

INDOOR AIR QUALITY FOR HEALTH AND COMFORT: MODELING ASPECTS OF INDOOR POLLUTANT CONCENTRATION PREDICTION

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***Abstract.** This papers deals with the influence of reactive processes, homogeneous reactions in the gas phase of indoor spaces or heterogeneous effects at the interface between the gas volume and solid surfaces or walls, in the prediction of indoor exposure to pollutants. The various physical or chemical phenomena to be taken into account in modeling these reactive effects are described in detail, and are introduced in an Indoor Air Quality model. The results of elementary case studies demonstrate clearly how significant are these processes in predicting the indoor air concentrations in pollutants. In a first test we consider the influence of gas phase chemistry on the prediction of indoor concentration in atmospheric pollutants and we show discrepancies of about 50% on the prediction of ozone concentration with or without taking into account the reactive processes. In a second part we discuss the influence of the diffusion and sorption effects of VOCs (Volatile Organic Compounds) at the interface solid/gas and we show clearly their influence either in the source effect (emission of a pollutant by desorption) or in the sink effect (adsorption and release of a pollutant by a wall).*

Keywords: *Indoor Air Quality, Modeling, Pollutant emission, Exposure prediction.*

1. INTRODUCTION: the Right to Healthy Indoor Air

Indoor air quality (IAQ) is an important determinant of population health and wellbeing. People in modern societies spent most of their time (80 to 90% in western countries) in indoor spaces such as at home, work, school and in vehicles. Exposure to the hazardous airborne agents present in many indoor environment causes adverse effects such

as respiratory disease, allergy and irritation of the respiratory tract. Improperly ventilated combustion appliances pose still a real risk of acute poisoning by carbon dioxide, indoor exposure to radon or tobacco smoke increase significantly the risk of lung cancer. Many chemicals encountered indoors cause adverse sensory effects, giving rise to a sense of discomfort and other symptoms. Even the productivity of workers has been shown to be strongly dependant of the IAQ. A recent study in USA (Fisk, 2000) evaluates the economic benefits of productivity in increasing IAQ to 40 to 200 billion US dollars.

Despite of these strong effects on health, wellbeing and productivity, until now most of national regulations on air quality have been focusing on outdoor air quality and a too small attention has been paid on IAQ. This is the reason why a WHO (World Health Organization) working group has formulated on may 2000 a set of statements on "The right to healthy indoor air" (WHO, 2000) under the principles of the human right to health.

In this context, a strong effort has been asked to building physicists in order to predict human exposure to indoor pollutants (Allard et Al. 1999). Various research programs have been launched at national or international level to improve the physical knowledge of the various phenomena involved in the emission of indoor pollutants. During the last 20 years, many single-zone or multi-zone simulation tools have been developed in order to estimate human exposure to indoor pollutants (Austin et Al. 1992 and Weir et Al. 1990). These tools can be categorized into two broad groups: the codes that are using probabilistic models (Ott et Al. 1988, Haghghat et Al., 1988) and the codes that are using deterministic models such as CONTAM (Axley, 1988), BILGA (Creuzevault et Al., 1990), COMIS (AIVC, 1990), CBSAIR (Haghghat et Al., 1991) or MACROMODEL (Traynor et Al., 1989). While probabilistic models aim to best fit statistical data with random or typical environmental and population groups characteristics input by the user, deterministic models first seek to predict the indoor air pollutant concentrations from mass balances of each building room. Most of the existing models have included internal sources and mass transport in their code. In these models however, the gas-phase chemistry process as well as mass transfer with solid elements are not fully implemented. The shortcoming of the approach is that the first-order assumption is not always valid and if it is validated the user has to guess the value.

In the present paper we propose to deal with the basic elementary phenomena of homogeneous reaction within the gas phase and heterogeneous reactions at the interface between gas and solid walls in order to show their specific contribution in the prediction process of indoor pollutant concentrations.

2 - PREDICTION OF HOMOGENEOUS REACTIONS

2.1 - Modeling Principles: Fundamentals of atmospheric chemistry

In the field of outdoor air pollution analysis, the detailed mechanisms of gas-phase atmospheric chemistry are divided into two broad categories which are photochemical and thermal reactions.

