

A Critical Review on Studies of Volatile Organic Compound (VOC) Sorption by Building Materials (RP-1097)

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ABSTRACT

The purpose of the investigation is to analyze the pros and cons of existing sorption models, as well as experimental methods. In addition, it summarizes existing sorption data in order to understand the phenomenon of VOC sorption on building materials better and to direct future research. We found that existing sorption models can be generally classified into either first order adsorption/desorption rate models or equilibrium-interface models. The adsorption/desorption rate models are simple and easy to use, but the model parameters have to be obtained from experimental data by curve fitting. The parameters of the equilibrium-interface models can be obtained through independent measurements, but lacking material property data and high computing costs hinder their applications. The sorption of VOCs on building materials is usually measured by the widely used small-scale chamber test method. The sink strength can be affected by the chemical properties of VOCs, physical properties of materials, and environmental conditions, such as temperature, humidity and air velocity.

KEYWORDS: VOC, sorption, modeling, diffusion, indoor air quality

INTRODUCTION

Indoor air pollution has been identified as one of the top environmental risks to the health of building dwellers (U.S. EPA 1987, 1995). The high risk from exposure to indoor air pollution reflects the elevated concentrations of indoor contaminants, the large number of people exposed to indoor air pollution, and the amount of time spent indoors (U.S. EPA 1995). Our building interiors, once thought of as providing safe havens from the pernicious effects of outdoor air pollution and harsh climates, may actually be more polluted than the surrounding ambient environment (Spengler and Chen, 2000).

Volatile organic compounds (VOCs) constitute an important class of indoor-air contaminants. Building materials, which have been developed mainly from the viewpoint of construction and energy conservation, have been acknowledged as major VOC sources indoors. Evidence from a variety of investigations and systematic studies suggests that building materials can also affect the transport and the removal of indoor VOCs by sorption and desorption. The re-emission of sorbed VOCs can dramatically elevate VOC concentrations in the indoor environment for months or years after a source event (Tichenor et al. 1988; Berglund et al. 1988; Sparks et al. 1994). Materials capable of depositing, adsorbing, and/or accumulating pollutants can influence indoor air quality (IAQ) during the entire service life of a building (Nielsen 1987). Therefore, an accurate characterization of the sorption by building materials and the sorption impact on IAQ is important. VOC sorption by building materials is a well-known, but poorly understood phenomenon. Though it can be detected by well-designed experiments, the design of indoor air quality needs a sorption model.

This paper will first review fundamental processes of sorption since it is crucial for understanding the complicated sorption by building material. The fundamental studies have led to the development of sorption models and experimental methods for modeling and measuring the sorption. The pros and cons of these models and methods will be discussed in the paper as well. A good sorption model and experimental method should take the influence of key parameters (such as the chemical properties of VOCs, physical properties of building materials and environmental parameters) on sorption into account. The paper will also review the importance of these parameters.

FUNDAMENTALS OF THE VOC SORPTION PROCESS AND SORPTION MODELS

Figure 1 shows three mass transfer regions associated with the VOC sorption by a piece of building material (Yang et al. 2001). First, the VOCs are transferred from the bulk air phase to the material-air interface. At the interface, VOCs in both the air phase and material phase are exchanged dynamically. Such a process may involve physisorption and chemisorption (Clark 1997). In physisorption, the adsorbate (the adsorbing VOC molecule) is

bound to the adsorbent (the solid surface) by intermolecular van der Waals forces or electrostatic forces. Chemisorption is thought to result from the formation of a chemical bond between the adsorbent and the adsorbate. Generally, physisorption is considered a relatively reversible process while chemisorption is not. Finally, for a permeable material, the VOCs at the surface of the material can also diffuse into the interior of the material.

Due to the complex nature of the sorption process, not all the sorption mechanisms are readily modeled. Frequently, the sorption process is assumed to be totally reversible and only the mechanism that dominates the sorption is considered. Different mathematical models have been developed based on different assumptions used.

Some researchers view surface sorption as the dominant mechanism. Two distinct processes, adsorption (whereby the VOCs are adhered onto the material surface) and desorption (whereby the VOCs are released from the material surface to the air), are assumed to occur simultaneously. The adsorption rate and desorption rate are assumed to be respectively proportional (i.e., first order) with the VOC concentration in the air and the VOC mass in the material surface. This type of models is called *first order adsorption/desorption rate models*. In these models, VOC diffusion inside the material may or may not be considered.

Other researchers assume that an equilibrium condition always exists at the material-air interface, and the sorption rate is governed by the relatively slow diffusion process within the material, which can be modeled by well-established mass transfer principles based on Fick's law. For the interfacial mass transfer, a constant coefficient (partition coefficient) is used to represent the ratio between the interfacial VOC concentrations for the two phases. Thus, this type of models is called *equilibrium-interface models*.

Figure 2 shows schematically the approaches used by the first order adsorption/desorption rate models and equilibrium-interface models. Table 1 compares some typical models in terms of governing equations, major assumptions used, determination of model parameters, and applicability for IAQ prediction. The development of both types of models and their strengths and weaknesses are now discussed.

First order adsorption/desorption rate models

The linear Langmuir model (Tichenor et al. 1991) is probably the most widely used sorption model. The following equation is used to represent the dynamic adsorption and desorption process at the interface:

$$dM/dt = k_a C_a - k_d M \quad (1)$$

Where dM/dt is the net mass rate of change of VOC adsorbed on the material surface, $k_a C_a$ represents the VOC adsorption rate onto the material surface, and $k_d M$ represents the VOC desorption rate from the material surface. An equilibrium coefficient, $k_c (=k_a/k_d)$, is defined to represent the sink strength of the material at equilibrium (i.e., $dM/dt=0$). Since the adsorption and desorption process occur simultaneously, it is very difficult to measure directly the adsorption coefficient (k_a) and the desorption coefficient (k_d). They are usually determined by fitting the model to experimental data. For example, combining Equation (1) with the mass balance equation for the small-scale chamber test, Tichenor et al. (1991) obtained the following solution, which can be fitted to the test data during the desorption period to determine k_a and k_d .

$$C(t) = C_e \frac{(N - r_2)e^{-r_1 t} - (N - r_1)e^{-r_2 t}}{r_1 - r_2} \quad (2)$$

One limitation to the linear Langmuir model is that it considers only the relatively fast surface sorption process while ignoring the relatively slow in-material diffusion, which may be significant for complex materials, such as carpet (Tichenor et al. 1991). Some empirical models have been developed to overcome this problem. For example, the two-sink model (Colombo et al. 1993a,b; De Bortoli et al. 1996) assumes that the material is composed of a fast sink (the adsorption and desorption coefficients are represented by k_3 and k_4 .) and a slow sink (the adsorption and desorption coefficients are represented by k_5 and k_6 as shown in Table 1.). The use of the slow sink, which adds two more parameters in the model, makes it fit the long-term sorption data better. Similar to the two-sink model, the K-diffusion model (Dunn et al. 1993) and the sorption-diffusion hybrid model (Dunn et al. 1993) have been proposed to explicitly represent the in-material diffusion. The difference between the K-diffusion model and the sorption-diffusion hybrid model is that the former considers only the in-material diffusion while the latter considers both the surface sorption and the in-material diffusion. The consideration of the in-material diffusion may help the first order adsorption/desorption rate models predict the long-term sorption process better, but it also needs more model parameters.

