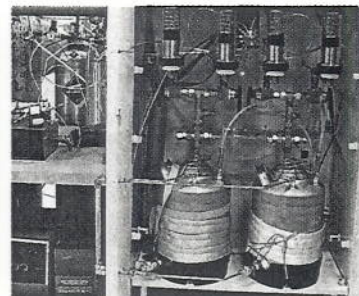
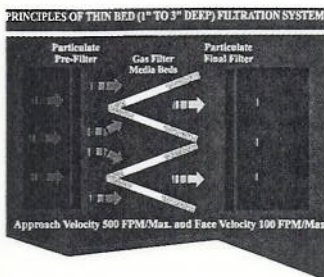


Photo 1. I.M. Pei designed Pyramid at Louvre—See article on page 17. (left)

Photo 2. Air Filtration Schematic—See article on page 24. (center)

Photo 3. Carbon Adsorption Test Bed—See article on page 34. (right)



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### The Front Cover

The cover photograph shows Kathleen Owen, a chemical engineer, setting the temperature and RH on the apparatus being used for ASHRAE Research Project RP-792 for "Evaluation of Test Methods for Determining the Effectiveness and Capability of Gas Phase Air Filtration Equipment for Indoor Air Application—Phase II: A Laboratory Study to Support the Development of Test Methods." Cover photo courtesy of Research Triangle Institute, Center for Environmental Technology, Research Triangle Park, North Carolina. The apparatus was developed with the US Environmental Protection Agency under CR-817083.

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Bill:

Here is a revised copy of Doug VanOsdell's and my paper "Carbon Adsorption for Indoor Air Cleaning" accepted for publication by the ASHRAE Journal. This copy includes revisions requested by the ASHRAE peer reviewers and editors.

Thanks for your help.

Les



## **Carbon Adsorption for Indoor Air Cleaning**

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L. E. Sparks, Radon Mitigation Branch, Air and Energy Engineering Research Laboratory, U. S. Environmental Protection Agency, Research Triangle Park, NC, 27711.

### **Introduction**

Gas-phase air filtration equipment (GPAFE) has been applied for many years to control industrial gaseous contaminants. Interest in cleaning recirculation air to provide ventilation without the need to condition excessive outdoor air (as suggested in ASHRAE 62-1989) has promoted increased interest in GPAFE as indoor air control devices. The removal of volatile organic compounds (VOCs) using granular activated carbon (GAC) is the focus of this paper. Hundreds of VOCs have been identified indoors, with each compound generally being present at a very low concentration. Brown et al. (1994) reviewed the indoor air literature and reported mean concentrations for various classifications of office buildings to range from 0.180 to 4.15 mg/m<sup>3</sup>, with a new building having a total VOC concentration as high as 39.3 mg/m<sup>3</sup>. Further, they reported that the mean concentration of individual compounds was about 0.05 mg/m<sup>3</sup>, with most being present at below 0.005 mg/m<sup>3</sup>. Mølhave et al. (1986) found that a mixture of 22 VOCs, when presented to test subjects at total concentrations in the range of 5 to 25 mg/m<sup>3</sup>, produced irritation of the nose, eyes, and throat. Thus, indoor VOCs have a potential health impact on building occupants and also present a highly

varied challenge to GPAFE.

The most common adsorbents for indoor air applications are GAC, activated alumina impregnated with potassium permanganate, and GAC impregnated to improve its performance against specific contaminants. GAC is generally specified to control VOCs. Granular media can be utilized in a number of different physical arrangements (ASHRAE, 1991), most of which consist of multiple beds of GAC in a zig-zag or Z arrangement with the airflow roughly perpendicular through each bed. Commercial bed depths generally range from 1.25 to 7.5 cm, with nominal bed residence times of 0.025 to 0.1 s. Activated carbon for indoor air applications is also available as activated carbon fibers and carbon coated fibers.

Direct testing of GAC in indoor air cleaner applications requires very long test times, and most testing has been done at elevated concentrations. Consequently the lifetime of GAC has been a subject of discussion in the literature (ASHRAE, 1991; Graham and Bayati, 1990; Ramanathan et al., 1988; Liu, 1990). The central question was whether radical differences in performance existed between the relatively high concentrations at which most data have been collected (400 to 4000 mg/m<sup>3</sup>) and the relatively low indoor concentrations at which GPAFE would be applied (0.4 to 2 mg/m<sup>3</sup>). Little published information is available on actual field trials of GPAFE.

This paper addresses GAC performance in two directions. The first part of the paper presents performance measurements for GAC at low challenge VOC concentrations that might be encountered indoors. Unlike previously reported tests, these were continued long enough to directly determine the GAC's expected lifetime.

The results suggest that test results obtained at high challenge concentrations may be extrapolated to low, indoor concentrations. Further study is needed, but these data are encouraging. In the second part, the implications of these performance measurements for the use of GAC to remove VOCs and improve indoor air quality (IAQ) are explored using an indoor air building simulation model.

