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CHARACTERIZATION OF PARTICULATE AND ORGANIC EMISSIONS FROM MAJOR INDOOR SOURCES

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Abstract

As our understanding of human exposure to air pollutants improves, it is becoming increasingly evident that indoor environments play a critical role in determining exposures. However, it is not possible at the present time to establish the relative contribution of indoor and outdoor sources to personal exposures, nor can the contribution of specific indoor emissions be quantified. To address these issues, a chamber experiment was initiated to measure particulate and organic emissions from important indoor sources. Data on particle size distributions, morphology, mutagenicity, and elemental and chemical composition, as well as information about volatile organic emissions, were collected for each source. Results of the study will be used to determine the feasibility of using source-receptor techniques for indoor source apportionment.

Introduction

Available information indicates that concentrations of many pollutants are routinely elevated in nonindustrial indoor environments. It is known, for instance, that indoor respirable particle (RSP) levels are often significantly higher than corresponding outdoor values. Similarly, a broad spectrum of toxic and hazardous air pollutants have been measured at greater concentrations indoors than outdoors. While more data are needed, evidence on hand suggests that for a large segment of the U.S. population, exposure to many air pollutants is due primarily to indoor, rather than outdoor sources.

An understanding of the chemical and physical properties of indoor air contaminants as well as their origins is a necessary first step in developing effective mitigating measures and in designing appropriate control strategies. Sources need to be identified and the contribution of each to measured contaminant concentrations must be quantified. Two categories of source-apportionment methods have been applied to outdoor air: dispersion techniques and source-receptor techniques. In the former case, emission rates and meteorological parameters are combined with a mathematical dispersion model to predict concentrations downwind.

In the latter instance, receptor models are used to calculate the contribution of various sources to measured pollutant levels at specific monitoring sites.

While both approaches are theoretically applicable to indoor air pollution, the source-receptor technique is potentially more useful. In order to apply the dispersion method, data are required about indoor source strengths, ventilation rates, indoor mixing conditions, physical and chemical removal rates, and outdoor pollutant concentrations. Unfortunately, such detailed information is rarely, if ever, available. The source-receptor method, on the other hand, uses actual air measurements and knowledge of emission characteristics from individual sources to calculate contributions from various emission categories. To apply this approach, it is useful to have detailed data on the nature of emissions from each potential source. In addition, emissions must differ in some respect from source to source in order to distinguish between them. The calculation generally involves the assumption that emission components combine linearly at the receptor (i.e., conservation of mass).

This paper presents preliminary results from a chamber study designed to characterize particulate and organic emissions from major indoor sources. Information obtained from this research project will be used to construct a "source signature" for individual sources; an essential first step towards development of a generalized receptor model to identify and apportion indoor aerosols and vapors.

Experimental

Individual sources (i.e., cigarette smoking, gas-cooking stove) were placed in an environmental chamber and operated periodically for 4-6 hours. Data were collected on particle size distributions, morphology, mass concentrations, organic and elemental carbon, mutagenicity, and elemental and organic composition. Gaseous organic compounds were collected on Tenax and subsequently subjected to thermal desorption and analysis by gas chromatography/mass spectrometry (GC/MS). A summary of sampling parameters and analytical techniques is given in Table 1.

Experiments were carried out at the Indoor Air Quality Research House located at the University of California Richmond Field Station. The research house is a two-story, wood frame building, which contains a 128 m^3 three-room environmental chamber. Extensive weatherization has reduced air infiltration in one room to below 0.1 air changes per hour. Individual emission sources were isolated in that room and closed off from the rest of the chamber. The interior of the testing room measures 3.4 m x 4.6 m x 2.3 m and is constructed of plasterboard (3 walls and ceiling) and plywood sheeting (1 wall). The floor is covered with sheet vinyl and all surfaces have been painted. Temperature and relative humidity probes are located near the center of each room. Simultaneous measurements of all parameters listed in Table 1 were made in the testing room and in the outer enclosure.