Another major concern regarding the use of first order adsorption/desorption rate models lies in their statistical nature in determining model parameters. Two or more model parameters have to be obtained by fitting the model to experimental data using a nonlinear regression method. It has been shown that with different initial values of model

parameters, the nonlinear regression might lead to multiple solutions (An et al. 1997; De Bortoli et al. 1996). In the worst-case scenario, the values of model parameters may differ as much as 100 times in two sets of solutions due to different initial values of model parameters when the same set of data are used in the regression. The model parameters obtained thereby may not be reliable.

Equilibrium-interface models

Equilibrium-interface models consider that the time scale for the surface sorption at the material-air interface to reach equilibrium is much smaller compared with the time scale for the VOC diffusion process within the material; therefore, the interface is assumed to be always at equilibrium. Considering low VOC concentrations indoors, Henry's law can be applied to the surface sorption at the material-air interface:

$$C_m = K_{ma} C_a^* \quad (3)$$

Based on Equation (3), Axley (1991) developed the simplest equilibrium-interface model in which a uniform concentration in both the air (C_a^*) and the material phase (C_m) was assumed. The model uses only one parameter, partition coefficient K_{ma} , and it only gives a rough idea about the sorption capacity of the material because the assumption of the uniform air phase concentration (C_a^*) and uniform material phase concentration (C_m) cannot be met under most situations. To improve the model, Axley (1991) further developed the boundary layer diffusion model to consider the VOC transport through the air boundary layer above the material surface. The mass transfer coefficient of VOC across the boundary layer can be obtained from the following empirical correlations (Axley 1991):

$$Sh_L = h_m L / D_a = 0.664 Re_L^{1/2} Sc^{1/3} \quad (Re_L < 500,000) \quad (4)$$

$$Sh_L = h_m L / D_a = 0.037 Re_L^{4/5} Sc^{1/3} \quad (Re_L > 500,000) \quad (5)$$

However, VOC diffusion in the material is sometimes more important than VOC mass transfer across the boundary layer. Therefore, the diffusion-controlled sink model (Little et al. 1996) has been proposed to consider the diffusion process inside the material. The model combines the equilibrium-interface sorption with the in-material diffusion described by Fick's law. The governing equations of the "diffusion-controlled sorption model" are listed in Table 1. Little et al. (1996) also gave the following analytical solution for analyzing the VOC concentration within a small-scale chamber with the presence of a sorptive material:

$$C_a(t) = 2\alpha D_m \sum_{n=1}^{\infty} \left\{ \frac{\exp(-D_m \lambda_n^2 t) (\lambda_n^2)}{[L(\alpha - \beta \lambda_n^2)^2 + \lambda_n^2 (L + \beta) + \alpha] \cos(\lambda_n L)} \cdot \int_0^t \exp(D_m \lambda_n^2 \tau) \cdot C_{in}(\tau) d\tau \right\} \quad (6)$$

The parameters used by the model are the physical properties of materials (partition coefficient and diffusion coefficient). Such property data may be obtained by independent measurements (Cox et al. 2000; Zhao et al. 1999; Bodalal et al. 1999). In addition, Yang et al. (2001) has developed a more comprehensive numerical model that considers VOC transport in the air, surface sorption, and diffusion in the material. This numerical model can be used for detailed simulation of sorption and IAQ in buildings.

Compared to the first order adsorption/desorption rate models, the parameters used in equilibrium-interface models have more distinct physical meanings and can be obtained independently. This allows the possibility of validating a sorption model rigorously, without resorting to the curve fitting routine as used by first order adsorption/desorption rate models. This is important because in order to evaluate a sorption model, the experimental data used for validation should be different from those used for developing the model. Otherwise, the results would show only how good the curve fitting rather than validating the model.

On the other hand, at present the material property data for a large number of materials are still not available. Furthermore, equilibrium-interface models tend to be more complex and computationally intensive compared to first order adsorption/desorption models. Their applicability to a wide range of materials is still yet to be validated. To validate different sorption models, different experimental methods have been used. The following will discuss different experimental methods in detail.

EXPERIMENTAL METHODS

The sink effect of building materials can be directly detected by well-designed experiments. The experimental data are also used for developing or validating sorption models. Thus, the development of experimental methods is closely related to that of sorption models. In this section, we will first discuss the widely used small-scale test chamber method, then other experimental methods, especially the latest research efforts in the direct measurement of partition coefficient and diffusion coefficient required by the equilibrium-interface models.

Environmental chamber test method

A small-scale stainless steel or glass chamber (such as the one recommended by the ASTM, 1997), or a full-scale stainless steel chamber (such as the one constructed in US EPA, NRC in Canada or CSIRO in Australia) can be used to measure the sorption effect of a material specimen. The test is usually conducted under a set of specific environmental conditions (e.g., 23°C, 50% relative humidity, and a ventilation rate of one air change per hour). The experiment usually consists of two periods: the dynamic adsorption period and the dynamic desorption period. Figure 3 shows an example of a small-scale chamber test system. During the dynamic adsorption period, compounds generated from the VOC generator (C) are carried by the conditioned, clean air from the conditioner (airflow, temperature and humidity controller, B) to the chamber containing the test specimen. The VOC concentration in the chamber is obtained by analyzing air samples taken from the chamber exhaust (E). After the system reaches an apparent equilibrium (the concentration at the chamber exhaust does not increase any more), the dynamic desorption period starts whereby the VOC supply is stopped while the chamber is continuously flushed out by the clean air. When no sinks are present in the chamber, the VOC concentration for the chamber air can be described by the following mass conservation equation (assuming perfect air mixing inside the chamber):

$$V \frac{dC_a}{dt} = Q(C_{in} - C_a) \quad (7)$$

However, because of the sorption of VOC by the test specimen, the actual VOC concentration in the chamber will differ from the no-sink curve represented by Equation (7). The difference between the measured concentrations and the no-sink curve indicates the sorption by the test specimen. Figure 4 shows an example of measured sorption effect by a carpet sample using a small-scale chamber (An et al. 1999).

