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Tracing the Sources of Indoor Aerosols Using Evolved Gas Analysis

Joan M. Daisey* and Lara A. Gundel
Indoor Environment Program, Applied Science Division, Lawrence Berkeley Laboratory, Berkeley, CA 94720

An exploratory study was conducted to determine whether microanalytical evolved gas analysis for measuring carbon and nitrogen could be used to distinguish aerosols from different indoor sources. The method requires < 20 μg of particulate matter and it is rapid and inexpensive. Thermograms (rate of evolution of CO_2 and NO_x versus temperature) were determined for samples of environmental tobacco smoke (ETS), kerosene soot, indoor aerosol from woodburning homes, and outdoor aerosol from New Jersey. The thermograms were used to calculate the amounts of carbon and nitrogen for five temperature intervals from 50 to 600°C normalized to particle mass.

The thermograms from different sources showed distinct differences. ETS and kerosene soot had the highest mass percentages of carbon and nitrogen (37% C, 4% N and 27% C, 10% N, respectively), although their nitrogen thermograms differed substantially. Organic nitrogen dominated the ETS samples while most of the nitrogen in kerosene soot was $(NH_4)_2SO_4$. The kerosene soot samples also had the highest percentages of black (elemental) carbon and could thus be distinguished from woodsmoke. These results on a limited number of samples suggest that, with additional research, this method may be useful for distinguishing the contributions of ETS, kerosene soot, and woodsmoke in indoor atmospheres.

INTRODUCTION

Cigarette smoke and other indoor combustion sources such as woodburning stoves and kerosene heaters can be major contributors to concentrations of airborne particulate matter in indoor environments (Spengler and Sexton, 1983). Infiltration of outdoor air also contributes to indoor concentrations of airborne particulate matter and, in some instances, may be the major source of indoor aerosol (Lioy et al., 1988). Indoor combustion sources can substantially increase the indoor concentrations of particulate organic matter and its mutagenic activity (Lofroth et al., 1983, Traynor et al., 1983, Lewtas et al., 1987, Daisey et al., 1989). Both the chemical composition and the biological activity of particles vary, depending upon their sources (Albert et al., 1983), and their health impacts are also expected to differ. Thus, it would be useful to be able to estimate the contributions from various sources. The relative contributions of outdoor air and various indoor sources to indoor concentrations of particles, however, are not easily determined because there are large differences in source usage and ventilation and infiltration rates among homes.

Sexton and Hayward (1987) have suggested that receptor source apportionment models might be useful for estimating the contributions of various sources to indoor concentrations of airborne particles. If tracers or patterns of tracers could be found for each major indoor source type, then receptor source apportionment modeling could provide the means to determine the contributions of each of these sources to indoor concentrations or to personal exposures to airborne particulate matter. Such information would be useful for identification of the sources most in need of mitigation with respect to human health and materials dam-

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*To whom correspondence should be addressed.

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age. Particulate emissions from most of the indoor sources are largely organic in nature; thus, analyses for the many individual organic compounds in these source emissions may offer the potential for distinguishing sources. Such analyses, however, are expensive and typically require sampling hundreds of cubic meters of air. Thus there is a need for a more rapid and inexpensive analytical method for measuring source tracers, particularly for use for large surveys. Such a method should require only a small sample of indoor particulate matter since there are constraints on the volume of air that can be sampled in a given time period in indoor environments without perturbing the concentrations of the species being sampled.

In this study, we investigated the applicability of the microanalytical evolved gas analysis (EGA) method developed by Dod and Novakov (1982) to receptor source apportionment modeling in indoor air. Samples of emissions from indoor sources or source-influenced particulate matter were obtained from various investigators and analyzed by EGA for organic and inorganic carbon and nitrogen. Temperature intervals were identified for the isolation of different features in the thermograms from the EGA and the composition profiles from each source type were compared.