### **Adsorption of VOCs on GAC**

The adsorption of VOCs on GAC is a complex function of the contaminant, the relative adsorptivity of multiple contaminants, the temperature and relative humidity, airflow rate and adsorbent bed size, and the characteristics of the GAC. Informed design of GPAFE requires that the performance of GAC for control of indoor contaminants be explored over the full range of conditions that might be encountered. This is an overwhelming task, and the research described below is only a beginning.

#### Experimental Conditions for Tests of GAC

Because the performance of activated carbon as a GPAFE is significantly affected by the contaminant, the choice of test contaminant is important. A very large number of VOCs have been detected indoors, and indoor environments differ significantly in their VOC content. Outdoor air is similarly complex. VanOsdell (1994) compared the VOCs detected indoors by a number of investigators and found only moderate overlap with no single compound that stood out as representative. Based on its physical properties, toluene has been recommended as a reasonable surrogate for indoor VOCs that can be used for testing GAC (Liu, 1990). Toluene was used as the single challenge VOC in these tests. GPAFE manufacturers provide application data



that can be used to relate probable GAC performance with other chemicals to that obtained with toluene. The relative performance of GAC types measured at one concentration may be reversed at a lower concentration, so such comparisons must be made with caution. Mixtures of challenge contaminants, such as are actually found in indoor air, further complicate the picture. The contaminants compete for adsorption sites, and an easily adsorbed chemical can displace one for which GAC has less affinity. The significance of this effect in the complex indoor air application is not clear. Test method development using a mixture of VOCs having a range of functional groups is currently underway as part of ASHRAE research project RP-792, but the research is incomplete. The tests described below were all conducted with toluene as the single contaminant.

Increasing relative humidity generally reduces the collection of VOCs on GAC, may affect some compounds more than others, and becomes more important at low VOC challenge concentrations (Nelson et al., 1976). These characteristics complicate GAC testing at indoor concentrations.

#### Small-Scale Tests of GAC Challenged with Toluene

All test results were obtained using a test gas consisting of clean air at 25°C and 50% relative humidity (RH) mixed with toluene at the indicated concentration. In the scenario analysis that follows, the behavior of toluene was taken to be representative of all VOCs that might be collected on GAC air cleaners. The GAC test bed was 4.83 cm in diameter and held a nominal 2.54 cm deep bed containing approximately 25 g of GAC. The GAC was virgin coconut shell carbon, 8 x 16 mesh, with a carbon

tetrachloride activity of 62.5% per ASTM D3467 (ASTM, 1988). All tests were conducted at a constant flow rate of 25.6 L/min, which gave a velocity through the bed of 23 cm/s and a nominal residence time of 0.11 s. The pressure drop through the test bed was approximately 127 Pa (0.5 in. H<sub>2</sub>O). The challenge toluene concentration was constant throughout the test, and toluene was the only contaminant.

The results of six tests with the indicated challenge concentrations are shown in Figure 1. The curves display the expected shape of a breakthrough curve for VOCs on activated carbon -- a period of essentially 100% collection, during which time the carbon is loading as an adsorption wavefront moves through the bed, followed by a relatively rapid breakthrough period as the wavefront reaches the downstream side of the carbon bed. As the challenge concentrations approach indoor air concentrations, the test times become extremely long. This is good news for users, but makes direct testing at indoor conditions a severe challenge. The variation in the curve shapes arises primarily in the termination point of the different tests. The extended tests in which the fractional penetration reaches approximately 1 and begins to decrease (274, 96, and 4.0 mg/m<sup>3</sup>) show the behavior of a saturated carbon bed after the challenge has been removed. In these cases, the curves begin to decline just after the toluene challenge is turned off and clean air continues to sweep through the bed, desorbing some of the toluene.

The performance of GAC can be characterized in a number of ways. Breakthrough time,  $t_b$ , is defined as the elapsed time between beginning the challenge and the time at which the penetrating concentration reaches the specified breakthrough

fraction [10 or 50% breakthrough times ( $t_{10\%}$  and  $t_{50\%}$ , respectively) are useful measures]. Breakthrough times can be obtained directly from Figure 1. Another performance measure is the activity of the carbon, often expressed as the equilibrium mass of contaminant collected relative to the mass of the carbon bed, expressed as a percentage. Still another is retentivity, which is similar to activity except that, following loading, the GAC is allowed to equilibrate in clean air. Thus activity is the maximum that the GAC can hold in contaminated air, while retentivity is the maximum the GAC can retain in clean air. Neither is readily obtained from the data shown in Figure 1. For these tests, the mass of contaminant collected on the carbon from its initial condition to a particular breakthrough time was estimated by integrating the performance curve from start to the breakthrough time of interest using the equation:

$$M = CQ \int [1 - P(t)] dt \quad (1)$$

where:  $M$  = Mass of contaminant collected,  
 $C$  = Challenge concentration,  
 $Q$  = Test flowrate,  
 $P$  = Fractional penetration through test bed, and  
 $t$  = Time.