Other than the dynamic adsorption tests and the dynamic desorption tests, the static adsorption tests have also been used (Colombo et al. 1993a,b; Kephelopoulos et al. 1996; De Bortoli et al. 1996). For a static adsorption test, a known quantity of VOC is injected into the clean chamber-specimen system without ventilation. The VOCs in the chamber will be gradually adsorbed by the material until a steady state is reached. The sink effect of the material can be determined by measuring the decay of the VOC concentration in the chamber.

In addition to using the small-scale chambers, the FLEC (Field and Laboratory Emission Cell)-technique has also been used for sorption tests (Tirkkonen et al. 1997 and Hansson et al. 1998). The FLEC, originally developed by Wolkoff et al. (1993), has a volume of 35 ml and a test surface area of 177 cm². The principles and procedures of the FLEC test method are the same as that of the small chamber method. The only difference is that the FLEC can be used in the field by attaching the test material directly to the FLEC without cutting the test material. Therefore, the FLEC method is convenient to use; however, the loading factor (defined as the ratio of the effective surface area of the material to the volume of the test chamber) in the FLEC method is very high, about 500m²/m³, compared with the loading factor of 1-2 m²/m³ in the small chamber method. Furthermore, the geometry of the FLEC may severely disturb the boundary layer, which in turn will affect the mass transfer between the bulk air and the material surface (Uhde et al. 1998). Therefore, the applicability and the accuracy of the FLEC method should be further examined.

The principle of the environmental chamber test method is relatively simple. Several VOCs can be tested at the same time if there are no chemical reactions among them. However, the method does not differentiate the distinction of the physical processes involved (i.e. the surface sorption and/or the internal diffusion) (Meininghaus et al. 1999). Thus, the test data cannot be used directly to determine the model parameters of equilibrium-interface models (e.g., partition coefficient and diffusion coefficient).

Methods for measuring partition and diffusion coefficients

Many efforts have been made to directly measure the VOC diffusion coefficient and partition coefficient of materials as required by equilibrium-interface models. Borazzo and Davidson (1990) measured the partition coefficient of fiber material with the use of a “packed column” method. The basic principle of the method is that the retention time of a VOC through a column packed with the test specimen is directly related with its partition coefficient. Therefore, by measuring the retention time of a specific VOC, its partition coefficient can be calculated. The limitation of this method is that material is required to be cut into small pieces to put into the column, which may significantly change the material structure. Bodalal et al. (1999) developed another method to measure the partition coefficient and diffusion coefficient of dry materials. Their method requires a specially designed “diffusimeter” consisting of two stainless steel chambers with the test material in between the two chambers. At the beginning of the test, certain amount of VOCs are injected into one chamber while allowing the VOCs in that chamber to diffuse through the material into the other chamber. The diffusion coefficient and partition coefficient of the material can be inferred from the monitored concentration versus time profiles in both chambers. Fick’s law is used to solve the diffusion through the material.

Researchers have also used a microbalance to measure the material properties (Kirchner et al. 1996; Zhao et al. 1999). In such a method, the material specimen is placed on a microbalance inside a small-scale chamber. The chamber is then flushed by an inert gas containing a known concentration of VOCs. The sorption is measured through recording the weight increase of the test specimen as a function of time. The advantage of the microbalance test method is that no VOC sampling of the chamber air is necessary. This may significantly reduce the labor costs as well as sampling errors because auto-recording of the weight over time profile is available. However, due to the limited resolution of the microbalances (about 10^{-7} - 10^{-5} g), the experiments can only be done under VOC concentration much higher than that in indoor air. Otherwise, the change in the weight of the specimen will be too small to be detected. Another disadvantage is that the material sample has to be cut into very small pieces (< 1mm in thickness) to fit into the range of microbalance. This may significantly affect the surface characteristics of the test materials. In addition, only a single compound can be tested using the microbalance method.

Other than the methods mentioned above, simpler procedures for measuring the diffusion coefficient of indoor materials have also been developed. The cup method (Kirchner et al. 1999) and CLIMPAQ method (Kirchner et al., 1999) are two examples of the simpler procedures. For the cup method, a cup containing saturated VOC liquid and vapor is covered by the test specimen to allow VOC diffusion through the material driven by constant concentration gradient. By measuring the rate of weight loss of VOC inside the cup (essentially the steady VOC diffusion rate across the material), the diffusion coefficient of the material can be obtained by applying Fick's law. However, it should be noted that the partition coefficient was not considered in the cup method. The CLIMPAQ method is similar to the cup method except that the two CLIMPAQ chambers are used to create a constant concentration difference on two sides of the material.

With the experimental methods discussed above, many VOCs have been tested for their sink effects on different building materials and it was found that the sink strength is affected by many parameters. To better direct the development of future sorption models, the following will discuss the available sorption data and the effects of key parameters on sorption in detail.

EXISTING SORPTION DATA AND THE EFFECTS OF KEY PARAMETERS ON SORPTION

Over 40 VOCs have been tested for their sink effects on different building materials. Example materials tested include ceiling tile, wallboard, latex caulk, blown vinyl, polyurethane foam, cotton curtain, wool carpet, vinyl carpet and its pad, etc. Table 2 lists typical sorption data from literature, analyzed by the linear Langmuir model. Table 3 lists some typical partition and diffusion coefficients from literature. Data listed in both tables (Table 2 and Table 3) give the normal ranges of the model parameters (the linear Langmuir model or the equilibrium-interface models), which can be used in validating an unknown model or can be used in the nonlinear regression process as a restriction of the parameters in order to obtain reasonable values for model parameters.

The experimental data, mostly analyzed by the linear Langmuir model, indicate that the sink strength (characterized by the equilibrium coefficient k_e defined in the linear Langmuir model) depends on many parameters including:

- Chemical properties of VOCs
- Physical properties of building materials
- VOC concentration
- Temperature
- Humidity
- Air velocity

The following section summarizes the results of previous studies on the effects of the above key factors on sorption.

