

Breakthrough time of activated-carbon filters used in residential and office buildings – Modelling and comparison with experimental data

Razvan Stefan Popescu¹⁻², Patrice Blondeau¹, Eric Jouandon³ and I. Colda²

¹ LEPTAB, University of La Rochelle, La Rochelle, France

² Bucharest Technical University of Construction, Bucharest, Romania

³ DGA/CTSN, Toulon, France

Corresponding email: patrice.blondeau@univ-lr.fr

ABSTRACT

While being used for years in industrial applications, gaseous contaminant sorption units such as activated-carbon filters remain marginal in office and residential buildings. This study deals with models that could help building designers and administrators to design and maintain activated-carbon filters depending on the environmental conditions (pollution load, airflow rate, temperature, humidity) they operate. More precisely, as a necessary first step of the development, the emphasis is put here on demonstrating the relevance of the models by comparing the predicted and measured breakthrough curves of various contaminants (isolated or as a mixture in the air) in dry and isothermal conditions. The results globally show a good agreement between predicted and measured data, while highlighting some interesting trends for future model developments. Beyond the question of model validation, the results also underline great discrepancies in the breakthrough time of the filter from one contaminant to another: the filter breaks within few tens of hours for some species when it breaks after several months for others. Considering the number of contaminant found in indoor settings, this shows that the question of the efficiency of the filter cannot be answered globally, and that the question of how often the filtering medium must be changed is not as simple as one can think.

INTRODUCTION

In the context of climate change, reducing drastically the energy consumption and related CO₂ emission of buildings has become a really challenging problem for most countries. For instance, the European directive on Energy Performance in Buildings (EPBD) estimates that the energy consumption of buildings should be reduced by 22% by the year 2010. Since ventilation can represent from 30 to 40% of the energy loss in new buildings, reducing the air change rate by using air cleaning systems may be an interesting solution to save energy while maintaining an acceptable indoor air quality. Two broad air cleaning technologies exist: oxidation techniques such as photocatalysis or cold plasmas have the advantage of decomposing organic compounds but face the problems of possible deactivation of the catalyst as well as possible yield of harmful secondary chemicals. On the other hand, adsorption systems such as activated carbon filters only trap gaseous species which means that the adsorption medium must be changed periodically to keep the filter efficiency. Then the question becomes to know how often the medium must be changed depending on the conditions the filter operates. Most existing methods are intended for industrial applications and cannot correctly account for the conditions encountered in buildings, especially time-varying mixtures of contaminants at low concentrations in the air, and in most cases fluctuating temperature and humidity at the filter inlet. The present paper addresses this problem. More precisely, the results presented here are part of a larger study aimed at developing physically-based models capable of predicting the dynamic change in the

efficiency of a packed-bed activated carbon filter operating in such conditions. As a necessary first step of the development, the emphasis is put on demonstrating the ability of the models to accurately fit the breakthrough curves of any gas. After describing the models for isolated contaminants as well as for mixtures of contaminants in the supply air-flow, the paper presents a comparison between the predicted and measured breakthrough profiles of six contaminants pertaining to different chemical classes.

MODELS DESCRIPTION

The adsorption filter models used for the study have been developed based on contributions from Axley [1] and Yu and Neretnieks [2] in the early nineties, as well as theories and equations proposed for the dynamic modeling of filters used in industrial applications (see for instance Do [3] or Tien [4]). All these studies consider that the sorption dynamics in the filtering medium are governed by four elemental transport phenomena, namely advective transports (contaminants are carried by the airflow passing through the filter), diffusion through the boundary layer separating the bulk air-phase from the adsorbent surface, porous diffusion in the adsorbent, and adsorption/desorption processes at the pore surfaces. The existing models have first been extended for a better representation of the sorption equilibrium of individual species in the field of low concentrations. Then, a multi-component adsorption isotherm model was implemented to account for the interactions between species at the pore surfaces.