Photochemical reactions depend on the absorption of a photon by an atom, molecule, free radical or ion which leads to its activation. Of all the photolytically activated reactions possible, photolytic dissociation is the most common; it consists of the fragmentation of the original species into its components. If A and A^* respectively represent the reactive element in its electronic ground and excited states, the whole process can thus be summarized by the following unimolecular reaction (Seinfeld, 1986):



where B and C are dissociation products, n_B and n_C the number of units of each product formed, and $h\nu$ represents the energy added by the photon.

For low concentrations of A , the resulting rates of change of molecular concentration of both reagent and products are given by the following equations:

$$\frac{d[A]}{dt} = -k_I[A] = -\frac{1}{n_B} \frac{d[B]}{dt} = -\frac{1}{n_C} \frac{d[C]}{dt} = \dots \quad (2)$$

where the first order rate constant k_I (s) depends on the integral of the solar energy density $I(\lambda)$ (photons.m⁻³.s⁻¹) which corresponds to the radiant intensity per unit wavelength, the absorption cross section $\sigma_A(\lambda, T)$ of molecule A at wavelength λ (m) and temperature T (K), and the probability $\Phi_A(\lambda, T)$ that molecule A decomposes on absorbing radiation of wavelength λ at temperature T :

$$k_1 = \int_{\lambda_1}^{\lambda_2} \sigma_A(\lambda, T) \Phi_A(\lambda, T) I(\lambda) d\lambda \quad (3)$$

Seinfeld (1986) indicated that in the lower atmosphere, the wavelength range of interest is approximately [280-730 nm]. As done by Finlayson-Pitts and Pitts (1986), Seinfeld also proposed some values or temperature-dependent relationships for the absorption cross sections and probabilities of dissociation of the more important reactive species.

Bimolecular and trimolecular thermal reactions result from the collision of two or three atoms, molecules or ions which, under favorable thermodynamic conditions, combine to form new chemical species. They can be represented by the following transformations:



where k_2 (m³.molecule⁻¹.s⁻¹) is the rate constant for bimolecular reaction, k_3 (m⁶.molecule⁻².s⁻¹) is the rate constant for trimolecular reaction and n_C , n_D , n_E , n_F , etc ... are the stoichiometric coefficients.

In both cases, the molecular rates of production and removal are functions of the rate constant, the stoichiometric coefficients and the reagent concentrations in the reaction:

$$\frac{1}{n_C} \frac{d[C]}{dt} = \frac{1}{n_D} \frac{d[D]}{dt} = -k_2[C][D] = \frac{-1}{n_E} \frac{d[E]}{dt} = \frac{-1}{n_F} \frac{d[F]}{dt} = \dots \quad (6)$$

$$\frac{1}{n_E} \frac{d[E]}{dt} = \frac{1}{n_F} \frac{d[F]}{dt} = \frac{1}{n_G} \frac{d[G]}{dt} = -k_3[E][F][G] = \frac{-1}{n_H} \frac{d[H]}{dt} = \frac{-1}{n_I} \frac{d[I]}{dt} = \dots \quad (7)$$

Seinfeld (1986) and Atkinson et al (1992) suggested rate constant values were a function of temperature for the reactions involved in the most important atmospheric pollutant transformations.

2.2 - Integration in an IAQ simulation tool

In most IAQ simulation tools, concentrations as well as airflow rates are expressed in mass units rather than in molecular units. If the rate expressions presented above in molecular kinetic format are converted into mass generation or removal rate expressions, the contributions of ventilation (or infiltration), internal sources (or sinks) and homogeneous chemistry to the concentration change of each element p of a well mixed single zone, can be represented by the following mass conservation equation (Blondeau, 1996) :

$$\rho V \frac{dC_p}{dt} = Q_m (C_{o,p} - C_p) + S_p + \sum_{i=1}^{n_p} \left(K_{ip} \prod_{j=1}^{n_j} C_j \right) \quad (8)$$