EXPERIMENTAL

Method

The method of analysis was temperature-programmed evolved gas analysis in oxygen (Dod and Novakov, 1982; Dod et al., 1984). Thermograms (rate of evolution of CO_2 and NO_x as a function of temperature) were determined for particulate samples of environmental tobacco (cigarette) smoke (ETS), kerosene heater emissions, indoor air samples from woodburning homes, and outdoor aerosol from a site in New Jersey. The thermograms were determined by heating a 1.7-cm^2 disk of each loaded filter at a constant rate (10°C/min) in oxygen from 50 to 700°C. The evolved gases resulting from the volatilization, decomposition, and combustion of the sample are passed over a hot platinum catalyst to convert all carbon to CO_2 and all nitrogen to NO_x. The CO_2 and NO_x are monitored continuously, by using two gas specific detectors: a nondispersive infrared CO_2 analyzer and a chemiluminescence NO_x analyzer. The use of pure oxygen greatly minimizes the pyrolysis of organics (Cadle et al., 1983). The resultant thermograms are plots of rate of evolution of gas versus temperature; the integrated areas of the curves are proportional to the carbon and nitrogen content of the sample. Quantitation was effected through calibration with gases of known concentrations and by measuring the gas flow rate through the system. The calibration was verified by analyzing samples of known elemental content. Teflon-coated glass fiber filters (with fabric backing removed) were only heated to about 450°C and, consequently, yield only lower limits for concentrations of elemental carbon. The percentages of evolved carbon and nitrogen in the particulate matter on the filters were then calculated from the thermograms. All data were blank corrected for the appropriate filter medium. Particle mass on the filters was determined gravimetrically.

The EGA method requires only a very small sample of particulate matter. For this study, particle masses on the 1.7-cm^2 filter disks ranged from 30 to 400 μg. However, EGA analysis can be done with as little as 10 μg of material.

Samples

Table 1 presents a summary of the samples used in this exploratory research, their sources, type of filter, and the loading on each filter. The environmental tobacco smoke samples were provided by J. Lewtas, EPA, Health Effects Research Laboratory,

TABLE 1. Nature of Samples, Filter Media, and Filter Loading

Samples	Filter type	Loading ($\mu g/cm^2$)
Cigarette smoke	Quartz	120–231
Kerosene soot	Quartz	19–32
Woodsmoke	Teflon-coated glass fiber[a]	59–142
Outdoor aerosol	Glass fiber	220–252

[a] The fabric backing was removed before analysis; EGA to ~ 450°C.

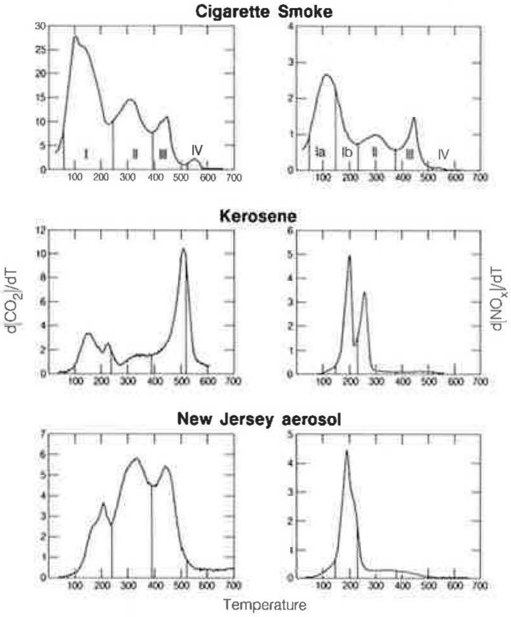
and were generated in an environmental chamber. Samples of kerosene soot were provided by P. M. Boone and B. Leaderer, John B. Pierce Foundation, Yale University School of Medicine and were generated in an environmental chamber and collected at the ceiling level. Samples were obtained for a convective/radiant heater, a convective heater, and a radiant heater. The woodsmoke samples were provided by one of the authors (J. M. Daisey) and were collected in homes in Wisconsin with woodburning stoves but no other indoor combustion sources. Details of sample collection have been reported by Daisey et al. (1989). The woodsmoke samples would also include any particulate matter from other sources in the house such as infiltration of outdoor aerosol and resuspension of deposited dust. Particles from these other sources generally contain less organic matter than woodsmoke (Daisey et al., 1989). The outdoor samples were collected in Ringwood, N.J., a rural, wooded site in northern New Jersey, as part of the ATEOS Study (Lioy and Daisey, 1986). Since the Ringwood samples were collected on preheated glass fiber filters, which produce sulfate artifacts during sampling, the filters were not preweighed. The geometric mean summertime mass concentration at this site, 27.8 ± 64.7 $\mu g/m^3$ (geometric standard deviation) determined from samples collected during two summers on Spectrograde filters (to eliminate sulfate artifact), was used to estimate the mass concentrations for these samples.