Table 1. S

Parameter Measured	
PAH composi airborne pa (< 15 µm d$_a$	
Organic/ele total carbo composition airborne pa (< 15 µm d$_a$	
Mutagenicit associated airborne pa (< 2.5 µm d	
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Table 1. Summary of In-Chamber Air Measurements and Analytical Methods.

Parameter Measured	Sampling Medium	Sampling Rate	Analytical Methods
PAH composition of airborne particles (< 15 μm d_a)	Fiber glass filter	8 Lpm	High performance liquid chromatography
Organic/elemental/ total carbon composition of airborne particles (< 15 μm d_a)	Quartz/silica filter	8 Lpm	C_T by coulometric analyzer C_E by reflectance & light absorption C_O by difference
Mutagenicity associated with airborne particles (< 2.5 μm d_a)	Teflon filter	50 Lpm	Modified Ames test
Morphology and elemental composition of individual particles (> 1 μm d_a)	Nuclepore filter	14 Lpm	Computer-controlled scanning electron microscope with energy-dispersive x-ray analysis
Morphology and elemental composition of individual particles (< 1 μm d_a)	Beryllium/ carbon grids (collection by electrostatic precipitator)	6 Lpm	Computer-controlled scanning electron microscope with energy-dispersive x-ray analysis
Volatile organic compounds	Tenax tubes	0.5 Lpm	Gas chromatography/ mass spectrometry

Instruments installed in the environmental chamber provide real-time data on particle concentrations and important environmental parameters and allow for programmable control over critical experimental parameters. Instrumentation used to measure airborne particles includes; a condensation nuclei counter, an electrostatic classifier (particle size and concentration data $0.01 < d_a < 0.3$ μm), an optical particle counter (particle size and concentration data $0.1 < d_a < 3$ μm) and a Piezobalance real-time aerosol monitor (mass concentration).

The two indoor combustion sources examined were a gas cooking stove (natural gas) and tobacco smoking. The gas range was located along one wall and operated by a computer located outside the chamber. A cigarette-smoking machine was positioned near the middle of the room. The duration of cigarette combustion was controlled by a timer that extinguished the cigarette after a preset interval (usually 6 minutes).

To obtain sufficient quantities of material for analysis, the gas range was operated for 15 minutes every half hour. Two burners were turned on simultaneously and both were covered with aluminum plates to quench flame temperatures. During the course of gas range experiments (4-6 hours), temperatures in the chamber typically increased from 20°C to 34°C and relative humidity increased from 60% to 70%. In-chamber particle concentrations remained relatively high throughout each run, with values consistently $\geq$ 50,000 particles per cm^3.

The cigarette smoking machine was connected to an external timer, so that after manual ignition of the cigarette, the chamber was not entered again during the testing sequence. A standard smoking rate of two 35 mL puffs per minute was used and both "mainstream" and "sidestream" tobacco smoke was emitted directly into the testing room. A typical six-minute cigarette burn consumed ~ 600 mg of tobacco and produced a peak concentration of 100,000 to 200,000 particles per cm^3. This corresponds to a peak mass concentration of ~ 400 $\mu g/m^3$.

Results and Discussion

Results of carbon, mutagenicity, and elemental analyses are summarized in Table 2. Values given are for matched samples inside and outside the testing room (chamber) on February 1, 1984 (gas range) and February 7, 1984 (cigarette smoke). Data on PAH and volatile organic compounds were not available as of this writing.

Measured carbon concentrations given in Table 2 show that both organic and elemental carbon levels were higher outside the chamber during the gas stove experiment. Infiltration of outside air establishes baseline concentrations inside the chamber. Therefore, lower in-chamber values indicate that gas stove emissions are not a significant source of either particle phase organic or elemental carbon. The situation was similar for elemental carbon from cigarette smoke. Tobacco smoke is definitely a source of particle phase organic carbon, with in-chamber levels seven times higher than outside values.

