United States
Environmental Protection
Agency

Environmental Monitoring
Systems Laboratory
Research Triangle Park, NC 27711

Research and Development

EPA/600/S4-88/028 Sept. 1988

EPA

Project Summary

Field Evaluation of Sampling and Analysis for Organic Pollutants in Indoor Air

J. C. Chuang, G. A. Mack, J. W. Stockrahm, S. W. Hannan, C. Bridges, and M. R. Kuhlman

The objectives of this study were to determine the feasibility of the use of newly developed indoor air samplers in residential indoor air sampling and to evaluate methodology for characterization of the concentrations of polynuclear aromatic hydrocarbons (PAH), PAH derivatives, and nicotine in residential air.

Residential air sampling was conducted in Columbus, Ohio, during the winter of 1986/87. The residences were selected on the basis of these characteristics: electric/gas heating system, electric/gas cooking appliances, and amount of cigarette smoking in the home. The indoor air sampler was equipped with a quartz fiber filter to collect particles followed by XAD-4 resin to trap vapors. A modified EPA medium volume sampler with an identical sampling module was used for outdoor air sampling. Eight homes were sampled; two homes were sampled twice. The indoor air was sampled in the kitchen and the living room over consecutive 8-hr periods. Each outdoor sample was a single 16-hr sample taken simultaneously with the indoor samples.

The levels of PAH detected in outdoor and indoor air samples ranged from 4000 ng/m^3 (naphthalene) to less than 0.1 ng/m^3 (cyclopenta[c,d]-pyrene). The PAH derivatives were found at much lower levels than their parent PAH. Higher average indoor levels of all but three target compounds were found compared to outdoor levels. The higher outdoor levels of these three compounds (naphthalene dicarboxylic acid anhydride, pyrene dicarboxylic acid anhydride, and 2-nitrofluoranthene) are probably due to atmospheric transformation. Cigarette smoking was identified as the most significant contributor to indoor levels of PAH and PAH derivatives. Homes with gas heating systems appeared to have higher pollutant levels compared to homes with electric heating systems. However, homes with electric cooking appliances were associated with higher pollutant levels than homes with gas appliances, but the true effects of heating and cooking systems cannot be accurately known because of the small sample sizes and the lack of statistical significance for most pollutant differences.

This Project Summary was developed by EPA's Environmental Monitoring Systems Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

Recently many research and monitoring efforts have focused on assessing and improving the quality of air. Several studies have demonstrated that some polynuclear aromatic hydrocarbons (PAH) and nitrated PAH (NO_2-PAH) found in ambient and indoor air are potent carcinogens, mutagens, or

both There is an increasing concern over human exposure to these compounds in indoor air from the home, workplace, and school, because we spend more than 80 percent of our time indoors.

Generally, the sampling methodology used for indoor air sampling has not been fully established. The existing sampling devices used in outdoor ambient air sampling are not suitable for indoor sampling applications because of size, lack of portability, and noise. In a pilot residential indoor air study, the sampler pumping unit was located outside the house in an insulated housing to minimize the noise level. Depending upon the structure of each house sampled, there were different modifications made to allow the tube connecting the sampling module to the pumping unit to pass through an open window. This sampling device significantly added to the sampling costs and to the levels of inconvenience experienced by the residents of the sampled house. Therefore we have developed a prototype indoor air sampler that is quiet, transportable, and relatively unobtrusive. This prototype device can be operated at a flow rate (8 cfm) sufficient to collect enough particulate-bound and vapor phase organic compounds over a period of eight hours for chemical analysis and microbioassay analyses.

To determine the feasibility and advantages of using this newly-developed prototype air sampler in residential sampling, a pilot field study was conducted in eight homes in Columbus, Ohio, during the winter of 1986/1987. The objectives of this study were to determine the effectiveness of the prototype sampler in collecting residential air samples and to evaluate methodology for characterization of the concentrations of nicotine, polynuclear aromatic hydrocarbon (PAH), and PAH derivatives in residential air. An investigation of the correlations among levels of PAH, nicotine, and numbers of cigarettes smoked was performed. The correlations between three potential PAH markers phenanthrene, pyrene, and fluoranthene and other PAH were also investigated. This study consisted of the following subtasks:

1. Developing a study design for the field study,
2. Performing the field study in the winter of 1986/1987 according to the study design,
3. Conducting chemical analyses of the collected samples and preparing samples for microbioassay analyses,
4. Conducting statistical analyses of the collected data.