Filter model for single contaminants

Under isothermal conditions, the sorption dynamics of a single contaminant in a packed-bed of spherical activated-carbon pellets is described by two equations, one representing the mass balance of the contaminant in the inter-pellet air-phase, and the other representing the mass balance within the pellet:

$$\frac{dC}{dt} = D_x \frac{d^2C}{dx^2} - \frac{d(uC)}{dx} - \frac{1}{r_{air}} h_m A_s (C - C^*) \quad (1)$$

$$r_{air} D_e \left(\frac{d^2 C_p}{dr^2} + \frac{2}{r} \frac{dC_p}{dr} \right) = r_s \frac{dC_s}{dt} + r_{air} e \frac{dC_p}{dt} \quad (2)$$

In equation (1), the first term of the right-hand side represents the contribution of turbulent axial dispersion. D_x ($m^2.s^{-1}$) is the axial dispersion coefficient, x (m) the axial dimension and C (kg/kg_{air}) the inter-pellet concentration. The second term, where u ($m.s^{-1}$) stands for the interstitial (inter-pellet) air velocity, represents the contribution of advective transports. Finally, the last term represents the contribution of boundary layer diffusion; r_{air} ($kg.m^{-3}$) is the air density ($kg.m^{-3}$), h_m ($m.s^{-1}$) the convective mass-transfer coefficient of the contaminant, A_s (m^2) the surface area of the pellets exposed to the bulk air, and C^* (kg/kg_{air}) the contaminant concentration at the pellet surface.

In equation (2), the left-hand side represents the radial diffusion within the adsorbent pellet. The terms on the right-hand side represent the contaminant accumulation in the air-phase of the pores, and the contaminant accumulation at the pore surface due to adsorption/desorption. C_p (kg/kg_{air}) is the pore air-phase concentration at radius r (m), D_p ($m^2.s^{-1}$) the diffusion coefficient in the pores, C_s ($kg.kg^{-1}$) the sorbed-phase concentration, and r_s ($kg.m^{-3}$) the activated-carbon density.

The continuity between the two equations is given by $C_p(r=R) = C^*$, where R (m) is the pellet radius. Moreover, since the adsorption/desorption transports at the pore surfaces are much faster than the diffusion transports, C_s and C_p can be considered to be always in equilibrium and related by the adsorption isotherm of the contaminant and adsorbent system:

$$C_s = f(C_p) \text{ or } C_p = f^{-1}(C_s) \quad (3)$$

The Dubinin/Radushkevich equation is usually taken as the adsorption isotherm model for its predictive characteristics (*i.e.* the model parameters for any gas can be assessed from its physical properties, and the temperature dependence of the sorption equilibrium is included in the model). However, this model is not suitable for many indoor air applications since it isn't defined at $C_p = 0$. It will therefore fail in representing the behavior of filters at the first times and/or when the contaminant air-phase concentrations approach zero. To get round this problem, as well as probably better fit reality, the simulations presented hereafter were carried out considering a linear adsorption isotherm model, $C_s = K_p C_p$, where K_p (kg_{air}/kg) is the so-called partition coefficient of the contaminant / adsorbent system.

