Characterizing Sources of Indoor Air Pollution and Related Sink Effects

BRUCE A. TICHENOR
editor

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Foreword

The Symposium on Methods for Characterizing Indoor Sources and Sinks was held 25–28 September 1994, in Washington, DC. The symposium was sponsored by ASTM Committee D22 on Sampling and Analysis of Atmospheres and its Subcommittee D22.05 on Indoor Air. Bruce A. Tichenor formerly of the U.S. Environmental Protection Agency served as both chairman of the symposium and editor of this publication.

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Overview

Exposure to indoor air pollution is recognized by environmental professionals as a major contributor to human health risk. Levels of pollutants, especially volatile organic compounds (VOCs), are generally higher indoors than outdoors. Since people spent 80 to 90% of their time indoors, these elevated pollutant levels cause high exposures. Depending on the specific pollutants, these exposures may increase the risk of acute and chronic health effects. In order to quantify these increased risks, data and information are needed on the sources of indoor air pollutants.

Indoor air pollutants are emitted by a variety of products and materials. Since the early 1980s, scientists and engineers have been developing methods for characterizing these emissions. As the research on sources advanced, attention was also placed on the interaction between the source emissions and other indoor surfaces (or sinks). It was found that VOCs can be adsorbed on sink surfaces and later re-emitted. Thus, understanding both source and sink behavior is important for fully describing the exposures to indoor air pollutants.

As the research on sources and sinks advances, it is essential that information transfer within the scientific community be maximized. Thus, in September of 1994 a "Symposium on Methods for Characterizing Indoor Sources and Sinks" was held in Washington, DC. The symposium, sponsored by ASTM Committee D22 on Sampling and Analysis of Atmospheres and its Subcommittee D22.05 on Indoor Air, brought together over 100 researchers and practitioners representing public and private interests in the United States, Canada, and a number of European countries. The purpose of the symposium was to review and discuss approaches for characterizing emissions from indoor materials and products, including the interaction of these emissions with indoor sinks. Papers were presented by indoor air quality (IAQ) researchers and practitioners covering current approaches, as well as methods under development. The symposium was designed to provide a forum for discussion among IAQ professionals involved in evaluating indoor sources and sinks. This Special Technical Publication (STP) contains papers presented at the symposium. The papers contained herein are more detailed and comprehensive that the oral and poster presentations from which they were developed. Thus, the STP provides an opportunity for in-depth examination of the issues presented and discussed at the symposium. It is intended to benefit the symposium attendees as well as the wider audience interested in indoor sources and sinks.

The first paper in the STP provides the views of the symposium chairman on the status of source and sink test methods. He reviews the types of methods and facilities typically used to determine emissions from indoor sources, as well as innovative approaches. Protocols for evaluating indoor sink adsorption and desorption processes are also discussed, with a focus on volatile organic compounds (VOCs). Emphasis is placed on developing models for predicting VOC emissions. IAQ models used to predict the exposures of building occupants to VOC emissions are demonstrated and discussed. The paper concludes with a plea for standardization of test methods.

The main body of the STP is organized in four sections:

1) Test Chambers and Facilities,
2) Testing Methods and Protocols,

3) Models for Predicting Source and Sink Behavior, and
4) Interpretation and Application of Test Results.

The remainder of this overview provides summaries and highlights of the papers in each of these sections.

Test Chambers and Facilities

This section contains papers on the design, construction, characterization, and operation of source and sink testing facilities. A wide variety of test facilities are described and discussed. The chamber sizes span six orders of magnitude, ranging from the 3.5×10^{-5} m^3 FLEC micro-chamber to a 55 m^3 large dynamic chamber. Systems to evaluate emissions of several classes of indoor pollutants, including VOCs, particles, and biocontaminants, are presented. While most of the systems presented use chemical or physical measurements to determine emissions, a chamber designed to be used in conjunction with biological response (mouse bioassay) is also presented.

VOC emissions from many sources are controlled by gas-phase limited mass transfer processes. For these sources, the velocity at the surface of the source is an important variable. Zhang and Haghighat describe a novel, flow-through test chamber with provisions for controlling the velocity and level of turbulence. The 0.02 m^3 stainless steel chamber uses two fans for air flow control. Data are provided that demonstrate the regulation of velocity and turbulence at selected levels. The effect of velocity on the evaporation rate of water is also demonstrated. This paper highlights the need to test materials using reasonable mass transfer regimes.

