

EVALUATING MATERIALS FOR HEALTHY BUILDINGS

6229

B. A. Tichenor
U.S. Environmental Protection Agency
Air and Energy Engineering Research Laboratory
Indoor Air Branch (MD-54)
Research Triangle Park, NC 27711
USA

Evaluating materials to ensure healthy buildings requires source characterization coupled with exposure assessment (see Table 1). This paper discusses the approaches currently used by indoor air quality (IAQ) researchers and practitioners to characterize the emissions from indoor materials, including the interaction of these emissions with indoor sinks. Procedures for analyzing chamber test data to produce emission rates and adsorption/desorption rate constants are discussed, as is the use of these results in IAQ models to predict occupant exposure.

Table 1 - EVALUATING MATERIALS FOR HEALTHY BUILDINGS

SOURCE CHARACTERIZATION

- Determine Pollutant Emission Rates from Indoor Sources
- Determine Sink Adsorption/Desorption Rates
- Determine Biological Response to Source Emissions

EXPOSURE ASSESSMENT

- Determine Human Exposure to Source Emissions/Sink Re-emissions
 - Determine Impact of Exposure on Human Health and Comfort
-

SOURCES AND SINKS

There are myriad sources of indoor air pollution, including building materials, furnishings, consumer products, combustion (e.g., environmental tobacco smoke [ETS], cooking, heating appliances), and outdoor air. All sources are of concern when considering the design and operation of a healthy building. As a first step, the building designer needs to determine the indoor pollutant load to be imposed by the construction materials and building furnishings. Indoor sources can be broken down into dry and wet materials.

Dry Materials - Dry materials, which include the majority of materials used to construct and furnish residential and commercial environments, are characterized by relatively low emission rates which decay slowly. Such materials include: wood

products, floor coverings (e.g., carpet, vinyl), wall coverings (e.g., wallpaper, fabric), ceiling materials (e.g., acoustic tiles, "blown" gypsum), and insulation (e.g., fiberglass, rigid foam). Heating, ventilating, and air-conditioning (HVAC) systems also include potential indoor air pollution sources, such as duct liners. Furnishings, composed of pressed wood products and/or upholstery, are additional potential sources.

Wet Materials - Modern construction techniques rely heavily on a wide variety of architectural coatings (e.g., paints, stains, varnishes), adhesives, caulks, and sealants. Such materials are applied "wet" and their emissions (mostly petroleum based solvents) are relatively high and decay rapidly.

Sinks - Indoor surfaces act as sinks by adsorbing and later re-emitting vapor-phase organic indoor air pollutants. As shown later in this paper, indoor sinks play a major role in determining the concentration vs. time history associated with indoor sources, especially wet sources. Indoor sinks of interest include: floors (particularly carpets and rugs), walls, ceilings, HVAC systems (including supply and return ducts and filters), and furnishings.

Factors Affecting Emissions - In developing and using methods for determining the emission characteristics of indoor sources, the governing physical and chemical processes, as well as the important variables, need to be considered (1). The important factors are presented in Table 2.

Table 2 - FACTORS AFFECTING SOURCE EMISSIONS AND SINK ADSORPTION/DESORPTION

MASS TRANSFER PROCESSES

- Evaporation (from wet products)
- Adsorption (to/from indoor sinks)
- Diffusion (in air, in material)
- Convection (bulk flow)

ENVIRONMENTAL VARIABLES

- Temperature (affects rate of evaporation, adsorption, diffusion)
- Humidity (affects emission rates of formaldehyde)
- Air Exchange Rate (affects indoor concentration via dilution/flushing)
- Boundary Layer (controls rate of gas-phase mass transfer)
 - Velocity, Turbulence

MATERIAL CHARACTERISTICS/COMPOSITION

- Amount Used (affects emission rate and total emissions)
- Surface Roughness (affects mass adsorbed to sink)
- Number and Type of Chemical Constituents
 - Vapor Pressure, Diffusivity (affect emission rates)

SOURCE TESTING METHODS -- CHEMICAL EMISSIONS

A variety of methods are used by IAQ investigators to determine the chemical emissions from indoor materials and furnishings (see Table 3). Each of these methods requires the use of appropriate techniques for sampling (e.g., syringe, canister, sorbent) and analysis (e.g., gas chromatography -- GC) of the organic chemicals of interest.