Effects of chemical properties of VOCs

Physiosorption, a relatively reversible process, has been assumed as the dominant mechanism in the surface sorption process. Physiosorption at the material-air interface results from van der Waals interactions including: (a) dipole/dipole interactions between polar molecules, (b) dipole/induced-dipole interactions between polar and non-polar molecules, and (c) induced-dipole/induced-dipole interactions between non-polar molecules. The strength of the interactions decreases from type (a) to type (c). The polarity of the compounds used to describe the interaction

strength between molecules is a key factor during adsorption-desorption processes. Therefore, the molecular polarities are usually taken into account when characterizing sink effects in the indoor environment. However, the polarities of VOCs are not quantitatively available. They are commonly represented by the vapor pressure or the boiling point temperature of the compounds. Polar compounds have relatively higher boiling point temperatures or lower vapor pressures than corresponding non-polar compounds (An et al. 1997).

The dependence of the sink strength on the vapor pressure or boiling point of a vapor compound has been frequently observed (Levsen et al. 1993; Chang et al. 1994; An et al. 1999; Corsi et al. 1999). Figure 5 compares the relationship between the sink strength (represented by the equilibrium coefficient k_e) with the vapor pressure obtained from several researchers. The results from these studies show that the logarithm of k_e decreases linearly with the increase of the logarithm vapor pressure. An exception, however, was given by Corsi et al. (1999) who found that a less volatile compound (1,2,4-trichlorobenzene) does not follow the linear trend. The reason for that was not explained.

Other chemical properties of VOCs, such as the oxidation status of the compounds, may also affect the sorption effect. Chang et al. (1998) found that the sink strength of carpet and gypsum board toward oxygenated polar compounds (propylene glycol, ethylene glycol, 2-butoxyethoxy ethanol, and Texanol) is orders of magnitude higher than those toward other polar and non-polar VOCs such as tetrachloroethylene, ethylene, and alkane species. The reason may be that the oxidation status of those oxygenated polar compounds plays a more important role in determining sink behavior than the polarity effect.

Effects of physical properties of building materials

Material surface roughness, essentially reflecting the efficient surface area, can strongly affect the sorption capacity of the material. The specific area (SA), defined as the efficient surface area per gram material (m^2/g), is usually used to quantify the material surface roughness. Kircher et al. (1996) measured the specific area of gypsum board and of acoustic tiles by using nitrogen adsorption technique. The results showed good agreement with the chamber experimental data, that is, the greater the specific area, the larger the equilibrium coefficient. However, the data on wall paper did not seem to fit well in this trend (Kephelopoulus et al., 1996). A given specific area should provide information on the VOC sorption capacity of the materials. However, the specific area value represents only the surface area information, whereas the VOC sorption capacity depends also on material porosity, pore size, diffusion rate, as well as the mass or the thickness of the materials. To overcome the limitations of the specific area, the “equivalent area” (EqA) (European Commission, 1997) was introduced as:

$$\text{EqA} = \text{specific area} \times \text{weight of the sample} \quad (8)$$

The equivalent area shows better agreement with the previous chamber test data than SA does, as indicated in Table 4.

Besides the specific area or the equivalent area of the material, the material loading factor in the test chamber (defined as the ratio of the surface area of the material to the volume of the test chamber) has also been studied regarding its impact on the sink strength. However, whether the impact of loading factor on the sorption strength is significant or not, is in fact, determined by the ratio of the specimen surface area to the inner surface area of the test chamber (approximately proportional to the loading ratio) because of the sink effect of the chamber walls. To minimize the impact of the loading factor on the test results, a higher loading factor is recommended (Kephelopoulus et al., 1996; Van Der Wal et al. 1998; Jorgensen et al., 1999).

Effects of VOC concentration in air

Tichenor et al. (1991) studied the sorption of tetrachloroethylene on carpet under different concentrations (from 5 mg/m^3 to 50 mg/m^3) and found that the calculated equilibrium coefficient (k_e) is independent of the concentrations tested in the chamber. Kephelopoulus et al. (1996) also obtained similar results when they investigated the sorption of n-decane on carpet under two largely different concentrations. Experiments with realistic indoor concentrations of VOCs, which were conducted by Jorgensen et al. (1999), also showed no significant difference between low ($300 \mu\text{g/m}^3$ toluene and $160 \mu\text{g/m}^3$ α -pinene) and high VOCs concentration (5g/m^3 , Tichenor et al. 1991). The conclusion has also been confirmed by Corsi et al. (1999), who studied the sorption of eight VOCs (methyl tert-butyl ether, cyclohexane, iso-propanol, toluene, tetrachloroethene, ethylbenzene, o-dichlorobenzene, and 1,2,4-trichlorobenzene) on the carpet under three different inlet concentrations (2.5, 5, and 15 ppm). All those studies seem to indicate that VOC concentration has no significant influence on the sorption capacity of a material-VOC combination. This suggests that sorption tests may be preferably conducted under relatively higher concentrations since high concentrations can be measured more accurately than low concentrations.

Effects of air temperature

Tichenor et al. (1991) examined the temperature effect on the sorption process. They found that the adsorption and desorption rate of tetrachloroethylene on the carpet at 35°C was significantly higher than those at 23°C. However, because the desorption rate increased more rapidly than the adsorption rate, the equilibrium coefficient decreased with the increase of temperature from 23°C to 35°C. Van der Wal et al. (1998) also found that M/C_0 (approximately proportional to k_e) was smaller at a higher temperature for most compounds tested (mesitylene, n-decane, n-undecane, tetrachloroethene, limonene ethylene glycol monobutylether, 2,2,4-trimethyl-1,3 pentanediol diisobutyrate). This confirms that the sorption was weaker under a higher temperature. However, toluene, m+p-xylene and 2-ethylhexanol were exceptions because of experimental or other unknown factors. In a study of VOC sorption by particles from a combustion source, Goss et al. (1997) used the following equation to describe the dependence of the sorption coefficient k_e on absolute temperature:

$$\ln k_e = (\Delta U_{des}/RT) + \text{const.} \quad (9)$$

Equation (9) also agrees with the trend suggested by Tichenor et al. (1991) and Van der Wal et al. (1998). That is, the equilibrium coefficient (k_e) decreases with the increase of temperature.

