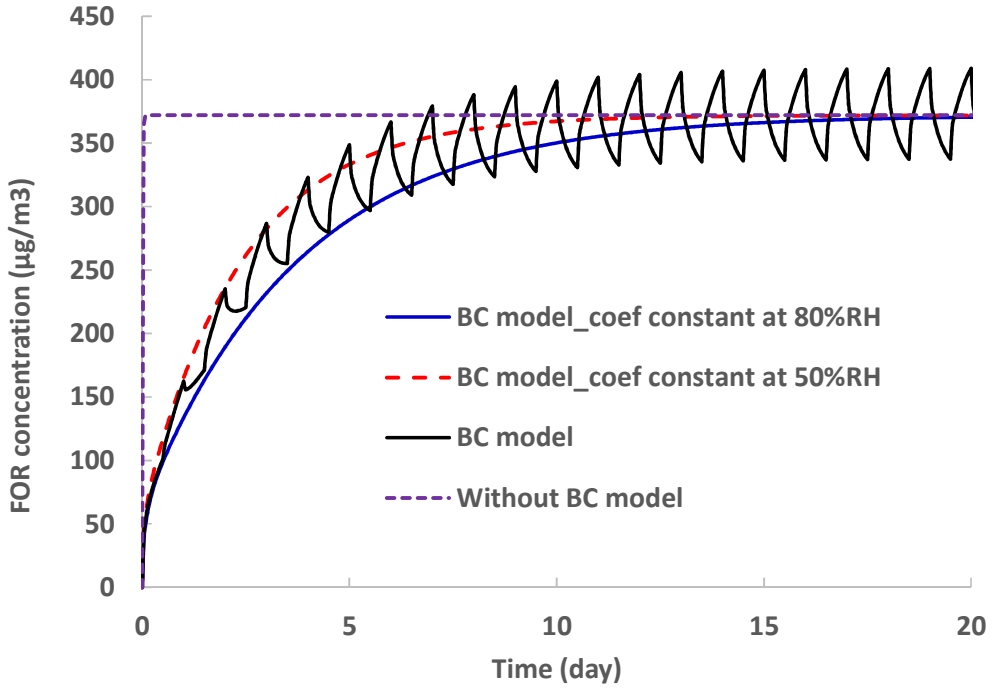


Figure 19: Impact of RH variation on indoor formaldehyde concentration

The impact of RH daily variation on formaldehyde concentration in the airtight chamber is shown in Figure 19. In addition to **BC** and **Without-BC** models, the simulations have been done with two additional cases: **BC** model with $K_{m,FOR}$ and $D_{m,FOR}$ are constant and determined at 50% and 80% RH. Note that for **Without-BC** model and **BC** model with constant coefficients, we have the same observation as the one in the previous section because the impact of RH variation is neglected. However, very interesting results have been obtained for **BC** model showing that the indoor formaldehyde changes and varies significantly with the variation of RH at the equilibrium state. The results obtained are very important to understand the indoor formaldehyde concentration in buildings subjected to real conditions of humidity and such experimental works deserve to be carried out. At the equilibrium state, the indoor formaldehyde concentration obtained by **BC** model varies between 338 and 409 µg/m^3 depending on RH variation compared to a constant value of 372 µg/m^3 obtained by other

models. The results should be explained by the impact of RH on the formaldehyde properties ($K_{m,FOR}$ and $D_{m,FOR}$) of calcium silicate which are shown in Figure 20 and Figure 21. Note that, due to the RH variation, the partition coefficient at the air-material interface ($K_{m,FOR}$) varies between 2770 and 3460 (a difference of 20%) which results in a decreased/increased formaldehyde concentration in the test chamber (see Figure 20).