RESULTS AND DISCUSSION

Analysis of the Thermograms

Figure 1 presents examples of thermograms obtained for three source types. In order to compare different samples, the thermograms were compared to each other and to thermograms of standards such as NH_4HSO_4, $(NH_4)_2SO_4$, NH_4NO_3, and selected organic compounds. The temperature intervals were chosen by comparison of thermogram structural features for the various samples and they represent reasonable choices which captured the features of each thermogram while emphasizing common features for all. Temperature intervals were selected independently for the carbon and nitrogen thermograms but differ by no more than 10°C. The first temperature interval for nitrogen was subdivided to separate highly volatile organic nitrogen in the thermograms (region

FIGURE 1. Thermograms of particulate matter samples from various sources. Left side, carbon; right side, nitrogen. The four temperature intervals are indicated by the numerals I, II, III, and IV for the two thermograms at the top of the figure.



Ia) from the inorganic nitrogen of NH_4NO_3 and the first peak in the thermogram of $(NH_4)_2SO_4$ (region Ib). A second peak for $(NH_4)_2SO_4$ and a single peak for NH_4HSO_4 are found in region II (Dod and Novakov, 1982). Based on these comparisons the following temperature intervals were integrated:

I. 50–240°C: Thermally volatilizable organic carbon; 50–235°C: organic and inorganic nitrogen.
   Ia. 50–150°C: Organic nitrogen of high volatility and low molecular weight;
   Ib. 150–235°C: Organic nitrogen of intermediate volatility, inorganic ammonium salts, NH_4NO_3, $(NH_4)_2SO_4$.
II. 240–390°C: Organic carbon of intermediate volatility and molecular weight; 235–380°C: organic nitrogen and ammonium salts, NH_4HSO_4 and $(NH_4)_2SO_4$ (second peak).
III. 390–520°C: Black (elemental) (Rosen et al., 1978, 1980) carbon and some high molecular weight organic carbon; 380–520°C: low volatility organic nitrogen, probably part of the black carbon matrix;
IV. 520–600°C: Black (elemental) carbon, if large particles are present or sample is nearly all black carbon; nitrogen associated with black carbon.

The temperature intervals defined for black carbon correspond to the combustion of the strongly optically absorbing component of combustion-derived particles, above 390°C (Dod and Novakov, 1982). Urban atmospheric particles, combustion source samples and polycrystalline graphite have similar Raman spectra (Rosen et al., 1978, 1980). Black carbon corresponds to what many investigators call elemental carbon. In an interlaboratory comparison, EGA in oxygen gave values for black carbon which were within ± 10% of the mean of all 17 of the methods of analysis for elemental carbon (Groblicki et al., 1983).

It is clear from Figure 1 that the different source types have different thermograms. The CO_2 and NO_x thermograms for the ETS were very similar to each other but differed for the other source types. Pure nitrogen-containing organic compounds also have similar CO_2 and NO_x thermograms (Dod et al., 1984). Each source type shows CO_2 peaks in approximately the same regions but the relative amounts in the regions differ.

The NO_x thermograms show two distinct patterns: inorganic nitrogen, principally $(NH_4)_2SO_4$, gives sharp peaks (regions Ib and II) as seen in kerosene soot. Inorganic nitrogen peaks were also seen in region Ib for woodsmoke (not shown) and outdoor aerosol samples. For cigarette smoke, all of the peaks correspond to organic nitrogen, including the unusual feature at high temperature (region III). The nitrogen in kerosene soot is almost wholly inorganic, with little or no evidence of organic nitrogen.

The differences among source types were quantified and normalized to particle mass. The averages for each source type and each temperature interval were then calculated. The results are presented in Tables 2 and 3 as percentage of carbon or nitrogen ± standard deviation, for each of the temperature intervals of the thermograms. The results of the analyses of the samples from each of the individual kerosene heaters are presented because each represents a different type of kerosene heater and there were substantial differences in the compositions of the particulate emissions among the different heaters.

Organic Carbon. Cigarette smoke had the highest percentage of organic carbon (OC) (regions I and II), particularly in the lowest temperature interval, I. The kerosene soot samples also showed high percentages of OC when compared to the woodsmoke and New Jersey outdoor aerosol samples. Some of the differences in region I may be due to differences in filter face velocities and