To allow integration of Equation (1), the fractional penetration data for each test were fit to an exponential function (beginning when the downstream measurement became greater than zero and stopping at  $t_{50\%}$ ). The fit was excellent over this range. The mass collected at a particular breakthrough time was then divided by the GAC bed mass to obtain a capacity at a given breakthrough time, expressed as a percentage. Table 1 gives these performance values for the tests shown in Figure 1.



### Comparison to Reported Tests of GAC

Direct comparison of the test results to published data are not possible because of the scarcity of published data at low concentrations and differences between the experimental conditions. In summary, several authors have estimated that the lifetimes of GAC for indoor VOC control are on the order 1000 h and up, depending on concentration (Graham and Bayati, 1990; Liu, 1990).

Table 1. Performance of GAC Challenged with Toluene

Challenge Conc., ppm	0.44	1.1	9.2	9.7	71.7	72.7
Challenge Conc., mg/m <sup>3</sup>	1.7	4.0	34	36	270	274
Carbon bed mass, g	24.1	23.1	23.4	21.9	25.8	27.3
t <sub>10%</sub> , h	625	344	72	66	11.9	11.8
t <sub>50%</sub> , h	750	422	88	82	15.0	14.7
Capacity at t <sub>10%</sub> , %	6.6	9.2	16.2	16.5	18.8	18.0
Capacity at t <sub>50%</sub> , %	7.9	11.3	19.7	20.6	23.8	22.3

The retentivity value reported for toluene on GAC at 2860 mg/m<sup>3</sup> is 17% (ASHRAE, 1991). The capacity values in Table 1 have a linear relationship with the logarithm of challenge concentration, as does retentivity (ASHRAE, 1991). Extrapolation of these data to 2860 mg/m<sup>3</sup> shows that the values given in Table 1 are in general agreement, considering that the GACs are different and that desorption occurs when retentivity is measured.

The regular spacing in time (logarithmic coordinates) of the Figure 1 curves for

roughly decade-sized steps in concentration suggests that the relationship between breakthrough time and concentration might be exponential. This behavior has been previously demonstrated (Nelson and Harder, 1976) for higher concentrations, but its extension to challenge concentrations near those in indoor air had not been previously shown. This relationship is expressed by Equation (2) and as a ratio of breakthrough times at two different concentrations in Equation (3):

$$t_b = AC^B \quad (2)$$

$$t_b|_{C_2} = t_b|_{C_1} (C_2/C_1)^B \quad (3)$$

where:  $A$  = Constant for a given carbon and contaminant,  
 $B$  = Slope of breakthrough time versus contaminant concentration plot in logarithmic coordinates,  
 $C_1$  = Challenge concentration 1, and  
 $C_2$  = Challenge concentration 2.

Figure 2 shows the breakthrough times as a function of concentration for the test runs shown in Figure 1. For these data, the exponent,  $B$ , was -0.78 when computed for  $t_{10\%}$ . Nelson and Harder (1976) reported an average value for  $B$  of  $-0.67 \pm 0.17$  with a range of -0.395 to -0.937 for nine VOCs at concentrations from about 200 to 7500 mg/m<sup>3</sup> on three different GACs. The relationship shown in Figure 2 is encouraging from the standpoint of testing GAC adsorbers for indoor air applications, because it suggests that useful information may be obtained from tests at relatively high concentrations, which is much faster, easier to do, and requires less sophisticated instrumentation. Such an approach must be used cautiously, however. The wide range in the value of  $B$

for different VOCs and different GACs means that the rank order of adsorptivity for different VOCs on a single GAC might not be preserved from high to low concentrations. The same may be true for the same VOC when collected on different GACs. Competitive adsorption effects with a multiple VOC challenge are likely to be even more complex. Overall, the considerations above suggest that conducting tests using more than a single contaminant is important if GAC performance at low indoor concentrations is to be estimated from test data collected at higher VOC concentrations that require conveniently short test times.

### **Simulation of Indoor Air GPAFE Applications**

Rational design of GAC control devices for indoor applications has been greatly hindered by the variety of VOC compounds present and the lack of GAC performance data at indoor concentrations. The balance of this paper utilizes a building simulation model to evaluate the use of GAC to control VOCs in three application scenarios that might cause elevated VOC levels in buildings. The simulation provides estimates of the effect of the air cleaner on the contaminant concentration in the building as the ventilation system and/or air cleaner removes it. The requirements placed on the GAC are compared to the performance data presented above. The three scenarios are:

- Spilling a solvent in the copy/storage room (high rate, short duration),
- Waxing the reception area floor (high rate, short duration), and
- Painting the conference room (medium rate and duration.)

While not separately modeled, the effect of GAC on low-rate, long-term sources



such as building furnishings is evident from the scenario modeling after the elevated concentrations have decayed.