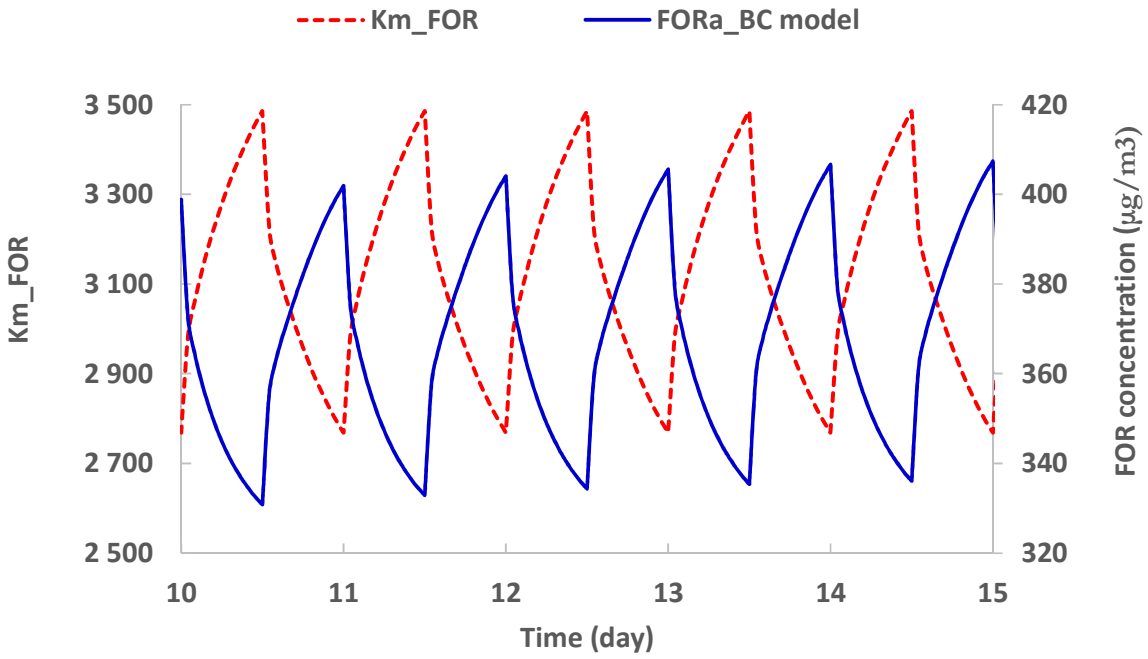


Figure 20: Impact of $K_{m,FOR}$ on indoor FOR variation in the airtight chamber

Note that, due to the initial formaldehyde concentrations in CS board which are set to 0 ($\mu g/m^3$) and formaldehyde diffusion process inside the material, the indoor formaldehyde concentration needs much more time to reach the equilibrium state compared to indoor RH in which the initial RH in material were set to 50% RH. Regarding the diffusion coefficient D_m of water vapor and formaldehyde, the ones at the surface (x=L) of the CS board are shown in Figure 21. As can be seen in this figure, the diffusion coefficients D_m of both water vapor and formaldehyde vary inversely with the variation of RH: D_m decreases/increases when RH increases/decreases. It can be explained by equation (8) for water vapor diffusion coefficient (increasing slope of sorption curve with increased relative humidity) and equation (25) for formaldehyde diffusion coefficient. Although the variation tendency of D_m for formaldehyde and water vapor was similar, the variation magnitude is different: at the equilibrium state, the coefficient $D_{m,wv}$ varies between 3.65×10^{-9} and 7.63×10^{-9} (m^2/s) compared to 9.58×10^{-10} and 1.20×10^{-9} (m^2/s) for $D_{m,FOR}$ coefficient when the RH (at the surface x=L) varies between 56.3 % and 71.4 % RH.

