

Transient analysis of Indoor Air VOC-concentration Numerical tool for dynamic simulations

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Abstract

A dynamic simulation tool, specially constructed for system analyses in the field of indoor air quality (IAQ), is presented in this paper. This tool (called VOC-Tools) calculates the distribution of volatile organic compounds (VOC) in indoor air, taking into account the presence of building materials, furniture, appliances, ventilation systems and other dynamical sources and sinks of VOC's normally present in a building. Results of simulations are concentrations and duration times of VOC's in indoor air and building construction elements, which are necessary parameters for the IAQ assessment.

Besides the program description, two examples of validation tests are presented in this work. The first test concerns small scale laboratory measurements, and the second one large scale measurements on a house in use. In both tests, the program has shown a high degree of reliability. As an illustration of other program capabilities, an example of VOC's calculations with variable ventilation strategies is provided.

VOC-Tools is an open source and freely available on the Internet and any researcher or practitioner can use, expand and develop further this program.

Key words: Indoor Air Quality, Volatile Organic Compounds, Modeling, Dynamic Simulations, Sorption, Diffusion.

Practical Implications

VOC-Tools offer possibilities to assess concentration and duration of VOC's in the indoor air with respect to; surrounding building materials, sink effects in building materials, ventilation strategies, outdoor conditions and indoor sources of VOC's. Concentration of VOC's in indoor air could be a design criterion for ventilation systems. Building materials could strongly influence the duration and concentration of VOC's in indoor air. Consequently could selecting of indoor surface materials be a design criterion with respect for VOC concentration in the indoor air.

1 Introduction

A number of different volatile organic compounds (VOC) are present in indoor air. These gaseous pollutants are partly responsible for the reduced indoor air quality (IAQ). Headache, weakness, throat and eye irritation are some of the typical symptoms of sicknesses associated with the presence of VOC's (Mølhave et al., 1987). It has been proven that some of the symptoms are present even at low concentration levels of VOC's (Ten Brinke et al., 1998).

The cleanness of the indoor air and the way it is perceived by humans are fundamental considerations when a qualitative assessment of the indoor air is to be performed (Nilsson, 2003). Concerning VOC's and their impacts on human health, the qualitative assessment relates concentrations and exposure time to VOC's, in order to secure an acceptable IAQ and thereby a healthy indoor environment. It is desirable to perform this assessment under the design phase of a building.

Distribution of VOC's in indoor air is a rather complex problem. Building materials, furniture, appliances, ventilation systems, occupants, processes, etc. act as dynamical sources and sinks of VOC's. Field measurements are one way of obtaining quantitative assessments of IAQ, i.e. concentration and duration of VOCs in indoor air. Results from measurements are important because they give an insight in the real situation, but they can be done only after the building is constructed. An alternative is to perform calculations of VOC's in indoor air by means of a simulation tool. Calculations can be

done during the design phase of a building, when different design solutions and ventilation strategies can be explored.

There are two fundamentally different types of calculation models. Empirical or statistical models use parameters that are based on laboratory experiments, and then extend the application of these parameters to large-scale models, using the rule of similarity or proportionality. Since parameters do not have a clear physical meaning (they are defined only to explain one single experiment with mathematics), consequently, these models fail to assess the behavior when conditions and geometry are changed in relation to the original experiment. For example, it is impossible to simulate a whole building with an empirical emission model.

In recent years, several physical calculation models have been proposed (Yang et al., 2001; Huang et al., 2002; Zhao et al., 2002; Hansson, 2003; Murakami et al., 2003; Haghghat et al., 2003). These models are based on a number of fundamental mass transfer processes, where material transport properties are determined in experiments (Meininghaus, 1999; Hansson, 2003). Physical models have a clear physical meaning. As a consequence, it is possible to assess the behavior of a whole room (or a house), even when geometry and included building materials are changed.

In the initial approach, physical models were quite simple, limited to only one solid material component (Yang et al., 2001a; Zhao et al., 2002). Possibilities of modeling multi-layered and multi-component structures are needed when the situation in real

rooms are considered (Zhang et al., 2004). Physical models that were more realistic and practical useful were recently proposed (Haghighat et al., 2003; Zhang et al., 2004; Karlsson, 2004).

This paper proposes a numerical computer software (VOC-Tools), in which the framework is a physical calculation model. The strategy is to build a structured and highly flexible “toolbox”, rather than a predetermined software, to facilitate handling of complex systems. It must be stressed that the indoor environment is considered as a complex dynamic system in this study. Components from the toolbox are used to assemble a simulation model that represents the indoor environment.

When a real indoor environment is simulated, the toolbox must handle some features in order to be practically useful. These are mainly; handling of a multiple set of VOC's, multi-component structures (e.g. walls, floor, ceiling etc.) and sandwich components (e.g. layered wall consisting of more than one material). Handling of arbitrary boundary conditions is also an important factor that is included as a result of the toolbox approach.

In practice, VOC-Tools offer possibilities to assess concentration and duration of VOC's in the indoor air with respect to; initial concentration of VOC's in surrounding building materials, sink effects in building materials, ventilation strategies, outdoor conditions and indoor sources of VOC's.

2 Distribution of VOC's in indoor environment – The dynamic system

Distribution of VOC's in indoor environment should be analyzed on a building as a system, taking into account interactions between building envelope, building services, occupants and surrounding environment. This is a complex system that changes in time, as it is shown in Figure 1. Each interaction represents a specific physical process or a set of such processes and a dynamical behavior of the system as a whole depends on type and strength of these interactions. As a result, different sources of VOC's appear in indoor environment.

Quantities of VOC's can enter or exit the system passing, for example, the ventilation system, as it is shown in Figure 1. The strength of this process is defined by the ventilation airflow and the difference in concentrations between indoor and outdoor air. Occupants and equipment are also sources of VOC's. The characteristic of each source is, of course, highly individual. They differ not only in type of released gases but also in rates at which they are released, and these can be also variable in time. Finally, surrounding building materials are also an essential part of the system, since many of them contain primary emissions that sooner or later can be emitted to the indoor air, or have a significant storage possibility. Thus, only mass transfer of VOC's or both mass transfer and storage of VOC's can be addressed within such system.

3 Physical processes associated to mass transfer and sorption of VOC's in building materials

Mass transfer of VOC's between visible surfaces of surrounding materials and the indoor air is defined by boundary layer diffusion. There is always a resistance for mass transfer at the material surface, which is a function of the boundary conditions and material properties, but independent of the direction of the transport (into or out of the wall). Mass transfer within building materials is governed by molecular diffusion in vapor phase. Fick's first empirical law describes diffusion of gases in still air. The same type of equation is used to describe the diffusion within a pore system of the material.

The primary definition of sorption is the microscopic mass transfer of VOC from the very near surrounding air phase to a solid surface of a material (Tiffonnet et al., 2002). In this application the sorption represents a process of bounding VOC molecules at a solid surface. The strength of sorption is determined by the nature of the adsorbent (solid material) and the nature of the adsorbate (VOC molecules). VOC can be adsorbed at surfaces that are visible from the indoor air and at internal pore surfaces inside the material. Consequently there are two types of sorption processes: surface sorption and internal sorption.

4 Governing equations for the mass balance of VOC's

Some basic conditions that are used in this mathematical model:

- The amount of air that is within the room is assumed to be well mixed.
- No chemical reaction between different VOC's besides sorption and desorption.
- No phase changes for VOC's besides sorption and desorption.
- Immediate and full reversible sorption process, Henry's law describes both sorption and desorption with a linear partition coefficient.
- Isothermal conditions and a constant moisture content in the entire system (actually constant material properties for a normal thermal and hygroscopic condition).
- One-dimensional mass transfer within building materials.

Mass balance equations for the indoor air, the surface of a building material and the inside of a material are presented hereafter. These equations are time dependent partial differential equations.

The quantity of VOC's that are present in the indoor air are determined by the ventilation rate, the relation between concentration in indoor and outdoor air, the emission from sources and the interaction between indoor air and the surface of building materials. The mass balance equation for the indoor air is presented in Equation 1:

$$V \cdot \frac{\partial C_{zone}(t)}{\partial t} = \sum_{n=1}^{Sysnr} R_{A,n}(t) \cdot (C_{supply,n}(t) - C_{zone}(t)) + \sum_{n=1}^{Snr} \beta_{voc,n} \cdot A_n \cdot (C_{s,n}(t) - C_{zone}(t)) + \sum_{n=1}^{Gnr} J_{gain,n}(t) \quad (1)$$

where V (m^3) is the volume of the indoor air, C_{zone} (kg_{voc}/m^3_{air}) is the concentration in the indoor air, R_A (m^3_{air}/s) is the ventilation rate, C_{supply} (kg_{voc}/m^3_{air}) is the concentration in the supply air, β_{voc} (m/s) is the mass transfer coefficient for a specific construction, A (m^2) is the area for a specific construction, C_s (kg_{voc}/m^3_{air}) is the concentration at the surface for a specific construction and J_{gain} (kg_{voc}/s) is the mass flux from a gain within the indoor air. $Sysnr$ (-) (System number) is the total number of ventilation systems that is connected to the indoor air. Snr (-) (Surface number) is the total number of internal surfaces that is connected to the indoor air. Gnr (-) (Gain number) is the total number of gains within the indoor air. The mass balance equation for the visible surface of a construction is:

$$K_s \cdot \frac{\partial C_s(t)}{\partial t} = -\beta_{voc} \cdot (C_{zone}(t) - C_s(t)) + D \cdot \frac{\partial C_{int}(x=0,t)}{\partial x} \quad (2)$$

where K_s (m) is the partition coefficient at the surface, D (m^2/s) is the diffusion coefficient for the material closest to the surface, $C_{int}(x,t)$ (kg_{voc}/m^3_{air}) is the vapor phase concentration inside the material and x (m) is the depth measured from the surface towards the material. For this case, the concentration of VOC's at the surface is determined by the mass flux from the indoor air, the mass flux from the material and the amount of sorbed VOC at the surface.

Mass balance inside the material is determined by Equation 3:

$$K_{int} \cdot \frac{\partial C_{int}(x,t)}{\partial t} = D \cdot \frac{\partial^2 C_{int}(x,t)}{\partial x^2} \quad (3)$$

where K_{int} (-) is the internal partition coefficient. Concentration of VOC in a specific point inside a material is determined by the mass flux from each side of the point and the sorption at the same location as the point.