Table 3 - SOURCE TESTING METHODS -- CHEMICAL EMISSIONS

LABORATORY STUDIES

- **EXTRACTION AND DIRECT ANALYSIS**
 - Provide Information on Material Composition
 - Do Not Provide Emissions Composition or Emissions Rate Data

- **STATIC HEADSPACE**
 - Provide Information on Emissions Composition
 - Do Not Provide Emissions Rate Data

DYNAMIC CHAMBER STUDIES

- **SMALL CHAMBERS**
 - Provide Emissions Composition and Emissions Rate Data under Controlled Environmental Conditions
 - Chamber Size May Limit Use for Some Material Sources (e.g., furniture, work stations)

- **LARGE CHAMBERS**
 - Provide Emissions Composition and Emissions Rate Data under Controlled Environmental Conditions
 - May Be Required for Evaluating Emissions During the Application Phase of Wet Materials

FULL-SCALE STUDIES

- **TEST HOUSES**
 - Provide Emissions Composition and Emissions Rate Data under "Semi-controlled" Environmental Conditions; Sink Factors Must Be Considered
 - Very Useful for Validating Chamber Emissions Test Results Using IAQ Models

- **FIELD STUDIES**
 - Provide Integrated Emissions Profile of All Sources and Re-emitting Sinks under Uncontrolled Conditions
 - Emission Rate Determinations Generally Not Possible
 - Differentiating Between Source and Sink Emissions Extremely Difficult

Dynamic Chamber Studies - While laboratory extraction and headspace studies provide information on the composition of emissions, dynamic flow-through chamber testing is needed to develop emission rate data. Both small and large chambers are commonly used to conduct such testing.

Small Chambers - Small ($< 5 \text{ m}^3$) environmental test chambers are used throughout the world to evaluate emissions from indoor materials (2,3,4). A typical small chamber facility includes: a clean air delivery system, one or more well mixed test chambers (built with non-adsorbent interiors), environmental controls (temperature, humidity, air flow rate), and sampling and analysis equipment. Emissions testing is conducted by placing a sample in the chamber and measuring the concentration (individual compounds or total organics) at the chamber outlet. The sample size is usually determined by the "loading factor" (i.e., the ratio of the test specimen area to the chamber volume). Generally, the loading factor would be set equal to the surface area to volume ratio one would expect for normal use of the material in full-scale environments. Concentration data are collected over a sufficient time interval to adequately describe the time history of the emission rate. While small chamber testing methods are still being improved, the technology has matured enough to result in an ASTM standard guide (5) and a Commission of the European Communities guideline (6).

Small chambers have obvious limitations. Normally, only samples of larger materials (e.g., carpet) can be tested. Small chambers may not be applicable for testing complete assemblages (e.g., furniture, work stations). For some materials, small chamber testing may provide only a portion of the emission profile of interest. For example, the rate of emissions from the application of paints and coatings via brushing, spraying, rolling, etc. is higher than the rate during the drying process. Small chamber testing cannot be used to evaluate the application phase of the coating process.

Large Chambers - Large, room sized (e.g., $15 - 30 \text{ m}^3$) chambers are used to overcome the limitations of small chambers noted above (4,7). As with small chamber testing, careful control of the environmental variables is necessary to ensure accurate results from large chamber testing. Emissions testing procedures using large chambers are essentially the same as with small chambers, except in large chambers the sample is usually collected at one or more locations in the chamber instead of in the outlet flow.

Full-Scale Studies - While dynamic chamber studies are useful for determining emission rates of indoor materials under controlled conditions, full-scale test house and/or field studies are necessary to validate the chamber data. Full-scale studies also provide the opportunity to evaluate the interaction of source emissions with indoor sinks. In addition, evaluation of such factors as variable air exchange rates, operation of heating/cooling systems, room-to-room air movement, and occupant activities is possible with full-scale studies.

Test Houses - IAQ test houses are used to investigate a variety of indoor air pollution research questions, including the behavior of sources and sinks (8). Test houses are generally unoccupied and are provided with instruments and equipment for monitoring a variety of variables, including: temperature, humidity, air exchange rate, and operation of the heating/cooling system. Systems are installed to allow

indoor air samples to be collected at various locations within the house. Both on-site and off-site analytical instruments are used to quantify indoor pollutant levels. IAQ test houses are generally single family residences, with construction features typical of the area where they are located. Since they are unoccupied, test houses can be used to investigate the behavior of single sources without the confounding effects of occupant activities. Unlike chamber studies, precise control of the environmental variables is difficult, especially the air exchange rate which is controlled by the weather. In addition, the multitude and complexity of interior surfaces make it imperative that the interaction of sources and sinks be considered during data analysis. Consideration must also be given to the pollutant levels in the outdoor air and the background levels in the test house prior to any experiments. In spite of these complications, IAQ test houses are extremely valuable research tools for investigating sources and sinks in a realistic manner.