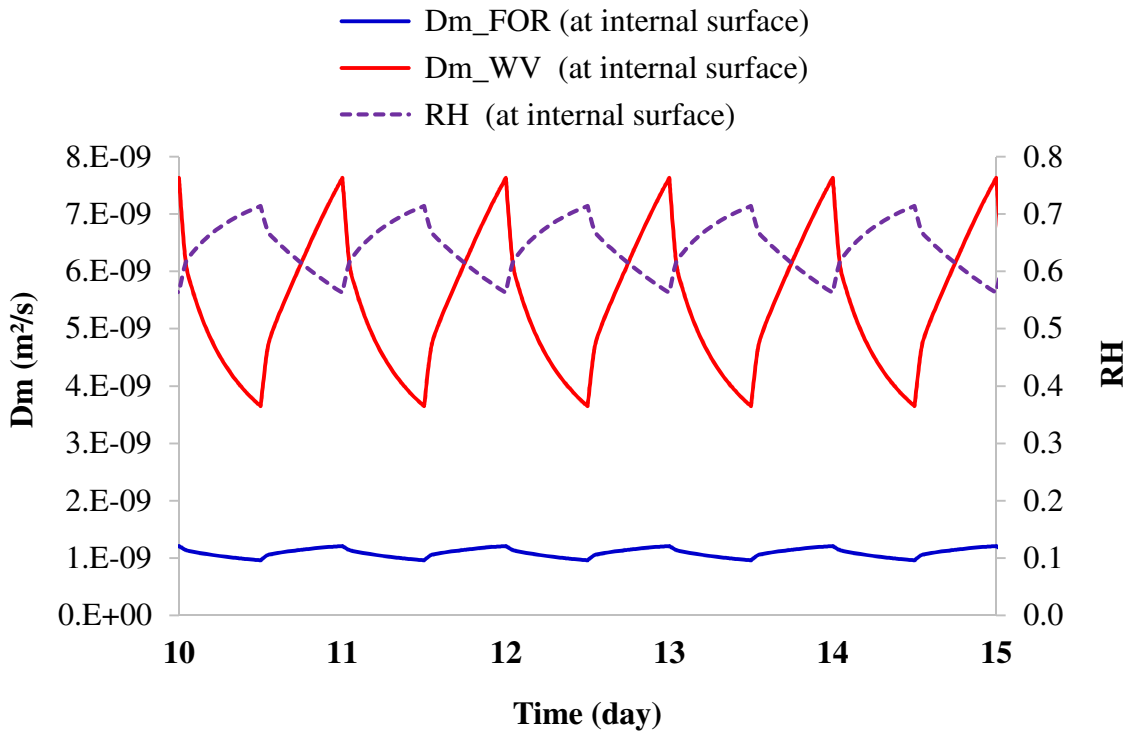


Figure 21: Impact of RH variation on $D_{m,FOR}$ and $D_{m,wv}$ (for formaldehyde-FOR) in the surface (x=L) of the CS board

The results obtained in this second numerical study confirmed that the formaldehyde concentration in real indoor air is influenced by humidity variation and cannot be simply calculated from the emission rate of building materials and air exchange rate as usually done in the literature. It suggested that the numerical tools for predicting IAQ could underestimate or overestimate the formaldehyde concentration in buildings if the impact of RH variation on formaldehyde diffusion and storage properties of building materials is neglected.

5. Conclusion

The objective of the paper is to fulfil the gap in the literature concerning the transient effect of humidity on formaldehyde sorption of building materials, the influence of the formaldehyde and moisture buffering capacity of hygroscopic porous material on indoor RH and formaldehyde concentration. In addition, new empirical formulas ($K_{m,FOR}$ and $D_{m,FOR}$ coefficients as functions of humidity) have been proposed based on experimental data found in the literature. A coupled moisture and VOC transport simulation model is presented and explained. The similarity between the moisture and pollutant transport and storage

coefficients of porous building materials has been clearly established. The physical model has been implemented in the environment SPARK (Simulation Problem Analysis and Research Kernel) which is suited to complex problems. The present model, after having been validated with the experimental data for the calcium silicate sample, is applied to study its moisture and formaldehyde buffering capacity. The results obtained in this paper are very interesting and many conclusions can be drawn as following:

