



Project Summary

The Particle Team (PTEAM) Study: Analysis of the Data

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EPA and the California Air Resources Board sponsored a study of human exposure to inhalable particles in the Los Angeles Basin. Results were reported in Volumes I and II; this is a summary of the third and final volume, dealing with statistical analysis and physical models. Nicotine and air exchange results are presented and analyzed. A model was developed to estimate the penetration factors and decay rates for inhalable (PM_{10}) and fine ($PM_{2.5}$) particles, for 15 elements associated with each size fraction, and for polyaromatic hydrocarbons (PAHs). The model was also used to estimate source emission rates for particles and elements produced by cigarettes and cooking, and also for PAHs produced by cigarettes.

This Project Summary was developed by EPA's National Exposure Research 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

In 1986, Congress mandated that the US EPA undertake a study of exposure to particles. EPA's Atmospheric Research and Exposure Assessment Laboratory (AREAL), now part of the National Expo-

sure Research Laboratory (NERL), joined with California's Air Resources Board to sponsor a study in the Los Angeles Basin. The study was carried out primarily by the Research Triangle Institute (RTI) and the Harvard School of Public Health, with additional support from Lawrence Berkeley Laboratory (LBL), Acurex, and AREAL. Small portable personal monitors were designed to measure inhalable particles (aerodynamic diameter less than $10\ \mu m$, or PM_{10}). In addition, stationary microenvironmental monitors were designed to sample both PM_{10} and $PM_{2.5}$ (fine particles $<2.5\ \mu m$ in diameter). The personal and indoor samplers were equipped with filters to collect nicotine. Monitors were also developed to measure PAHs and phthalate esters. Air exchange rates in each home were measured using perfluorotracer (PFT) techniques. A total of 178 residents of Riverside, CA, took part in the study in the fall of 1990.

The results of the study are presented in three volumes. Volume I (Pellizzari *et al.*, 1993) provides a full description of the procedures and presents summary population-weighted statistics for particles and elements. A Project Summary for Volume I is also available (Pellizzari *et al.*, 1993). Volume II (Sheldon *et al.*, 1993) presents summary population-weighted statistics for PAHs and phthalate esters. Volume III presents summary statistics for air ex-



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change rates and nicotine concentrations, and also provides more detailed statistical analyses and physical models for all parameters measured. This document is the Project Summary for Volume III.

Procedure

Measurement Methods

A personal exposure monitor (PEM) was designed to collect PM_{10} using a sharp-cut impactor with a circular set of holes 1.9 mm in diameter. Particles are collected at a constant flow rate of 4 L/m on a 37-mm Teflon filter mounted below a greased impactor plate. The PEM consists of a soft canvas bag containing the pump and battery pack that can be worn on the hip, stomach, lower back, or over the shoulder. Nearly identical monitors were employed for concurrent indoor and outdoor sampling of PM_{10} and $PM_{2.5}$. For the personal and indoor samples, a second filter treated with citric acid to collect nicotine was placed behind the first filter.

The monitor for the PAHs and phthalate esters included a glass cartridge containing XAD-2 sorbent preceded by a quartz fiber filter. A box containing four Medo pumps sampled air at a constant flow rate of about 18 L/m during the 12-h monitoring periods. Both the filter and the cartridge were sonically extracted with methylene chloride. Analysis employed gas chromatography/mass spectrometry (GC-MS) in the selected ion mode.

Air exchange rates were measured using perfluorotracer (PFT) methods. Three continuously emitting sources of PFT in small boxes heated to 40°C were placed in each home 24 h before sampling began. Volumes of the homes were measured at this time. At each of the subsequent visits (the beginning of each of the two 12-h measurement periods), collector tubes containing activated charcoal were placed in the home at three sites. Two tubes were in the main living area, one in the bedroom, and one near the center of the home.

Study Design

The main goal of the study was to estimate the frequency distribution of exposures to PM_{10} particles for all nonsmoking Riverside residents aged ten and above. A second major objective was to estimate the frequency distribution of concentrations of PM_{10} and $PM_{2.5}$, PAHs, and phthalate esters in residences and nearby outdoor air, e.g., back yards. Other objectives included identifying important indoor sources and estimating their emission rates, determining the effect of outdoor air on indoor concentrations, and estimating

the contribution of personal activities to exposure.

A three-stage probability sampling procedure was adopted. In the first stage, 36 areas within Riverside were selected for study with a probability proportional to population size in each area. Areas were characterized and stratified by income to ensure a wide socioeconomic representation. In the second stage, an attempt was made to contact every household within these areas and administer a short questionnaire to determine eligibility for participation and the frequency of certain stratification variables such as employment and passive smoking. In the final stage, respondents were selected for monitoring. Respondents represented 139,000 \pm 16,000 (S.E.) nonsmoking Riverside residents aged ten and above.

Each participant wore the PEM for two consecutive 12-h periods. (Actual times monitored depended on participant activities and ranged from 8-14 h.) Concurrent PM_{10} and $PM_{2.5}$ samples were collected by the stationary indoor monitor (SIM) and stationary outdoor (ambient) monitor (SAM) at each home. This resulted in 10 particle samples per household (day and night samples from the PEM_{10} , SIM_{10} , $SIM_{2.5}$, SAM_{10} , and $SAM_{2.5}$). Air exchange rates were also calculated for each 12-h period. At a subset of 125 homes (and 65 outdoor areas near the residences), monitors to measure indoor and outdoor PAHs and phthalate esters were operated for each 12-h period.

Participants were asked to note activities that might involve increased particle levels (nearby smoking, cooking, gardening, etc.). Following each of the two 12-h monitoring periods, they answered an interviewer-administered recall questionnaire concerning their activities and locations during that time.

Up to four participants per day could be monitored, requiring 48 days in the field. A central outdoor site was maintained over the entire period (Sept. 22–Nov. 9, 1990). The site had two high-volume samplers (Wedding & Assoc.) with 10- μ m inlets (actual cutpoint about 9.0 μ m); two dichotomous PM_{10} and $PM_{2.5}$ samplers (Sierra-Andersen) (actual cutpoints about 9.5 and 2.5 μ m); one PEM and one SAM_{10} (actual cutpoint about 11.0 μ m, as measured in laboratory studies); and one $SAM_{2.5}$ (actual cutpoint = 2.5 μ m).