Field Studies - Literally hundreds of field studies have been conducted to investigate indoor air pollution problems. Field studies often provide insight into the "source" of the IAQ problem. For example, finding excessive levels of a compound associated with a specific source (e.g., paradichlorobenzene from moth repellent) can enable the source to be identified. Unfortunately, many indoor sources (e.g., solvent containing products) share common emission profiles in terms of the compounds emitted. Thus, isolating the source of a common indoor pollutant based on indoor measurements may be impossible. In addition, re-emissions from indoor sinks can cause elevated indoor concentrations of some pollutants to exist long after the original source of the pollutant has been depleted. Thus, field study results generally provide an integrated assessment of IAQ due to the emissions from a multitude of sources and re-emitting sinks under uncontrolled conditions, and using field study results to determine the emission rates of individual sources is extremely difficult if not impossible.

EVALUATION OF INDOOR SINKS

Methods used to evaluate indoor sinks with respect to their adsorptive and desorptive behavior parallel the source characterizations methods described above.

Packed Columns - The principles of gas/solid chromatography have been used to evaluate the sink characteristics of indoor materials (9). In this technique, samples of material (e.g., fibers) are packed into columns and the retention times of the organic compounds are determined at various temperatures. The retention times are related to the partition coefficients which represent the equilibrium mass adsorbed on the material being tested. This method furnishes information on the equilibrium conditions, but does not provide kinetic data on the adsorption or desorption rates.

Dynamic Chamber Tests - Dynamic flow-through chambers can be used to evaluate the sink rates (adsorption/desorption) for indoor surfaces (10,11). Samples of the sink material are placed in chambers and exposed to known concentrations of pollutants. As with source testing, concentration vs. time data are collected. These data are then analyzed, using appropriate sink models, to determine the mass adsorbed and the adsorption and desorption rates.

Test House Studies - IAQ test house studies can be used to evaluate the validity of chamber derived adsorption and desorption sink rates (11). Concentration vs. time data collected in test house studies are evaluated using IAQ models containing equations describing the sink behavior. To date, such experiments have been only partially successful in validating dynamic chamber sink results.

ANALYSIS OF DYNAMIC CHAMBER TEST RESULTS

As noted above, dynamic chamber testing is the most common method being used to determine: a) source emission rates, b) mass adsorbed on sinks, and c) sink adsorption and desorption rates. Computational techniques have been developed to analyze dynamic chamber test data to produce source and sink rates.

Source Evaluations - Source emission factors are determined by fitting appropriate source models to chamber concentration vs. time data. The model selected is based on the source behavior.

For sources with constant emission rates, the calculation of the source emission factor is straightforward:

$$EF = C(N/L)$$

where EF = emission factor (mg/m²-hr); C = chamber concentration at equilibrium (mg/m³); N = chamber air exchange rate (hr⁻¹); and L = chamber loading (m²/m³).

For sources with decaying emissions, a common approach is to assume a first order decay (3):

$$EF = EF_0 e^{-kt}$$

where EF₀ = initial emission factor (mg/m²-hr); k = first order rate constant (hr⁻¹); and t = time (hr). Another approach for some sources is to assume two first order rate constants (2). Source models have also been developed that are based on fundamental mass transfer processes (12).

Sink Evaluations - Methods for determining sink characteristics (e.g., mass adsorbed; adsorption and desorption rates) from dynamic chamber test data are not as well developed as the source evaluation methods. One approach uses two chambers: one with sink material, the other empty (10). Ratios of the concentrations from the two chambers are analyzed, assuming first order processes, to obtain empirical equations for adsorption and desorption. Another approach uses a single chamber and fits the concentration vs. time data using models based on adsorption/desorption theory (e.g., Langmuir isotherms) (11). This approach provides adsorption and desorption rate constants. Both approaches provide estimates of mass adsorbed by calculating the difference between "sink" and "no sink" concentration vs. time profiles.