with: ρ : Density of air ($\text{kg}_{\text{air}} \cdot \text{m}^{-3}$),
 V : Room volume (m^3),
 C_p : Mass concentration of element p ($\text{kg}_p \cdot \text{kg}_{\text{air}}^{-1}$),
 C_j : Mass concentration of element j which is a reagent in reaction i ($\text{kg}_p \cdot \text{kg}_{\text{air}}^{-1}$),
 t : Time (s),
 Q_m : Ventilation or infiltration airflow rate ($\text{kg}_{\text{air}} \cdot \text{s}^{-1}$),
 $C_{o,p}$: Outdoor concentration of p ($\text{kg}_p \cdot \text{kg}_{\text{air}}^{-1}$),
 S_p : Internal emission or removal rate of p ($\text{kg}_p \cdot \text{s}^{-1}$),
 K_{ip} : Composite term containing the rate constant of reaction i ,
 n_p : Number of reactions involving p ,
 n_j : Number of reagents involved in reaction i .

Each term of the sum in Eq. 8 represents the dynamic change in concentration resulting from one chemical reaction involving species p . The kinetic rate constant, the molecular quantities for unit conversion and the sense of the contribution (+ if production and - if removal) are included into the composite term K_{ip} whose units depend on the type of reaction i .

All the concentrations (contaminants and non-contaminants) are calculated by solving the set of differential equations within which bimolecular and trimolecular kinetics introduce non-linearities.

2.3 - Case study

This model was used (Blondeau et Al., 1998) to investigate the need of taking into account the gas-phase reactions in the frame of an health-related IAQ analysis. In these case studies, only 5 inorganic contaminants that are frequently used as atmospheric pollution indices are considered: nitric oxide (NO), nitrogen dioxide (NO₂), ozone (O₃), sulfur dioxide (SO₂) and carbon monoxide (CO).

The corresponding indoor chemical mechanism was defined from Atkinson et al (1992) atmospheric kinetic mechanism by: a) assuming zero concentrations of organic

species, b) selecting the reactions involving at least one of the five contaminants, c) eliminating all thermal reactions that cannot take place under temperature and pressure conditions of the lower atmosphere, and d) eliminating all photochemical reactions that present zero absorption cross sections or quantum yields in the wavelength band [280-350 nm] (the solar radiation cutting threshold of the window is assumed to be 350 nm). It is made up of 22 reactions and involves 5 additional species which interact with the 5 selected contaminants. They are the oxygen atom (O), oxygen molecule (O₂), nitrate radical (NO₃), dinitrogen pentoxide (N₂O₅) and sulfur trioxide (SO₃). As the O₂ concentration in air can be considered to be constant, accurate and dynamic prediction of NO, NO₂, O₃, SO₂ and CO concentrations thus requires that 9 different species be handled.

The simulations were carried out by considering a 24-day simulation period (July 15 - August 8 1994) and using the real climatic and contaminant (average hourly outdoor concentrations) data of La Rochelle in France. The outdoor concentrations of the non-pollutant species were assessed from the pseudo steady-state approximation. Indeed, O, NO₃, N₂O₅ and SO₃ are so highly reactive chemical elements that it may be assumed their rate of formation continuously equals to their rate of disappearance. A mass balance between their formation and removal reactions thus gives a good approximation of their equilibrium concentration as a function of the known chemical species concentrations. In these studies, the outdoor chemical mechanism was defined from the indoor one by taking into account the additional photochemical dissociation reaction of nitrogen pentoxide which cannot occur inside the building because of the solar radiation cutting threshold of the window.

The reference building is a single-zone office building (floor area 20 m² and volume 50 m³) with a 2.5 m² window facing south. The inside surfaces short-wave reflection coefficient of the walls is 0.5 and the HVAC plant allows the airflow rate to vary from 0.5 ach to 3 ach. Two extreme cases are considered for the occupancy scheme and the resulting indoor emission rates: in the first case, no indoor pollution sources exists while in the second scenario, the room is occupied by 8 smokers between 8-12 am and between 2-6 pm. The corresponding tobacco smoke, CO and NO₂, emission rates are given by Cohas (1994), see Table 1. In both cases, the indoor temperature is assumed to be 25°C and to remain constant during the occupancy.