All PEM, SIM, SAM, and dichotomous sampler filters (about 2500) were analyzed by energy-dispersive x-ray fluorescence (EDXRF) for a suite of 42 elements (Dzubay *et al.*, 1988). The analysis was carried out at EPA's AREAL in Research

Triangle Park, NC. Some filters were analyzed twice under blind conditions. A subset of about 100 filters was analyzed by the LBL for quality assurance purposes. An additional set of about 600 citric-acid treated filters from personal and indoor samplers was analyzed for nicotine.

All filters were weighed onsite. Replicate weighings were required to be within ± 4 μ g/filter. Blank filters were weighed, sent out with field samples, and reweighed along with the field samples. Duplicate indoor and outdoor samples were collected at 10% of the homes. Duplicate SAM and PEM samples were also collected at the central site. Duplicate PEM samples were also collected by EPA, RTI, and Harvard scientists while onsite.

Results

Of 632 permanent residences contacted, 443 (70%) completed the screening interview. Of these, 257 were asked to participate and 178 (69%) agreed. More than 2750 particle samples were collected, about 96% of those attempted.

Quality of the Data

Blank PEM and SIM/SAM filters ($N = 51$) showed consistent small increases in mass averaging 9.5 ± 8.4 μ g; this value was subtracted from each field sample. Limits of detection (LODs), based on three times the standard deviation of the blanks, were on the order of 7 to 10 μ g/m³. All field samples exceeded the LOD.

Duplicate samples ($N = 363$) showed excellent precision for all types of samplers at all locations, with median relative standard deviations ranging from 2% to 4%.

The collocated samplers at the central site showed good agreement, with correlations between the three types of samplers ranging from 0.96 to 0.99. As had been previously noted, the central-site PEMs collected about 12% more mass than the dichotomous samplers, perhaps due to their higher cutpoint (11 μ m compared to 9.5 μ m) or to a particle "bounce" effect, measured in the laboratory at less than 9%.

Background levels of elements on laboratory and field blanks were low. Analyses of standard reference materials (SRM 1832 and 1833) were within 7% of the correct values for all 12 elements contained. Median relative standard deviations (RSD) for duplicates analyzed blindly by the principal laboratory were better than 15% for all 15 prevalent elements. Median RSDs for duplicates analyzed by the principal laboratory and by the quality assurance laboratory (LBL) were less than 21% for all elements except manganese (76%) and

copper (27%). The LBL reported 10% to 20% higher average values for 13 of 14 elements.

For benzo-a-pyrene, the quantifiable limit was 0.08 ng/m³, with a median precision of 3%, a mean amount on the field blanks of 0.0 ng, and a mean recovery of 95 ± 19%. Similar results were obtained for the other PAHs and phthalate esters.

Concentrations

Concentrations of particles and elements have been reported (Clayton *et al.*, 1993; Özkaynak *et al.*, 1993; Pellizzari *et al.*, 1993; Wallace *et al.*, 1993). Population-weighted daytime personal PM₁₀ concentrations averaged about 150 µg/m³, compared to concurrent indoor and outdoor mean concentrations of about 95 µg/m³. The overnight personal PM₁₀ mean was much lower (77 µg/m³) and more similar to the indoor (63 µg/m³) and outdoor (86 µg/m³) means. About 25% of the population was estimated to have exceeded the 24-h National Ambient Air Quality Standard for PM₁₀ of 150 µg/m³. Over 90% of the population exceeded the California Ambient Air Quality Standard for PM₁₀ of 50 µg/m³.

Mean values of the fine (PM_{2.5}) particle mass were 48 and 49 µg/m³ for daytime indoor and outdoor samples, and 36 and 50 µg/m³ for the overnight indoor and outdoor samples, respectively. Thus, fine particles accounted for about 50% of the total PM₁₀ mass both indoors and outdoors during the day and about 60% both indoors and outdoors at night.

Nicotine

A total of 334 valid measurements were obtained for the personal samples, and 230 for the indoor samples. About 30% (176) of the 564 analyzed nicotine samples exceeded the LOD of 0.15 µg/filter (corresponding to a nominal value of about 0.05 µg/m³). Most of these were from personal or indoor samples associated with exposure to cigarette smoke. Mean personal and indoor nicotine concentrations were on the order of 1 µg/m³ for those samples associated with reported exposure to tobacco smoke but were below the LOD for those samples with no reported exposure.

A regression of indoor nicotine concentrations on the number of cigarettes smoked in the home during the monitoring period indicated that indoor nicotine values increased by about 0.12 µg/m³ for each cigarette reported smoked during the monitoring period. The *R*² value is 35.4% (*N* = 227).

A regression of personal nicotine levels on minutes exposed to cigarette smoke suggested that personal nicotine exposures increased by about 0.013 µg/m³ per minute of reported exposure to cigarette smoke. The *R*² value is 36.6% (*N* = 334).

PAHs

Median indoor and outdoor concentrations ranged between 0.1 and 2 ng/m³ for all but the two most volatile 3-ringed PAHs: acenaphthylene (day and night medians of 3.5 and 3.8 ng/m³ indoors and 1.8 and 6.9 ng/m³ outdoors) and phenanthrene (16 and 15 ng/m³ indoors and 8.8 and 12 ng/m³ outdoors). Little difference was seen between indoor and outdoor concentrations.

Phthalate Esters

Median indoor values for four phthalate esters ranged between 30 and 400 ng/m³; they were below the detection limit for di-*n*-octylphthalate. Median outdoor levels were often below the detection limit, with the highest value being 28 ng/m³ for di-2-ethylhexylphthalate. Indoor levels of four phthalates were about 2 to 15 times higher than outdoor levels.

Air Exchange

A total of 1010 12-h average air samples were collected. There were 273 duplicate pairs and 464 single observations. Two observations were outliers, resulting in 735 values after averaging the duplicates. About 20% of the samples had PFT amounts below the LOD.