These scenarios were implemented in the hypothetical building shown in Figure 3. In all cases, the IAQ in the building was modeled using RISKBETA, a later version of the personal-computer based building IAQ simulation model described by Sparks et al. (1991). RISKBETA is a completely mixed room model incorporating source/sink behavior that can generate concentration and exposure estimates as functions of time. As VOC contaminants build up in the indoor air, they are adsorbed into the sinks (paint, carpets, upholstery, etc.). As the concentrations drop, the sinks become sources and re-emit the VOCs at low but significant rates. RISKBETA has been utilized to model a number of buildings, and has been found to generally provide good estimates of the contaminant decay rates. The building layout, heating, ventilation, and air-conditioning (HVAC) design and operating sequence, amount and source of ventilation, air cleaners (local and central), and presence and properties of sources and sinks can all be varied. The source emission rates and source/sink parameters utilized in the scenarios were presented by Sparks et al. (1991) and were derived from actual measurements of building materials' source/sink behavior.

The building and HVAC size and operating parameters utilized for this paper are summarized in Table 2. Each room has a single supply diffuser, and the building has a single return located in the reception area. The bathroom exhausts are the only exhausts in the building. Natural ventilation of the building occurred at 0.3 air changes per hour (ACH), and room-to-room flows, other than the HVAC flows, were zero. The

overall ventilation rate meets the requirements of ASHRAE Standard 62-1989 for an occupancy of 12 people. GPAFEs were located to clean the recirculating indoor air, and the outdoor air was assumed to be VOC-free. In all cases, the contaminants originate only as stated in the scenario and none are generated by people and furnishings (except when re-emitting adsorbed contaminants). The air cleaners were taken to be 100% efficient during the use period, as was true over most of the duration of the tests described above. The simulations thus assume that the air cleaners are changed or regenerated at the time of initial breakthrough, although in actual use some breakthrough would probably be accepted. This assumption of 100% efficiency is inadequate when the GAC reaches its retentivity limit for the mass of VOC adsorbed, which will occur at some (low) contaminant concentration.

Table 2. Building and HVAC Parameters

Total HVAC supply flow	2000 m <sup>3</sup> /h (1177 cfm, 5 ACH)
Bathroom exhaust	510 m <sup>3</sup> /min (300 cfm)
Building floor area	160 m <sup>2</sup> (1722 ft <sup>2</sup> )
Building volume	4000 m <sup>3</sup> (14,124 ft <sup>3</sup> )
Building air cleaner GAC mass comparable to test runs using 2.5 cm bed and same residence time	32.5 kg (72 lb) (for the GAC only, about \$75)
VOC mass at 6% GAC capacity	1950 g
VOC mass at 10% GAC capacity	3250 g

Spill in the Copy/Storage Room. Solvents are often stored in office buildings.

The spill scenario assumes that 2 kg (2.3L) of toluene is spilled in the copy/storage room. Half the solvent is cleaned up as a liquid, but half vaporizes and is carried throughout the building by the HVAC system. Figure 4 shows the predicted concentrations of toluene within the copy/storage room and the building average concentration with and without the GAC air cleaner. The concentrations in the copy/storage room build to very high levels, and load the sinks in that room to high levels. The concentrations then begin to decay and reach levels of below 0.1 mg/m<sup>3</sup> within less than 10 days even without the air cleaner. The difference between the decay curves with and without the air cleaner is comparatively small for the first 2 days. Then they begin to diverge because the air cleaner removes more contaminant than the sinks can re-emit. The results are similar for the rest of the building, although at much lower levels. For both the building and the room, the air cleaner is predicted to produce better IAQ more quickly. Much of its impact is due to the reduction in VOC adsorbed by the source/sinks in the building. A total of 0.8 kg (0.024 g VOC/g GAC) is removed by the air cleaner, well within the measured capacity of the GAC. As the VOC concentration continues to drop, the GAC will reach the retentivity limit for the incoming VOC at around 0.2 mg/m<sup>3</sup> (extrapolating the data in Table 1), and cease to collect. The continued sharp drop in the VOC concentration projected in Figure 4 will therefore not continue, and the curves will begin to parallel the 'no air cleaner' cases. At any moment in time, the capacity of GAC depends on its exposure history. Because the hypothetical building is reasonably clean and has clean outdoor air, routine operation



of the GAC air cleaner might not cause serious capacity degradation. On the other hand, routine loading with a VOC due to some operation in the building could degrade the performance of the GAC.

Despite the effect of the air cleaner on the concentrations, Figure 4 shows that air cleaning alone would not fully mitigate the spill. The building average remains above 5 mg/m<sup>3</sup> for about 15 h. Better cleanup, short-term increased ventilation, a separate exhaust system in the copy/storage room, and/or similar measures would all be required to reach acceptable levels quickly and prevent significant loading of the sources/sinks in the building.