Table 1: Emission rates of CO and NO₂ from a cigarette

Contaminant	Emission rate / cigarette (g.s ⁻¹)
CO	1.1 10 ⁻⁴
NO ₂	1.8 10 ⁻⁶

2.4 - Influence of chemical kinetics on the predicted concentrations

The need to take into account gas-phase chemical reactions in the IAQ simulation can be demonstrated by comparing the predicted indoor contaminant concentrations with and without reactive model, that is to say by considering or not considering the last term of the conservation equations, Eq. 8. As health-related air quality standards are typically based on risk assessment and are most of the time specified in terms of concentration Threshold Limit Values (TLV) for short-term and long-term exposures, the calculated differences between the two predicted concentrations (with reactive term and without reactive term)

were integrated into the definition of the following discrepancy index in order to show how neglecting gas-phase chemistry processes may affect IAQ analysis:

$$E_{\Delta t}^p = 100 \frac{C_{\Delta t,1}^p - C_{\Delta t,0}^p}{TLV_{\Delta t}^p} \quad (9)$$

- with: $E_{\Delta t}^p$: IAQ discrepancy index for pollutant p and exposure time Δt (%)
 $C_{\Delta t,1}^p$: Average concentration of pollutant p with reactive model for exposure time Δt ($\mu\text{g}\cdot\text{m}^{-3}$)
 $C_{\Delta t,0}^p$: Average concentration of pollutant p without reactive model for exposure time Δt ($\mu\text{g}\cdot\text{m}^{-3}$)
 $TLV_{\Delta t}^p$: Threshold Limit concentration Value for a Δt exposure time to pollutant p ($\mu\text{g}\cdot\text{m}^{-3}$)

The indices were calculated for 1-hour and 8-hour exposure times using the TLVs value recommended by the World Health Organization (WHO, 1988). As a result, Figure 1 shows the hourly values of the discrepancy index (averaged over the 24 day period) as a function of time for 4 scenarios: 0.5 air change with and without emission source, and 3 air change with and without emission source. It shows that ignoring the reactive term in the conservation equations of nitrogen oxides can either contribute to an overestimation ($E < 0$) or an underestimation ($E > 0$) of their hourly concentrations depending on the available amounts of chemically reactive species in the room. Nevertheless, one must remember that the case considering a 0.5 ach airflow rate when smoking is allowed does not represent a realistic situation (0.5 ach is low for 8 smokers) and indoor NO_2 concentrations reach peaks corresponding to three times the TLV. In more realistic situations, NO and NO_2 concentrations are systematically overestimated or underestimated by about 10% of their TLV. In contrast to nitrogen oxides, the most important reactions involving ozone lead to disappearance. Neglecting these reactions thus always leads to an overestimate of O_3 concentrations: the hourly E^{O_3} values vary from -20% to -45% depending on the airflow rate and the type of emission.