The 24-h average air exchange rates were calculated for 175 Riverside homes using the convention of assigning half the LOD to values below the LOD. The geometric mean of the air exchange rates was 0.97 h⁻¹, with a geometric standard deviation (GSD) of 2.18 (Figure 1).

Correlations

The central site appeared to be a moderately good estimator of outdoor particle concentrations throughout the city. Spearman correlations of the central-site concentrations measured by all three methods with outdoor near-home concentrations as measured by the new samplers ranged from 0.8 to 0.85 (*p* < 0.00001). Linear regressions indicated that the central-site readings could explain about 60% of the variability observed in the near-home outdoor concentrations (Figure 2).

Outdoor concentrations could explain about 25% to 30% of the variability observed in indoor concentrations (Figure 3). Spearman correlations of near-home out-

door concentrations with indoor concentrations ranged from 0.5 to 0.6. Spearman correlations of the central-site outdoor concentrations with indoor concentrations were reduced somewhat (about 0.45 to 0.55).

Outdoor concentrations were able to account for only about 16% of the variability in personal exposures (Figure 4). This is understandable in view of the importance of indoor activities such as smoking, cooking, dusting, and vacuuming on exposures to particles. The higher daytime exposures were even less well represented by the outdoor concentrations, whether measured near the home or at the central site.

Indoor concentrations accounted for about half of the variability in personal exposures (Figure 5). However, neither the indoor concentrations alone, nor the outdoor concentrations alone, nor time-weighted averages of indoor and outdoor concentrations could do more than explain about two-thirds of the observed variability in personal exposures. It appears that the remaining portion of personal exposure arises from personal activities or unmeasured microenvironments that are not well represented by fixed indoor or outdoor monitors.

One of the variables most highly correlated with particle levels in the home was an estimated "dirt level." The technicians' estimate of dirt level was made while visiting each house. Two technicians carried out all the measurements. They estimated dirt and dust levels on a 7-point scale, and "calibrated" themselves by experimenting on several Boston homes before going to Riverside. The 24-h averages of personal and indoor particles and also indoor nicotine were significantly associated with estimated dirt level.

The mean indoor concentrations of PAHs in homes with smoking appeared generally higher than in homes without smoking. Student *t*-tests showed that for 10 out of 12 PAHs (and for di-*n*-butylphthalate), the difference between the geometric mean concentrations in smoking vs. nonsmoking homes was statistically significant, usually at *p* < 0.0001.

Mass-Balance Model

A model developed in Koutrakis *et al.* (1992) was solved using nonlinear least squares to estimate penetration factors, decay rates, and source strengths for particles and elements from both size fractions. In this model, which assumes per-

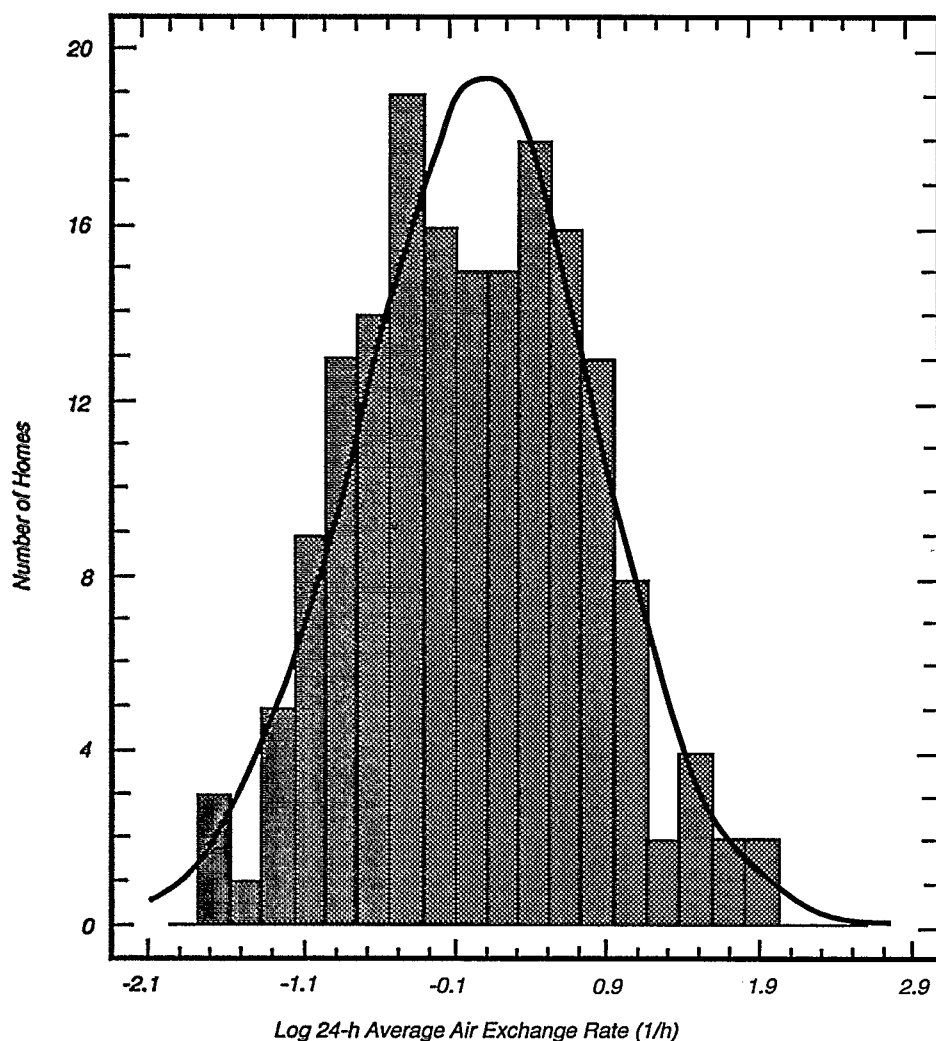


Figure 1. Normal fit to the logarithms of the overnight air exchange rates, assigning values of LOD/2 to all nondetected samples. The geometric mean = 0.97 h^{-1} .