TABLE 2. Carbon as a Percentage of Total Particle Mass

		Region				
Source	n	I	II	III	IV	SUM
Cigarette smoke	2	21.6 ± 2.6	10.5 ± 1.1	4.5 ± 0.8	0.57 ± 0.16	37.2
Wood smoke	2	1.9 ± 0.9	7.3 ± 1.8	2.9 ± 2.8	—[a]	12.2
Kerosene soot—convective	1	7.3	3.2	13.4	3.4	27.3
Kerosene soot—convective/radiant	1	9.2	5.9	20.4	1.4	37.0
Kerosene soot—radiant	1	6.6	2.6	5.1	0.91	15.2
New Jersey outdoor aerosol	2	2.1 ± 0.6	4.2 ± 0.6	3.1 ± 0.1	0.35 ± 0.13	9.8

Values given as mean ± standard deviation; n, number of samples.
[a]Not analyzed because of interference from Teflon in filter.

filter media. The woodsmoke (indoor) and New Jersey outdoor samples were all collected at high face velocities, i.e., about 17 cm s^{-1}. At higher face velocities, the semivolatiles tend to be lost from the filter, particularly for long sampling times (Schwartz et al., 1981). The ETS samples were collected at a lower face velocity of about 4 cm s^{-1}. The woodsmoke samples were collected on Teflon-coated glass fiber filters while the cigarette smoke and kerosene soot were collected on quartz filters. Several investigators have recently reported evidence that quartz filters pick up organic vapors during sampling (Fitz, 1987; McDow and Huntzicker, 1987).

The percentage of OC in the woodsmoke samples, which were collected in houses, averaged 9.2%. Stiles (1983) has reported an average of 29.2% OC for stack emissions from burning oak with a similar moisture content in a woodburning stove collected on quartz filters. Rau (1989) has recently reported values of 14 ± 4% OC for emissions from burning hardwood under hot burn conditions; OC averaged 57 ± 5% for cool burn conditions. Rau's samples were collected on quartz filters but a correction was made for filter adsorption of organic vapors during sampling.

The average concentration of OC (6.3%) estimated for the two New Jersey outdoor aerosol samples in this study is similar to the average concentration of 8.2% reported previously for the site using a different method and more samples (Daisey, 1987).

TABLE 3. Nitrogen as a Percentage of Total Particle Mass

		Region					
Source	n	Ia	Ib	II	III	IV	SUM
Cigarette smoke	2	1.6 ± 0.1	0.74 ± 0.01	0.89 ± 0.03	0.67 ± 0.04	0.05 ± 0.02	4.0
Wood smoke	2	0.12 ± 0.01	0.72 ± 0.58	0.59 ± 0.26	0.15 ± 0.15	—[a]	1.6
Kerosene soot—convective	1	0.32	5.3	3.62	0.55	0.10	9.9
Kerosene soot—convective/radiant	1	0.52	4.4	2.9	0.88	0.26	8.9
Kerosene soot—radiant	1	0.18	2.9	2.7	0.22	0	6.0
New Jersey outdoor aerosol	2	0.15 ± 0	1.6 ± 0.1	0.47 ± 0.15	0.20 ± 0.06	0.04 ± 0.03	2.5

Values given as mean ± standard deviation; n, number of samples.
[a]Not analyzed due to interference from Teflon coating of filter.

Black (Elemental) Carbon. The kerosene soot samples had the highest concentrations of black (and high molecular weight) carbon, 14.9 ± 8.1% of particle mass (regions III and IV). There was considerable variation among the individual samples from different types of kerosene heaters, as can be seen in Table 2. The convective/radiant heater emitted particles with the highest percentage of black carbon, 22%, followed by the convective heater with 17% and the radiant heater with only 5%. The average concentration of black carbon in the woodsmoke samples was only 3%. There may have been some additional black carbon in the woodsmoke samples which could not be measured because of the interference from the Teflon-coated filter in the highest temperature region. Stiles (1983) reported concentrations of elemental carbon in woodsmoke from oak to range from 1.6% to 7.2%, depending on the percent moisture in the wood. (The method used by Stiles to measure the elemental carbon differs from the method used here to measure black carbon but the two measurement methods should give very similar values.) Rau (1989) reported an average elemental carbon concentration for stack emissions of 16 ± 4% for hot burning hardwood and 5 ± 1% for cool burning hardwood.

The concentration of elemental carbon in the two outdoor aerosol samples from the Ringwood site in New Jersey was estimated to be 3.5%. The average concentration of elemental carbon in outdoor aerosol collected in the summertime in New Jersey at this same site was 1.5%, measured by another method for a different and larger set of samples (Daisey, 1987).