USING THE RESULTS OF SOURCE/SINK EVALUATIONS

The results of source/sink evaluations can be used to predict concentration vs. time and to evaluate occupant exposure.

Predict Concentration vs. Time - IAQ models are used to calculate indoor concentrations, in time and space, based on: 1) source/sink behavior; 2) time of use, amount, and location of sources; 3) type, area, and location of sinks; 4) outdoor air exchange; 5) number and dimensions of rooms; 6) room-to-room air movements; and 7) HVAC system operation. As discussed above, IAQ test house experimental data can be used with IAQ models to validate the chamber-derived source emission rates and sink adsorption and desorption rates (8). Figure 1 is an IAQ model prediction of VOC concentrations (due to wood stain) in a test house.

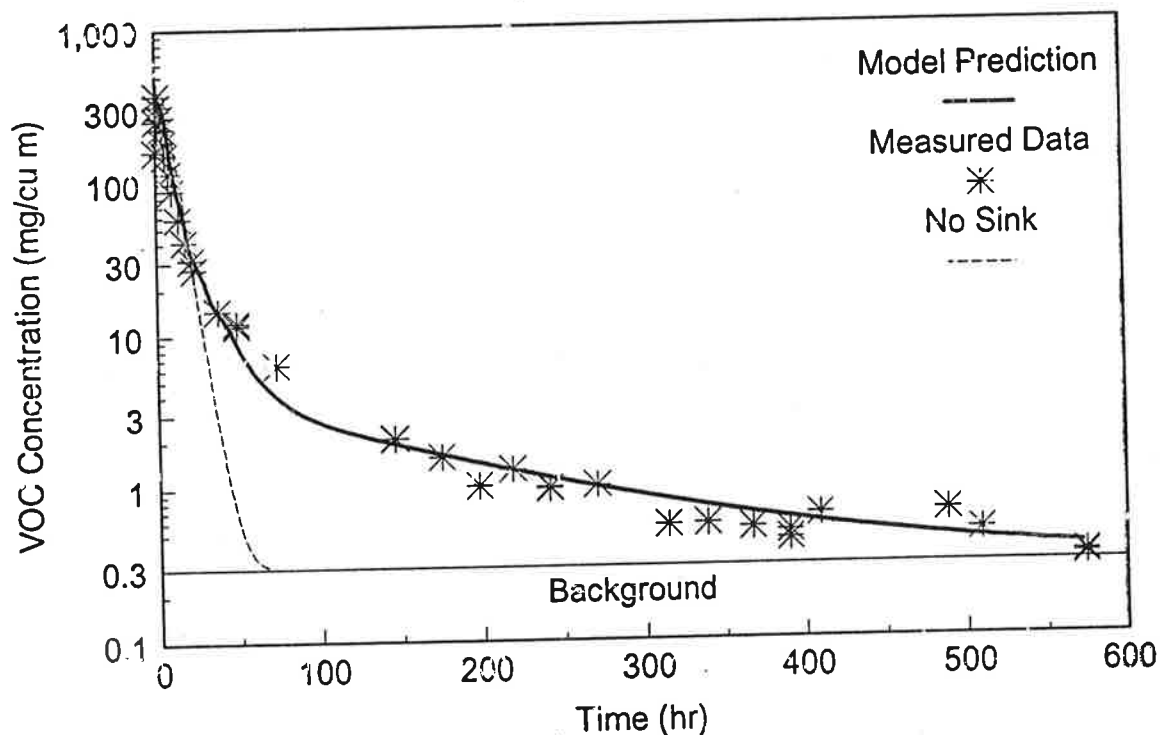


Figure 1. IAQ Model Predictions: Test House - Wood Stain

Several features of Figure 1 merit discussion:

1) IAQ models can accurately predict indoor concentrations if source and sink behavior is well defined and the indoor environment is well characterized. For the example shown in Figure 1, source and sink rates were based on dynamic chamber tests and the IAQ test house environment was well known, including measurements of outdoor air exchange during the experiment.

2) Indoor sinks can dramatically extend the time of elevated concentrations due to source emissions. The "No Sink" curve in Figure 1 shows that the concentration would be reduced to background in less than 100 hours in the absence of re-emissions from sinks. The actual data and the model predictions show that the concentrations exceeded background levels for at least 500 hours.