fect instantaneous mixing and steady-state conditions throughout each 12-h monitoring period, the indoor concentration of particles or elements is given by

$$C_{in} = \frac{P\alpha C_{out} + Q_{is}/V}{\alpha + k} \quad [1]$$

where

C_{in} = indoor concentration (ng/m^3 for elements, $\mu\text{g}/\text{m}^3$ for particles)

P = penetration coefficient

α = air exchange rate (h^{-1})

C_{out} = outdoor concentration (ng/m^3 or $\mu\text{g}/\text{m}^3$)

Q_{is} = mass flux generated by indoor sources (ng/h or $\mu\text{g}/\text{h}$)

V = volume of room or house (m^3)

k = decay rate due to diffusion or sedimentation (h^{-1})

From initial multivariate analyses, the most important indoor sources appeared to be smoking and cooking. Therefore the indoor source term Q_{is} was replaced by the following expression:

$$Q_{is} = (N_{cig} S_{cig} + T_{cook} S_{cook})/t + Q_{other}$$

where

t = duration of the monitoring period (h)

N_{cig} = number of cigarettes smoked during monitoring period

S_{cig} = mass of elements or particles generated per cigarette smoked (ng/cig or $\mu\text{g}/\text{cig}$)

T_{cook} = time spent cooking (min) during monitoring period

S_{cook} = mass of elements or particles generated per minute of cooking (ng/min or $\mu\text{g}/\text{min}$)

Q_{other} = mass flux of elements or particles from all other indoor sources (ng/h or $\mu\text{g}/\text{h}$)

With these changes, the equation for the indoor concentration due to these indoor sources becomes

$$C_{in} = \frac{P\alpha C_{out}}{\alpha + k} + \frac{N_{cig} S_{cig} + T_{cook} S_{cook}}{(\alpha + k)Vt} + \frac{Q_{other}}{(\alpha + k)V} \quad [2]$$

The indoor and outdoor concentrations, number of cigarettes smoked, monitoring duration, time spent cooking, house volumes, and air exchange rates were all measured or recorded. The penetration factor, decay rates, and source strengths for smoking, cooking, and all other indoor sources (Q_{other}) were estimated using a nonlinear model (NLIN in SAS software). The Gauss-Newton approximation technique was chosen to regress the residuals onto the partial derivatives of the model with respect to the unknown parameters until the estimates converge. On the first run, the penetration coefficients were allowed to "float" (no requirement was made that they be ≤ 1). Since nearly all coefficients came out close to one, a second run was made bounding them from above by one. The NLIN program provides statistical uncertainties (upper and lower 95% confidence intervals) for all parameter estimates. However, it should be noted that these uncertainties assume perfect measurements and are therefore underestimates of the true uncertainties.

Results are presented in Table 1 for the combined day and night samples. Penetration factors are very close to unity for nearly all particles and elements. The calculated decay rate for fine particles is $0.39 \pm 0.16 \text{ h}^{-1}$, and for PM_{10} is $0.65 \pm 0.28 \text{ h}^{-1}$. Each cigarette emits $22 \pm 8 \text{ mg}$ of PM_{10} on average, about two-thirds of which ($14 \pm 4 \text{ mg}$) is in the fine fraction. Cooking emits $4.1 \pm 1.6 \text{ mg}/\text{min}$ of

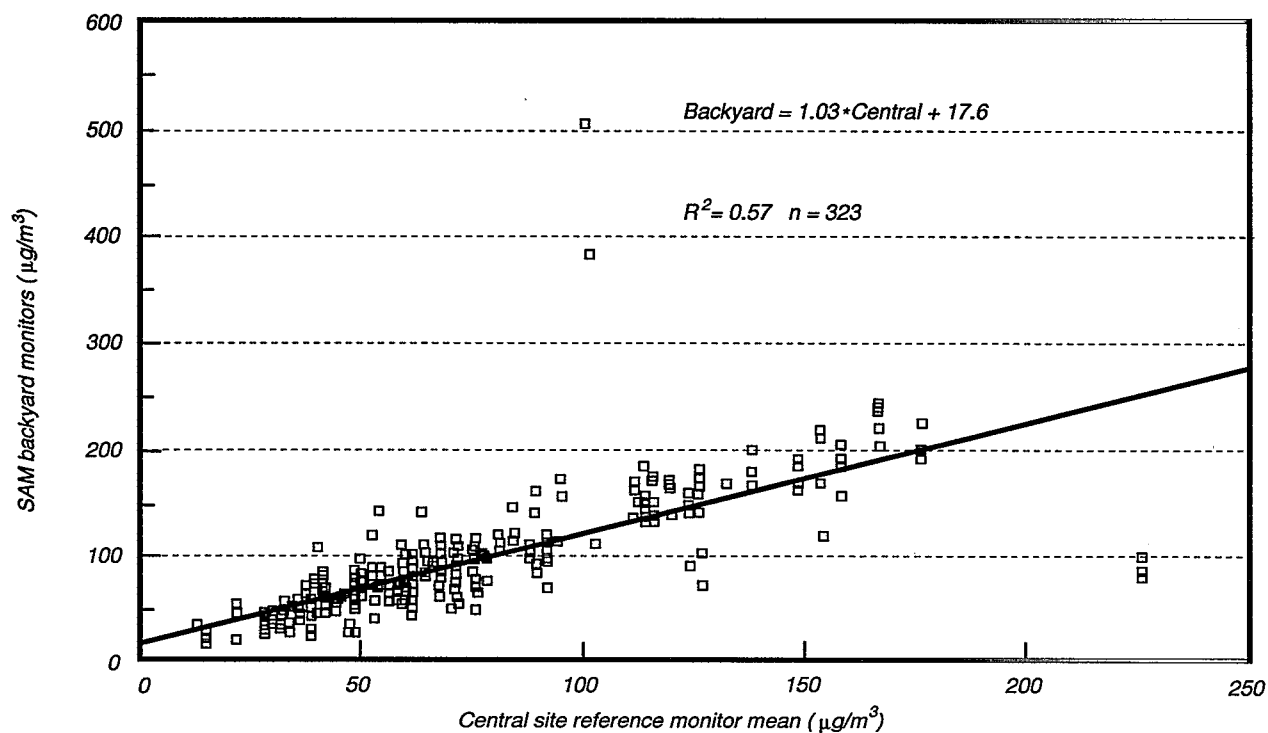


Figure 2. Central-site mean of two dichotomous samplers vs. residential outdoor monitors.
 $R^2 = 57\%$