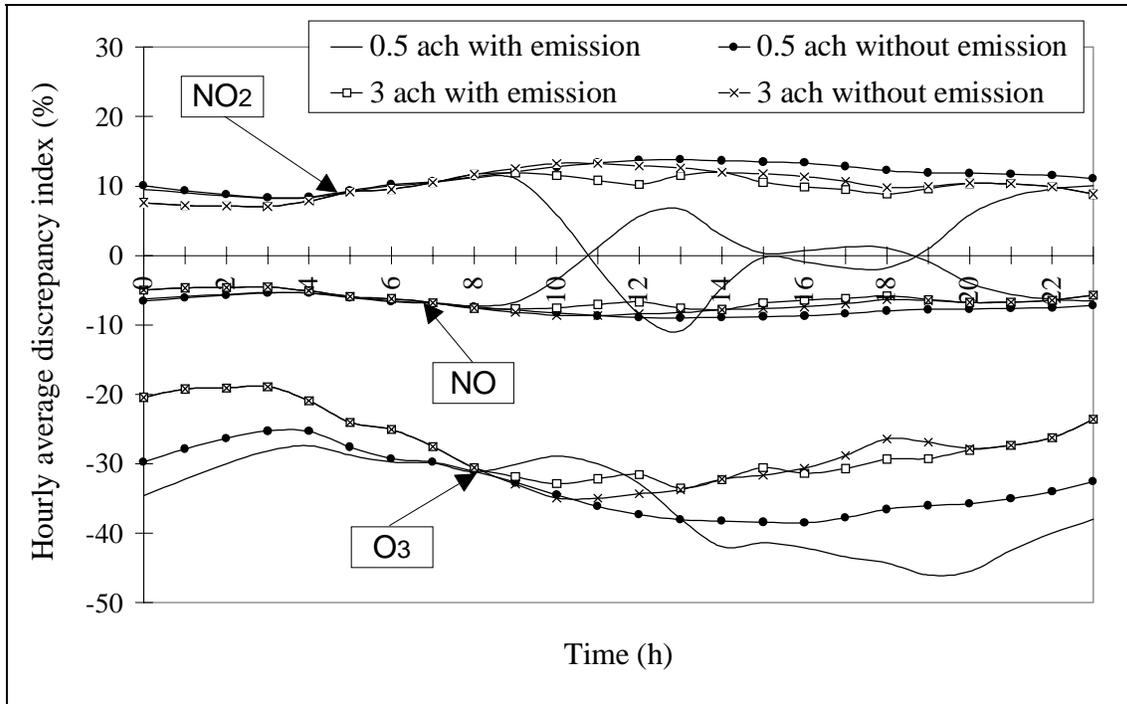


Figure 1: Average hourly discrepancy index as a function of time

3 - PREDICTION OF HETEROGENEOUS REACTIONS: Interactions with solid materials

3.1 – Physical analysis

Considering the exchange of a gaseous element between a porous solid and ambient air, four different phenomena can take place:

- Diffusion within the boundary layer developing along the solid material,
- Diffusion within the porous material,
- Sorption/desorption phenomena at the surface of the solid or within the pores if porous,
- Chemical reactions at the surface if the gaseous element can combined or dissociate at the contact of the solid phase.

We describe briefly these four physical phenomena in the following paragraph.

Diffusion within the boundary layer

The diffusion through the boundary layer developing along the solid material or external diffusion is directly related to the flow characteristics along the solid surface. The mass flow rate through out the surface is given by a newtonian law issued from the boundary layer theory (Bejan, 1995).

$$J = \rho_{air} A_s h_m (C_{int} - C^*) \quad (10)$$

where :

- ρ_{air} : density of air ($\text{kg}\cdot\text{m}^{-3}$)
- J : mass flow rate through out the boundary layer ($\text{kg}_{\text{gaz}}\cdot\text{s}^{-1}$)
- A_s : area of exchange (m^2)

- h_m : mass transfer coefficient ($\text{m}\cdot\text{s}^{-1}$)
 C_{int} : pollutant concentration in the gas out of the boundary layer ($\text{kg}_{\text{gaz}}/\text{kg}_{\text{air}}$)
 C^* : pollutant concentration at the solid surface ($\text{kg}_{\text{gaz}}/\text{kg}_{\text{air}}$)

The mass transfer coefficient h_m can be determined experimentally or given by correlations of natural or forced convection.

Diffusion within the porous material

Diffusion transport of gases in building materials involves a complex variety of processes including in particular molecular diffusion and Knudsen diffusion. Molecular diffusion occurs in the largest pores where collisions between gaseous molecules are predominant, while Knudsen diffusion occurs in the smallest pores where the mean free path of the diffusing species (i.e. the average distance travelled between molecular collisions) is limited by the pore dimensions. They may be modelled analogously to Fick's law through the introduction of an effective diffusion coefficient D_e (m^2/s) that may be calculated from the relation:

$$D_e = D^o \frac{\varepsilon}{\tau} \quad (11)$$

where :

D^o is the mean diffusion coefficient in the pores of the material (m^2/s);

ε (m^3/m^3) is the connected-pore volume fraction; it accounts for the real cross-sectional area offered to diffusion in the porous media since, in analysing macro pore diffusion, the transport is assumed to occur only through the pores and the flux through the solid phase can be neglected;

τ is the tortuosity factor of the porous network; it accounts for the geometric constraints which give a longer diffusion path and a reduced concentration gradient in the direction of flow.