Effects of relative humidity

Goss et al. (1997) found decreased sorption strength with increased humidity level over the range of 10-95% relative humidity when they studied the sorption of VOCs (n-Hexane, n-Octane, benzene, toluene, o-Xylene, chlorobenzene, dichloromethane, chloroform, trichloroethylene, ethyl ether, acetone) by particles from a combustion source. They used the following equation to account for the effects of relative humidity on the sorption coefficient k_e (C_2 is usually negative):

$$\ln k_e = C_1 + C_2 \times RH \quad (10)$$

Corsi et al. (1999) found that the sorption of the eight VOCs (methyl tert-butyl ether, cyclohexane, iso-propanol, toluene, tetrachloroethene, ethylbenzene, o-dichlorobenzene, and 1,2,4-trichlorobenzene) on unpainted gypsum board decreased with the increase of the relative humidity among three relative humidity levels (0%, 50%, 80%). Kirchner et al. (1996) also found that the sorption strength of 2-butoxyethanol on the gypsum board is weaker under higher humidity levels when they tested the sorption of 2-butoxyethanol on the gypsum board under dry (0% relative humidity) and humid (40% relative humidity) conditions. The results suggest possible competitions for sorption sites between water vapor and VOCs.

However, an opposite trend was observed for some materials. For example, Kirchner et al. (1996) studied the influence of humidity on the sink effect by performing tests in a 1 m³ chamber with a wall covering exposed to 500 µg of 2-butoxyethanol. The tests were conducted under five different humidity levels (15%, 39%, 58%, 60%, 91%). The results indicate that for 2-butoxyethanol, the sink strength increased with the increase of the humidity ratio in the chamber. Corsi et al. (1999) also gave similar results when they found the sorption of a highly soluble compound (iso-propanol) on a carpet increased with the increase of relative humidity. The possible reason might be that 2-butoxyethanol or iso-propanol was absorbed by water that condensed on the surface of the wall covering or the carpet.

Interestingly, Corsi et al. (1999) found that for seven VOC (methyl tert-butyl ether, cyclohexane, toluene, tetrachloroethene, ethylbenzene, o-dichlorobenzene, and 1,2,4-trichlorobenzene) and material (carpet, vinyl and wood flooring, upholstery and fiberglass shower stall) combinations, no apparent impact of relative humidity on the sorption was observed. Similar results were given by Jorgensen et al. (1993) who found no measurable effect for the sorption of toluene and α -pinene on the wool carpet or the nylon carpet when the relative humidity varied from 20 to 50%.

The above review suggests that the effects of relative humidity on the sorption strength depend on the VOC-material combinations. For future studies, the specific VOC properties (e.g., solubility) and material properties (e.g., possible reaction with water) should be quantified and correlated with the sorption effect using an empirical or a theoretical relationship.

Effects of air velocity

Kjaer and Tirkkonen (1996) found that the air velocity had an influence on the desorption rate of a mixture of 17 compounds on a painted gypsum board. The higher the air velocity, the higher the desorption rate. Guo (1993) also reported an increase in both the adsorption coefficient and the desorption coefficient for the sorption of ethylbenzene on wallboard with the increase of the air movement inside the chamber. However, these results may not be generalized to other materials such as carpet and PVC. For example, Jorgensen et al. (1999) showed that the air velocity did not influence the sorption of α -pinene and toluene on wool carpet at air velocities of 0, 0.1 and 0.2

m/s (measured at 0.01 m above the material). For a chamber that has no special design for controlling air velocity over the material surface, the air velocity in the test chamber is directly related to the air change rate. Some researchers, therefore, controlled air change rate instead of air velocity. Corsi et al. (1999) did experiment under three different air change rates (0.5, 2.1, and 3.2/hr) and found no apparent effects of air change rate on sorption process of eight VOCs (methyl tert-butyl ether, cyclohexane, iso-propanol, toluene, tetrachloroethene, ethylbenzene, o-dichlorobenzene, and 1,2,4-trichlorobenzene) on carpet and its padding. From the mass transfer point of view, the question to be answered is when the air phase mass transfer becomes an influencing factor compared to other mechanisms. Further experimental and theoretical work is necessary to clarify this question.

The above review of sorption data shows that despite some progress, many questions still remain unanswered regarding the impacts of environmental conditions on the sorption process. Some conclusions based on one particular material may not be generalized to other materials. Therefore, current sorption models, either first order adsorption/desorption rate models or equilibrium-interface models, cannot consider the effects of the above key factors on the sorption process. More systematic studies are necessary to uncover the underlying mechanisms of the effects of those influencing factors on sorption.

CONCLUSIONS

1. The sorption models can be classified into first order adsorption/desorption rate models and equilibrium-interface models according to the different approaches used in describing the surface sorption. First order adsorption/desorption rate models assume that adsorption and desorption occur at the same time on the material surface and the adsorption rate is proportional (i.e., first order) to the concentration in air while the desorption rate is proportional (i.e., first order) to the mass adsorbed at the surface. They require that both adsorption and desorption rate coefficients be determined from the same set of experimental data by curve fitting, which may not be reliable. Equilibrium-interface models assume that the equilibrium condition always holds at the interface between the air and solid surface, while the rate of adsorption or desorption is solely governed by the mass transfer principles. The model parameters used by equilibrium-interface models have clear physical meanings compared to those used by the first order adsorption/desorption rate models, but their applications are currently hindered by lacking material property data and high computing costs.

2. The environmental chamber test method has been the most widely used method to study the VOC sorption by building materials. Its principle is simple. However, it can not differentiate the physical processes involved, e.g. surface sorption and/or in-material diffusion process, which limits the use of its data for estimating parameters of sorption models. Methods to measure the partition and diffusion coefficients have been proposed to overcome the limitations of environmental chamber tests.

3. Previous sorption tests, mainly conducted with the small-scale chamber and analyzed using the linear Langmuir model, indicate that the strength of the sorption effect is affected by many factors including the chemical properties of VOCs, physical properties of building materials, and environmental conditions such as temperature, humidity, and air velocity. The sink strength usually decreases with the increase of the vapor pressure of the VOC and decreases with the increase of the environmental temperature. However, the impacts of humidity and air velocity still remain inconclusive. Sorption models considering the impacts of the above key factors are still not available. Further studies are needed for improving experimental methods, obtaining more VOC sorption data from building materials, and developing better sorption models.