Waxing the Floor in the Reception Area. In this scenario, the floor of the reception area (48 m<sup>2</sup>) is waxed, with the measured VOC emission rate for the wax being that reported by Sparks et al.(1991). Figure 5 shows, for the reception area and the overall building, the effect of a GAC on indoor VOC concentrations. As before, the air cleaner causes a large reduction in the VOC concentrations in the building, particularly after the initial concentration spike has been removed. In its impact on building IAQ, waxing a floor with a wax having these emission characteristics is similar to a spill. The 515 g of VOC collected on the air cleaner (0.016 g VOC/g GAC) is well within the capacity of the GAC. Because floor waxing occurs periodically, the capacity of the GAC must be considered. If it were the only challenge, the GAC could reasonably handle about four waxing operations. This scenario points out the need for routine maintenance of GAC air cleaners and maintenance plans that consider building operations if residual capacity is important.

Painting a Room. This scenario postulates that 50 m<sup>2</sup> of the walls of the conference room are painted with a low emission latex paint. The total VOC emission is about 98 g over 30 days, with the air cleaner, when used, collecting 76 g (0.002 g VOC/g GAC), well within its capacity. Figure 6 shows the concentrations in the conference room and in the rest of the building with and without the air cleaner. Again in this case, the air cleaner reduces the concentrations more quickly, prevents the sources/sinks from loading up, and thus reduces the time elevated concentrations are present. Because the paint is a long-lived source, the central air cleaner does not have as great an impact on the room concentration as has been true under the earlier scenarios for high emission rate sources.

Low-level, long-term emissions. The performance of centrally located GAC air cleaners to control long-term low-level emissions will depend on the emission rates and the building circulation rates. Contaminants must enter the HVAC system to be removed. If the contaminants are entering the space more slowly than they are removed by the GAC, their concentration will drop until a steady state is reached. From the standpoint of substituting recirculated air for outdoor air in ventilation, the decision rests on the economics of each building. At a constant 0.18 mg/m<sup>3</sup> of total VOC (the low end of the range given by Brown et al., 1994), an air cleaner in constant use would have to remove about 3150 g of contaminant in a year, requiring a retentivity of about 10%. However, the indoor concentrations given by Brown et al. (1994) are not steady state values and would be lower were an air cleaner installed, and the emission rates are not known. Thus the life of a GAC air cleaner cannot be predicted from these

data. In any case, the air cleaner must be maintained properly if the benefits are to continue.

## **Conclusions**

The performance of GAC, when challenged with toluene in air at 50% RH, can be measured at a relatively high concentration and usefully extrapolated to low concentrations that approach those encountered in indoor air control applications. The toluene holding capacity of GAC diminishes as the challenge concentration declines, but appears to remain adequate to control VOC emissions for extended periods. Tests with other VOCs and mixtures of VOCs continue, and the knowledge base will expand rapidly.

GPAFE technologies have a role in the improvement of IAQ in buildings. Building designs must consider a range of options, beginning with source control, to arrive at a solution that meets the needs of the occupants and is economical and energy efficient.

Standard test methods would provide the engineer with the data needed to rationally design GPAFE for use indoors. ASHRAE is currently sponsoring a research program (RP-792) to develop such a test method. The first phase of the research is complete (VanOsdell, 1994), and laboratory development is underway.

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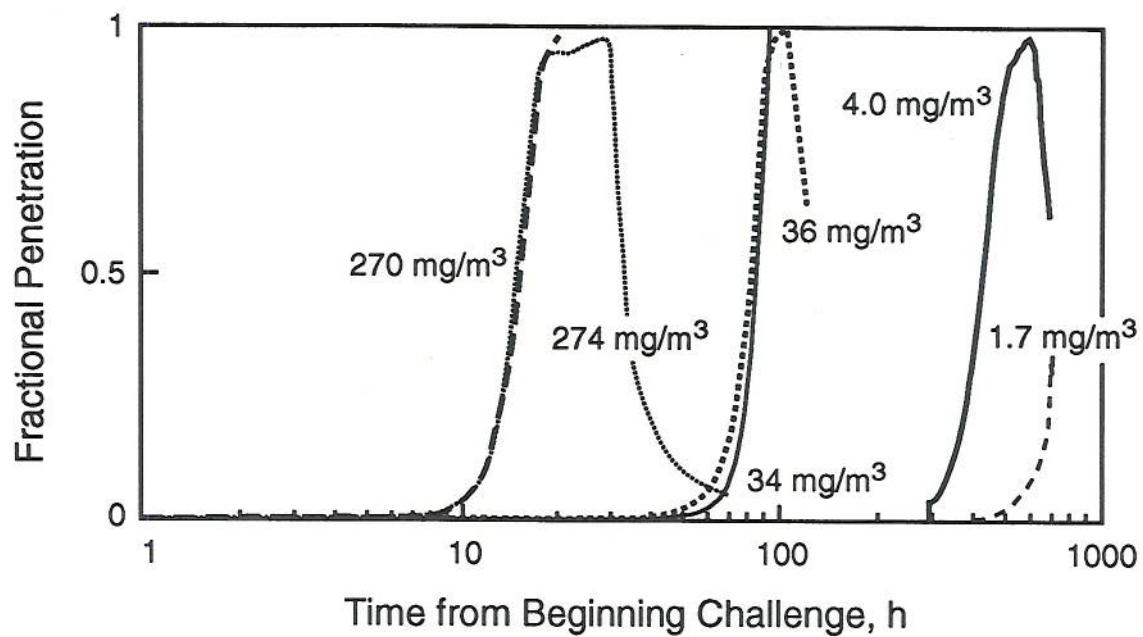


Figure 1. Measured penetration of toluene through 2.54 cm GAC at 25 °C and 50% RH.

