AIR CLEANERS FOR VOLATILE ORGANIC COMPOUNDS IN INDOOR AIR

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ABSTRACT

The feasibility of activated carbon adsorption for the control of volatile organic compound (VOC) emissions at indoor air concentration levels (ppb) was investigated. Adsorption data for VOC at concentrations below 10 ppm are not publicly available. Adsorption isotherms for benzene, acetaldehyde, and 1,1,1-trichloroethane in the 100- to 200-ppb range were obtained on three different carbons using a microbalance. Two different models were used to predict in-duct carbon filter lifetimes based on the experimental data. The results show that the small quantities of carbon used in panel and duct filters appear to have limited usefulness in indoor VOC control.

INTRODUCTION

Indoor air environments, where humans spend the majority of their time, can contain significant levels of volatile organic compounds (VOC). Because many of these compounds are potentially toxic, it is necessary to determine ways to control these compounds through source removal and heating, ventilating, and air-conditioning (HVAC) system modification, including the incorporation of air cleaners. This paper presents the results of a study to determine the feasibility of carbon adsorption for the control of VOC in indoor air.

BACKGROUND

Sources

There is a wide variety of sources of VOC in indoor air. These sources vary tremendously depending on the nature of the facility, but often include contributions from outdoor air that enters the building through leaks and through the HVAC system. Other typical sources include combustion, smoking, building materials, and office machines. Various studies of indoor air quality (IAQ) have identified more than 250 organics at levels greater than 1 ppb (Sterling 1985) with concentrations expected in the few hundred ppb range but not exceeding a few ppm (Wallace et al. 1987; Wang 1975; Tucker 1986).

Controls

In evaluating the feasibility of different control strategies, a maximum allowable level for each pollutant is needed. Unfortunately, toxicity data are lacking for many of the compounds. Therefore, arbitrary criteria, such as odor, are often used. Depending on the compound, these may or may not be adequate from a health standard perspective.

Source removal and ventilation have received the most attention as control strategies. However, source removal is not always practical because, for example, offices need photocopy machines to be readily accessible. Also, increasing the

TABLE 1
Carbon Properties

Carbon/property	Coal base	Pinewood charcoal base	Coconut shell base	
Activation method	Steam	Steam	Steam	
CCl ₄ Adsorption wt. % (min)	60	65	60	
Surface area m²/g	1,050-1,150	1,150	1,050-1,150	

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ventilation rate can result in prohibitively high HVAC costs.

Therefore, it is apparent that practical methods for the removal of organic vapors need to be developed. Adsorption, absorption, incineration, and catalytic conversion are removal techniques for pollutant gases. All have demonstrated applicability in conventional pollution control applications and could be incorporated into an HVAC system with minimal retrofit. However, adsorption on carbon seems to be the most popular technique currently in use for indoor applications. A review by Turk (1977) discusses this applicability in detail.

Two commercially available air-cleaning devices incorporating activated charcoal as the removal technique for VOC were studied by Daisey and Hodgson (1988). Two other devices were examined in the same study, but these used additional techniques (such as catalytic oxidation) to reduce the VOC level. The two carbon-only devices showed extremely low efficiencies (from 0% to approximately 6%) for VOC removal (which indicate low adsorption). Deitz (1988) also suggests that there may be problems with the use of activated carbon for gas and vapor removal in HVAC systems. These results show the need for additional research into the efficacy of carbon adsorption for indoor air VOC control. It is especially important to test the carbon by itself to determine, without question of interference, the potential of activated carbon for use in IAO control devices.

PURPOSE OF THE STUDY

The majority of adsorption systems currently marketed for indoor air pollution applications deal with odor control. These systems consist primarily of in-duct carbon filters.* Quantifiable performance data are not available for any of the systems surveyed. The carbon in each unit is assumed to be spent when occupant complaints increase. Obviously, this method of bed lifetime estimation has drawbacks, particularly when dealing with potentially toxic compounds that have relatively high odor thresholds. Adsorption isotherm data in the

^{*}More detailed information on the vendor survey can be obtained from the authors.