NOMENCLATURE

A	Specimen surface area (m ²)	M	VOC mass adsorbed per unit sink surface area (mg/m ²)
C _a	VOC concentration in the bulk air (mg/m ³)	N	Air change rate (h ⁻¹)
C _a *	Near-surface, air-phase concentration (mg/m ³)	Q	The supply air flow rate for the chamber (m ³ /h)
C _e	VOC equilibrium concentration in the chamber (mg/m ³)	R	Universal gas constant
C _{in}	VOC concentration of the supply air (mg/m ³)	r ₁	={(N+k _a L+k _d)+ [(N+k _a L+k _d) ² - 4Nk _d] ^{1/2} }/2
C ₀	Initial VOC concentration in the chamber before the desorption test (mg/m ³)	r ₂	={(N+k _a L+k _d)- [(N+k _a L+k _d) ² - 4Nk _d] ^{1/2} }/2
C ₁ , C ₂	Coefficients obtained by curve fitting	Re _L	= UL/v
C _m	VOC concentration in the sink material (mg/m ³)	RH	Relative humidity (%)
D _a	The molecular diffusivity of the air (m ² /s)	S _c	= v/D _a
D _m	Diffusion coefficient (m ² /s)	t	Time (hr)
h _m	The average film mass transfer coefficient over the adsorbent surface (m/h)	T	Temperature (K)
k ₃ , k ₅	Adsorption coefficient of the fast/slow sink (h ⁻¹)	U	Mean fluid velocity (m/s)
k ₄ , k ₆	Desorption coefficient of the fast/slow sink (h ⁻¹)	V	Chamber volume (m ³), excluding the volume of the test specimen
k _a	Adsorption coefficient (m/h)	W ₁ , W ₂	Mass of VOC adsorbed in the fast/slow sink (mg)
k _d	Desorption coefficient (h ⁻¹)	W(t), W _s	Mass of VOC adsorbed on the surface of the material at time t (mg)
k _e	Equilibrium coefficient, mg/m ² / mg/m ³	X	Position in the material (m), 0 means the surface of the material, L means the bottom of the material
λ _n ^S	Positive roots of λ _n tan(λ _n L) = α - βλ _n ²	ρ	Air density (g/m ³)
K _{ma}	Material-air partition coefficient (mg/m ³ of VOC in material/mg/m ³ of VOC in air)	ΔU _{des}	Internal energy of desorption
L	The thickness of the material (m)	α	= $\frac{Q}{AD_m K_{ma}}$
L	The length of the surface in the direction of the flow (m)	β	= $\frac{V}{AK_{ma}}$
		v	The kinematic viscosity of the air phase (m ² /s)

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Table 1. Comparison of typical VOC sorption models*

Models		Governing equations	Parameters	⌒	⌒	⌒	-		Reference
First order adsorption/desorption rate models	Linear Langmuir	$dM/dt = k_a C_a - k_d M$	k_a, k_d	N	Y	N	C-F	Y	Tichenor et al. 1991
	Two-sink model	$dW_1/dt = k_3 VC_a - k_4 W_1$ $dW_2/dt = k_5 VC_a - k_6 W_2$	k_3, k_4, k_5, k_6	N	Y	N	C-F	Y	Colombo et al. 1993a,b De Bortoli et al. 1996
	K-diffusion model	$\frac{\partial C_m(x, t)}{\partial t} = D_m \frac{\partial^2 C_m(x, t)}{\partial x^2}$ $C_m(+0, t) = C_a(t)$	D_m	N	N	Y	C-F	Y	Dunn et al. 1993
	Sorption-diffusion hybrid model	$\frac{\partial C_m(x, t)}{\partial t} = D_m \frac{\partial^2 C_m(x, t)}{\partial x^2}$ $\frac{dW(t)}{dt} = k_3 VC_a(t) + AD_m \left. \frac{\partial C_m(x, t)}{\partial x} \right _{x=+0} - k_4 W(t)$ $C_m(+0, t) = \frac{W(t)}{A}$	D_m, k_3, k_4	N	Y	Y	C-F	Y	Dunn et al. 1993
Equilibrium-interface models	Boundary layer diffusion model	$W_s = h_m \rho A (C_a - C_a^*)$ $C_m = K_{ma} C_a^*$	K_{ma}	Y	Y	N	M	Y	Axley 1991
	Diffusion-controlled sorption model	$\frac{\partial C_m(x, t)}{\partial t} = D_m \cdot \frac{\partial^2 C_m(x, t)}{\partial x^2}$ $C_m(0, t) = K_{ma} \cdot C_a^*(t)$ $\left. \frac{\partial C_m(x, t)}{\partial x} \right _{x=L} = 0$	D_m, K_{ma}	N	Y	Y	M	Y	Little et al. 1996
	Numerical model	$\frac{\partial C_m(x, t)}{\partial t} = D_m \cdot \frac{\partial^2 C_m(x, t)}{\partial x^2}$ $C_m(0, t) = K_{ma} \cdot C_a^*(t)$ $C_m(L, t)=0$	D_m, K_{ma}	Y	Y	Y	M	Y	Yang et al. 2001

⌒ Consideration of VOC transport in the air

⌒ Consideration of surface sorption

⌒ Consideration of VOC diffusion in the material

- Determination of model parameters: C-F means by curve fitting. M means by direct measurements

Application for IAQ prediction and coupling with CFD code

* See nomenclature for notation

Table 2. Selected sorption data from literature (analyzed by the linear Langmuir model)