Mutagenic density (revertants per m^3) in *Salmonella typhimurium* strain TA98 was determined for particulate extracts using a microsuspension Ames test. A positive response without S9 (-S9 in Table 2) represents the effects of direct-acting mutagens, such as alkylating agents and many nitro-PAH, that are mutagenic in the absence of mammalian metabolic activity. The addition of a rat liver extract (+ S9 in Table 2) is used to test for indirect-acting mutagens (promutagens), such as benzo(a)pyrene, whose metabolites are mutagenic.

As shown in Table 2, mutagenicity values were comparable inside and outside the chamber for the gas stove experiment, suggesting that particulate emissions are not highly mutagenic. During the cigarette smoke experiment, the -S9 value was lower in the chamber, while the mutagenicity value for +S9 was significantly elevated. These data

nd Analytical Methods.

Analytical Methods
High performance liquid chromatography
C_T by coulometric analyzer
C_E by reflectance & light absorption
C_O by difference
Modified Ames test
Computer-controlled scanning electron microscope with energy-dispersive x-ray analysis
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confirm previous studies which show that passive tobacco smoke ("sidestream" plus "mainstream" emissions) is a source of airborne particulate matter containing indirect-acting mutagens.

Table 2. Summary of Carbon, Mutagenicity, and Elemental Analyses for Gas Stove and Cigarette Smoke Emissions.

Parameter Measured	Gas-Cooking Stove		Cigarette Smoke	
	Outside the Chamber	Inside the Chamber	Outside the Chamber	Inside the Chamber
CARBON (μg/m³)				
organic	24.4	18.8	27.0	195
elemental	2.3	1.3	10.4	11.9
total	26.7	20.1	37.4	207
MUTAGENICITY (rev/m³)				
- S9	175	215	245	85
+ S9	125	100	270	1870
% PARTICLES CONTAINING SPECIFIC ELEMENT				
Na	3.0	4.4	2.6	3.2
Mg	1.3	2.0	2.6	1.1
Al	9.1	8.9	9.5	6.0
Si	8.5	59.1*	6.8	11.3
P	0.2	--	0.5	0.2
S	3.2	11.3	1.1	1.2
Cd	2.0	2.0	0.5	0.4
K	2.2	3.5	2.1	3.0
Ca	0.4	--	1.1	0.2
Fe	1.3	--	1.1	--
Pb	--	--	--	--

*probable instrument-induced artifact

Elemental composition of individual particles was determined by energy-dispersive x-ray analysis. Data for selected elements are presented in Table 2. The percentage of particles in the chamber containing sodium, magnesium, aluminum, phosphorus, cadmium, potassium, calcium, iron and lead was not significantly different than outside. Although silicon-containing particles were elevated in the chamber, extremely low concentrations were measured in a large number of

particles. It is therefore likely that observed values are the result of instrument-induced artifact and do not represent the contribution of gas combustion. Sulfur-containing particles, in contrast, were definitely a component of gas stove emissions. Since sulfur is a constituent of natural gas, it is reasonable to expect that combustion will produce particles containing sulfur.

Because a major fraction of tobacco smoke is composed of liquid aerosol, single particle analysis by automated SEM techniques will be unable to detect elevated elemental constituents in most instances. Thus particles measured during the cigarette smoke experiment are likely to represent background values. This is confirmed by the similarity of inside and outside measurements on February 7, 1984.

Conclusions

Preliminary findings from a chamber study indicate that definite differences exist between particulate emissions from a gas cooking stove and cigarette smoking. Gas stove emissions (natural gas) are not a significant source of elemental and particle phase organic carbon, are not highly mutagenic in a modified Ames test, and are a source of sulfur-containing particles. Cigarette smoke is high in particle phase organic carbon, and tests positive for mutagenicity on the +S9 modified Ames test.

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