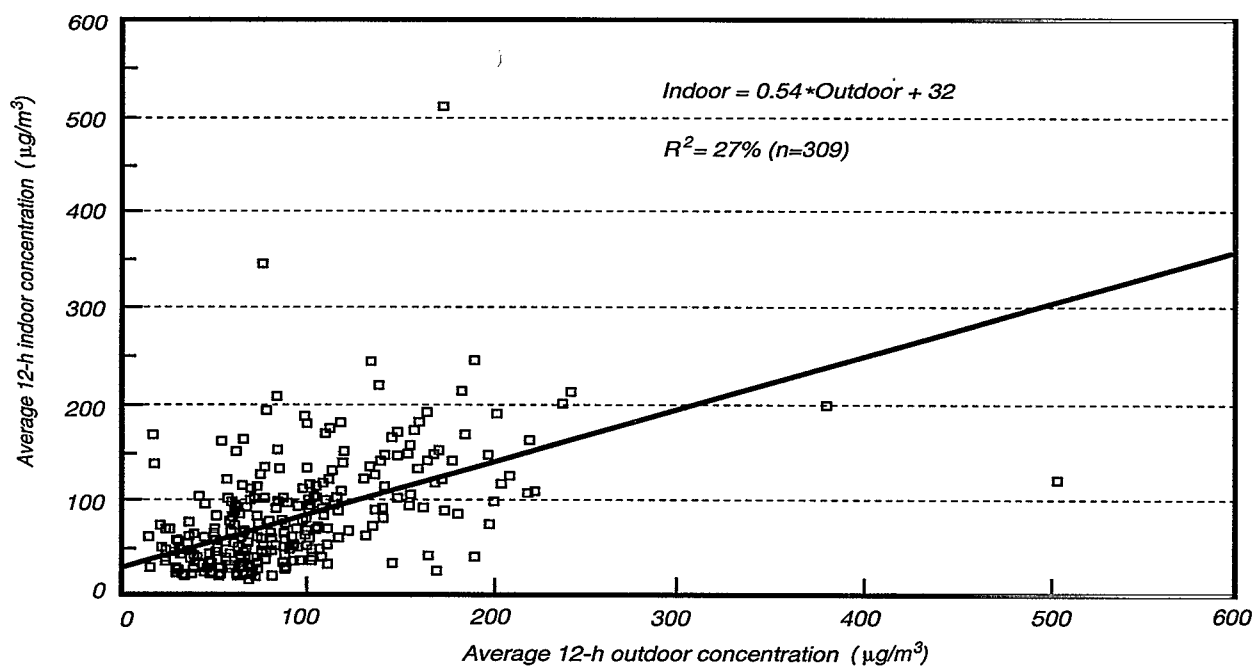


Figure 3. Indoor vs. outdoor PM_{10} concentrations.
 $R^2 = 27\%$

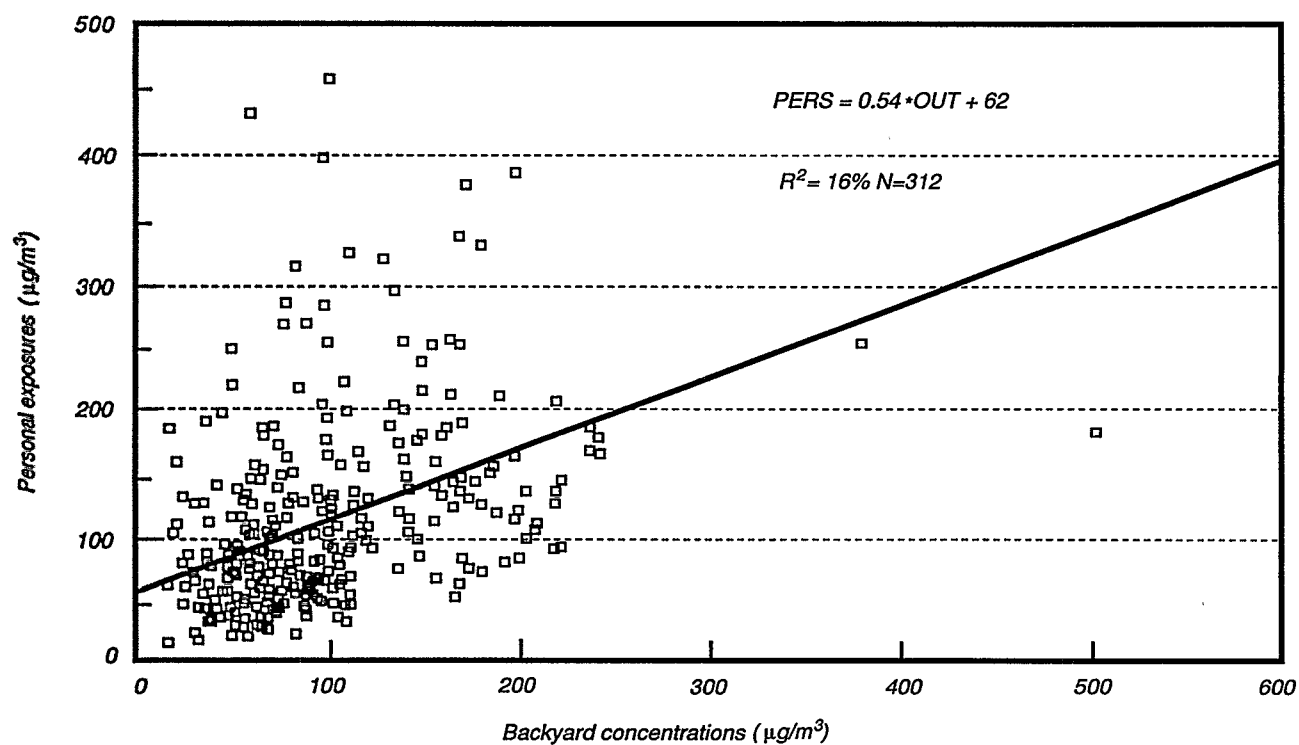


Figure 4. Personal exposures vs. residential (backyard) outdoor PM_{10} concentrations.
 $R^2 = 16\%$