3) Indoor sources, especially wet sources, can produce vapor-phase organic

concentrations well above the levels presumed to cause irritation. In the experiment described by Figure 1 (where only 6 m² of oak flooring was stained in a house with a volume of 300 m³), concentrations of organic vapors reached several hundred mg/m³. In another study, concentrations of several thousand mg/m³ were measured after parquet wood floors were refinished with a two-component polyurethane coating (13). These concentrations are well above the 3 - 25 mg/m³ "discomfort range" and the > 25 mg/m³ "toxic range" proposed by Molhave (14).

4) Vapor-phase organic concentrations from wet sources vary widely over time. As shown in Figure 1 and reported by others (13), concentrations can change by several orders of magnitude in hours or days. This time varying behavior is not consistent with the constant concentration exposures used in evaluating irritation and other human health effects (15). In addition, even though the concentrations decay over time, they are well in excess of the concentration of total organics (25 mg/m³) for periods much longer than the exposure time (2.75 hr) of the human subjects evaluated by Otto and his colleagues (15). Thus, irritation and other effects are being evaluated at concentrations and exposure times well below those which are caused by the use of common indoor sources of vapor-phase organics.

Evaluate Occupant Exposure - By combining occupant activity patterns with concentration/time profiles, occupant exposure to source emissions can be estimated. The U. S. Environmental Protection Agency has developed an IAQ exposure model that is used to determine both instantaneous and cumulative individual exposure to vapor-phase organic compounds based on inhalation (16). Figure 2 shows the cumulative exposure calculated by the model for three people in the test house environment illustrated by Figure 1. Person A is assumed to have applied the wood stain over hours 0 - 4 and then left (still exposed to a background of 0.3 mg/m³); person B was in the house the whole time; and person C entered the house after the staining was complete (4 hours) and stayed.