Four papers provide information on the design and operation of large, dynamic test chambers. Lawless et al. present test results from an 8 m^3 chamber, including particle emissions from vacuum cleaner operation and VOC emissions from a powdered carpet cleaner. The paper presents a mathematical technique (deconvolution) for separating the temporal patterns of the source and chamber in order to better quantify the source emissions. A large 55 m^3 chamber designed for measuring VOC emissions from building materials and other indoor products is described Zhang et al. The chamber design includes provisions for control of air velocities and distributions to simulate various ventilation scenarios. Buttner and Stetzenbach describe a 35 m^3 experimental room designed to study airborne particles and microorganisms. They report on a study of glass fibers released from fiberglass ductboard. Data on the effectiveness of four fungal spore collectors are presented, along with data on the dispersal of fungal spores due to vacuuming a contaminated carpet. A 14.5 m^3 chamber developed for studying biocontaminant growth and dispersion, as well as particle air cleaners, is described by VanOsdell et al. The chamber, constructed inside a clean room, includes a ventilation system designed to prevent escape of the test microorganisms. Chamber decontamination procedures are discussed.

Static chambers (0.06 m^3) designed to evaluate biocontaminant growth are described by Foarde et al. The paper presents a test method submitted to ASTM as a "Standard Guide for Evaluating the Ability of Indoor Materials to Support Microbial Growth." Data are presented on the effect of humidity on the growth of *Penicillium glabrum* mold on ceiling tiles and on the tile moisture content. Humidity control is achieved by saturated salt solutions.

Two papers provide information on very small chamber systems. Roache et al. present the results of a study comparing the Field and Laboratory Emission Cell—FLEC (3.5×10^{-5} m^3) to traditional dynamic chambers (0.05 m^3) when both were used to determine VOC emissions from floor wax on a glass plate and latex paint on gypsumboard. The emission rates of total VOC (TVOC) from dried floor wax compared well between the two chambers. For latex paint, differences in emission rates were observed between the two system. The discrepancies were most pronounced for ethylene glycol, a major VOC component. The differences were due, in part, to the low velocity and high air exchange rate in the FLEC which provide a mass transfer regime quite different from the small chamber. Leigh presents the results from an evaluation of a 50 mL (5×10^{-5} m^3) flow through tube packed with insulation material. Clean, humidified air is passed through the tube and collected for measurement of formaldehyde and other VOCs. Emission rates are estimated based on the mass of pollutants collected. While the emission rates determined from this system are only qualitatively comparable to data from more traditional methods, this technique can be used as a screening tool to compare emissions among similar products.

While most indoor air source characterization systems provide data on the chemical (for example, VOC) emissions, Mason et al. present a chamber system designed to be used in conjunction with a mouse bioassay (ASTM E 981-84) for determining the irritation potential of the emissions. The paper describes a 0.034 m^3 flow-through glass chamber that contains the material to be evaluated. This source chamber is connected directly to a 0.002 m^3 chamber where the mice are exposed to the emissions. The source chamber is leak free and has no detectable background VOC emissions. It was designed as a replacement for a crude fish aquarium "chamber" used to evaluate carpet emissions. The aquarium "chamber" had significant leakage and produced high background emissions.

Testing Methods and Protocols

This section presents information on experimental methods and protocols for determining emission factors and for evaluating sink adsorption and desorption rates. Papers are presented on the effect of experimental variables on emissions, and recommendations are provided for improving existing test methods. Sink testing protocols are discussed, including an examination of particles caused by re-emitting sinks. Several methods used to determine formaldehyde emissions from wood products are discussed and compared.