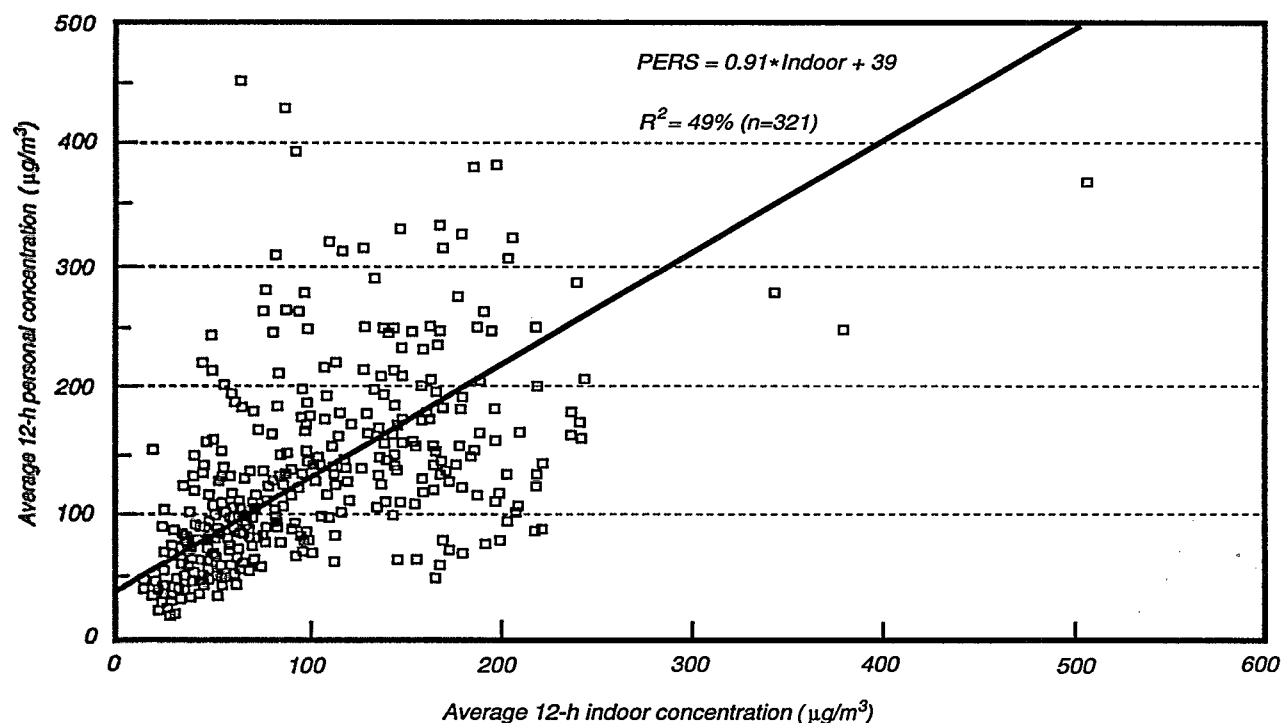


Figure 5. Personal exposures vs. residential indoor PM_{10} concentrations.
 $R^2 = 49\%$

Table 1. Penetration Factors, Decay Rates, and Source Strengths: Nonlinear Estimates

VAR	Penetration			Decay Rate (1/h)			Other Sources (µg/h)			S_cook (µg/min)			S_smoke (µg/cig)		
	mean	195	µ95	mean	195	µ95	mean	195	µ95	mean	195	µ95	mean	195	µ95
PM2.5*	1.00	0.89	1.11	0.39	0.22	0.55	1.1	0.0	2.1	1.7	1.0	2.3	13.8	10.2	17.3
Al	1.00	0.95	1.05	0.03	-0.03	0.09	3.0	-3.7	9.8	0.9	-1.4	3.1	9.0	-2.5	20.5
Mn	0.87	0.78	0.95	0.23	0.07	0.38	0.5	0.2	0.9	0.1	-0.1	0.2	0.2	-0.4	0.8
Br	0.90	0.81	0.99	0.28	0.15	0.41	0.6	0.3	0.9	0.1	0.0	0.2	1.9	1.3	2.5
Pb			fail to converge												
Ti			fail to converge												
Cu	1.00	0.56	1.44	1.63	0.38	2.88	3.8	1.4	6.3	0.6	0.0	1.2	3.7	0.2	7.2
Sr	0.97	0.93	1.01	0.07	0.01	0.12	0.1	0.0	0.2	0.0	0.0	0.0	0.1	-0.1	0.2
P	1.00	0.98	1.02	0.04	0.02	0.06	0.8	0.2	1.3	0.1	-0.1	0.3	2.0	1.1	2.9
Si	0.98	0.75	1.20	0.54	0.04	1.05	57.3	12.5	102.0	6.1	-8.6	20.9	14.4	-58.3	87.2
Ca	1.00	0.65	1.35	0.61	-0.02	1.25	34.1	3.4	64.8	11.9	-0.6	24.4	165.6	72.0	259.1
Fe	1.00	0.76	1.24	0.70	0.11	1.29	23.8	1.8	45.7	4.5	-3.3	12.3	23.8	-16.3	63.9
K	1.00	0.81	1.19	0.16	-0.04	0.37	8.9	-0.5	18.3	0.0	-4.4	4.4	121.3	85.7	156.9
S	1.00	0.97	1.03	0.16	0.12	0.19	4.0	-3.7	11.7	1.0	-3.9	5.9	27.1	2.4	51.7
Zn	0.71	0.57	0.86	0.78	0.31	1.25	7.5	4.2	10.9	0.4	-0.5	1.2	2.9	-1.5	7.4
Cl	0.50	0.28	0.72	0.64	0.05	1.24	20.6	7.2	34.0	5.9	0.1	11.6	102.6	54.0	151.2
PM10*	1.00	0.85	1.15	0.65	0.36	0.93	5.6	2.6	8.7	4.1	2.6	5.7	21.9	13.6	30.2
Al	1.00	0.80	1.20	0.80	0.38	1.21	154.5	52.0	257.0	69.5	16.6	122.4	97.6	-159.0	354.2
Mn	1.00	0.80	1.20	0.69	0.30	1.07	1.2	-0.2	2.6	0.9	0.1	1.7	1.1	-2.7	4.9
Br	1.00	0.90	1.10	0.21	0.11	0.32	0.4	0.1	0.6	0.1	0.0	0.3	1.8	1.2	2.5
Pb	1.00	0.89	1.11	0.14	0.01	0.26	0.0	-0.6	0.6	0.0	-0.3	0.3	2.1	0.4	3.9
Ti	1.00	0.80	1.20	0.60	0.22	1.08	10.3	2.6	18.1	4.0	0.3	7.8	10.0	-8.4	28.4
Cu	0.83	0.62	1.05	0.77	0.18	1.36	3.2	1.3	5.1	0.5	0.0	1.1	3.5	0.4	6.5
Sr	1.00	0.83	1.16	0.62	0.28	0.97	0.9	0.3	1.5	0.3	0.0	0.5	2.6	1.2	3.9
P			fail to converge												
Si	1.00	0.81	1.19	0.62	0.26	0.97	237.8	16.1	459.6	149.3	26.9	271.8	296.4	-239.9	886.6
Ca	1.00	0.68	1.32	0.63	0.06	1.20	107.6	-27.1	242.3	118.7	37.3	200.1	800.0	329.0	1271.0
Fe	1.00	0.80	1.20	0.66	0.26	1.06	51.5	-15.5	118.5	46.7	8.5	84.8	73.0	-109.8	255.9
K	1.00	0.83	1.17	0.46	0.17	0.75	43.6	8.6	78.5	17.6	0.1	35.2	215.7	116.9	314.5
S	1.00	0.96	1.04	0.21	0.17	0.26	22.7	10.4	34.9	6.8	-0.7	14.3	68.0	29.3	106.7
Zn	1.00	0.81	1.19	0.37	0.10	0.64	7.4	3.4	11.4	1.2	-0.2	2.5	4.0	-3.0	11.0
Cl	0.94	0.44	1.43	2.36	0.48	4.24	148.4	49.4	247.4	45.7	17.6	73.9	320.2	107.0	533.4