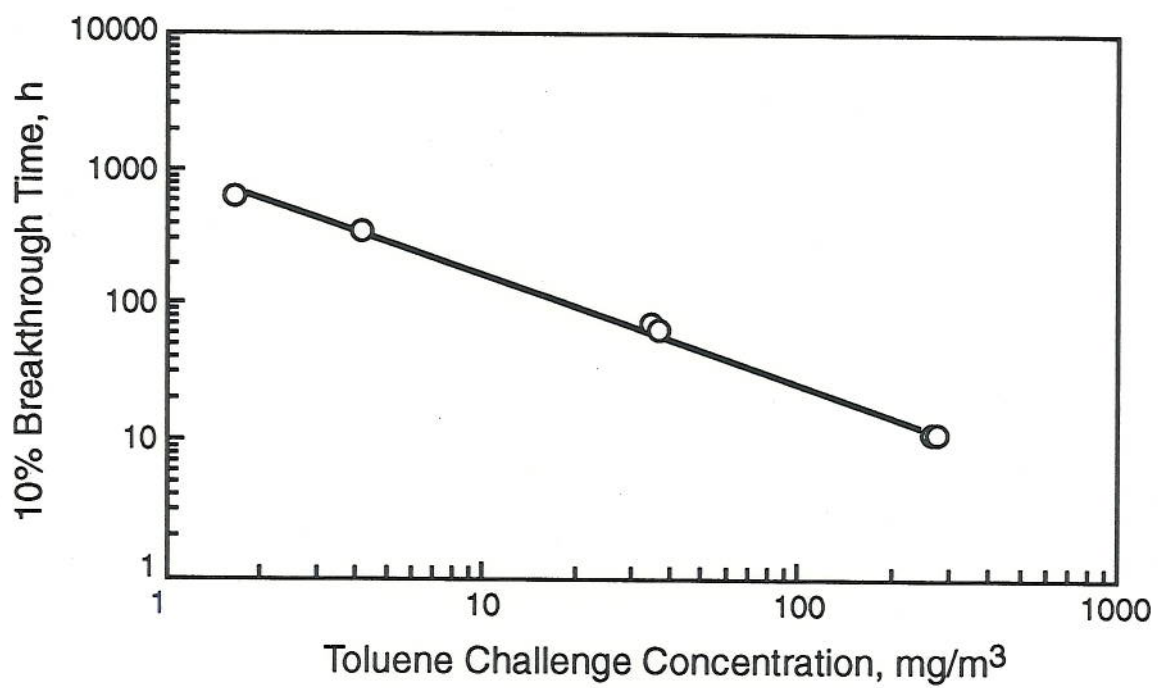


Figure 2. Effect of challenge concentration on 10% breakthrough time.