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TABLE 2 Summary of Adsorption Results

Compound		Uptake* (×107)	
Acetaldehyde	Wood-base	Coal-base	Coconut shell-base
Concentration (ppb)	mol CH₃CHO/g C	mol CH₃CHO/g C	mol CH₃CHO/g C
119	3.39	3.52	10.0
153	4.17	10.00	10.0
Benzene	Wood-base	Coal-base	Coconut shell-base
Concentration (ppb)	mol C₅H₅/g C	mol C₀H₀/g C	mol C ₆ H ₆ /g C
101	2.42	1.24	4.30
119	4.30	6.37	5.60
176	10.00	7.27	6.33
1, 1, 1-Trichloroethane	Wood-base	Coal-base	Coconut shell-base
Concentration (ppb)	mol CH₃CCl₃/g C	mol CH₃CCl₃/g C	mol CH ₃ CCl ₃ /g C
115	0.749	1.15	2.04
183	1.33	2.80	1.91

^aEach number represents the average of 3 to 4 data points.

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low concentration regime (<10 ppm) are not publicly available, even for simple compounds, on any carbons. This led to our effort to obtain adsorption isotherms in the ppb range in order to determine the applicability of activated carbon adsorption for indoor air pollution control.

EXPERIMENTAL DESIGN

Three types of activated carbon were tested: a coal-based carbon, a wood-based carbon, and a coconut-shell-based carbon. Table 1 lists some of the physical characteristics of these carbons.

The pollutants chosen were relatively simple, low molecular weight compounds representative of three different classes of organic compounds: benzene (aromatic hydrocarbon), acetaldehyde (oxygenated hydrocarbon), and 1,1,1-trichloroethane (halogenated hydrocarbon). Isotherms were measured in the 100- to 200-ppb range for each compound. This concentration range was chosen based on measurements of VOC concentrations reported in the literature (Sterling 1985; Wallace et al. 1987; Wang 1975).

Materials and Equipment

The low concentrations were generated with clean, dry airstreams flowing over permeation tubes with certified permeation rates. These tubes had permeation rates of 71.3 ng/min for benzene, 148 ng/min for 1,1,1-trichloroethane, and 32.5 ng/min for acetaldehyde. Nitrogen was used as a purge gas during degasing. All gases used had purities greater than 99.999%.

The experimental setup is shown in Figure 1. It consisted of two mass flow controllers with flow ranges of 0 to 1000

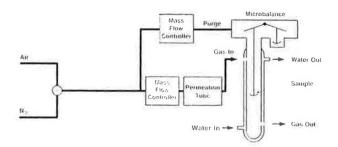


Figure 1 Schematic of experimental setup

cm³/min and 0 to 100 cm³/min, a microbalance, and a thermostatted water bath for the permeation tubes and balance jacket.

The microbalance was used for the adsorption studies. It is capable of weighing up to 100 g with an accuracy of $+0.5 \text{ }\mu\text{g}$. This device may be operated under low-temperature, high-vacuum conditions for the determination of BET surface area and is ideally suited to make adsorption isotherm measurements under different pressure and temperature conditions.

Experimental Procedure

Gravimetric measurement of adsorption isotherms can be carried out in either a static or dynamic mode. The static mode, usually under a vacuum, has the advantages of a high signal-to-noise ratio and a fairly good control of partial pressure of adsorbate. However, equilibration times can be very long, particularly at low concentrations, and the conditions are not representative of real systems, where air is present with the adsorbate and may affect its adsorptivity. The authors chose to operate in the dynamic (flow) mode, at atmospheric pressure, in order to reduce experimental time and measure adsorption under conditions more representative of streams challenging real-world filters.

The carbon sample (150 to 200 mg) was degased under flowing nitrogen while it was heated with an infrared lamp. Degasing was assumed to be complete when the weight remained constant for 10 to 15 minutes. The sample was then allowed to cool in nitrogen to 30°C before switching to the airstream and introducing the permeation tube. Although this temperature is high relative to HVAC applications, temperature control at lower temperatures was found to be very difficult. We do not believe that this will qualitatively affect the conclusions reached in this study. Flow rates were adjusted to give the desired concentrations. Equilibrium was assumed when the weight gain (on the 100- μ g scale) leveled off (rate of weight gain < 4 μ g/h).

RESULTS

It is no accident that there is a paucity of scientific data on the effectiveness of activated carbon for organic vapor removal at the ppb level. Experiments to measure uptakes, either through changes in gas-phase concentrations or gravimetric techniques, are extremely laborious and can be very expensive. It is only very recently that concern about potential health effects at low concentration levels has begun to justify the effort and expense involved. While there are various reports about the capacity of carbons in use for controlling different emissions (Turk 1977), details about the composition and concentrations of the streams tested are often lacking.

It must be admitted, prior to discussion of the results, that there are many potential sources of error in the low concentration/low uptake experiments described in this study. Extreme care was taken to minimize errors in measurement. Replicate runs were used to confirm the values obtained. While there may be some question as to the absolute values of uptake measured, the authors believe that any error incurred was not significant enough to materially affect the conclusions of the study.