* Mass units in mg.

Inhalable particles, of which about 40% (1.7 ± 0.6 mg/min) is in the fine fraction. All elements emitted by cooking were limited almost completely to the coarse fraction. Sources other than cooking and smoking emit about 5.6 ± 3.1 mg/h of PM_{10} , of which only about 1.1 mg/h ± 1.0 (20%) is in the fine fraction.

Decay rates for elements associated with the fine fraction were generally lower than for elements associated with the coarse fraction, as would be expected. For example, sulfur, which has the lowest mass median diameter of all the elements, had calculated decay rates of 0.16 ± 0.04 and 0.21 ± 0.04 h⁻¹ for the $PM_{2.5}$ and PM_{10} fractions, respectively. The crustal elements (Ca, Al, Mn, Fe), on the other hand, had decay rates ranging from 0.6 to 0.8 h⁻¹.

A similar model was developed for the PAHs and phthalate esters. However, the model differs in not including a cooking source term because initial regression analyses did not identify cooking as an important source of PAHs. Also, because of a much lower sample size for the PAH and phthalate measurements (only 60 homes with outdoor measurements, compared to 160 for the particles), the parameter estimates show much larger uncertainties.

Results showed that penetration factors for most PAHs were very close to one (Table 2). Because of the very large uncertainties involved in trying to estimate four unknown parameters (penetration coefficients, decay rates, smoking source strengths, and flux from other indoor sources), the penetration coefficients were all set equal to one and the remaining three parameters in Table 2 were calculated using the nonlinear algorithm.

The estimated average decay rates for PAHs ranged from 0.4 to 1.6 h⁻¹ with sizeable variation. No apparent dependence on volatility was noted. Estimated decay rates for the phthalates had very large uncertainties.

Estimated indoor source strengths for smoking for benzo(a)anthracene, chrysene, benzo(e)pyrene, benzo(a)pyrene, benzo(ghi)perylene, and coronene were 122, 192, 86, 264, 244, and 245 ng/cigarette, respectively. Smoking contributed 20% to 40% of the total concentrations of eight PAHs in homes reporting smoking. Considering all homes, outdoor air contributed more than half the total concentrations of six PAHs, mostly the less volatile ones, and "other" (unidentified) indoor sources contributed more than half of three volatile PAHs.

These other indoor sources were responsible for 97% to 99% of the total

concentrations measured for diethyl phthalate and di-n-butyl phthalate. Smoking was not indicated as an indoor source for phthalates.

Other (unidentified) indoor sources were found to be very important for both of the phthalates and for the volatile PAHs, but the estimates of source strengths were highly uncertain.

The physical models had reasonably good fit to particulate PAHs, and the correlation between predicted and observed concentrations averaged about 0.7.

Outdoor air was the major source of indoor particles, providing about three-fourths of fine particles and two-thirds of inhalable particles in the average home. It was also the major source for most elements, providing 70% to 100% of the observed indoor concentrations for 12 of the 15 elements. Only copper and chlorine were predominantly due to indoor sources in both the fine particle and inhalable particle fractions.

Unidentified indoor sources accounted for most of the remaining particle and elemental mass collected on the indoor monitors. The nature of these sources is not yet understood. They do not include smoking, other combustion sources, cooking, dusting, vacuuming, spraying, or cleaning, since all these sources together account for less than the unidentified sources. For example, the unidentified sources accounted for 26% of the average indoor PM_{10} particles, whereas smoking accounted for 4% and cooking for 5%.