Nitrogen. The average percentages of nitrogen in the particles, reported in Table 3, were lower than those of carbon. The cigarette smoke and the kerosene soot showed the highest average percentages of nitrogen, 4% and 8%, respectively. Based on the thermograms in Figure 1, however, the chemical nature of the nitrogen from these two sources appears to be different. For the kerosene soot, the two peaks with maxima at 190°C and 270°C in regions Ib and II are characteristic of $(NH_4)_2SO_4$. The shape of the thermogram for the cigarette smoke is quite different, with a broad peak in region Ia having a maximum at about 120°C. Thus, by further dividing region I into two subregions from 50 to 150°C and 150 to 235°C, these two sources can better be distinguished. The woodsmoke samples (not shown) also had region Ia maxima indicating the presence of organic nitrogen compounds (Dod et al., 1984).

Source Discrimination

The purpose of this research was to provide preliminary information on the ability of the EGA method to differentiate among various sources and estimate their contributions to airborne particulate matter. Because readily available samples of source emissions or source-influenced aerosol were used, there has been no standardization of sampling method or filter medium. Despite this, the thermograms show clearly different patterns for the different source types. Cigarette smoke and kerosene had the highest total concentrations of both carbon and nitrogen. For cigarette smoke, the fraction of nitrogen in the smoke was 2.5 times greater than that in the woodsmoke and 1.6 times that in the outdoor aerosol. The distribution of the nitrogen in the cigarette smoke was similar to that of the carbon while the other source profiles showed little or no correlation in distributions of carbon and nitrogen (Figure 1). These differences should enhance discrimination of cigarette smoke from these sources.

There were differences in region Ia and Ib/II nitrogen that should also enhance source discrimination among the tobacco smoke, the kerosene soot and the woodsmoke. Cigarette smoke had a high concentration of organic nitrogen in region Ia

(1.65%) while the kerosene soot had only one-sixth to one-ninth the concentration and woodsmoke had even less. The kerosene soot was enriched in $(NH_4)_2SO_4$ (regions Ib and II; 5.6% to 8.9%) while the tobacco smoke had only 1.6% nitrogen in these regions. The kerosene soot also differed from the cigarette smoke in black carbon composition. The carbon concentration in regions III and IV was about three times higher, on average, for the kerosene soot than the cigarette smoke. The woodsmoke aerosol could be discriminated from cigarette smoke by region III nitrogen which was about four times higher in the ETS than in the woodsmoke aerosol. There may also be significant differences in region I nitrogen between these two source types, although differences in sampling method for the two samples prevented any clear conclusions. However, very little nitrogen would be expected in this region for a sample of woodsmoke collected at the same face velocity and on the same filter type as the ETS because wood has only about one-tenth as much nitrogen as tobacco (Arola, 1978; Surgeon General, 1979).

It is also possible to combine the data from EGA with other tracer measurements to increase discrimination among sources. Trace metal analysis, for example, is now done very routinely and inexpensively on very small samples of aerosol. Source profiles for outdoor sources typically include trace element concentrations plus organic and elemental carbon. For indoor sources, it may be useful to develop profiles consisting of EGA data combined with selected trace elements such as K, S, and Fe.

There is another possible contributor to indoor aerosol, i.e., cooking. Volatilization of fats during cooking and their subsequent condensation would be expected to produce an aerosol with high concentrations of VOC. Burning food will produce pyrolysis products including soot. Some organic nitrogen aerosol may also be produced during the cooking of meats. This source type may be much more variable than the other types, and therefore, its contribution to aerosol levels may be difficult to estimate. It may be that its overall contribution for a 24-h sample collected in a house is not very large (although exposures to the cook might be quite high). In a chemical mass balance source apportionment model, this source may simply end up in the residual or unattributable aerosol mass.

CONCLUSIONS AND RECOMMENDATIONS

This preliminary research, despite limitations in the samples available for analysis, indicates that the EGA method may be useful for discriminating the contributions of various sources of indoor aerosol in a source apportionment model. The method has the advantages of being simple, rapid and inexpensive. In addition, very little sample is needed for the analysis. Additional experimental data, however, are needed to prove the utility of the method. The following specific recommendations are made:

1. Source emissions samples should be collected on the same filter medium, at the same face velocity and then analyzed and compared;
2. Mixtures of aerosols from two or three sources should be generated and analyzed by this method to determine how well the method works for "real world" mixtures; further refinement in the thermogram analysis protocol might also be required;
3. Once good source emissions profiles have been generated, the data should be subjected to the single value decomposition analysis recommended by Henry (1982) to evaluate multicollinearities and determine which sources can be distinguished accurately using EGA composition profiles, taking into account source variabilities.

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