Table 2 summarizes the results for the uptakes of organic pollutants on the various carbons. It is apparent that the capacities (mol/g carbon) do not vary much with either compound or carbon in the concentration range studied. Capacities ranged from 10⁻⁸ to 10⁻⁶ mol/g. Acetaldehyde uptake is about two to three times higher than the other compounds studied. Figures 2, 3, and 4 represent the isotherms over the concentration ranges studied. The magnitude of isotherm slopes in the concentration range studied decreased from benzene to 1,1,1-trichloroethane to acetaldehyde. Higher isotherm slopes indicate more favorable adsorption. The data for benzene, for which three data points are available, were linearized in order to obtain an equation for use in a predictive packed bed model. Although the data (Figure 2) do not appear to fit a straight line through the origin very well, we hoped that this exercise would give us a feel for the applicability of carbon adsorption to indoor air pollution control. Table 3 lists the linearized isotherms for benzene adsorption on the three carbons in this study.

It is possible that, at the extremely low concentrations studied, chemisorption may be a major contributor to measured uptake. The fact that carbons used in indoor applications are generally very difficult to regenerate lends some support to this hypothesis. Significant chemisorptive effects may be the explanation for certain apparent anomalies in the observed results, such as lower uptakes for 1,1,1-trichloroethane than for acetaldehyde.

Isotherm data are useful for carbon screening and pro-

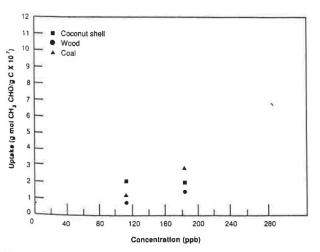


Figure 2 Adsorption of 1,1,1-trichloroethane on activated carbons

vide an upper bound on the capacity of the adsorbent, but they do not provide direct practical information on the applicability of the technique. For this reason, estimates of the carbon filter size were carried out using a method recommended by the manufacturer of two of the carbons and a model developed by RTI. This is a standard fixed-bed breakthrough model that makes the assumption of (1) a linear isotherm, (2) plug flow, and (3) no radial or axial dispersive effects (EPA 1987). Two methods were used in order to compare results. The superficial velocity of gas across the filter was assumed to be 12.7 cm/s, corresponding to a flow of 100 cfm (2830 lpm) across a 2 ft by 2 ft (61 cm by 61 cm) filter. An exit concentration of 50 ppb was set as the level corresponding to useful bed life.

The first method was applied to the data for all three compounds, with challenge concentrations varying from 115 to 183 ppb. The lifetime for 6-in-thick (15-cm-thick) filters, such as those typically marketed for odor control applications, was determined. The results in Table 4 show that breakthrough occurs rather quickly, bringing into question the applicability of a once-through filter even in the absence of other pollutants or humidity effects, both of which would cause performance to deteriorate further.

The second method, the RTI program, was applied to the benzene data because it probably represents the best case

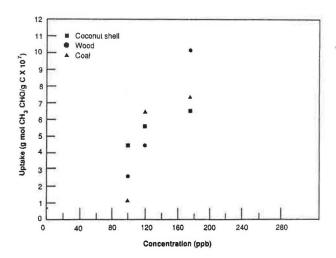


Figure 3 Adsorption of benzene on activated carbons

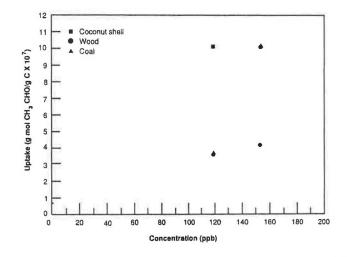


Figure 4 Adsorption of acetaldehyde on activated carbons

TABLE 3
Linear Equations for Benzene®

Carbon type Wood-base	Uptake		
	1.0 × 10 ⁻⁸ C _b	-7.7×10^{-7}	mol C ₆ H ₆ /g C
Coal-base	$6.5 imes10^{-9}C_b$	-3.6×10^{-7}	mol C₀H₀/g C
Coconut shell- base	$2.4 \times 10^{-9} C_b$	$+2.3 \times 10^{-7}$	mol C ₆ H ₆ /g C

^aC_b = Benzene concentration in ppb.