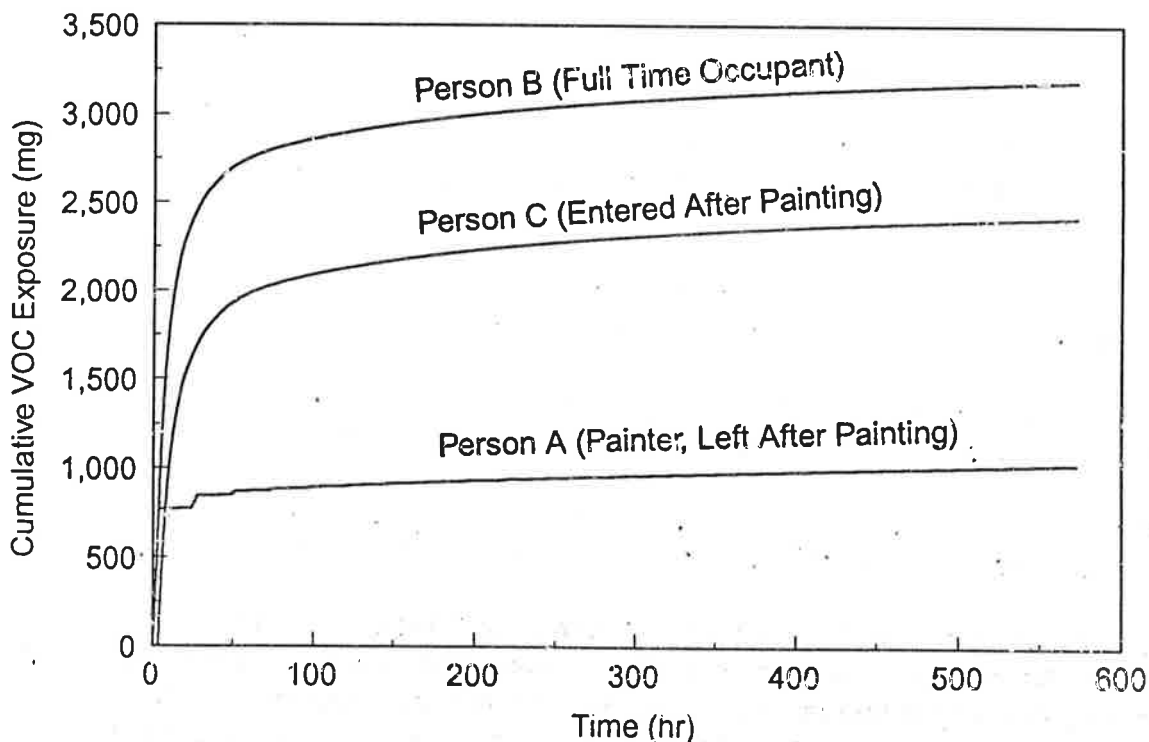


Figure 2. Exposure Predictions: Test House - Wood Stain

LINKING BIOLOGICAL RESPONSES TO SOURCE/SINK CHARACTERISTICS

So far, this paper has discussed methods for determining the emissions characteristics of indoor sources and sinks. Models for determining indoor concentrations and individual exposure by coupling the source/sink behavior to environmental parameters have also been addressed. The next obvious step in the source evaluation process is to link the impact of sources/sinks on IAQ to human health and comfort. Following is a brief discussion of how indoor sources and sinks might be linked to human health and comfort.

Use Available Health Effects Data - Data on the health effects of many chemicals are available. For example, the Registry of Toxic Effects of Chemical Substances (RTECS) supplies information on thousands of chemicals (17). Effects such as skin and eye irritation, mutation, reproductive effects, toxicity, and cancer are presented for a variety of exposure scenarios, including inhalation. The data in RTECS are presented for single compounds and are based, for the most part, on animal exposures to constant concentrations.

Use Published Guidance - Several organizations publish guidance on exposure to indoor air pollutants. The American Conference of Governmental Industrial Hygienists (ACGIH) publishes occupational exposure guidance for a number of chemical substances and physical agents (18). While the ACGIH guidance is widely used by industrial hygienists, its application to non-industrial environments is questioned by many in the IAQ research community. Guidance specific to IAQ is available from the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) (19), the World Health Organization (WHO) (20), and the Canadian government (21). The guidance published by these organizations is for specific chemicals and constant concentration. Some guidance is provided for dealing with mixtures (18).

SOURCE TESTING METHODS -- BIOLOGICAL RESPONSES

Due to several factors, using guidance on indoor concentrations to suggest limits on source emissions provides only an indirect link between these emissions and a biological response:

- 1) Guidance on indoor concentrations is generally limited to single compounds, while indoor source emissions usually involve complex mixtures.
- 2) Guidance on indoor concentrations is based on averages over a given time interval, while most indoor source emissions vary over time (e.g., see Figure 1).
- 3) Data on many of the health effects endpoints (e.g., sensory irritation) do not exist for a majority of the compounds emitted from many indoor sources.

In order to overcome these limitations, testing methods are needed to directly determine the biological response to source emissions. Such biological response based methods might include, for example: 1) gas-phase bioassays using microorganisms; 2) animal tests for sensory irritation (e.g., mouse respiration) (22), inhalation toxicity, and eye and skin irritation; and 3) human evaluations of sensory

reaction (e.g., Fanger's olf panels) (23), respiratory irritation (e.g., jar tests by sensitive individuals), skin and eye irritation, and a wide variety of health and comfort effects via exposure chamber studies.

CONCLUSIONS

Much is known about the behavior of indoor sources and sinks, and methods are available, or are being developed, for determining the source/sink chemical emission rates. IAQ models can use these rates to predict indoor concentrations of the pollutants emitted. Unfortunately, the available information on human health and comfort is not consistent with the complex behavior of indoor sources (e.g., multiple pollutants and time varying emissions). Thus, methods are needed to directly link source/sink emissions to human health and comfort.

REFERENCES

1. Tichenor, B. 1992. Characterizing Material Sources and Sinks: Current Approaches. in: Sources of indoor Air Contaminants - Characterizing Emissions and Health Impacts, Annals of the New York Academy of Sciences. Vol. 641: 63-78.
2. Colombo, A., M. De Bortoli, H. Knöppel, H. Schauenburg & H. Vissers. 1990. Determination of volatile organic compounds from household products in small test chambers and comparison with headspace analysis. Indoor Air '90, Toronto, Proceedings of the 5th international Conference on Indoor Air Quality and Climate. Vol. 3: 599-604.
3. Tichenor, B. 1989. Indoor Air Sources: Using Small Environmental Test Chambers to Characterize Organic Emissions from Indoor Materials and Products. EPA-600/8-89-074 (NTIS No. PB90-110131/AS), U. S. Environmental Protection Agency, Research Triangle Park, NC.
4. Black, M. 1990. Environmental chamber technology for the study of volatile organic compound emissions from manufactured products, Indoor Air '90, Toronto, Proceedings of the 5th international Conference on Indoor Air Quality and Climate. Vol. 3: 713-718.
5. American Society for Testing and Materials. 1991. Standard Guide for Small-Scale Environmental Chamber Determinations of Organic Emissions from Indoor Materials/Products. ASTM D5116-90, Philadelphia, PA.
6. Commission of the European Communities. 1991. Guideline for the Characterization of Volatile Organic Compounds Emitted from Indoor Materials and Products Using Small Test Chambers. COST Project Report No. 8. Office for Official Publications of the European Communities, EUR 13593, Luxembourg.