4.1 Numerical methods (Discretized form of mass balance equations)

There is no a general analytical solution to a problem defined by equations 1-3, instead, a numerical method is used and details are presented hereafter in brief. Numerical calculations are based on the control volume method and an explicit time discretization scheme. The first step in this method is to divide the domain into discrete control volumes. The storage capacity of the volume is lumped in a calculation node that is assigned to it. A mass balance equation (Equation 3) for a control volume of internal material with thickness d (m) gets the following form after the discretization:

$$C_{int}^{n+1} = \frac{(C_{int, left}^n - C_{int}^n)}{K_{int} \cdot d \cdot (R + R_{left})} \cdot \Delta t + \frac{(C_{int, right}^n - C_{int}^n)}{K_{int} \cdot d \cdot (R + R_{right})} \cdot \Delta t + C_{int}^n \quad (4)$$

where R (s/m) is the resistance to mass transfer inside the material due to molecular diffusion and upper indices (n and $^{n+1}$) correspond to the discrete time step, Δt (s)

Notations in Equation 4 are illustrated in Figure 2.

For the entire mathematical model, there are on the whole three different types of calculation nodes: one for the indoor air, one for the surface and one for the control volume of interior material. Similar calculation schemes, as exemplified in Equation 4, exist for the surface node and the indoor air node.

When the entire system is analyzed, all calculation nodes are connected in a system which is shown in Figure 3, following their physical connections from Figure 1. Using an electrical network analogy, the connection between nodes is represented by resistances (e.g. mass transfer coefficients, diffusion in materials or ventilation rate).

5 VOC-Tools

VOC-Tools is a library of software packages that are developed for modeling the distribution of VOC in an indoor environment. Using the graphical programming language Simulink (MathWorks), VOC packages are designed as block diagrams, see Figure 4. A block diagram is a common term for the basic elements used during the modeling process. Modeling in Simulink is a process of assembling more complex blocks from the standard ones. Simulink has a modular structure that is hierarchical and open on every level. This means that each of the block diagrams may hide other subsystems. The structure and the graphical approach facilitate handling and control of very complex interaction between different parts of the model. As a part of Matlab package, Simulink has built-in state-of-the-art ordinary differential equation (ODE) solvers, which are automatically configured at run-time of the model. Therefore, only a physical model needs to be implemented, and not the solver.

Each block diagram in the VOC-Tools library represents a part of the dynamic system from Figure 1. Blocks contain calculation nodes with corresponding calculation schemes, which are defined by the mathematical model. The total network is established when all blocks are coupled together, see Figure 3 and 4.

At present VOC-Tools library contains five categories of block diagrams. They are:

- Zone (Indoor air)

- Surface material
- Material (Internal material of constructions)
- Systems (e.g. ventilation systems)
- Gains (e.g. internal sources of VOC)

VOC-Tools block diagrams communicate over data arrays. Each of the arrays includes necessary physical states and properties at the boundaries of the block diagrams. There are all together five different arrays, structured as it is shown in Table 1 and Figure 4. The defined interface allows anyone to contribute with a new block diagram to the existing library. New blocks could be based on other modeling theories (e.g. non linear sorption isotherms) or could have other features (e.g. wet coating materials).

Material properties are arranged in a material database in the form of “structures”. A structure is a type of a multidimensional array in Matlab, with data containers called “fields”. A database can contain properties for different materials, but they must be organized in the same types of fields, see Table 2.

Keeping the same structure, and especially field names, allows users to create an individual database of materials. Any other properties can be added to the database, just with another describing name. This feature allows users and developers to use their preferable material property matching their models.

VOC-Tools is a part of “The International Building Physics Toolbox” (IBPT)

(www.IBPT.org). One of the cornerstones in IBPT is that the software should be open on every level so that anyone could develop and adjust the tool to fulfill their needs. The software should also be freely available on the IBPT website. These two cornerstones are also incorporated in VOC-Tools.

6 Empirical validations of VOC-Tools

Two experiments are selected for the validation of VOC-Tools. The first experiment deals with diffusion and sorption of toluene in a gypsum board, in a small-scale experimental chamber. The second experiment shows the distribution of toluene in a full-scale house with authentic indoor environment. Using the standard block diagrams from the VOC-Tools library, numerical models for both experimental setups are designed and simulations performed.

6.1 Small-scale validation: Dynamic sink effect experiment

This experiment is originally performed by Hansson (2003), for studying the processes of diffusion and sorption of toluene in a gypsum board. The experiment is also called the “Twin Compartment method” because the experimental setup consists of two steel chambers, labeled A and B in Figure 5, separated by the test material.

Before the experiment starts, each compartment is well ventilated with clean air to insure that there is no toluene in the compartments or in the test material. The experiment starts (at $t = 0$ min) when a certain amount of toluene ($J_{\text{supply}} = 2.83 \times 10^{-9}$ kg_{voc}/s) is added to the supply air of the compartment A. Compartment B is ventilated with clean air only. Gypsum board is permeable to toluene and therefore some amount of toluene will diffuse to compartment B. The concentration of toluene in the supply and the exhaust air from both compartments is continuously measured and recorded using a quadruple mass spectrometer. Results from measurements are presented in Figure 6 and Figure 8.

Toluene is added to the supply air of the compartment A until the mass transfer to the compartment B reaches a steady state condition. At this moment the first phase of the experiment, called the adsorption phase, is completed.

The next phase of the experiment starts with a second step change in supply air concentration of toluene. From this moment ($t = 140$ min) both compartments are again ventilated with clean supply air. This phase of the experiment is called the desorption phase.

6.1.1 Input data to VOC-Tools simulations

The dynamic sink effect experiment is simulated in VOC-Tools. The model consists out of one Material block and two Zone blocks, to each Zone block there is a System block coupled. Basic conditions for the twin compartment are illustrated in Table 3. Dimensions and physical properties for the gypsum board are illustrated in Table 4.

The spectrometer gives data points of the concentration in each compartment as a function of time. From the mass balance at the steady state, it comes out that 98.73 % of the total mass of toluene that is introduced in compartment A is exhausted from compartment A and B (Hansson, 2003). The cause of a disappearance of 1.27 % of mass of toluene in the experiment is not known, but the author (Hansson, 2003) discussed different influencing factors. One way to recompense the missing quantity in the simulations is to introduce an irreversible sink in the model (Equation 1), which will

subtract 1.27 % of the introduced mass of toluene in order to achieve a mass balance, (Hansson, 2003).

In the corresponding VOC-Tools simulations another approach is used to correct the unfulfilled mass balance at the steady state. The total mass transfer resistance (the diffusion coefficient) over the gypsum board is selected so to minimize the relative difference in compartment concentration between measurements and simulations, in each compartment at the same time. The relative mass balance differences in each compartment are also set to be equal.

Partition coefficients, K_{int} (-) and K_s (m), are determined to achieve best fit to the measured data points at the transient part of both the adsorption phase and the desorption phase (Hansson, 2003).

6.1.2 Results of simulation with VOC-Tools

Results of simulations for the adsorption phase are demonstrated in Figure 6 and Figure 7. Figure 6 shows the concentration of toluene in each compartment as a function of time. Figure 7 illustrates the distribution of toluene across the gypsum board at some points in time. The desorption phase is presented in the same way in Figure 8 and Figure 9.

The relative difference between the measured and the simulated concentrations under the steady state condition is in the order of 1 %, the simulated concentrations are higher than the measured ones. When the transient part of the experiment is studied, the discrepancy

is generally higher than for the steady state condition. The discrepancy is larger in compartment B than in compartment A.

6.2 Large-scale validation: Field test in a whole house

This experiment was originally performed for the validation of a computational model “Surface Sink Model” (SSM), Won et al. (2001a). The test house has a total interior volume of 500 m³ divided in two floors and one basement. An open container of a pure toluene is placed in the basement. For the next nine hours a constant source of 539 mg/h toluene contaminates the indoor air. The total air exchange rate of the house was 0.16 h⁻¹ constantly. All doors inside the house were opened throughout the experiment.

The concentration of toluene in indoor air is measured at several locations in the house and results are shown in Figure 10. According to these results, the air inside the house can be considered as well mixed, which is one of the assumptions behind the VOC-Tools.

6.2.1 Input data to VOC-Tools simulation

Walls and roof materials and their individual areas are specified in Table 5. Apart from gypsum board and concrete, there are also large areas (floors) covered by painted wood. Since authors (Won et al., 2001b) could not detect any sink effect of the wood for toluene, consequently these areas are disregarded in the VOC-Tools simulations.

The wall type 1 is modeled as a 13 mm thick gypsum board with no coating at the visible surface and sealed for the transport of toluene on the rear side. The wall type 2 represents an internal wall with two visible surfaces towards the indoor air. It consists of acrylic

paint (50 μm), gypsum board (13 mm), still air (0.1 m), gypsum board (13 mm) and acrylic paint (50 μm). The only specification available for wall type 3 is the paint on the internal side of a concrete wall, (Won et. al., 2001b). However, in VOC-simulations this wall type is treated as a 0.1 m concrete wall, painted on the side towards interior (acrylic, 50 μm) and sealed for the transport of toluene on the other side.

Transport properties for the specified materials are given in Table 6. Properties for gypsum board and acrylic paint are collected from Hansson (2003). Since the transport properties of toluene in concrete were not available, the transport properties of n-Octane in concrete were used (Meininghaus, 1999). These material properties are believed to be the best available and are therefore used as a rough estimation for toluene and concrete.

6.2.2 Results of simulation with VOC-Tools

Concentration of toluene in the indoor air increases rapidly during the first nine hours, as it is shown in Figure 10. The rate of the increasing concentration is almost entirely determined by the strength of the source and the air exchange rate. Compared to the total amount of toluene that is released to the indoor air, only a small quantity is sorbed by the surrounding materials, Figure 12. It reaches a maximum of 0.576 g after 9.53 hours, which is only 11.9 % of the entirely released. The rest of toluene is dissolved in the indoor air and taken away by the exhaust air. One can notice that surrounding building materials act as sinks of toluene during the first period of the experiment.

After 9.53 h, the surrounding materials act as sources of toluene. Wall type 1 and 2 release the sorbed quantity of toluene very fast due to the high permeable gypsum board,

Figure 12. For the late desorption phase, after about 30 hours, the concentration of toluene in the indoor air is almost entirely determined by the mass transfer (internal diffusion) from the concrete wall (wall type 3). The strength of the toluene source from the concrete wall is large enough to extend the duration of toluene in the indoor air for more than 100 hours.