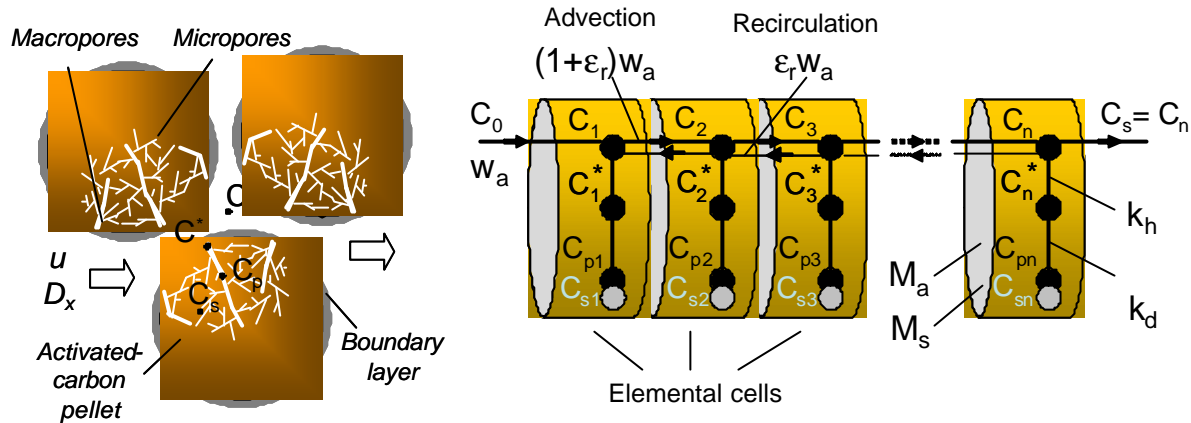


Figure 1: Schematic representation of the discrete filtering medium

To correctly account for the concentration gradients in the direction of the airflow on the one hand, and within the spherical pellets on the other hand, equations (1) and (2) are first discretized by decomposing the filter into n elemental cells connected in series (Figure 1); the concentrations C , C^* , C_p and C_s are assumed to be uniform within a same cell, but are of course different. Then, by using the linear driving force (LDF) assumption to describe the contaminant diffusion in the pores, equations (1) and (2) transform to the following equations where the only unknowns are the inter-pellet air-phase concentrations and the sorbed-phase concentrations in each cell, C_i and C_{si} , respectively:

$$\left\{ \begin{array}{l} M_{a1} \frac{dC_1}{dt} = w_a C_0 - w_a (1 + e_r) C_1 + w_a e_r C_2 - \frac{K_d K_h}{K_d + K_h} [C_1 - f^{-1}(C_{s1})] \\ M_{ai} \frac{dC_i}{dt} = w_a (1 + e_r) C_{i-1} - w_a (1 + 2e_r) C_i + w_a e_r C_{i+1} - \frac{K_d K_h}{K_d + K_h} [C_i - f^{-1}(C_{si})] \quad \text{for } i=2, n-1 \\ M_{an} \frac{dC_n}{dt} = w_a (1 + e_r) C_{n-1} - w_a (1 + e_r) C_n - \frac{K_d K_h}{K_d + K_h} [C_n - f^{-1}(C_{sn})] \end{array} \right. \quad (4)$$

$$M_{si} \frac{dC_{si}}{dt} = \frac{K_d K_h}{K_d + K_h} [C_i - f^{-1}(C_{si})],$$

M_{si} (kg) and M_{ai} (kg) are the mass of adsorbent and the inter-pellet mass of air in cell i , respectively; w_a (kg_{air}/s) is the airflow rate passing through the filter, and C_0 (kg/kg_{air}) the contaminant concentration at the filter inlet. Given that concentrations are thought to be very low compared to industrial applications, only the possible air recirculations between adjacent cells are considered for axial turbulent diffusion. The latter are characterised through e_r , a user-defined parameter ranging from 0 to infinity. $K_h = r_{air}A_s h_m$ and $K_d = 15M_s D_p \mathbf{s} / R^2$, with \mathbf{s} being the slope of the adsorption isotherm at concentration C_p , ($\mathbf{s} = K_p$ for a linear isotherm), represent the boundary layer diffusion rate and the diffusion rate in the pores of the pellets, respectively [1]. Each of the filter cells described by equations (4) was implemented as a mass transport component using the dynamic system simulation program Matlab/Simulink.