Materials	VOCs	Ads. Coeff. k_a (h^{-1})	Des. Coeff. k_d (h^{-1})	Eq. Coeff. k_e (m)	Reference
Carpet	Perchloroethylene	0.13	0.13	0.97	Tichnor et al. 1991
Carpet	Ethylbenzene	0.08	0.08	0.95	Tichnor et al. 1991
Wallboard	Perchloroethylene	0.21	1.5	0.14	Tichnor et al. 1991
Wallboard	Ethylbenzene	0.45	1.5	0.30	Tichnor et al. 1991
Ceiling tile	Perchloroethylene	0.10	0.61	0.16	Tichnor et al. 1991
Ceiling tile	Ethylbenzene	0.24	0.59	0.41	Tichnor et al. 1991
Vinyl tile	Ethylbenzene	0.26	2.74	0.095	An et al. 1999
Vinyl tile	Cyclohexanone	0.19	1.19	0.16	An et al. 1999
Vinyl tile	1,4-dichlorobenzene	0.25	0.83	0.30	An et al. 1999
Vinyl tile	Benzaldehyde	0.14	0.56	0.25	An et al. 1999
Vinyl tile	Dodecane	0.11	0.129	0.85	An et al. 1999
Painted drywall	Ethylbenzene	0.25	5.43	0.046	An et al. 1999
Painted drywall	Cyclohexanone	0.076	0.238	0.32	An et al. 1999
Painted drywall	1,4-dichlorobenzene	0.32	0.283	1.13	An et al. 1999
Painted drywall	Benzaldehyde	0.18	0.144	1.25	An et al. 1999
Painted drywall	Dodecane	0.076	0.026	2.93	An et al. 1999
Ceiling tile	Ethylbenzene	1.13	3.65	0.31	An et al. 1999
Ceiling tile	Cyclohexanone	1.23	1.12	1.10	An et al. 1999
Ceiling tile	1,4-dichlorobenzene	1.40	0.92	1.53	An et al. 1999
Ceiling tile	Benzaldehyde	1.08	0.783	1.38	An et al. 1999
Ceiling tile	Dodecane	0.97	0.191	5.07	An et al. 1999
Carpet	Ethylbenzene	0.67	0.638	1.05	An et al. 1999
Carpet	Cyclohexanone	0.44	0.205	2.15	An et al. 1999
Carpet	1,4-dichlorobenzene	0.60	0.158	3.79	An et al. 1999
Carpet	Benzaldehyde	0.57	0.092	6.22	An et al. 1999
Carpet	Dodecane	0.42	0.019	22.2	An et al. 1999
Carpet	Methyl-tert-butyl ether	0.76	4.9	0.156	Corsi et al. 1999
Carpet	Cyclohexane	0.39	1.7	0.229	Corsi et al. 1999
Carpet	Iso-propanol	0.75	1.1	0.682	Corsi et al. 1999
Carpet	Toluene	0.49	0.29	1.69	Corsi et al. 1999
Carpet	Tetrachloroethene	0.44	0.25	1.76	Corsi et al. 1999
Carpet	o-dichlorobenzene	0.60	0.08	7.5	Corsi et al. 1999
Carpet	1,2,4-trichlorobenzene	1.5	0.46	3.26	Corsi et al. 1999
Painted gypsum board	Methyl tert-butyl ether	0.07	0.12	0.58	Corsi et al. 1999
Painted gypsum board	Cyclohexane	0.01	0.18	0.056	Corsi et al. 1999
Painted gypsum board	Iso-propanol	0.08	0.24	0.33	Corsi et al. 1999
Painted gypsum board	Toluene	0.10	0.60	0.167	Corsi et al. 1999
Painted gypsum board	Tetrachloroethene	0.06	0.45	0.133	Corsi et al. 1999
Painted gypsum board	o-dichlorobenzene	0.26	0.25	1.04	Corsi et al. 1999
Painted gypsum board	1,2,4-trichlorobenzene	0.50	0.29	1.72	Corsi et al. 1999

Table 3. Selected data on the diffusion coefficient and the partition coefficient from literature

Material	VOCs	Partition coefficient K_{ma} (-)	Diffusion coefficient D_m (m ² /s)	Reference
Carpet backing material	Heptane	708.55	5.5×10^{-11}	Bodalal et al. 1999
Carpet backing material	Octane	6171.31	4.31×10^{-11}	Bodalal et al. 1999
Carpet backing material	Nonane	6216.05	2.83×10^{-11}	Bodalal et al. 1999
Carpet backing material	Decane	14617.24	5.42×10^{-12}	Bodalal et al. 1999
Carpet backing material	Undecane	24255.9	2.79×10^{-12}	Bodalal et al. 1999
Viny floor tile	Heptane	408.21	2.35×10^{-11}	Bodalal et al. 1999
Viny floor tile	Octane	1421.95	1.5×10^{-11}	Bodalal et al. 1999
Viny floor tile	Nonane	4821.88	7.22×10^{-12}	Bodalal et al. 1999
Viny floor tile	Decane	16072	2.74×10^{-12}	Bodalal et al. 1999
Viny floor tile	Undecane	61250	2.01×10^{-12}	Bodalal et al. 1999
OSB	Heptane	472.47	2.34×10^{-10}	Bodalal et al. 1999
OSB	Octane	998.94	1.12×10^{-10}	Bodalal et al. 1999
OSB	Nonane	2369.11	4.51×10^{-11}	Bodalal et al. 1999
OSB	Decane	12027.74	1.07×10^{-11}	Bodalal et al. 1999
OSB	Undecane	25931.86	7.24×10^{-12}	Bodalal et al. 1999
Gypsum board	Benzene	416	1.42×10^{-10}	Bodalal et al. 1999
Gypsum board	Toluene	941	6.38×10^{-11}	Bodalal et al. 1999
Gypsum board	Ethylbenzene	1360	2.77×10^{-11}	Bodalal et al. 1999
Gypsum board	Propylbenzene	4562	1.41×10^{-11}	Bodalal et al. 1999
Gypsum board	Butybenzene	14031	7.05×10^{-12}	Bodalal et al. 1999
Vinyl floor tile	Benzene	310	1.06×10^{-10}	Bodalal et al. 1999
Vinyl floor tile	Toluene	539	5.42×10^{-11}	Bodalal et al. 1999
Vinyl floor tile	Ethylbenzene	1522	2.02×10^{-11}	Bodalal et al. 1999
Vinyl floor tile	Propylbenzene	4520	8.03×10^{-12}	Bodalal et al. 1999
Vinyl floor tile	Butybenzene	13356	4.64×10^{-12}	Bodalal et al. 1999
Plywood	Benzene	184	2.08×10^{-11}	Bodalal et al. 1999
Plywood	Toluene	358	1.75×10^{-11}	Bodalal et al. 1999
Plywood	Ethylbenzene	2476	5.53×10^{-11}	Bodalal et al. 1999
Plywood	Propylbenzene	3249	2.16×10^{-11}	Bodalal et al. 1999
Plywood	Butybenzene	11918	7.55×10^{-12}	Bodalal et al. 1999
Particle board	Benzene	266	7.33×10^{-10}	Bodalal et al. 1999
Particle board	Toluene	968	2.68×10^{-10}	Bodalal et al. 1999
Particle board	Ethylbenzene	1237	1.05×10^{-10}	Bodalal et al. 1999
Particle board	Propylbenzene	4388	3.42×10^{-11}	Bodalal et al. 1999
Particle board	Butybenzene	18042	8.97×10^{-12}	Bodalal et al. 1999
Vinyl flooring	n-decane	3000	4.5×10^{-13}	Cox et al. 2000
Vinyl flooring	n-dodecane	17,000	3.4×10^{-13}	Cox et al. 2000
Vinyl flooring	n-tetradecane	120,000	1.2×10^{-13}	Cox et al. 2000
Vinyl flooring	n-pentadecane	420,000	6.7×10^{-14}	Cox et al. 2000

Table 4. Specific area, equivalent area and adsorption factor (European Commission, 1997)