Since transport properties of toluene in concrete are only estimated for this simulation, solutions with other magnitudes are studied in Figure 11. The graph that illustrates the resulting concentration of toluene in indoor air over a time has basically the same shape as in Figure 10. An increased diffusion coefficient D_{concrete} (for example) gives a higher concentration of toluene in the indoor air for the late desorption phase. The opposite behavior, a lowered concentration of toluene in the indoor air is observed when the diffusion coefficient is decreased. Changes of the internal partition coefficient give similar results.

Results of calculations based on SSM (Won et al., 2001a) are also given in Figure 10. Authors modeled the walls type 3 as gypsum boards. The rather bad correlation between the measured and the calculated data (SSM) can be explained by the neglect of the significant sink effect of the concrete.

7 Application of VOC-Tools: Simulation of a realistic room with variable ventilation airflow

This example illustrates additional simulation abilities of VOC-Tools, such as the investigation of a system dynamics under a multiple VOC release and a variable ventilation airflow rate. The model system represents an office room with constant sources of pollutants - Ethyl acetate and n-Octane. During working hours, when people are present in the room, the air exchange rate is at maximum, having 1 h^{-1} . During nights and weekends, the air exchange rate is reduced to 0.3 h^{-1} .

The investigation concerns the concentration of the selected gases in indoor air in time. In the first approach, the sorption effect of building materials is taken into account. In the second approach, which is described in relevant literature (Nilsson, 2003), the sink effect is disregarded and only a simplified dilution model is used. Resulting concentrations of VOC's for both cases are then compared in order to illustrate the power of the sink effect of the building construction. In addition, three different building construction solutions to the room enclosure are analyzed: concrete walls (massive), light sandwich walls (gypsum boards and an air gap in between) and light gypsum walls with an air barrier, each with the different sorption ability.

The source of VOC's together with the variable ventilation airflow is the origin to two equilibrium concentrations that will be present in the indoor air. A step change in the ventilation airflow is the start for a change in indoor air concentration of VOC's from one of these equilibriums to the others. How long this change takes place depends on the

area/volume ratio of the space, and also on transport properties of surrounding building materials, boundary conditions at the visible wall surfaces and the distribution of VOC inside the building construction (i.e. the time history of the indoor air concentration). The concentration of VOC's in indoor air during daytime is of special interest when the assessment of the IAQ is to be performed. Release of VOC's from massive walls is also an interesting aspect when IAQ is studied, since such walls could act as very large sinks/sources of VOC's in comparison to thin gypsum boards. The time that is needed to extract VOC's from these potential sources via ventilation of the zone is also investigated.

7.1 Input data to VOC simulations

The model room has dimensions comparable to a small office room, with an interior volume of 37.5 m^3 (5x3x2.5 m). All internal surfaces except the floor are open to VOC's (55 m^2). Inside the room there is a source of VOC's that emits n-Octane and Ethyl acetate at the same time and at the same and the constant rate of $900 \text{ } \mu\text{g/h}$. The emission rate is selected so to give reasonable equilibrium concentrations in the indoor air compared to normal mean concentrations of these gases.

The massive wall structure (0.1 m concrete) is highly tight to VOC's. The second structure represents highly permeable walls: 0.2 mm wallpaper with paste, 25 mm gypsum board, 0.1 m still air, 25 mm gypsum board, 0.2 mm wallpaper with paste. The last type of the construction consists of 0.2 mm wallpaper with paste, 25 mm gypsum board and an air (actually VOC) barrier on the rear side. Thus, this construction is

completely vapor tight but still able to admit a fast diffusion of VOC within the gypsum board.

Transport properties for selected materials are listed in Table 7. Data for the diffusion coefficient, D , were originally determined by Meininghaus (1999) under the assumption that a mass transfer resistance at the wall surfaces could be disregarded in comparison to the diffusion resistance inside the structure. Lee et al. (2000) and Haghghat et al. (2002) showed that this assumption was incorrect, especially if the material is highly permeable to VOC (i.e. gypsum board). Neglecting the mass transfer resistance will lead to an incorrect evaluation (to a low value) of the diffusion coefficient. In Haghghat et al. (2002) the corrected diffusion coefficients can be found, based on measurements made by Meininghaus (1999). The corrected values are used in this example.

The internal partition coefficient, K_{int} , for wallpaper is not available and therefore the wallpaper is included only as a resistance to VOC mass transport in the simulation. The partition coefficient at the surface (K_s) for all three materials originates from Hansson (2003).

Boundary conditions at all observable surfaces in the room are assumed to be similar, with the air velocity of 0.1 m/s parallel to the surface. Mass transfer coefficients in the air are calculated by using the model from Axley (1991). Based on diffusion coefficients in still air (6.0×10^{-6} m²/s for n-Octan, 8.5×10^{-6} m²/s for Ethyl acetate (Bolz et al., 1973)), the

following values are obtained and used in the simulations: 248×10^{-6} m/s for Ethyl acetate and 313×10^{-6} m/s for n-Octane.

For construction types one and two, mass transfer coefficients on the rear surfaces are the same as for the internal, and a zero VOC concentration is assumed in adjacent air volumes. For construction type three, the boundary condition at the rear side is a VOC tight surface (vapor barrier).

7.2 Results from simulations with VOC-Tools

Figure 13 shows variations in concentrations of Ethyl acetate and n-Octane in indoor air during one week, for the case with concrete walls. When the building construction is treated completely tight to VOC's (no sink effect), lower and upper equilibrium concentrations are achieved at $24 \mu\text{g}/\text{m}^3$ and $80 \mu\text{g}/\text{m}^3$ respectively. In the case that the building construction is open to VOC's, concentrations of Ethyl acetate and n-Octane remain all the time within these limits. There are two effects that prevent them to reach the limits: sorption and diffusion through the present walls. Certain quantities of VOC's are stored in walls and the rest leave the system (to adjacent rooms) governed by the proposed boundary conditions. The mass transfer of VOC's that leaves the system via diffusion through the concrete walls is 0.6 or 1.4 % (Ethyl acetate or n-Octane) of the mass transfer from the source. At the visible surfaces is the mass transfer larger, despite direction of mass flux. This mass transfer is in the range of 10-20 %, of the mass transfer from the source, when release is considered and 3-9 % for the uptake process. In Meininghaus (1999), the effect of mass transfer through the concrete walls is considered to be equal to zero for a similar room and at a steady state condition.

Duration curves for the concentrations of Ethyl acetate and n-Octane are given in Figure 14, together with the solution to the case with no sink effect. Concentrations of both gases are equal to one when the sink effect is neglected. Deviations from the unity, when the sink effect is present, tell about the differences in concentrations in percents, quantifying the sink effect at the same time.

According to Figure 14, concentrations of both Ethyl acetate and n-Octane are about 10 % higher during working hours than in the case without the sink effect. About a quarter of the day time period (2 h) is needed to reach the 10 % value, after the ventilation air flow is increased to 100 %. During nighttime and weekends, concentration levels are 6 % lower than in the case with no sink. The increments in concentrations during the first four hours (a quarter of this period) are caused by the dissolution of VOC in the indoor air before sorption to surface materials starts to be significant.

Figure 15 illustrates the active penetration depth for the current load from the internal side of the concrete wall. Changes in indoor air concentrations are visible to about 6 mm depth from the wall surface. Behind this so called penetration depth, the profile in the wall is mostly determined by the steady state condition, which is defined by the mean concentration in the indoor air and zero concentration on the rear side of the structure.

When the concentration profile across the concrete has the shape as the one from Figure 15, it means that a large quantity of Ethyl acetate is stored in the wall deep inside (behind

the first 6 mm). If the source of Ethyl acetate is suddenly removed from the room and the ventilation system remains working, the amount of Ethyl acetate that is released from the walls is enough to keep the mean concentration in the indoor air higher than $1 \mu\text{g}/\text{m}^3$ for nearly 50 days, Figure 16. A thing to keep in mind is that Ethyl acetate is also released to adjacent rooms. This contamination could be important when the long term aspects of depots are studied.

The second wall structure (gypsum boards and the air in between) is much more permeable than the concrete structure, and therefore the results are a bit different. In this case, the mass transfer through the wall is of more interest than for the other two (tight) structures – it reaches 10 to 15 % of the mass transfer rate from the source. For Ethyl acetate the mass transfer rate is larger than for n-Octane due to higher permeability of the selected materials. Mass transfer towards the adjacent rooms will change with the variations in the indoor air concentrations. During periods with low ventilation rates, when the concentration in indoor air is higher, this transfer is also larger. The difference between the resulting indoor air concentration for the case with the sink and without the sink is consequently larger for the specified period, see Figure 17.

Figure 18 illustrates results of simulation for structure type three (wall paper, gypsum board and a VOC tight barrier). In this case, only the effects from depots are studied, due to the presence of the VOC barrier. Mass transfer of VOC through the structure will not be active in this case. Concentration in the indoor air will be much higher during periods of low ventilation compared to the structure type two. The release of VOC's from the

structure increases the concentration in the indoor air up to 18 % during the periods of high ventilation rates, compared to the case with no sinks, see Figure 18.

8 Discussion

Meininghaus (1999) points out that the masonry of a house can play a substantial role in the context of the sink effect. Material properties for brick and concrete are advantageous when influence of the sink effect is considered. Constructions made of these materials are often thick and cover large areas in a house (e.g. wall and floor). The sink effect in this type of structures is illustrated in the large-scale validation and in the application example. The implication is that this type of constructions allows large depots of VOC's inside such a material. It is therefore important to determine accurate material properties for this kind of materials. As well as it is important to model the actual thickness of this type of materials when simulations are performed.

Many studies have focused on the relationship between moisture in buildings and IAQ (Bornehag et al., 2004; Bornehag et al., 2001; Sundell et al., 1994; Spengler et al., 1993). Other studies have pointed out that the moisture content in building materials could have a key function when VOC emissions from building materials are considered (Won et al., 2001b; Sjöberg, 2001; Haghghat et al., 1997; Kirchner et al., 1995). The moisture content in building materials are affected by seasonal changes in the climate as well as changes over the day. Material properties for both VOC diffusion and sorption could be affected by fluctuating moisture content in building materials. It could therefore be important to study the influence of moisture in building materials when VOC's in the indoor environment is considered. There is a mutual relationship between heat and moisture. For that reason, heat must be simulated coupled together with moisture. The thermal states in building materials also have a direct effect on diffusion and sorption of

VOC's (Van der Wal et al., 1997; Haghghat et al., 1997) since adsorption and desorption are exothermic and endothermic processes. Thick constructions made of concrete are again important when moisture and heat are considered, since they can contain large amounts of moisture.