Filter model for contaminant mixtures

Except in the case of water vapour, contaminant concentrations in the outdoor as well as the indoor air seldom exceed few tens of $\mu\text{g}/\text{m}^3$. Consequently, it can reasonably be assumed that species do not interact when diffusing, but only compete for sorption at the pore surfaces of the adsorbent. In the frame of a study dealing with the ability of activated-carbon filters to clean the fresh air in heavily-polluted environments, Blondeau et al [5] used the volume exclusion theory as multi-component adsorption isotherm model. This model is more or less an extension of the Dubinin/Radushkevich isotherm for single gases and thus also fails numerically when the concentrations approach zero. Here, multi-component facilities were added in the model described above by implementing the extended Langmuir equation in equations (4). This equation expresses that the sorbed-phase concentration of a given species j , depends on the air-phase concentrations of all components k of the contaminant mixture through the relation:

$$C_{sj} = f(C_{pk, k=1, n_p}) = \frac{C_{sj}^0 K_{Lj} C_{pj}}{1 + \sum_{k=1}^{n_p} K_{Lk} C_{pk}}, \quad (5)$$

where C_s^0 (kg.kg⁻¹) and K_L (kg_{air}.kg⁻¹) are the Langmuir equation parameters for the individual species. The slope of the adsorption isotherm for a contaminant j is then given by:

$$\mathbf{s}_j = \left(\frac{\partial C_{sj}}{\partial C_{pj}} \right)_{C_{pk, k \neq j}} = C_{sj}^0 K_{Lj} \left(\frac{1}{1 + \sum_{k=1}^{n_p} K_{Lk} C_{pk}} - \frac{K_{Lj} C_{pj}}{\left(1 + \sum_{k=1}^{n_p} K_{Lk} C_{pk} \right)^2} \right) \quad (6)$$

Practically, implementing the extended Langmuir equation thus contribute to couple the mass transport equations: for n elemental cells and n_p contaminants in the air, the problem to solve is a set of $2 \times n \times n_p$ coupled differential equations.

Model parameters

As described by equations (4) to (6), the parameters of the models are the boundary layer mass transfer coefficients, h_m , the diffusion coefficients, D_p , and the sorption coefficients, K_p (single contaminant model) or K_L and C_s^0 (multi-contaminant model).

Various correlations between dimensionless numbers can be found in the literature to determine the boundary layer diffusion coefficients in packed-beds of adsorbents. Two were considered here, the Ranz-Marshall (RM) and the Wakao & Smith (WS) correlations [4]:

$$\text{Ranz-Marshall : } Sh = 2.0 + 0.6Sc^{1/3}Re^{1/2} \quad \text{Wakao \& Smith : } Sh = 2.0 + 1.1Sc^{1/3}Re^{0.6} \quad (7)$$

In these expressions, the Sherwood (Sh), Schmidt (Sc) and Reynolds (Re) numbers are defined by:

$$Sh = \frac{2Rh_m}{D_m}, Sc = \frac{\mathbf{m}_{air}}{\mathbf{r}_{air}D_m} \text{ and } Re = \frac{2RG}{\mathbf{m}_{air}}, \quad (8)$$

where D_m ($\text{m}^2.\text{s}^{-1}$) is the molecular diffusivity of the contaminant in the air, \mathbf{m}_{air} ($\text{Pa}.\text{s}^{-1}$) the air viscosity, and G ($\text{kg}.\text{m}^{-2}.\text{s}^{-1}$) the mass flux per unit cross-sectional area of empty bed.

If neglecting both solid and surface diffusion transports, diffusion in the adsorbent pellets reduces to air-phase diffusion in the porous interstices. The latter is characterized by the effective diffusion coefficient of the contaminant/adsorbent systems, D_e ($\text{m}^2.\text{s}^{-1}$), which can be assessed from the relations [3]:

$$D_p = D_e = \frac{1}{t} \left(\frac{1}{D_m} + \frac{1}{D_k} \right)^{-1} \text{ and } D_k = 97r_p \sqrt{\frac{T}{M}}, \quad (9)$$

where D_m and D_k ($\text{m}^2.\text{s}^{-1}$) are the molecular and Knudsen diffusivities of the contaminant, respectively; T (K) stands for temperature, M ($\text{kg}.\text{mol}^{-1}$) for the molecular weight of the contaminant, r_p (m) for the macropore mean radius, and t for the tortuosity factor of the pores.