	Carpet	Wall paper	Gypsum board	Acoustic tile
k_a	0.45 - 0.57	0.44	1.16	1.29
SA (m ² /g)	0.28	1.19	1.8	2.3
EqA (m ²)	192-205	106	7200	4140

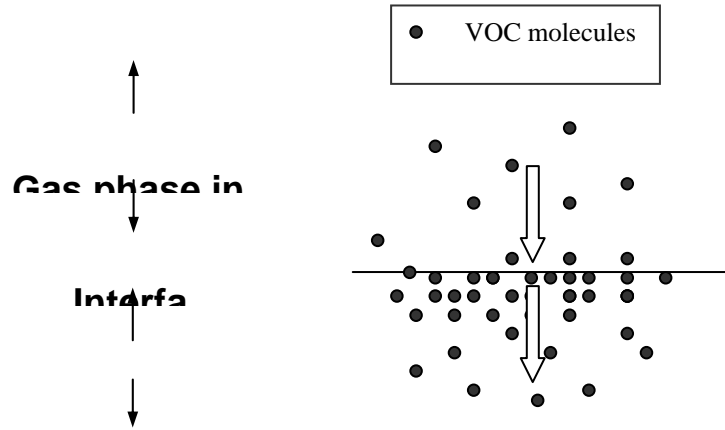


Figure 1. The schematic of the sorption process



Figure 2. Schematic of the sorption process considered by (a) first order adsorption/desorption rate models, and (b) equilibrium-interface models.

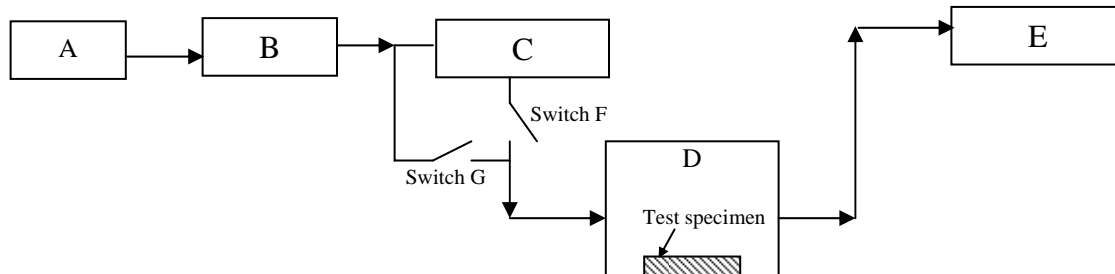


Figure 3. Schematic of a small-scale chamber sorption test system
 A: Clean air supply system; B: Airflow, temperature and humidity controller; C: Temperature controlled VOC generator containing VOC permeation tubes; D: Test chamber assembly; E: Air-sampling system at the chamber exhaust

Figure 3. Schematic of a small-scale chamber sorption test system

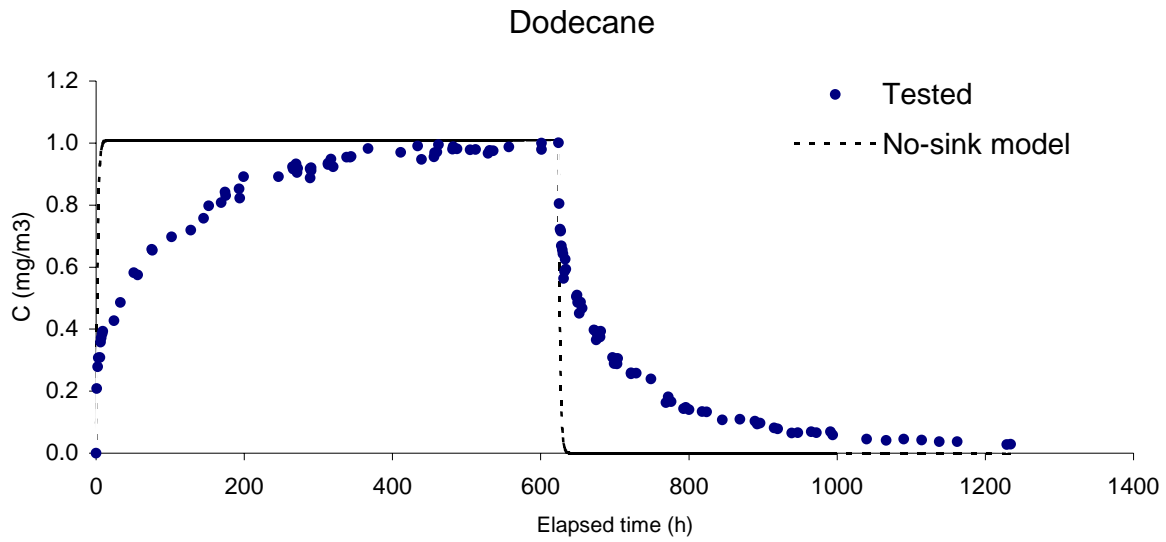


Figure 4. An example of measured sorption effect for carpet (An et al. 1999)

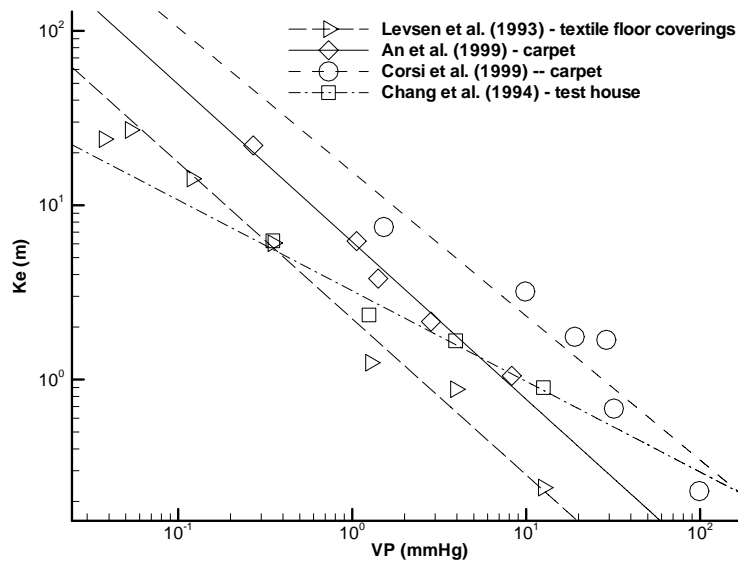


Figure 5. Relationship between equilibrium constant k_e and vapor pressure VP