Carniglia (1986) showed that the parameters of equation (11) can be measured (ε) or computed (D^o and τ) from porosimetry tests. Tiffonnet (2000b) has used this method and demonstrated a good agreement with classical diffusion tests.

Sorption/desorption phenomena at the surface of the solid or within the pores if porous

Adsorption may be defined as the separation of a substance from one phase (air in our case) and the accumulation of that substance on the surface of another phase (building materials here), the reverse phenomenon being defined as desorption.

The nature of sorption depends on the nature of the adsorbent, the nature of the adsorbate, and the interaction that may occur between them. Using the qualitative analysis, the sorption phenomena may be classified using three criteria. The first is based on the forces that occur between the sorbed species and the solid surface and leads to distinguish the physical adsorption of the chemical one. The second criterion is based on the conservation or non-conservation of the sorbed species identity, which correspond respectively to molecular and dissociative adsorption. Finally, the last criterion is based on the sorbed species distribution. The increase of the species concentration leads to three-step

emergence in sorption process: monolayer adsorption, multilayer adsorption, and condensation.

From a quantitative point of view, sorption kinetics are much faster than diffusion kinetics in the field of indoor air quality analysis. Then the sorption phenomenon may be considered like instantaneous and is governed by an equilibrium constraint. As a result, the sorbed-phase concentration (C_s) depends on its near surface air-phase concentration (C). The dependence law of C_s on C at a set temperature is called adsorption isotherm. It is often symbolised by the functional notation:

$$C_s = f(C) \quad (12)$$

Where f is a function depending on the temperature which unique for each couple species/solid.

Many adsorption isotherm models exist among which empirical models and physical models. Each one is based on a set of assumptions including the solid surface state and the interactions between sorbed species. Tiffonnet (Tiffonnet 2000c) has given various sorption isotherm types for various covering or building materials.

3.2 – Influence of gas/solid exchange in the prediction of indoor pollutant concentration.

The various phenomena described previously have been introduced in an IAQ model, and the physical parameters necessary have been measured by Tiffonnet (Tiffonnet, 2000a). They have been used to study the two main effects usually described as source and sink effects.

The source effect:

The source effect or “pollutant emission” by building materials or covering occurs when the pollutant concentration within the material is higher than it is in the indoor air volume. This case takes place mainly with new materials and has been strongly investigated (Saarela 2000). Even standards have been proposed to determine the emission rate of pollutant (mainly VOCs) of building materials.

The figure below (Tiffonnet, 2000a) illustrates the emission of acetone by two chipboard panels in direct contact with the indoor air (case 1) or covered by a paint (case 2). These 20 m² each walls are located in a 162 m³ room with a ventilation rate of 1 air change per hour. We can notice here that the presence of paint with a low porosity (low diffusion material) in case 2 leads to a strong modification of the concentration profile in the room.

The maximum concentration level is strongly decreased but after 72 hours, the concentration in case 2 becomes higher. In this scenario, the leading phenomena are the diffusion process through the covering material.