VOC-Tools have been developed with the structure of HAM-Tools (Heat, Air and Moisture) as a pattern. There is a plan to combine these tools in the future, in order to assess the influence of the HAM-states in building materials on the uptake and release of VOC's. A simulation of HAM-states together with VOC's means that material properties for diffusion and sorption of VOC's are dependent of the hygro-thermal condition in the material. This approach will even more increase the requirement of material properties and the way that they are determined.

9 Conclusions

A numerical model and a program structure for a dynamic simulation tool (called VOC-Tools) have been presented. The framework of the calculation model is based on fundamental mass transfer processes which describe distribution of VOC's in a building as a whole. Sources of VOCs that are different in nature, e.g. indoor air and contaminants, emission from walls, ventilation, etc., are modeled as separated calculation procedures (tools), giving the program a modular structure. As a result, the VOC model for the whole building is build-up in a similar way as the building itself – by coupling the modules in a system. The modular structure is maintained through a well defined interface, while numerical solvers and a graphical programming approach are inherited from Simulink (Mathworks), which is chosen as a developing environment.

VOC-Tools calculations are validated against two experiments. The first experiment accounts for a small-scale validation, where the sink effect of a gypsum plate is investigated in laboratory conditions. Gas concentrations in the testing chamber are appropriately assessed by VOC-Tools, and by that, the material properties of the gypsum board, which have been initially unknown, are estimated.

The second experiment is performed on a house in use (a large-scale validation) and therefore represents a more realistic indoor environment. VOC-Tools simulations of the considered house show very good correlation with the experiment. Material properties for this simulation are taken from literature. Results from Figure 11 and Figure 12 shows the sensitivity analyses on transport properties of the concrete. The selected properties for

concrete appear to be correct. The large-scale validation shows a significance of thick building materials and their capacity to act as potential depots for VOC's in indoor air. In the long-term study, the sink/source effect of a thick concrete dominates the one of the thin gypsum board.

The importance of VOC depots in a building is additionally illustrated in the application example. A VOC depot that builds up during nights and weekends when the air exchange rate is lowered, is studied. The release from the depot will increase the concentration of selected VOC's in the indoor air by 5 to 18 % compared to the case with no sinks in the system. The simulation also shows that it takes more than 50 days to "empty" the depot (0.1 m thick concrete wall), i.e. to release the stored amount of VOC's in indoor air and than to extract out by ventilation system.

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Symbols

A	m^2	Area of a construction element
C_{zone}	kg_{voc}/m^3_{air}	Concentration of VOC in indoor air
C_s	kg_{voc}/m^3_{air}	Concentration of VOC close to the material surface (Boundary layer)
C_{int}	kg_{voc}/m^3_{air}	Concentration of VOC in internal pores (air phase) of materials
C_{supply}	kg_{voc}/m^3_{air}	Concentration of VOC in supply air
D	m^2/s	Diffusion coefficient for VOC in building materials
D_a	m^2/s	Diffusion coefficient for VOC in still air
Gnr	-	Number of different gains within the indoor air
Gasnr	-	Total number of different VOC's that is included in one model
J_{gain}	kg_{voc}/s	Mass flux of VOCs that is generated from one single gain
K_{int}	-	Linear partition coefficient (Internal pores in material)
K_s	m	Linear partition coefficient (Surface of material)
L	m	Characteristic length for one surface
N	h^{-1}	Air exchange rate
R_a	m^3_{air}/s	Ventilation air flow rate
R	s/m	Resistance to mass transport (diffusion) of VOC in the interior of materials
R_s	s/m	Resistance to mass transport of VOC through surface material (If diffusion coefficient not known)
R_{right}	s/m	Resistance in positive X direction
R_{left}	s/m	Resistance in negative X direction
t	s	Time
Δt	s	Time step
U_{∞}	m/s	Undisturbed air flow speed (parallel material surface)
V	m^3	Volume of indoor air
x	m	Coordinate for depth in material (0=surface, positive direction towards the exterior)
β_{voc}	m/s	Mass transfer coefficient between indoor air and visible surface (over boundary layer)
ν	m^2/s	Kinematic viscosity for air

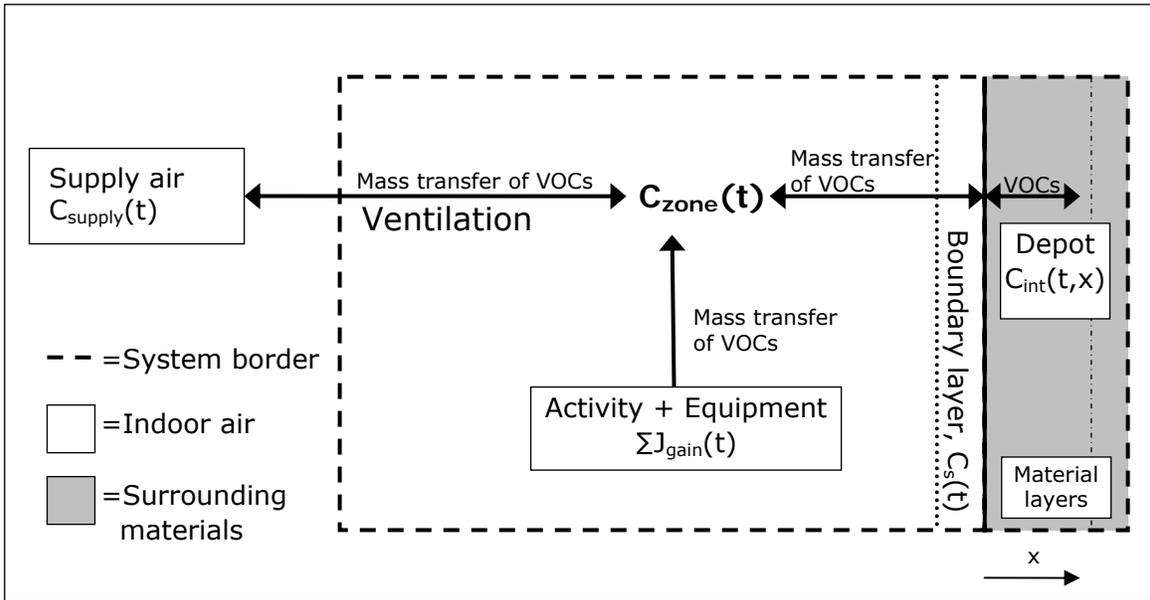


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Henrik Karlsson

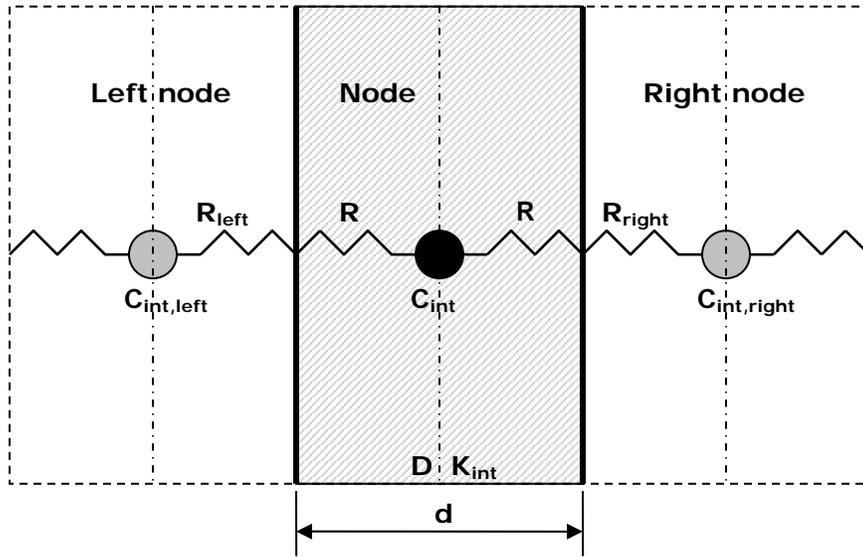


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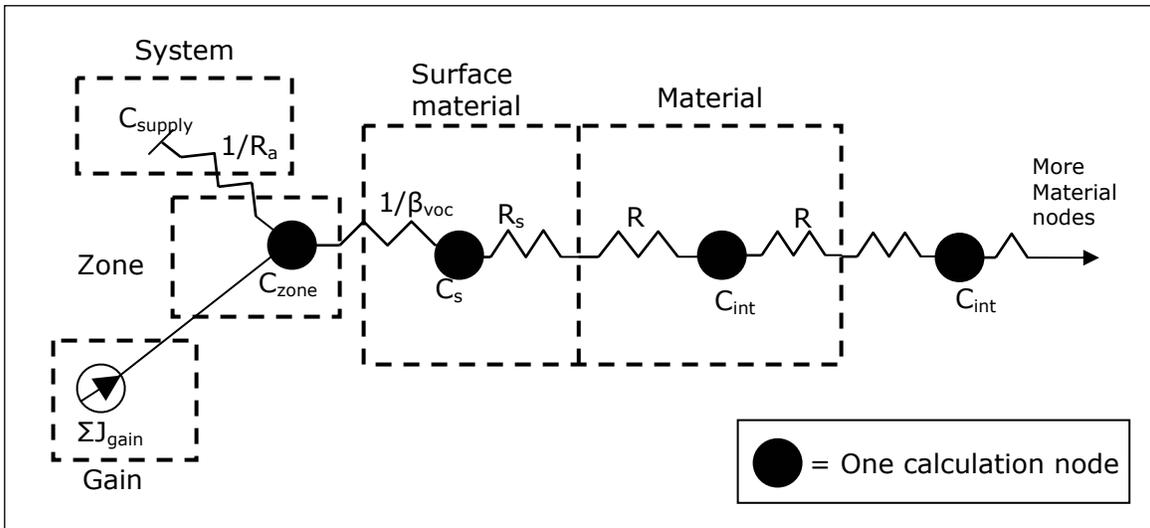