Information on the effect of experimental variables on VOC emissions is presented in three papers. Jensen et al. examine the effect of the test atmosphere on emissions from linoleum. Using a FLEC, linoleum was exposed to air and nitrogen. Relative to nitrogen, air exposure resulted in elevated levels of fatty acids. The authors conclude that this is caused by the oxidation of the aldehydes emitted from the linoleum. The importance of the velocity over the test surface is discussed by Zhang et al. They present data on the effect of velocity and turbulence on the emission rate of VOCs from wood stain applied to an oak board substrate. Using a gas-phase limited mass transfer model, the chamber data were used to obtain a mass transfer coefficient. Consistent with applicable theory, the mass transfer coefficient increased with increasing velocity. The paper by Guo et al. provides recommendations on how emissions testing protocols can be improved by properly accounting for the important experimental parameters, including velocity, test specimen size, and the time required to apply wet materials. A method for calculating emission factors directly from dynamic chamber data is presented. Finally, the authors recommend the development of "standard" emission sources to allow intercomparison of different test chambers.

Protocols for evaluating sink behavior are presented in two papers. Kjaer et al. describe an experiment where two test substrates (latex painted steel and carpet) were placed in a CLIMPAQ chamber and exposed to air from an office renovated six months earlier. The re-emissions of VOCs in the office air from the test substrates was measured by GC, and a model based on retention time (or boiling point) is proposed. A sensory evaluation of the sink re-emissions was also conducted. The authors conclude that the protocol needs further validation. A unique experiment involving particles from re-emitting sinks is presented by Johansson. He reports an increase in concentration of small particle in a test room after exposure to sidestream (SS) tobacco smoke. The author hypothesizes that the particles are caused by vaporization of gases from the walls followed by gas-to-particle conversion. This sink re-emission could increase the exposure time to SS tobacco smoke.

Two papers present techniques for evaluating contaminated indoor surfaces. Cole et al. describe a building study where concentrations of airborne bioaerosols (fungi and bacteria) were compared to surface samples collected from non-floor horizontal surfaces (e.g., table tops) and tile and carpet floor surfaces. Positive correlations were shown between: a) airborne fungi and bacteria and those collected from non-floor surfaces and b) carpet dust bacteria and airborne bacteria. The effect of deep carpet cleaning on airborne contamination is also reported. Vaccaro and Murphy present an original study on the exposure of children to the insecticide chlorpyrifos. The method involves dragging a weighted "sled" across a contaminated carpet or residential lawn to simulate crawling children. Exposure estimates caused by dermal contact, inhalation, and ingestion are provided. These estimates are used to calculate a child's dose of chlorpyrifos for both outdoor (turf) and indoor (carpet) activities.

Methods for determining formaldehyde emissions from pressed wood products are described in two papers. Liles et al. compare data from evaluations performed by two laboratories using different dynamic chamber test methods. One laboratory (lab A) used a 0.052 m^3 stainless steel chamber; the other (lab B) a 0.044 m^3 aluminum chamber. Different air exchange rates (N), loadings (L), and N/L ratios were also used. Two different analytical techniques were employed to measure formaldehyde emissions, and a wide variety of wood products were tested. The authors concluded that the systems did not compare well with respect to formaldehyde emissions. Crump et al. report on a study comparing three methods for measuring formaldehyde emission rates from particleboard and medium density fiberboard (MDF): a) the Dombey test using a 0.00445 m^3 chamber b) a 1 m^3 test chamber, and c) a 2.4 liter (0.0024 m^3) mini-chamber. All testing was conducted by the same lab using the same analysis method for formaldehyde. The study demonstrated good comparability between the three methods.

Models for Predicting Source and Sink Behavior

Models that predict the behavior or sources and sinks are needed as input to IAQ models used to predict exposure of indoor occupants to source emissions. Empirical source emissions models (for example, first-order decay) can be developed directly by fitting chamber test data. Fundamental models are developed for mass transfer processes: evaporation and diffusion for sources; adsorption and desorption for sinks. Chamber test data can be used to develop the appropriate coefficients for these mass transfer models. The papers in this section deal with these issues as well as providing insight into other IAQ modeling concerns.

Two papers by Evans explore mathematical techniques for developing IAQ and source emissions models. In "Linear Systems, Compartmental Modeling, and Estimability Issues in IAQ Studies" he applies linear system analysis techniques used in the field of biomathematics to typical IAQ modeling scenarios. The paper discusses how fitting an improper model to experimental data can provide redundant (non-unique) parameter estimates. Thus, one needs to select the right model *before* the experiment is conducted. In his other paper, "Development of Continuous-Application Source Terms and Analytical Solutions for One- and Two-Compartment Systems," Evans describes a set of models to be used for sources (for example, wall paint) that are applied over surfaces in finite periods of time. The paper also provides mathematical tools that allow these source models to be integrated into one or two compartment IAQ models.