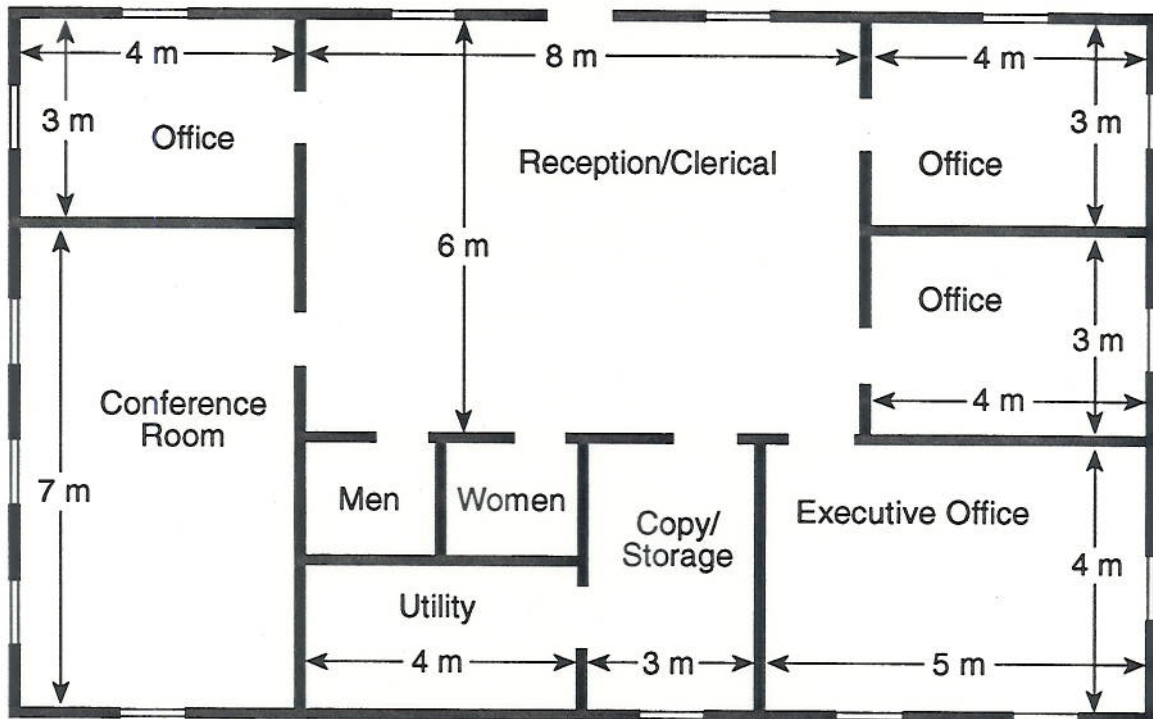


Figure 3. Floor plan of hypothetical office building.

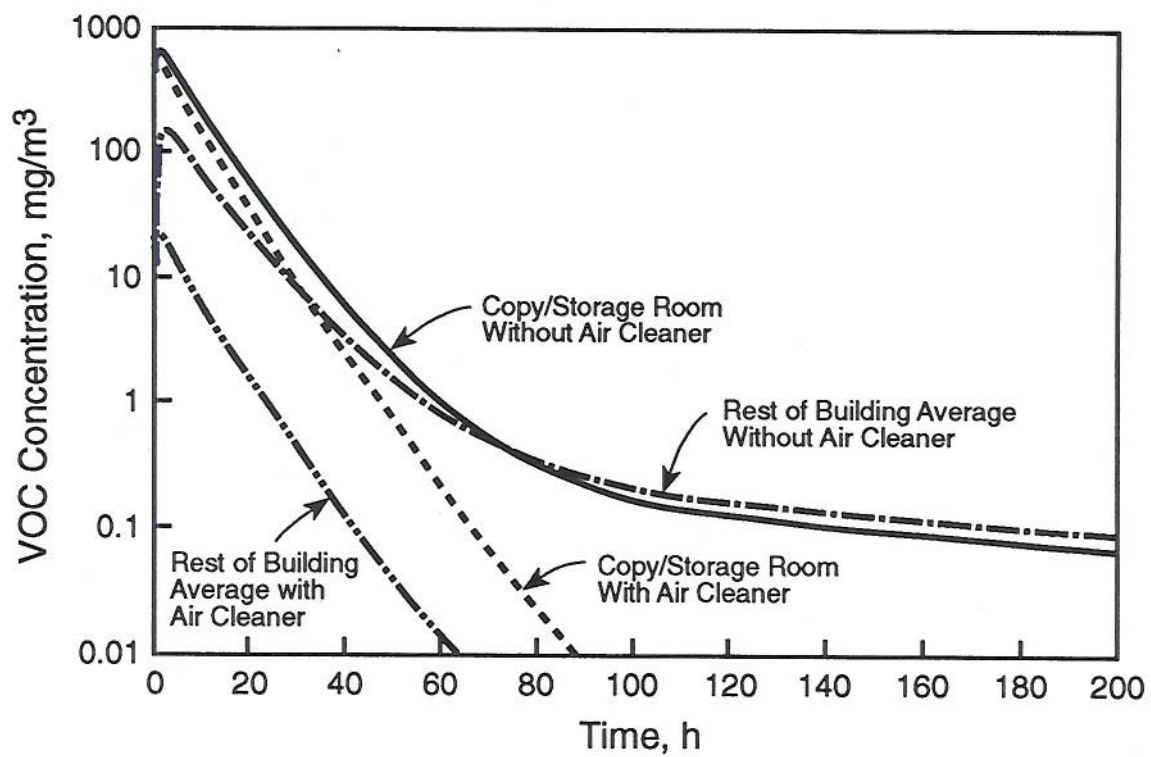


Figure 4. Effect of air cleaner on emissions from spill.