COMPARISON BETWEEN EXPERIMENTAL AND NUMERICAL RESULTS

As a way to assess the reliability of the models, and to identify the needs for future developments, experiments carried out on a packed-bed of commercial activated-carbon pellets (mean radius $R = 1.1$ mm) were simulated under Matlab/Simulink. These experiments were small-scale laboratory experiments aimed to determine the breakthrough curves of six contaminants pertaining to different chemical classes. The tests were first carried out with each gas isolated in the air, and then with all the gases in mixture. In all cases the airflow rate was 0.610 kg/h and the temperature was set to $20^\circ\text{C} \pm 2^\circ\text{C}$. In the absence of reliable sorption data for the activated-carbon tested, the partition coefficients K_p were determined from the sorption equilibrium obtained experimentally: after the filter breaks, the air-phase concentration is the same throughout the filter and equal to the concentration at the filter inlet, C_0 . The corresponding equilibrium sorbed-phase concentration, and subsequently the partition coefficients, can be assessed from the total mass of gas adsorbed by integrating over time the contaminant masses entering and leaving the filter, that is:

$$K_p = \frac{C_s}{C_0} = \frac{m_s / M_s}{C_0} = \frac{1}{C_0 M_s} \left(\frac{w_a}{\mathbf{r}_{air}} C_0 t_p - \frac{w_a}{\mathbf{r}_{air}} \int_0^{t_p} C(t) dt \right) \quad (10)$$

Equivalent K_L and C_s^0 values were also determined from the computed K_p to feed the multicomponent models. Table 1 summarizes all the model parameters corresponding to the single contaminant and multi-contaminant tests. To simulate the sorption dynamics of a new filtering media for each test, the concentrations in all cells were set to zero at time $t=0$ (initial conditions of the models). In all cases, the filtering medium was divided into 50 elemental

cells of same length in the direction of flow. The re-circulation factor was set to 0 (ideal piston flow). Given the strong uncertainty on 1) the convective mass transfer coefficients (the coefficients obtained from the RM correlation are about three-fold the ones returned by the WF correlation, see Table 1), 2) the actual convective surface area (A_s is lower than the surface area of a pellet times the number of pellets due to contacts between pellets), and 3) the sorption coefficients, five simulations were carried out for each isolated contaminant: first, the uncertainty on K_h was accounted for by considering the reference values of K_p (and resulting K_d , see Table 1), and repeating the simulations for $K_{h,max} = K_{h,ref} = r_{air} A_{s,max} h_m^{WF}$ and $K_{h,min} = r_{air} \frac{A_{s,max}}{2} h_m^{RM}$. Then, three other sets of simulations were carried out considering $K_h = K_{h,ref}$ and $K_p = K_{p,ref} \pm 25\%$ to account for the presumed uncertainty on the latter parameter. The predicted breakthrough curves as well as the experimental data for the six contaminants are depicted on Figure 2. For the multicomponent case, the simulations were carried out considering the K_h values computed from the RM correlation and the Langmuir coefficients presented in Table 1 (Figure 3).