Mage and Ott deal with the issue of nonuniform mixing in indoor environments and how it affects human exposure. First, they discuss the approach that uses an incomplete mixing factor and conclude that it fails to meet the conservation of mass principle. A new model is proposed that defines the indoor conditions during three time periods related to an episodic source: t_α, t_β, and t_γ; where t_α is the time the source is emitting, t_β is the time after the source stops but the concentration distribution is the space is nonuniform, and t_γ is the time from when the concentration becomes uniform to when it becomes nondetectable above background. The authors show that an assumption of uniform mixing (the "standard" assumption in most IAQ models) is reasonable if $t_\gamma \gg (t_\alpha + t_\beta)$. Experimental data are provided to support this conclusion.

Mass-transfer models for predicting source and sink behavior of carpet backing material are presented by Little and Hodgson. The models, applied to VOC diffusion within the polymer material, are based on fundamental theory and are validated with data from large chamber emissions tests of carpet. The authors propose the use of micro-balance experiments as a direct method of determining source emissions and sink adsorption/desorption. These simple experiments, coupled with data available in the literature, could supplant the more expensive and time consuming chamber test methods.

The paper by De Bortoli et al. presents models that describe the sink effect in small dynamic test chambers. They describe a new model that accounts for both fast and slow sinks. Data are presented from static and dynamic chamber tests using several VOCs in empty chambers, as well as typical indoor sink materials (carpet, vinyl wall covering, gypsumboard). The results show that the small sink effect of an empty chamber (glass or stainless steel surfaces) can be neglected when the chambers are used to evaluate typical indoor sinks.

Interpretation and Application of Test Results

The final section contains papers that provide insight on how test results can be used to make decisions on selecting indoor sources based on their emissions characteristics. Several approaches are discussed, including: bioresponse testing, product labeling, risk assessment, simplified screening methods, and product classifications based on exposure estimates.

Two papers discuss the evaluation of source emissions based on the sensory response (bioresponse) of the exposed individual. Tucker et al. provide a review of the US EPA research program on bioresponse based testing, with an emphasis on odor and sensory irritation of the eyes, nose and upper airways. They point out the limitations in traditional chemical testing methods and propose a higher level of cooperation between engineers, chemists, and biologists. The authors conclude that additional research is needed to develop practical bioresponse evaluation tools. This conclusion is supported by the paper of Hempel-Jørgensen et al. which describes a system and test protocol for measuring human eye exposure to determine sensory irritation. The paper reports on experiments conducted using CO_2 as a reference gas and concludes that further development of the method is needed before it can used to predict the potential of an indoor source to cause sensory irritation.

Two papers present different approaches for selecting indoor materials based on their emissions. Wolkoff and Nielsen report on a process for labeling building materials based on the time that their emissions can cause VOC concentrations above a known odor or irritation threshold. This method combines chemical emissions data (from FLEC evaluations) and odor and irritation responses based on human or animal testing. Source emissions models are used with standard exposure scenarios to predict exposure times. A more comprehensive, sequential approach is presented by Levin and Hodgson. They propose an initial screening test, using headspace analysis and a single 24 hour dynamic chamber test, that is used to identify emissions of compounds known to cause odor, irritation, toxic effects, or cancer. Materials "passing" such an evaluation would not require further evaluation. The materials with significant emissions based on the screening study would be subjected to more thorough evaluation, including chamber testing and model predictions of exposure.

Two papers are presented on the relationship between source emissions and risk assessment. Sparks et al. address source testing data requirements needed to conduct assessments of occupant exposure and risk. Models for conducting exposure assessments are discussed. Suggestions are made for dealing with poorly characterized sources, including recommendations on how to conduct chamber studies on sources with long term emissions. Johnston et al. review the US EPA's Indoor Air Source Characterization Project that is designed to score and rank classes of indoor products based on their potential as sources of exposure and risk. The method uses source emissions data in modeled exposure scenarios designed to represent typical product usage to assess both chronic and acute risk. The results will be used in a Source Ranking Database to allow comparison among classes or groups of indoor products.