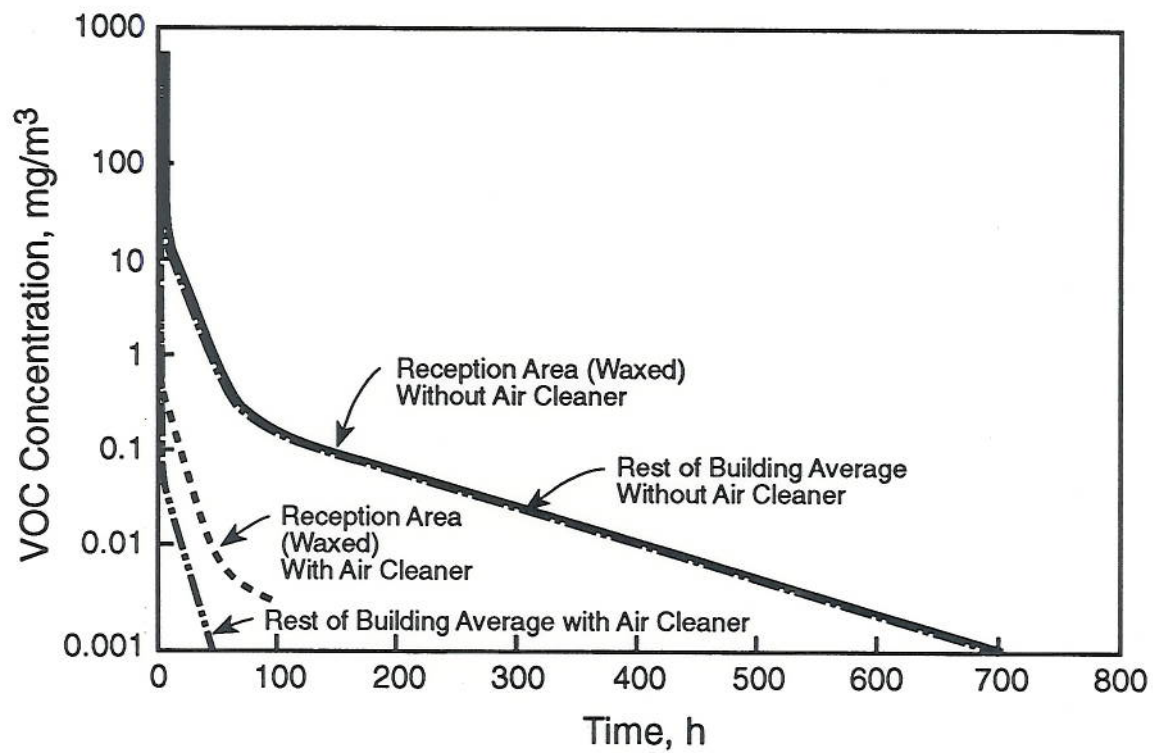


Figure 5. Effect of air cleaner on emissions from a waxed floor.



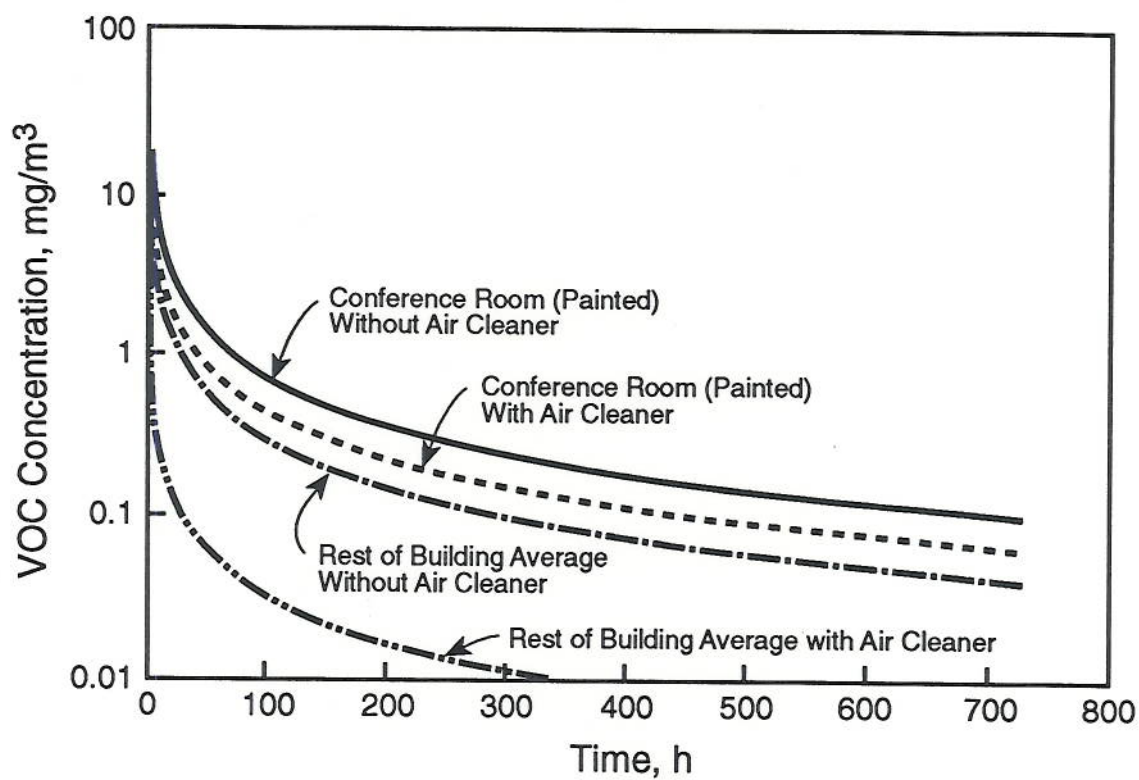


Figure 6. Effect of air cleaner on emissions from painted room.