- Formaldehyde and moisture buffering potential of hygroscopic material can moderate significantly indoor formaldehyde and RH variation, and reduce heath effect of pollution.
- Increasing/decreasing indoor RH results in decreased/increased indoor formaldehyde concentration and increased/decreased formaldehyde buffering capacity of the CS board (in our studied range of RH). The knowledge of moisture content in material is crucial to understand the formaldehyde behavior of porous building material.
- Concept of pollutant (formaldehyde)/moisture buffering capacity is only applied for the transient state. It suggested that a combination between formaldehyde/moisture buffering capacity of building material and ventilation system is necessary to ensure a good IAQ and thermal comfort in long-term pollutant and water vapor sources.
- Formaldehyde-surface sorption is the dominant mechanism affecting the formaldehyde buffering capacity.
- Formaldehyde concentration in real indoor air is influenced by humidity variation and cannot be simply calculated from the emission rate of materials and ventilation rate as usually done in the literature. The developed model allows to understand the impact of short and long-term variations of humidity on the prediction of the transient behavior of formaldehyde when the hygroscopic material is subjected to dynamic conditions of humidity and formaldehyde.

Finally, our paper showed that the reduction of indoor formaldehyde concentrations and indoor RH using sorption processes is a very interesting solution to improve IAQ and thermal comfort. Therefore, the model presented is very useful to optimize the use and design of clean and hygroscopic materials in buildings.

Acknowledgements

This study was carried out under the program Fulbright /Hauts-de-France which is supported by the Franco-American Fulbright Commission and the Hauts-de-France region, France.

Thanks to this financial support, it enabled Dr. Anh Dung TRAN LE to work at the BEESL of Syracuse University, USA for a period of six months. The authors wish to thank them.

The authors wish to thank the editor and reviewers for their constructive and useful comments on the previous version of this article.

NOMENCLATURE

A	Exposed area of the material	m^2
C	Concentration	kg/m^3
$C_{a,o}$	Outdoor ventilation air	kg/m^3
$C_{a,wv,e}$	Water vapor concentrations in the outside	kg/m^3
$C_{a,wv,i}$	Water vapor concentrations in the room air	kg/m^3
$D_{m,VOC}$	Diffusion coefficient of the VOC in the material	$m^2.s^{-1}$
$D_{m,wv}$	Mass transport coefficient associated to a moisture content gradient	$m^2.s^{-1}$
D_{VOC}^{air}	VOC diffusion coefficient in the free air	m^2/s
D_{wv}^{air}	Water vapor diffusion coefficient in the free air	m^2/s
ERFc	Cumulative Exposure Reduction Factor	%.h
$h_{m,VOC,e}$	Convective VOC transfer coefficient for the external surface	m/s
$h_{m,VOC,i}$	Convective VOC transfer coefficient for the internal surface	m/s
$h_{m,wv,e}$	Convective water vapor transfer coefficient for the external surface	m/s
$h_{m,wv,i}$	Convective water vapor transfer coefficient for the internal surface	m/s
$K_{m,VOC}$	Partition coefficient for VOC	-
$K_{m,wv}$	Partition coefficient for water vapor	-
L	Characteristic length (m)	m
PRF	Peak reduced factor	%
$P_{wv,sat}$	Saturation pressure of water vapor	Pa
Q	Flow rate	m^3/s
Re	Reynolds number	-
RH	Relative humidity	-
R_v	Gas constant for water vapor	J/(kg·K)
Sc	Schmidt number	-
Sh	Sherwood number	-

T	Temperature	K
t	Time	s
V	Volume space	m^3
w	Moisture content	$kg.kg^{-1}$
w_m	Monolayer moisture content	$kg.kg^{-1}$
x	Abscissa	m
υ	Kinematic viscosity of air	m^2/s
δ_{wv}	Water vapor permeability	kg.m-1.s-1 Pa-1
μ_{VOC}	VOC diffusion resistance factor	-
μ_{wv}	Water vapor diffusion resistance factor	-
θ	Moisture volumetric content	m^3/m^3

Subscripts

e= external

m= material

i= internal

wv= water vapor

VOC= Volatile Organic Compounds

FOR=formaldehyde

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