The papers presented in this book were prepared by the world's leading experts on indoor sources and sinks. The reader will find information on a wide variety of testing facilities, experimental methods, and modeling techniques. Guidance is also presented on how to use the data derived from source/sink evaluations. The symposium committee sincerely appreciates the contributions of the authors and ASTM staff that led to a successful symposium and made possible this valuable publication.

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Overview of Source/Sink Characterization

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Overview of Source/Sink Characterization Methods

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ABSTRACT: Methods for characterizing indoor sources and sinks are continuously being developed and improved. Test protocols (e.g., for chamber tests) are needed to specify equipment requirements, testing conditions, and data analysis methods. Protocols are being published by standards-setting organizations (e.g., ASTM), international groups (e.g., CEC), and individual countries (e.g., Denmark). Both empirical models and fundamental mass transfer models are being developed to predict the emission behavior of sources and sinks. These models can be validated using dynamic chamber data from properly executed experiments. Validated source emissions models can be used to evaluate options for material selection or labeling based on chemical emission characteristics and known human responses to these chemicals. Another approach involves measurement of human sensory response and animal irritation response to identify sources with potential problems. Chamber systems that combine chemical emissions determinations with sensory response have also been developed. Use of emissions testing to evaluate and select indoor materials and products is expected to increase as test methods become standardized and assessment techniques are agreed upon.

KEYWORDS: indoor sources, indoor sinks, source characterization, chamber tests, test methods, test protocols, empirical models, mass transfer models, source emission models

Source/Sink Characterization Systems and Methods

Systems and devices for characterizing indoor sources and sinks are continuously being developed and improved. Traditional methods for measuring emission rates of chemical contaminants employ dynamic, flow-through chambers [*1*]. Both large (room-size) and small (<5 m^3) chambers are used. Test protocols that specify equipment requirements, testing conditions, and data analysis methods are available, such as the ASTM Standard Guide for Small-Scale Environmental Chamber Determinations of Organic Emissions from Indoor Materials/Products (ASTM D 5116). Other approaches involve direct exposure of humans or animals to source emissions [2]. Table 1 illustrates the variety of methods being used by indoor air quality (IAQ) investigators to determine the chemical emissions from indoor materials and furnishings [*3*].

Laboratory Studies

Relatively simple laboratory studies can be conducted to determine material composition and emissions composition.

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TABLE 1—*Source testing methods.*

Laboratory Studies
Extraction and direct analysis:
Provide information on material composition
Do not provide emissions composition or emissions rate data
Static headspace:
Provides information on emissions composition
Does 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 emissions and sink re-emissions extremely difficult

Extraction—Solvent extraction (e.g., using methylene chloride) of dry indoor materials can furnish information on the organic chemicals contained in the material. Analysis of the extract by gas chromatography (GC) with mass spectrometry (MS) will identify the compounds, including the nonvolatile and semivolatile species. Such techniques have been used to evaluate the composition of carpet samples [*4*].

Direct Analysis—Some wet products (e.g., paints and other coatings) can be analyzed to determine their composition. Techniques are available for determining the mass of total volatile organic compounds (VOCs) or individual compounds per volume of coating. Generally, these techniques involve: (1) evaporation to dryness, or (2) dilution followed by GC analysis. EPA Reference Method 24 and associated American Society for Testing and Materials (ASTM) methods [*5*] are examples of such techniques.

Static Headspace—The composition of emissions from indoor materials can often be determined using static headspace analysis [*6*]. In this procedure, a sample of material is placed in a small (e.g., 1 L), airtight container lined with inert material. Samples of the air inside the container (i.e., headspace) are analyzed to determine the compounds emitted by the material. Static headspace analyses are normally conducted at ambient temperature (e.g., 23°C) and atmospheric pressure. In some cases, investigators use higher temperatures to increase the emission rate to ensure a high enough concentration to analyze. Also, inert gas, flow-through headspace analyses may be conducted on low-emitting materials (e.g., carpet) to provide an increased sample size [*4*].