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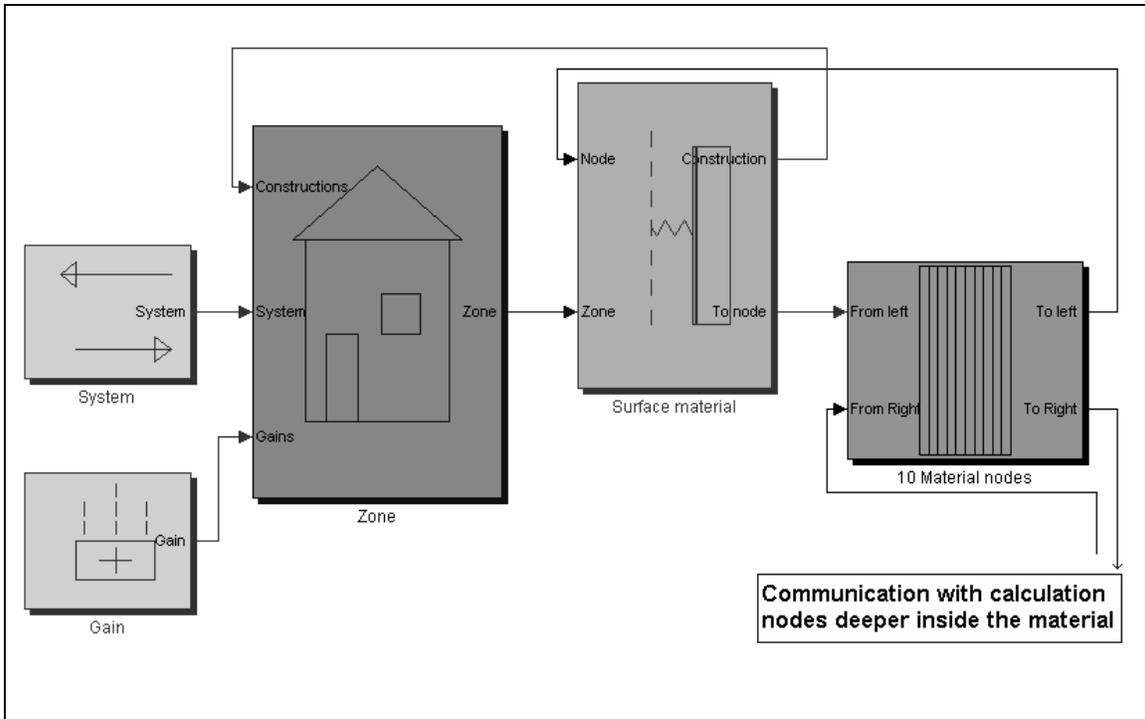


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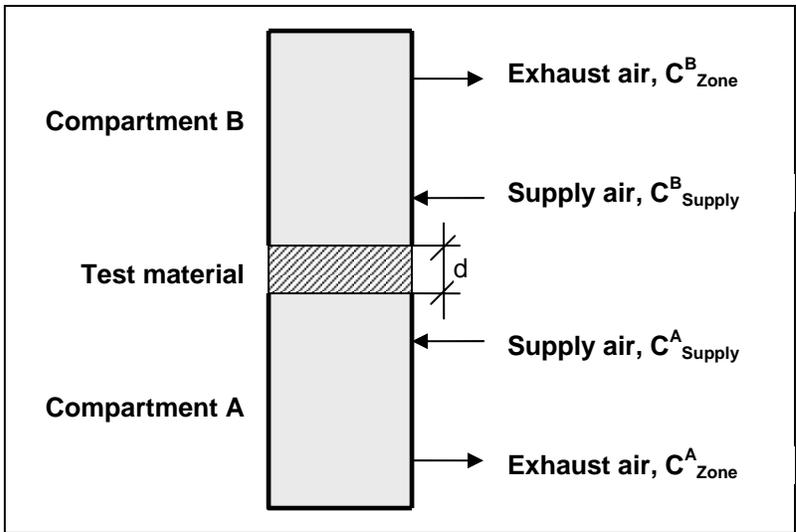


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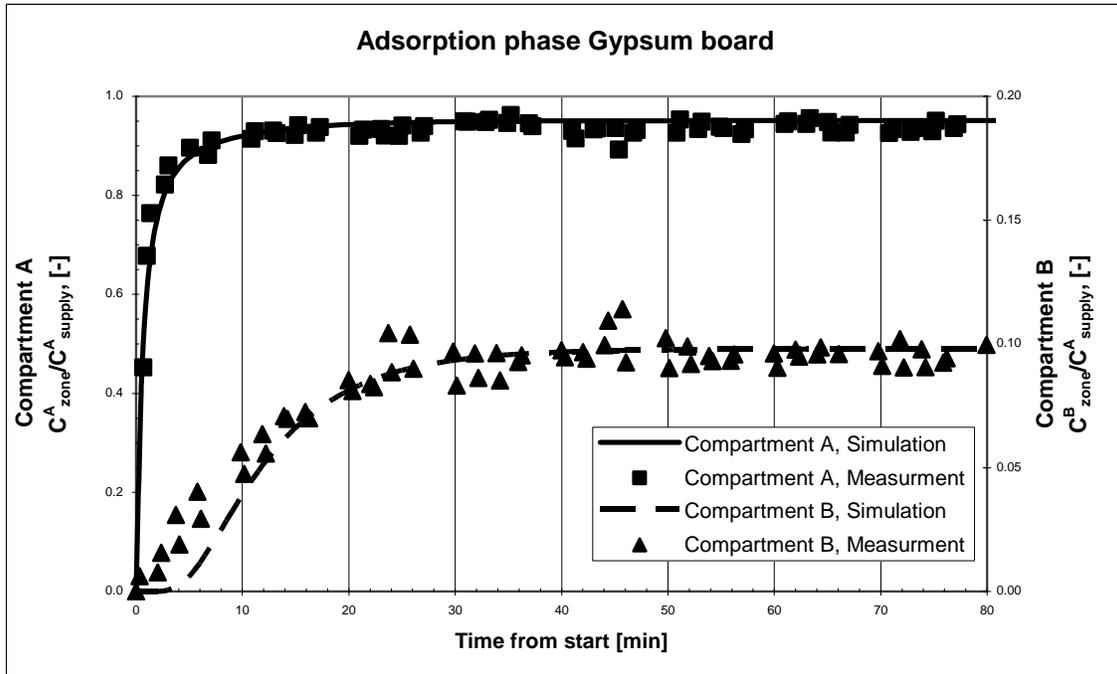


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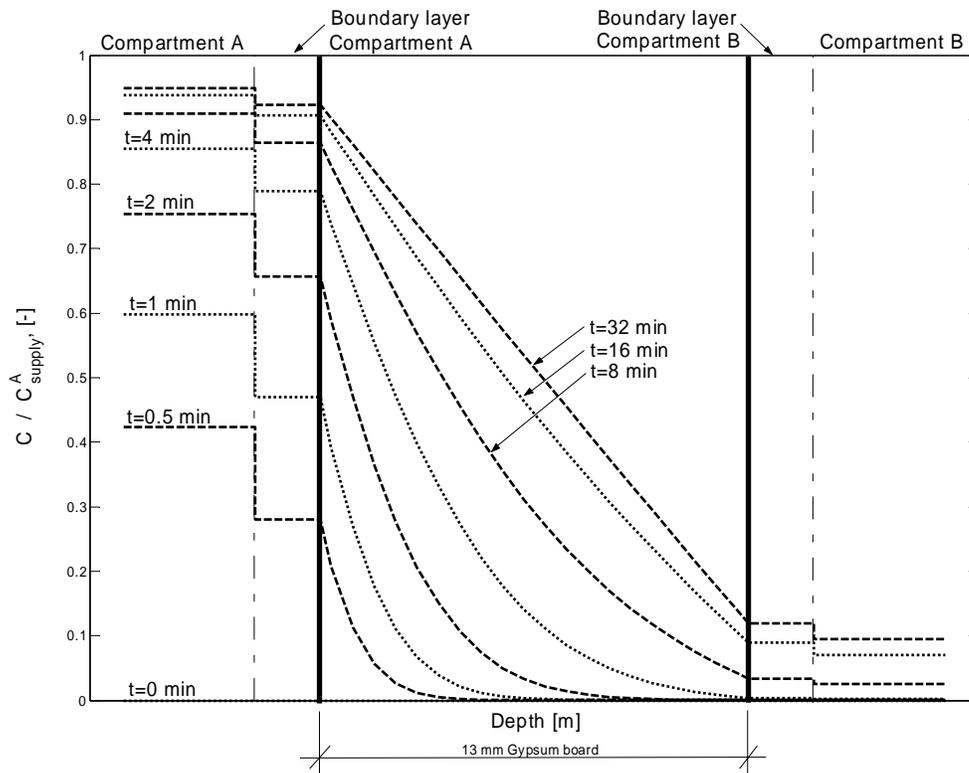


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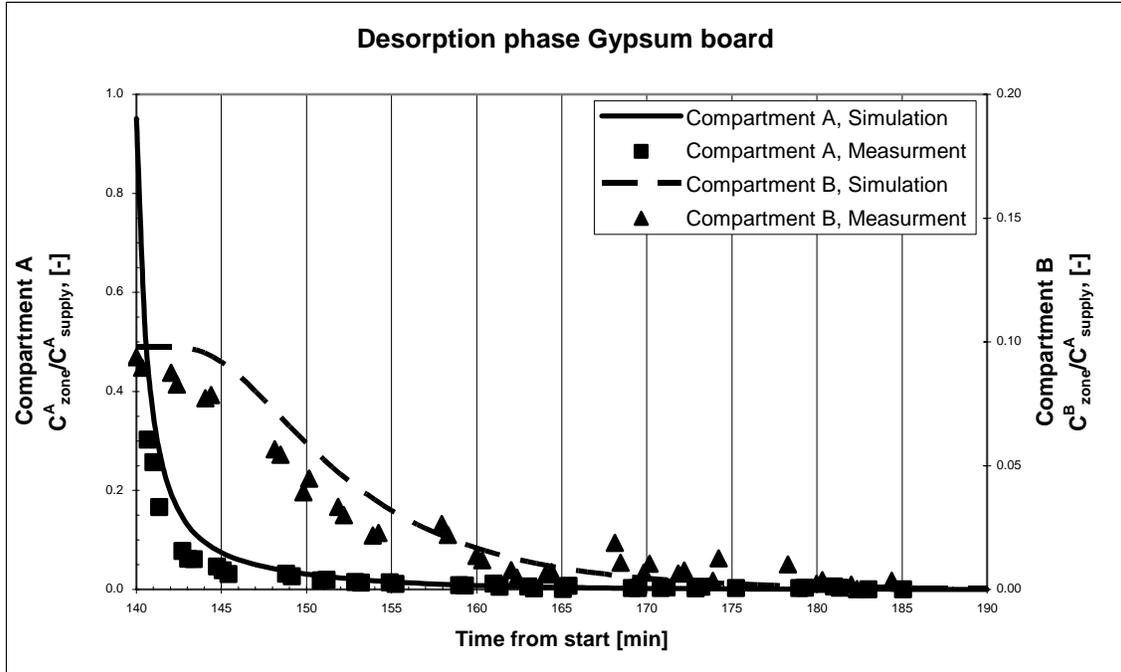