Table 1 : Model parameters corresponding to the tests

	R134a	Acetaldehyde	Ethanol	Acetone	Cyclohexane	Toluene
M (g.mol ⁻¹)	102	44	46	58	84	92
$C_0^{(1)}$ (mg/m ³)	2.80 (4)	1.08 (1)	15.65 (20)	1.06 (1)	1.25 (1)	1.07 (1)
D_m (m ² /s)	1.00 10 ⁻⁵	1.28 x 10 ⁻⁵	1.24 x 10 ⁻⁵	1.21 x 10 ⁻⁵	7.80 x 10 ⁻⁶	8.04 x 10 ⁻⁶
D_k (m ² /s)	2.60 x 10 ⁻⁴	3.96 x 10 ⁻⁴	3.87 x 10 ⁻⁴	3.45 x 10 ⁻⁴	2.86 x 10 ⁻⁴	2.74 x 10 ⁻⁴
D_e (m ² /s)	2.41 x 10 ⁻⁶	3.10 x 10 ⁻⁶	3.00 x 10 ⁻⁶	2.92 x 10 ⁻⁶	1.90 x 10 ⁻⁶	1.95 x 10 ⁻⁶
K_p (kg _{air} /kg _{AC})	86	1331	610	8314	25784	123642
C_s^0 (kg/kg _{AC})	0.542	0.2667	0.6372	0.3864	0.5315	0.7136
K_L (kg _{air} /kg)	260	5000	970	25000	50000	200000
Tests with the contaminants isolated in the air						
M_s (g)	21.35	22.3	16.3	16.3	8.41	8.43
M_a (g)	0.0168	0.0084	0.0116	0.0116	0.0072	0.0072
h_m (m/s) - WF	1.24 x 10 ⁻¹	1.49 x 10 ⁻¹	1.45 x 10 ⁻¹	1.43 x 10 ⁻¹	1.04 x 10 ⁻¹	1.06 x 10 ⁻¹
h_m (m/s) - RM	4.75 x 10 ⁻²	5.69 x 10 ⁻²	5.56 x 10 ⁻²	5.46 x 10 ⁻²	3.96 x 10 ⁻²	4.05 x 10 ⁻²
K_h (kg/s) ⁽²⁾ - WF	0.0193	0.0242	0.0172	0.0170	0.0064	0.0065
K_h (kg/s) ⁽²⁾ - RM	0.0074	0.0092	0.0066	0.0065	0.0024	0.0025
K_d (kg/s)	228	4710	1528	20328	20967	103886
Test with the mixture of contaminants – $M_s = 12.95$ g, $M_a = 0.0078$ g						
h_m (m/s) - RM	4.28 x 10 ⁻²	5.31 x 10 ⁻²	5.21 x 10 ⁻²	4.72 x 10 ⁻²	4.04 x 10 ⁻²	4.03 x 10 ⁻²
K_h (kg/s) ⁽²⁾ - RM	0.0040	0.0050	0.0049	0.0044	0.038	0.0038
K_d (kg/s)	<i>Calculated at each time-step as a function of the predicted concentrations</i>					

⁽¹⁾ Mean concentration measured during the tests - the concentration sets are put in brackets

⁽²⁾ Calculated based on $A_{s,max}$ (surface area of a pellet times the number of pellets)

If focusing first on the only numerical results, Figure 1 shows that decreasing K_h tends to level the predicted breakthrough curves. Increasing K_p also contribute to level the profiles but overall to delay the predicted breakthrough time of the filter. Then, it can be noted that the predicted concentrations at the filter outlet correctly fit the experimental data: the predicted breakthrough time agree with the measured ones, and the predicted concentrations are within the combined uncertainty range of K_h and K_p . It is important to emphasize here that only the sorption equilibrium state obtained experimentally were used to draw the K_p coefficients, and therefore the breakthrough profiles are not mathematical fits of the experimental data. The only contaminant for which the results aren't fully satisfactory is R134a: the breakthrough time is correct but the predicted concentration profile clearly do not resemble the measured one. Of all the possible explanations, the most likely is that the linear adsorption model fails for R134a due to its very high vapor pressure. Despite the strong uncertainty on the

parameters of the Langmuir adsorption isotherms, the predicted profiles also correctly fit the experimental data for the contaminants in mixture. Exceptions are acetone (breakthrough time overestimated of about 200 h) and acetaldehyde. In the latter case, it is interesting to note that the predicted breakthrough time seems to be correct but the filter surprisingly breaks at a concentration which is about half the inlet concentration. There are few doubts that this phenomenon originates from a binary surface reaction which contribute to consume acetaldehyde (and which is of course not accounted for in the model!). Unfortunately the tests were stopped too early to determine if toluene could be the other reactant involved in the reaction.