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TABLE 5

Variation of Exit Concentration with Bed Depth

T = 4,320 h (6 mo)		T = 30 h		
L (ft) ^b	C exit (ppb)	L ^b (ft)	C exit	
1,098.64	121	7.65	114	
1,097.67	115	8.15	55.6	
1,108.95	99.5	8.31	35.1	
1,137.29	67.2	8.41	20	
1,163.09	40.6			

a150 ppb benzene challenge
 12.7 cm/s superficial velocity
 3.0 mm particle size

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for the compounds studied. The linearized form of the equation on the coconut-shell-based carbon was used. The challenge concentration was assumed to be 150 ppb, and the other variables were set as before. Table 5 lists the results of this simulation. Again, breakthrough is shown to occur quickly.

This result reinforces the conclusion from the first method calculations that filter life would be extremely short, making these filters of negligible value as in-duct indoor air filters. However, it is important to note that the level at which acceptable breakthrough is set is critical to bed lifetime. In the absence of toxicological data, this may have to be set rather arbitrarily, erring on the side of caution.

The results of this study are also supported by the results reported by Daisey and Hodgson (1988). Both studies show that the small quantities of carbon used in panel and duct filters appear to have limited usefulness in controlling indoor air VOC at the extremely high face velocities often encountered. While it is true that most in-duct filters are used in multi-pass as opposed to once-through modes, these simulations represent the results for the first pass, where best performance would be expected. Carbon devices using more complex flow schemes (involving parallel beds with regeneration) may work, but they may be uneconomical except in very large-scale applications. Carbon adsorption may be viable in the event of major surges in concentration, such as spills or releases, which push the ambient concentrations into regimes where the shape of the isotherm is more favorable and sufficient driving force exists for a reasonable uptake. --

TABLE 4
Calculated Lifetime of a 6-inch Carbon Bed

Gas	Mole fraction $(\times 10^{-7})$		Lifetime (min)	Coconut shell
		Wood	Coal	
Benzene	1.01	1.02	0.52	1.82
	1.19	1.54	2.29	2.01
	1.76	0.24	1.76	1.54
Acetaldehyde	1.19	1.22	1.26	7.14
	1.53	1.16	2.79	2.79
1, 1, 1-Trichlo-	1.15	2.78	0.43	0.76
roethane	1.83	0.31	0.65	0.45

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CONCLUSIONS

The following conclusions may be drawn from this study:

- All of the carbons studied showed measurable uptakes of benzene, acetaldehyde, and 1,1,1-trichloroethane.
- The adsorption of all the compounds on the three carbons was similar—in the range of 10⁻⁸ to 10⁻⁶ gmol/g.
- A packed bed simulation showed that breakthrough occurs very quickly in the shallow (6 in or 15 cm) filters currently marketed for odor control.
- Carbon adsorption may be applicable for "event control,"
 i.e., sudden surges of concentration caused by spills or
 leaks.

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DISCUSSION

Carl N. Lawson, LRW Engineers Inc., Tampa, FL: In a computer room with laser printers, which emit very heavy ozone, which type of air cleaner would you recommend and where would you locate it?

K. Ramanathan, Research Triangle Institute, Research Triangle Park, NC: I am not sure. Carbon may or may not

Coconut-shell-based carbon b1 ft = 0.3048 m

work. You would need an efficient collection system, e.g., a hood, around the printer in order to draw the contaminated air into the cleaner.

Arthur E. Wheeler, Wheeler Engineering Co., Towson, MD: Please comment on the potential efficacy of carbon absorption in conjunction with or in lieu of bake out (90° to 100°F) to remove VOCs in new buildings prior to occupancy or following repainting or installation of new carpeting. VOC concentration data under such circumstances are unknown.

Ramanathan: It is difficult to say without knowledge of the exact concentrations and compounds. I would guess that carbon adsorption would definitely be more effective than in standard in-duct use.

Hal Levin, Hal Levin & Associates, Santa Cruz, CA: Was this research the same work reported by Dr. Ensor at IAQ '88?

Ramanathan: Yes. This is an expanded version of the VOC cleaner aspects of the work. Dr. Ensor's paper took a

broader view of air cleaner technologies and discussed this aspect briefly.

Levin: You suggested that proper design should be done to use carbon filters. Please describe proper design.

Ramanathan: Systems involving longer contact times between gas and carbon, e.g., pressure swing adsorption systems, might work. Systems based on short cycle time with alternate adsorption and regeneration may also work. Regeneration may present a problem at the low concentrations present in indoor air since chemisorption on the carbon may occur. All the above options will be capital- and energy-intensive and need to be investigated carefully.

Levin: What were the results for acetaldehyde and 1,1,1-trichloroethane?

Ramanathan: Capacities were low and, as the table showing Model I results indicates, breakthrough occurs very quickly.