Figure 8
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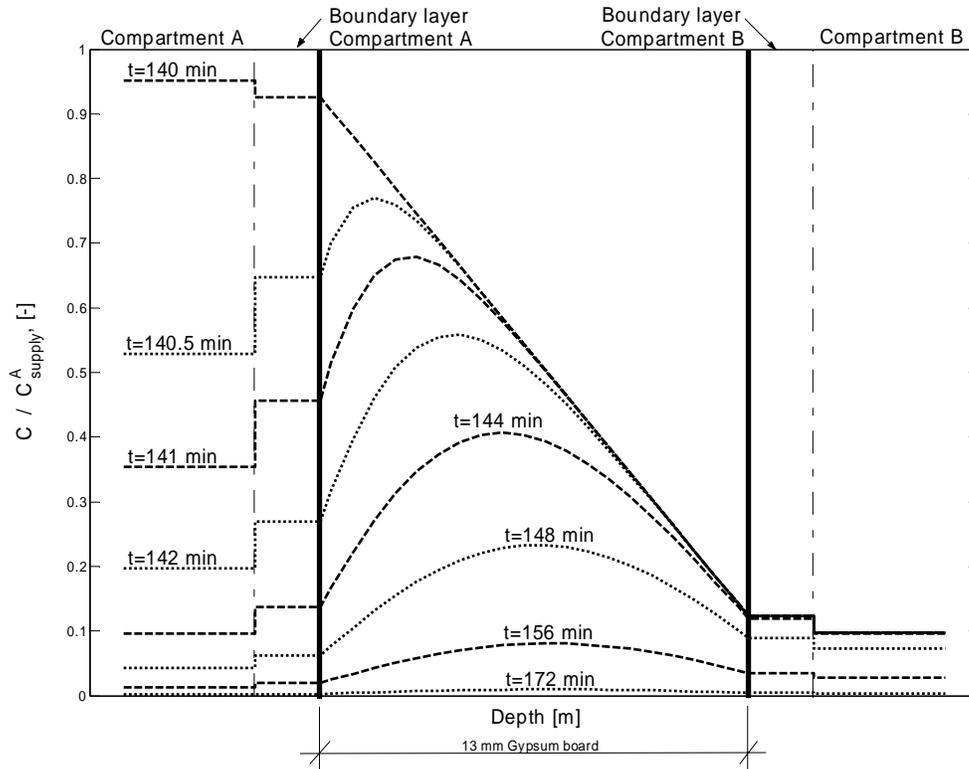


Figure 9
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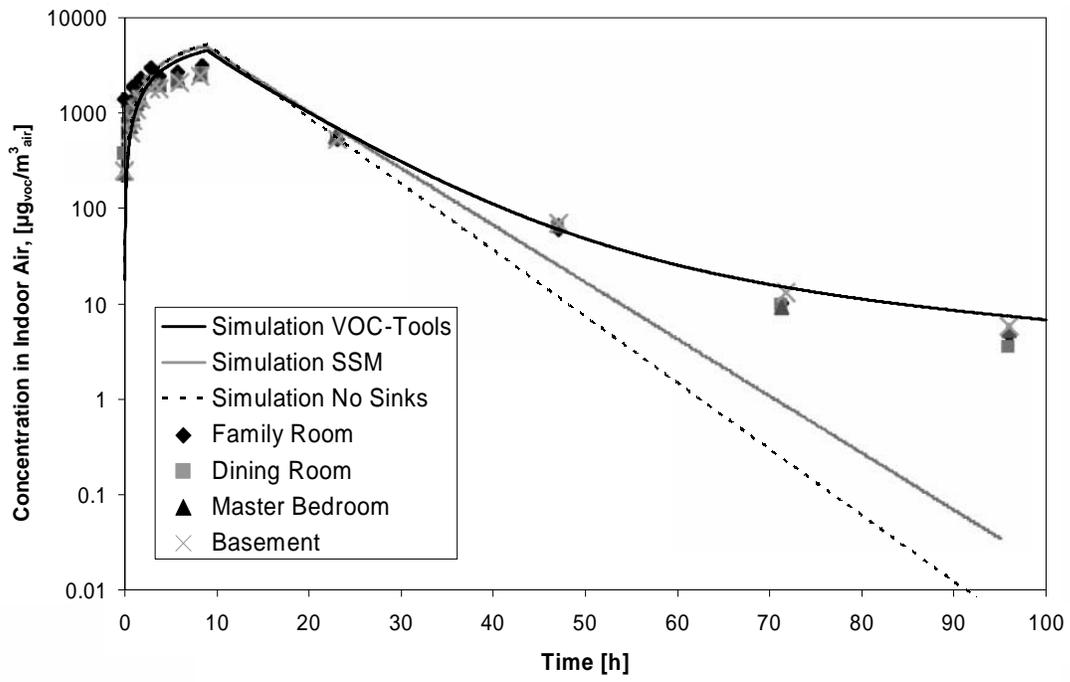


Figure 10
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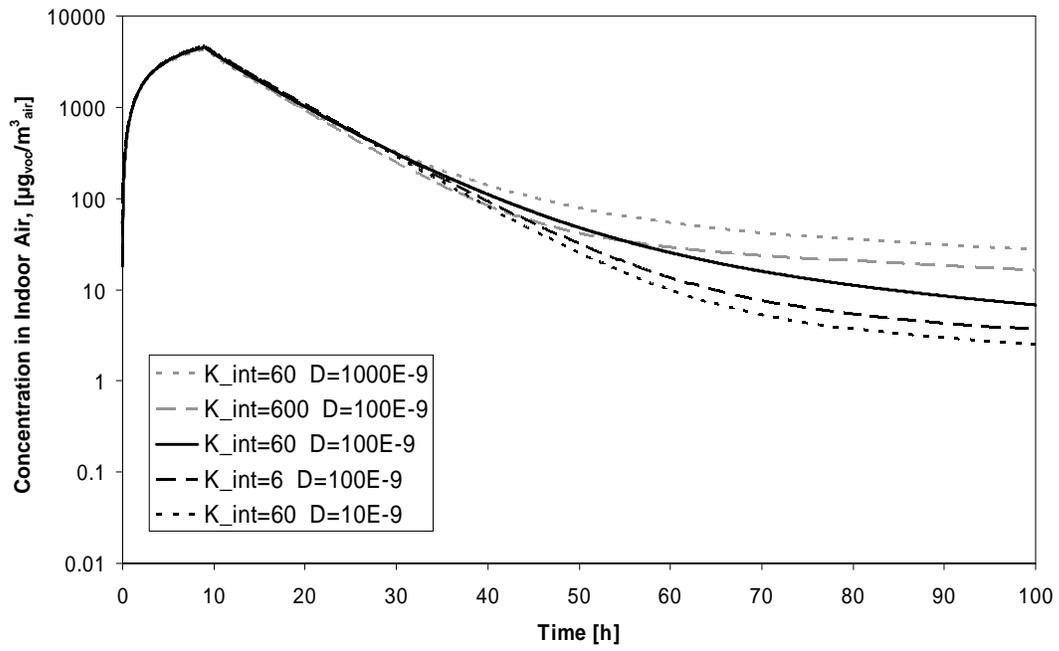


Figure 11
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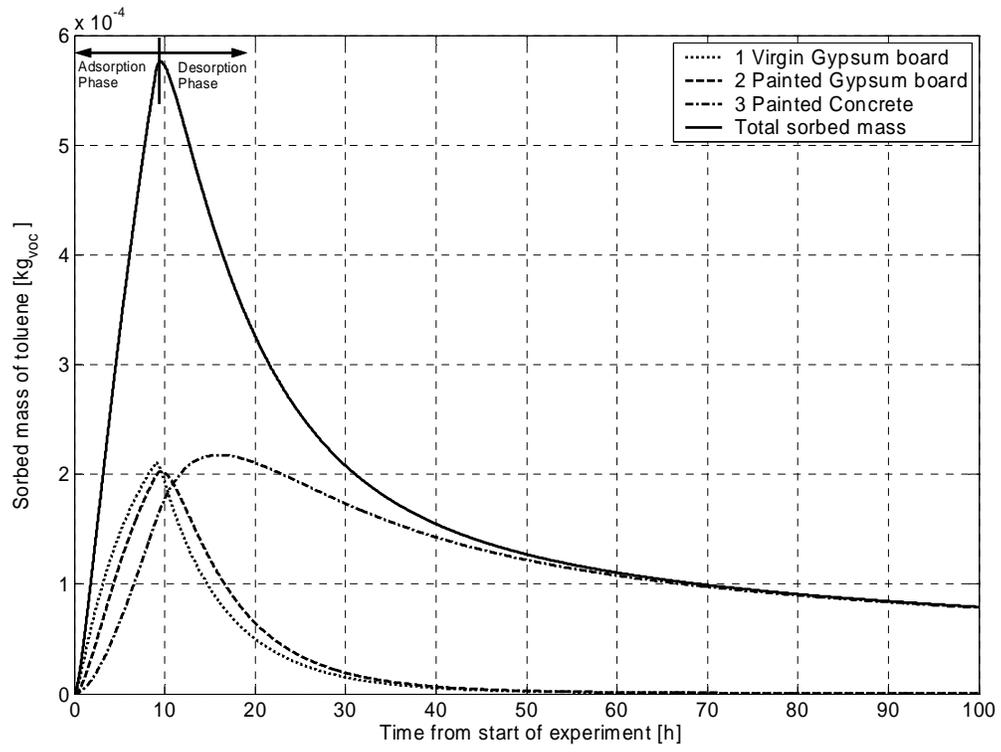