Procedure

A newly developed prototype indoor air sampler was used for the indoor air sampling, and a modified EPA medium volume sampler was used for the outdoor air sampling. The sampler was equipped with a quartz fiber filter followed by an XAD-2 trap to collect organic particles and vapors. The indoor samples were collected from the kitchen from approximately 0700 to 1500 eastern standard time (EST) and from the living room from 1500 to 2300 (EST). During the entire indoor air sampling, a single outdoor air sample was collected in the backyard of each house. The sampler flow rate was approximately 12.5 m^3/hr, which collected 100 m of air over 8 hr.

The sampling design is as follows:

Smoking	Heating	Cooking	No. of Homes
Yes	gas	gas	2
		electric	2
Yes	electric	electric	2
No	gas	gas	2
		electric	1
No	electric	electric	1

The homes selected were chosen to give the greatest variation in the number of cigarettes smoked. In addition, homes were selected so that nearly equal number of samples could be taken from each available combination of type of heating system and type of cooking appliance. The order in which the homes were sampled was randomized to remove potential source of bias that could result if some systematic order were used. No sampling was done either during or immediately following rain. No sampling was performed while the temperature was above 60°F. By not sampling during such periods of time we avoided gross inconsistencies due to the cleaning of the air by the rain or due to the shutting off of the heating system due to warm weather. All the homes are located in Columbus, Ohio, and the sampling was conducted in early March 1987.

The filter and XAD-4 samples were combined and extracted with dichloromethane for 16 hr. The extract was further extracted with ethyl acetate for an additional 8 hr. The dichloromethane extract and ethyl acetate extracts were combined and concentrated by Kuderna-Danish evaporation.

The extract was analyzed by positiv chemical ionization and negative chen ical ionization, gas chromatography/mas spectrometry to determine PAH, nitroge heterocyclic compounds. nitro-PAH ar oxygenated-PAH. Statistical analysis the data included the generation summary statistics, the comparison indoor and outdoor pollutant concentrations, and statistical modelin to assess the separate influence of th individual experimental factors.

Results

The extractable organic mas concentrations found in the XAD-4 ar filter combined extracts ranged fro 0.077 mg/m^3 to 0.57 mg/m^3 indoors ar 0.0091 mg/m^3 to 0.047 mg/m^3 outdoor Generally, higher levels of organic matt were found in the indoor air sample compared to that from the correspondir outdoor air samples. It appeared th cigarette smoking contributes to th higher levels of extractable organ matter found in the indoor air sample The highest concentrations of organ mass were found in home 5 in the livir room location where the highest numbe of cigarettes were smoked during th sampling period.

Concentrations of the individual PA measured in homes ranged from 0.1 *ng/m^3 to 4200 ng/m^3; naphthalene wa* most abundant and cyclopenta [c,d]pyrene, in general, was the lea abundant. The carcinogenic PAH such benzo[a]pyrene and indeno[1.2. c,d]pyrene were present individually concentrations from 0.18 to 3.3 ng/m The concentration of nicotine range from 45,000 ng/m^3 to 24 ng/m^3. Indo concentrations of quinoline an isoquinoline were 8.1 ng/m^3 to 11 ng/m^3. The NO_2-PAH and OXY-PA concentrations found indoors were 0.0 to 1.3 ng/m^3 and 0.021 to 40 ng/m respectively. For most of the targ compounds, higher indoor concentratio were detected compared to the outdo concentrations.