Beyond the problems of model validation and future numerical developments, all the results presented here underline the strong discrepancies in the breakthrough time of the filter from one contaminant to another. These breakthrough times vary from few hours for R134a and ethanol to several thousands of hours for toluene, which inevitably poses the question to know which contaminant to consider for maintenance issues. Should the life of the filtering media be based on high affinity contaminants such as toluene, with the consequence that the filter will be ineffectual in removing many other species contained in the air, or should it be based on lower affinity contaminants, with the consequence of much higher operating cost?

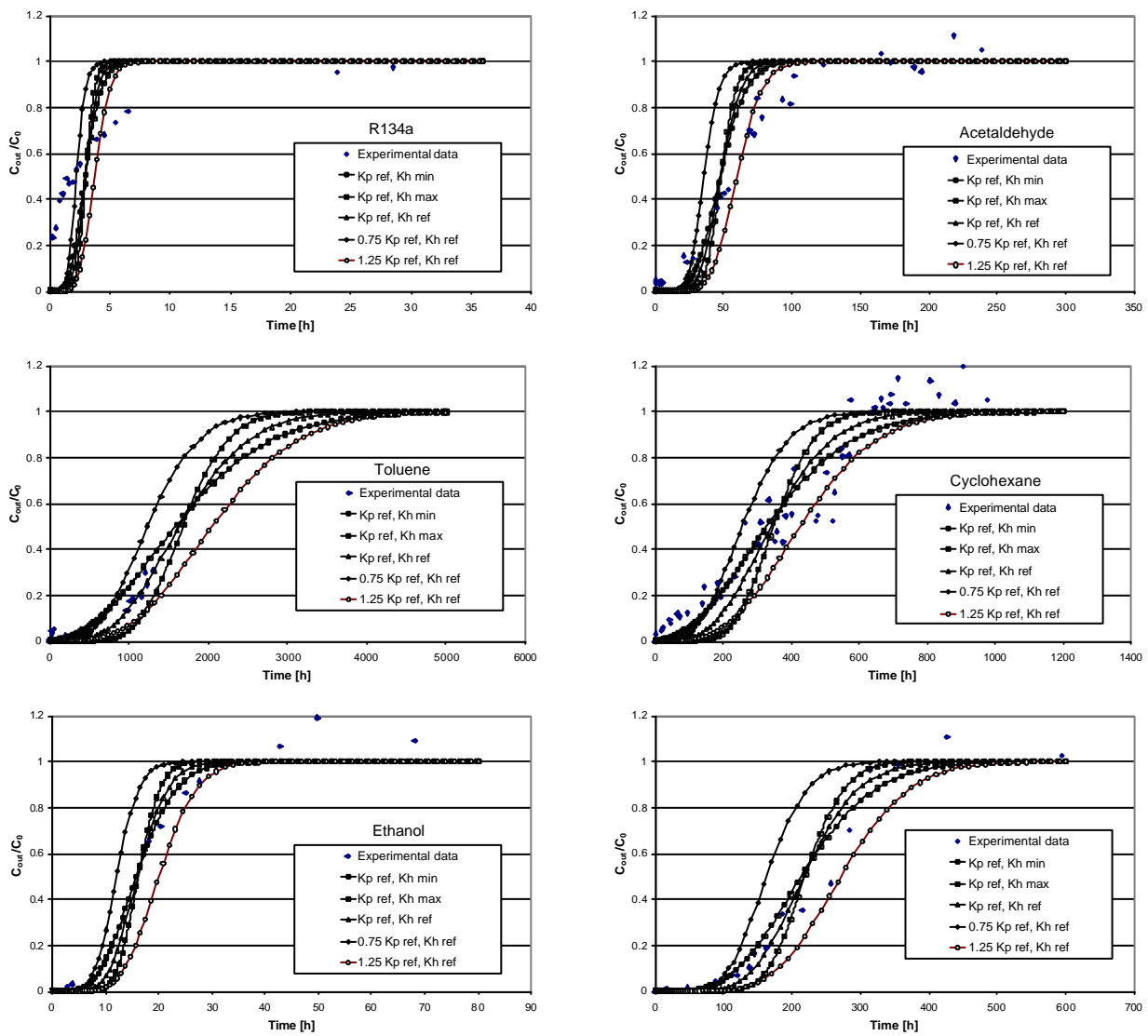


Figure 2: Breakthrough profiles of the contaminants isolated in the air

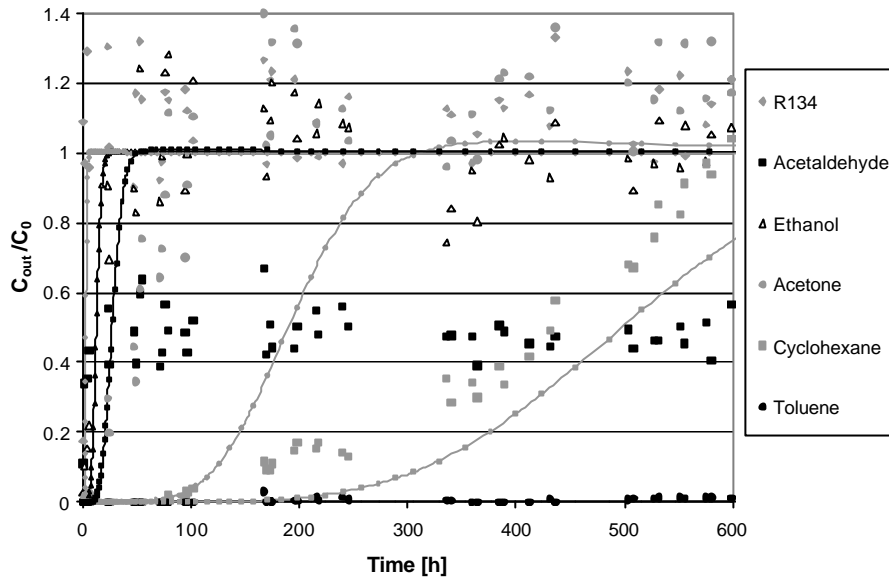


Figure 3: Breakthrough profiles of the contaminants as a mixture in the air

CONCLUSION

The results presented in this paper are a first step in developing dynamic adsorption filter models capable of accounting for all the elemental phenomena involved in the sorption process on the one hand, and the operating conditions characterizing building applications on the other hand. The predicted breakthrough profiles under dry and isothermal conditions globally fit the experimental data, which is a