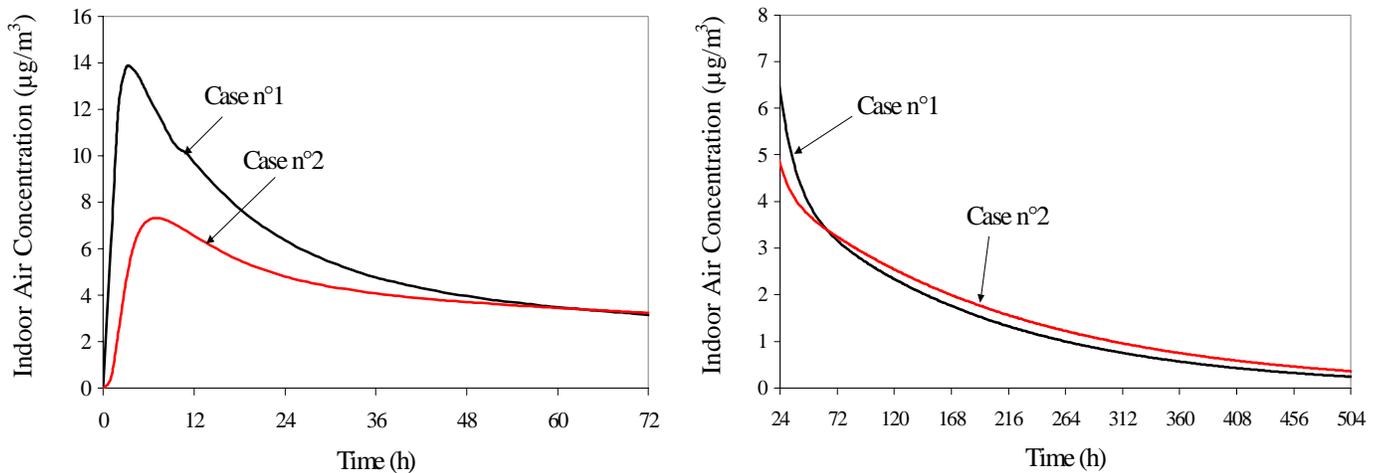


Figure 2 : Evolution of indoor acetone concentrations ($T^\circ = 25^\circ\text{C}$).

The sink effect:

In this second example, the acetone is emitted in the room by one chipboard wall (case 1 of Fig. 2), and we looked at the transfer to an other chipboard panel free of acetone and directly exposed to the indoor air. Fig. 3 shows the evolution of the mass flow at the interface between the second solid wall and the indoor air.

On Fig. 3 we can see that the chipboard behaves as a sink (negative value of the mass flow rate) during high indoor concentrations of acetone and then release the pollutant during a long period of time.

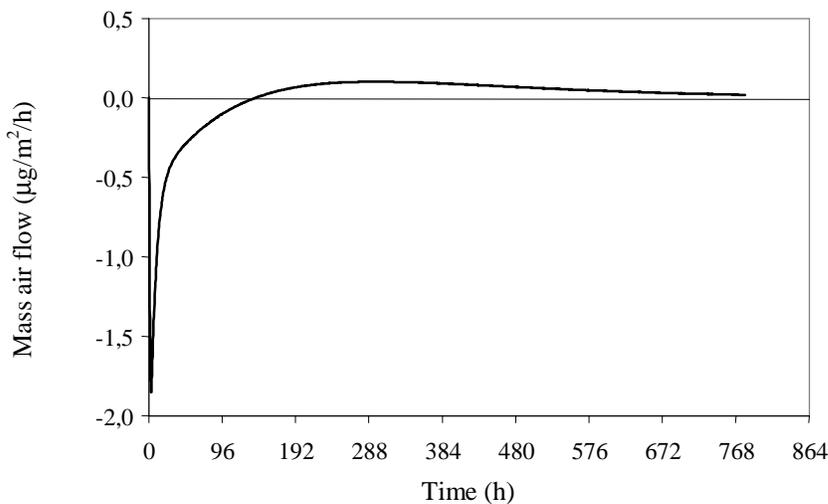


Figure 3: Acetone mass flow rate at the interface

In most studies on “material emission”, these phenomena are characterized as a global “emission rate”, but this simple example shows clearly the reversibility of the phenomena involved in this transfer process. Furthermore the proper selection of material could enable to reduce the peak level of concentration or delay its occurrence.

4. CONCLUSION

In the present paper, we show clearly that the influence of homogeneous reactions within the gas phase or heterogeneous reactions at the solid/gas interfaces cannot be neglected in predicting the indoor concentrations in pollutants.

On the one hand, these results are important because they can even suggest some way to passively limit the peak values of indoor pollutant concentrations by using appropriate materials, but on the other hand they also show clearly the lack of knowledge of the physical parameters necessary to describe the various phenomena to be taken into account.

The introduction of these reactive processes in a IAQ model has no formal difficulty but a tremendous research effort is then necessary to measure these phenomenological parameters to describe correctly these physical phenomena. That is the price to pay in order to improve the prediction of the dynamical evolution of pollutant concentrations in indoor spaces and evaluate new strategies improving the indoor air quality.

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