Figure 12
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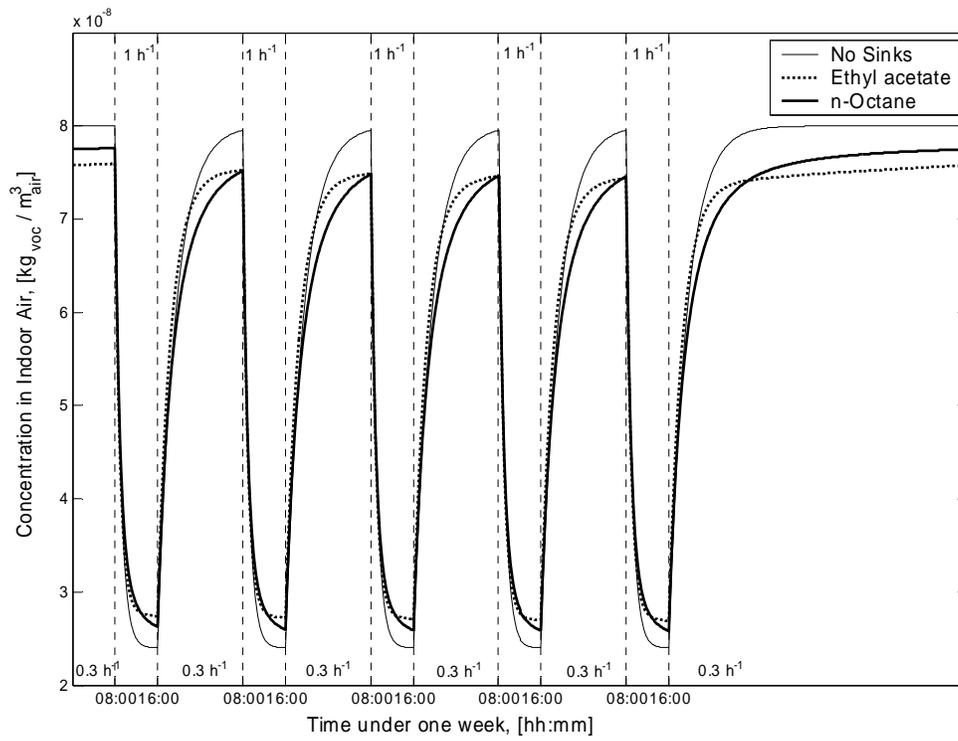


Figure 13
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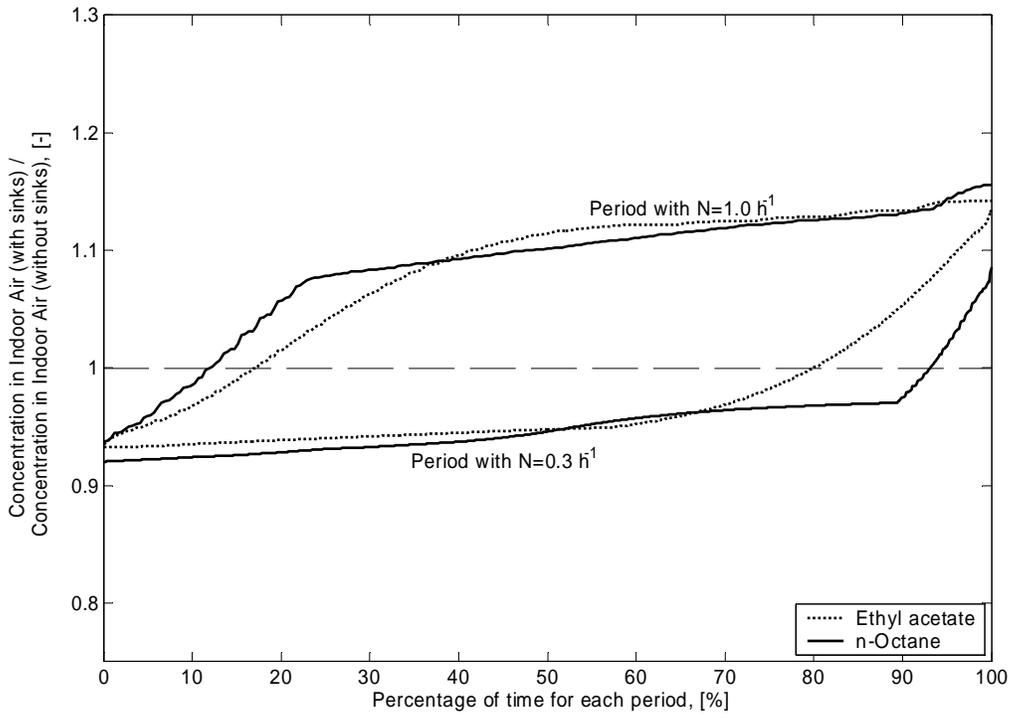


Figure 14
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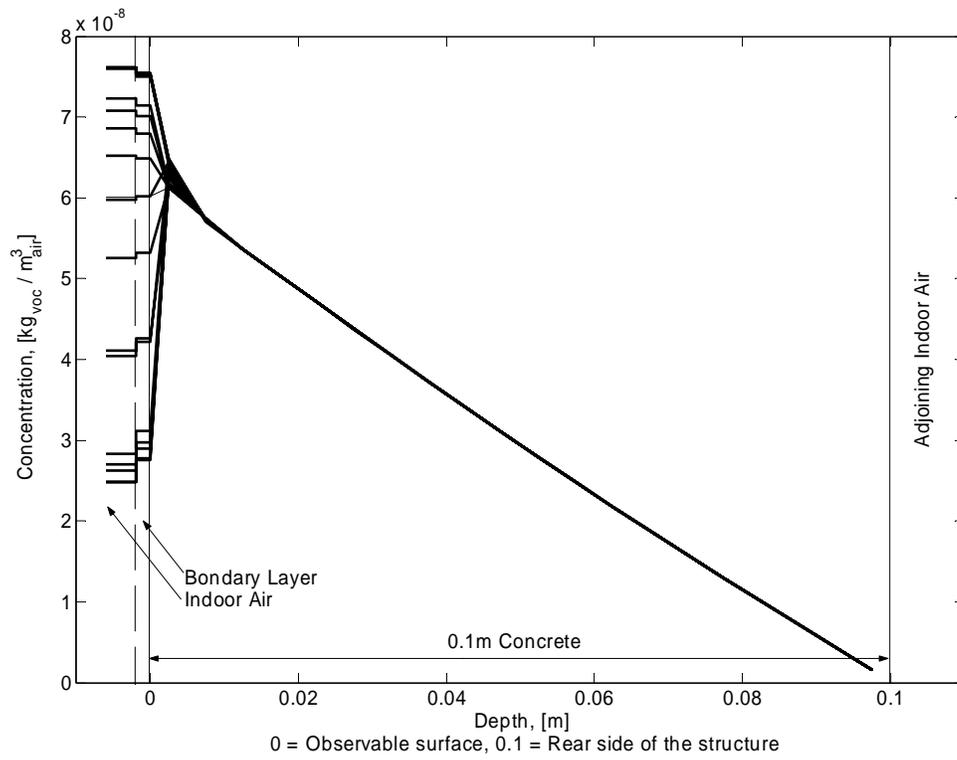


Figure 15
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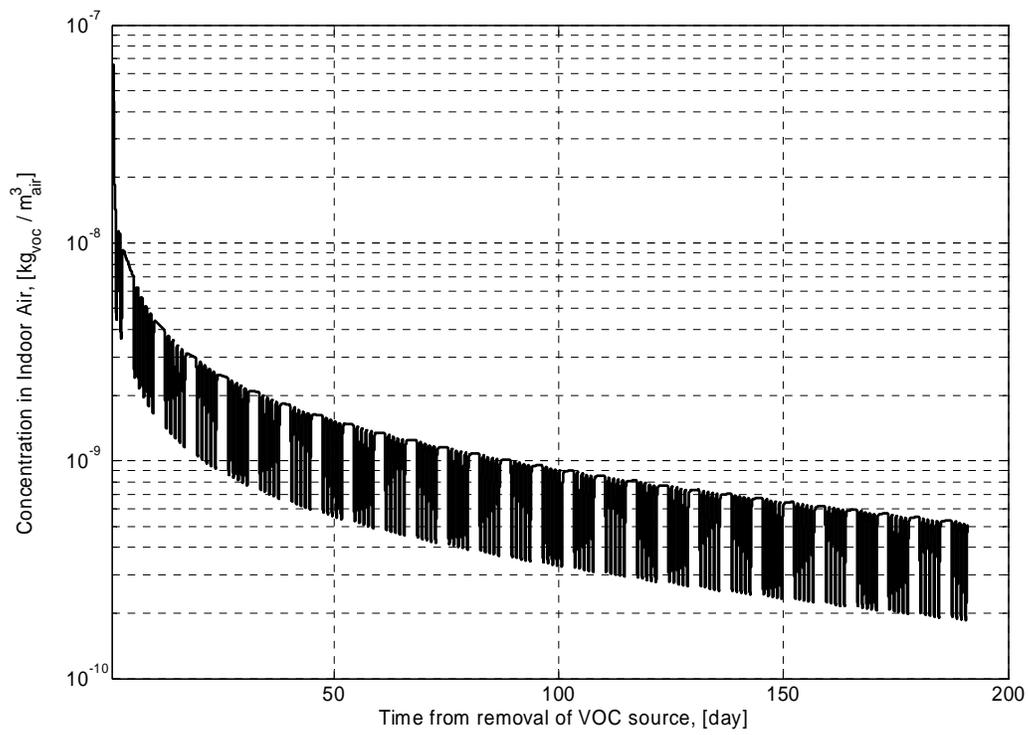