necessary condition for further developments of the models. Priority studies will now be directed toward coupling the mass –transfer equations with equations representing the thermal transfer in the filtering media, implementing functionalities to represent the influence of air humidity (absorption/desorption models, humidity-dependent diffusivities, ...), and adding surface chemistry models. It is important to note that progressing toward reliable design tools also needs more experimental data on surface reactions (models exist but very few is known about the species involved and the reaction kinetics) as well adsorption isotherms of contaminants in the range of (very) low concentrations.

REFERENCES

1. Axley, J.W. 1994. Tools for the analysis of gas-phase air-cleaning systems in buildings. ASHRAE Transactions, V. 100, Pt. 2.
2. Yu, J.W. and Neretnieks, I. 1993 The effect of a passive adsorption sheet on reducing organic pollutants in indoor air. Indoor Air 3(1), pp. 12-19.
3. Do, D.D. 1998. Adsorption Analysis: Equilibria and Kinetics. London: Imperial College Press
4. Yang, R.T. 1987 Gas separation by adsorption processes. Stoneham (MA): Butterwoths Publishers
5. Blondeau, P., Popescu, R., Colda, I. On the use of activated carbon filters in buildings. Proceedings of the 10th International Conference on Indoor Air Quality and Climate, Indoor Air 2005, pp.2935-2939