7. van der Wal, J., R. Steenlage & A. Hoogeveen. 1990. Measurement of organic compound emissions from consumer products in a walk-in test chamber, *Indoor Air '90, Toronto, Proceedings of the 5th International Conference on Indoor Air Quality and Climate*. Vol. 3: 611-616.
8. Tichenor, B., L. Sparks, J. White & M. Jackson. 1990. Evaluating sources of indoor air pollution, *Journal of the Air and Waste Management Association*, 40(4): 487-492.
9. Borazzo, J., C. Davidson & J. Andelman. 1990. Sorption of organic vapors to indoor surfaces of synthetic and natural fibrous materials. *Indoor Air '90, Toronto, Proceedings of the 5th International Conference on Indoor Air Quality and Climate*. Vol. 3: 617-622.
10. Matthews, T., A. Hawthorne & C. Thompson. 1987. Formaldehyde sorption and desorption characteristics of gypsum wallboard. *Environmental Science and Technology*. 21(7): 629-634.
11. Tichenor, B., Z. Guo, J. Dunn, L. Sparks & M. Mason. 1991. The interaction of vapor phase organic compounds with indoor sinks. *Indoor Air*. 1(1):23-35.
12. Guo, Z. & B. Tichenor. 1992. Fundamental Mass Transfer Models Applied to Evaluating the Emissions of Vapor-Phase Organics from Interior Architectural Coatings. Presented at 1992 EPA/A&WMA International Symposium on Measurement of Toxic and Related Air Pollutants, Durham, NC.
13. Schriever, E. & R. Marutzky. 1990. VOC emissions of coated parqueted floors. *Indoor Air '90, Toronto, Proceedings of the 5th International Conference on Indoor Air Quality and Climate*. Vol. 3: 551-555.
14. Molhave, L. 1990. Volatile organic compounds, indoor air quality and health. *Indoor Air '90, Toronto, Proceedings of the 5th International Conference on Indoor Air Quality and Climate*. Vol. 5: 15-33.
15. Otto, D., L. Molhave, G. Rose, H. Hudnell & D. House. 1990. Neurobehavioral and sensory irritant effects of controlled exposure to a complex mixture of volatile organic compounds. *Neurotoxicology and Teratology*. 12: 649-652.
16. Sparks, L. & W. Tucker. 1990. A computer model for calculating individual exposure due to indoor air pollution sources. *Indoor Air '90, Toronto, Proceedings of the 5th International Conference on Indoor Air Quality and Climate*. Vol. 4: 213-218.
17. U. S. Department of Health and Human Services. 1987. Registry of Toxic Effects of Chemical Substances: 1985-86 Edition. D. Sweet, Ed. Public Health Service, Centers for Disease Control, National Institute for Occupational Safety and Health. Washington, DC.
18. American Conference of Governmental Industrial Hygienists. 1988. Threshold Limit Values and Biological Exposure Indices. Cincinnati, OH.

19. American Society of Heating, Refrigerating and Air-Conditioning Engineers. 1989. Ventilation for Acceptable Indoor Air Quality. ASHRAE Standard 62-1989. Atlanta, GA.
20. World Health Organization. 1987. Air Quality Guidelines for Europe. WHO Regional Publication, European Series No. 23. Copenhagen, Denmark.
21. Health and Welfare Canada. 1987. Exposure Guidelines for Residential Indoor Air Quality. Ottawa.
22. Alarie, Y., L. Kane & C. Barrow. 1980. Sensory irritation: the use of an animal model to establish acceptable exposure to airborne chemical irritants. In Toxicology: Principles and Practice. Vol. 1: 48-92. John Wiley and Sons. New York, NY.
23. Fanger, P. 1988. Introduction of the olf and the decipol units to quantify air pollution perceived by humans indoors and outdoors. Energy and Buildings. 12(1): 1-6.