Figure 16
Henrik Karlsson

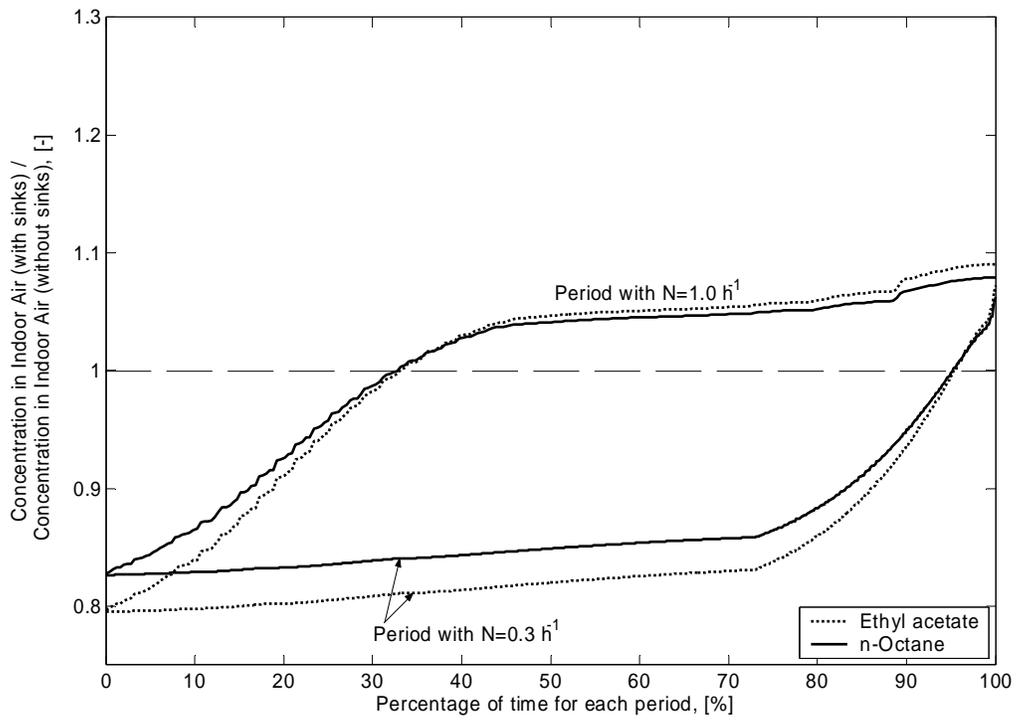


Figure 17
Henrik Karlsson

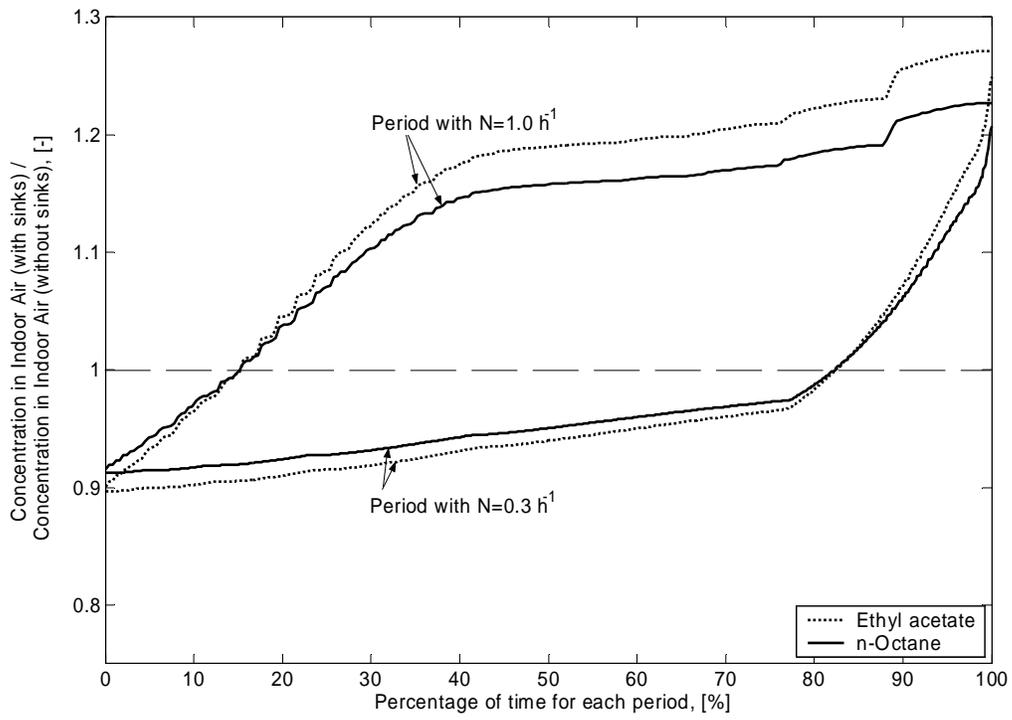


Figure 18
Henrik Karlsson

Figure captions

- Figure 1** The dynamic system illustrated together with the system border. All interactions between different parts of the system are illustrated with arrows.
- Figure 2** Discretization scheme inside a material.
- Figure 3** A network of calculation nodes together with the dividing of the network into block diagrams.
- Figure 4** VOC-Tools graphical environment. This figure also shows VOC-Tools five categories of block diagrams together with a complete set up of data arrays
- Figure 5** Sketch that illustrates the twin compartment.
- Figure 6** Compartment concentration for the adsorption phase, measurements and simulation from VOC-Tools.
- Figure 7** Simulated concentration distribution of toluene at the intersection from compartment A to compartment B under the adsorption phase. Each line represents a discrete point in time.
- Figure 8** Compartment concentration for the desorption phase, measurements and simulation from VOC-Tools.
- Figure 9** Simulated concentration distribution of toluene at the intersection from compartment A to compartment B under the desorption phase. Each line represents a discrete point in time.
- Figure 10** Comparison between simulated, both SSM and VOC-Tools, and measured levels of toluene in the indoor air.
- Figure 11** Sensitivity analysis of internal partition coefficient and diffusion coefficient in concrete. Simulated resulting toluene concentration in the indoor air, illustrated with different properties for concrete.
- Figure 12** Accumulated mass of sorbed toluene for each surface type and for the total structure of the house. The results are achieved from simulations with VOC-Tools.
- Figure 13** Concentration of Ethyl acetate and n-Octane in the indoor air under one week. The case with no sink is also included in order to quantify the sink effect.
- Figure 14** Duration curves that illustrates the concentration in the indoor air compared to the case with no sinks, for Ethyl acetate and n-Octane with structure 1 (0.1 m solid concrete).
- Figure 15** Concentration profiles for Ethyl acetate across the wall of solid concrete under one day (Monday week 50), each line represents the state in a discrete point in time (one hour between each line).
- Figure 16** Ethyl acetate concentration in the indoor air after removal of the source at time 0.
- Figure 17** Duration curves that illustrates the concentration in the indoor air compared to the case with no sinks, for Ethyl acetate and n-Octane with structure two (Carpet, 25 mm gypsum board, 10 cm air, 25 mm gypsum board and carpet).
- Figure 18** Duration curves that illustrates the concentration in the indoor air compared to the case with no sinks, for Ethyl acetate and n-Octane with structure three (Carpet, 25 mm gypsum board and vapor barrier).

Table 1 Specification for data arrays in VOC-Tools.

Data array	Dimension [Row, Column]	Specification, $(x)^{[Row, Column]}$			
Zone	[1, Gasnr]	$C_{zone}^{[1,1]}$	$C_{zone}^{[1,2]}$	→	$C_{zone}^{[1, Gasnr]}$
Construction	[2, Gasnr]	$C_s^{[1,1]}$	$C_s^{[1,2]}$	→	$C_s^{[1, Gasnr]}$
		$\beta_{voc}^{[2,1]}$	$\beta_{voc}^{[2,2]}$	→	$\beta_{voc}^{[2, Gasnr]}$
Gain	[1, Gasnr]	$J_{gain}^{[1,1]}$	$J_{gain}^{[1,2]}$	→	$J_{gain}^{[1, Gasnr]}$
System	[2, Gasnr]	$R_A^{[1,1]}$	$R_A^{[1,2]}$	→	$R_A^{[1, Gasnr]}$
		$C_{supply}^{[2,1]}$	$C_{supply}^{[2,2]}$	→	$C_{supply}^{[2, Gasnr]}$
Node	[2, Gasnr]	$C_{int}^{[1,1]}$	$C_{int}^{[1,2]}$	→	$C_{int}^{[1, Gasnr]}$
		$R^{[2,1]}$	$R^{[2,2]}$	→	$R^{[2, Gasnr]}$

Table 2 Definition of the set of fields for one specific material in VOC-Tools database for materials.

Field	Notation	Unit
name	-	-
partition_coefficient_interior	K_{int}	(-)
diffusion_VOC	D	(m ² /s)
partition_coefficient_surface	K_s	(m)
surf_mtrl_resistance	R_s	(s/m)

Table 3 Basic conditions for the twin compartment (Hansson, 2003).

Property	Notation	Unit	Compartment A	Compartment B
Air exchange rate	N	(h ⁻¹)	5806	2903
Volume	V	(m ³)	62x10 ⁻⁶	62x10 ⁻⁶
Concentration in Supply air, (adsorption phase)	C _{supply}	(kg _{voc} /m ³ _{air})	28.3x10 ⁻⁶	0
Mass transfer coefficient	β _{voc}	(m/s)	3x10 ⁻³	3x10 ⁻³

Table 4 Dimensions and physical properties for the gypsum board (Hansson, 2003).

Property	Notation	Unit	Value
Area	A	(m ²)	1.075x10 ⁻³
Thickness	d	(m)	0.013
Partition coefficient interior	K _{int}	(-)	20
Partition coefficient surface	K _s	(m)	0.0182
Diffusion coefficient	D	(m ² /s)	1.23x10 ⁻⁶

Table 5 Area and boundary condition for internal surface types.

Internal surface types	A (m ²)	β^*_{voc} (m/s)
1 Virgin gypsum board	170	377×10^{-6}
2 Painted gypsum board	150	377×10^{-6}
3 Painted concrete	145	377×10^{-6}

* Mass transfer coefficients are valid for the observable surface of the material. Given coefficients are calculated according to Axley (1991). Boundary condition and air properties: Diffusion coefficient for toluene in still air, $D_a=8.3 \cdot 10^{-6}$ (m²/s) (Hansson, 2003); Air velocity parallel to the surface (laminar flow), $U_\infty=0.1$ (m/s); Characteristic length of the surface, $L=2$ (m); Kinematic viscosity for air (20 °C), $\nu=15 \cdot 10^{-6}$ (m²/s)

Table 6 Specification of material properties (in combination with toluene) for the most frequent materials inside the test house.

Material	K_{int} (-)	K_s (m)	D (m^2/s)
Virgin gypsum board	20	0.0182	1580×10^{-9}
Acrylic paint	18	0.0021	3.83×10^{-9}
Concrete*	60	-	100×10^{-9}

* Properties for concrete/toluene are based on data for concrete/n-Octane (Meininghaus, 1999).

Table 7 Specification of material properties for materials in the model room.

	n-Octane			Ethyl acetate		
	$K_{int} (-)^1$	$K_s (m)^2$	$D (m^2/s)^3$	$K_{int} (-)^1$	$K_s (m)^2$	$D (m^2/s)^3$
Gypsum board	97.6	0.0182	1280×10^{-9}	124	0.0182	1770×10^{-9}
Solid concrete	59.4	0.0182	108×10^{-9}	1130	0.0182	51.3×10^{-9}
Carpet + Paste	-	0.0182	7.64×10^{-9}	-	0.0182	10.0×10^{-9}

¹ Properties obtained from Meininghaus (1999)

² Properties obtained from Hansson (2003)

³ Properties obtained from Haghghat et al. (2002)