The results of the statistical analyse revealed that cigarette smoking was th most significant contributor to PAH ar most PAH derivatives in indoor air. Th homes with gas heating systems a peared to have higher PAH and oth pollutant concentrations in indoor air, b the effect was not as important as th effect of smoking. The use of electr stoves appeared to result in higher indo pollutant levels compared to the use gas stoves. However, the true effect of electric cooking system is not well knov due to the small sample sizes and th

lack of statistical significance of the observed differences. In addition, further investigation of the data revealed that most of the electric appliance measurements were taken using Sampler C, while the gas appliance measurements were primarily taken using Sampler B. Sampler bias could be responsible for this difference. Comparisons of the effect of sampling locations revealed that the concentrations of the majority of the PAH and PAH derivatives were higher in the living room than in the kitchen, but the difference was statistically significant for only three of the compounds. Sources of variation not included in the statistical model constituted the largest single source of random variation for most target compounds. Home-to-home variation was second-largest for most of the compounds, while day-to-day variation was the smallest.

Conclusions and Recommendations

From this pilot field study, we concluded that the newly developed indoor samplers are quiet, transportable, and acceptable for residential indoor air sampling. The results of the current study were, in general, consistent with the results from the previous pilot study using a different sampling method.

The results demonstrated that cigarette smoking had the greatest effect on the levels of indoor PAH and PAH derivatives. However, the effect of gas/electric heating and cooking systems were not well established due to the small sample sizes and the dominance of the effects of cigarette smoking on PAH levels.

The statistical analyses also showed that there was strong evidence a correlation among the potential PAH markers (phenanthrene, pyrene and fluoranthene) and the other PAH and PAH derivatives. Phenanthrene and pyrene appeared to be better marker compounds than fluoranthene. It was noted that relatively poor correlations were found among PAH markers and PAH derivatives naphthalene dicarboxylic acid anhydride, pyrene dicarboxylic acid anhydride, and 2-nitrofluoranthene. The poor correlation may be because these compounds can be formed through secondary emission sources (e.g. atmospheric transformation). The investigation of correlations among a potential marker for cigarette smoking, nicotine, and other pollutants revealed that all correlations except one (naphthalene dicarboxylic acid anhydride) were positive and statistically significant. It was noted than quinoline indoor concentrations correlate very well with nicotine indoor concentrations. Therefore, quinoline can be a marker for the contribution of cigarette smoke to indoor PAH levels.

The following recommendations are based on the results of this study:

1. A study should be performed to improve the acoustic performance of the outdoor ambient samplers, because the noise levels produced by these units was found to be a source of irritation to the residents and neighbors in this study and would probably not be tolerated if sampling were performed when windows of the houses were open.
2. A pilot field study should be performed in the summertime to evaluate the sampler performance at warmer temperatures and to collect more data to assess the effect of cooking systems on indoor pollutant levels.
3. A small scale study of indoor PAH concentrations should be performed during a period of higher furnace use so that the relationship between amount of furnace use and PAH levels can be determined. In any such study it is important that the amount of furnace use be measured.
4. In the design of future residential air studies, it should not be necessary to sample different locations within the home. Fairly good air mixing occurs in most homes so that pollutant concentrations should be fairly uniform throughout the house. The samples are affected more by the residents' activities during the sampling period. Thus, "living room" samples reported here are likely to primarily reflect the cooking associated with dinner preparation while the "kitchen" samples reflect breakfast and lunch preparation activities. In future studies, the "location" variables should either be replaced by a "period of day" variable or by other variables that more specifically describe the residents' activities conducted during sampling.

J.C. Chuang, G.A. Mack, J.W. Stockrahm, S.W. Hannan, C. Bridges, and M.R. Kuhlman are with Battelle Columbus Division, Columbus, OH 43201-2693.
Nancy K. Wilson *is the EPA Project Officer (see below).*
The complete report, entitled "Field Evaluation of Sampling and Analysis for Organic Pollutants in Indoor Air," (Order No. PB 88-242 565/AS; Cost: $19.95, subject to change) will be available only from:
National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone: 703-487-4650
The EPA Project Officer can be contacted at:
Environmental Monitoring Systems Laboratory
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

United States
Environmental Protection
Agency

Center for Environmental Research
Information
Cincinnati OH 45268

EPA/600/S4-88/028