Of the identified indoor sources, the two most important were smoking and cooking (Figures 6 and 7). Smoking was estimated to increase 12-h average indoor concentrations of PM_{10} and $PM_{2.5}$ by 2 and 1.5 $\mu\text{g}/\text{m}^3$ per cigarette, respectively. Homes with smokers averaged about 30 $\mu\text{g}/\text{m}^3$ higher levels of PM_{10} than homes without smokers. Most of this increase was in the fine fraction. Cooking increased indoor concentrations of PM_{10} by about 20 $\mu\text{g}/\text{m}^3$, with most of the increase in the coarse particles.

Emission profiles for elements were obtained for smoking and for cooking. Major elements emitted by cigarettes were potassium, chlorine, and calcium. Elements associated with cooking included aluminum, iron, calcium, and chlorine.

Other household activities such as vacuuming and dusting appeared to make smaller contributions to indoor particle levels. An interesting finding was that commuting and working outside the home resulted in lower particle exposures than for persons staying at home.

As with the particle mass, daytime personal exposures to 14 of 15 elements were consistently higher than either indoor or outdoor concentrations. At night, levels of the elements were similar in all three types of samples.

Discussion

Source of Excess Personal Exposure

The more than 50% increase in daytime personal exposures compared to concurrent indoor or outdoor concentrations suggested that personal activities were important determinants of exposure. However, the nature of this "personal cloud" of particles has not yet been determined. Scanning electron microscopy was undertaken on 138 personal filters. Skin flakes were common on many filters. A preliminary analysis suggested that the average number of skin flakes per filter was 120,000 to 150,000. The mass of some personal filters may have been considerably increased by unusually large numbers of skin flakes. However, attempts to calculate the mass of skin flakes from estimates of their volume and density suggest an average contribution to the mass of only about 4 $\mu\text{g}/\text{m}^3$, less than 10% of the mass of the average personal cloud.

Another approach to the composition of the personal cloud is elemental analysis, using x-ray fluorescence. Analysis of all personal and indoor filters showed that 14 of 15 elements were elevated by values of 50% to 100% in the personal filters compared to the indoor filters (Figure 8). This observation suggests that a component of the personal cloud is an aerosol of the same general composition as the indoor aerosol. This could be particles created by activities, e.g., cooking, or reentrained household dust from motion (walking across carpets, sitting on upholstered furniture). House dust is a mixture of airborne outdoor aerosols, tracked-in soil and road dust, and aerosols produced by indoor sources. As such, it should contain crustal elements from soil, lead and bromine from automobiles, and other elements from combustion sources. This would be consistent with the observation that nearly all elements were elevated in personal samples. The fact that personal overnight samples showed smaller mass increases than the personal daytime samples is also consistent with the fact that the participants were sleeping for much of the 12-h overnight monitoring period, and were thus not engaging in these particle-generating or reentraining activities.

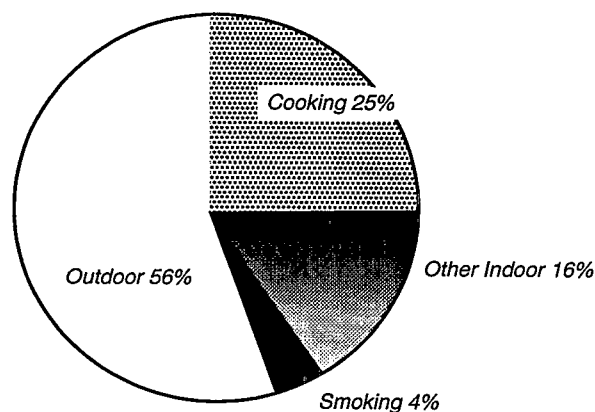
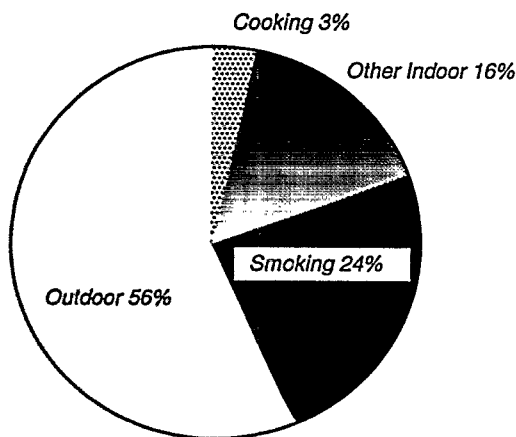
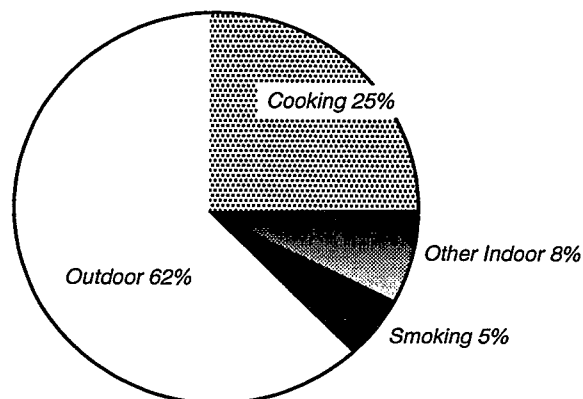
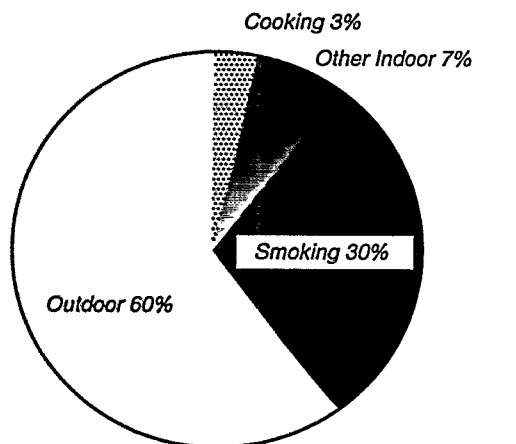
Table 2. Penetration Efficiencies, Decay Rates and Indoor Source Strengths for PAHs and Phthalates

Chemical	Penetration Factor P			r*	Decay rate k (per hour)			Source strength: smoking (ng/cigarette)			Flux from other indoor sources (ng/hour)		
	mean	L95%	U95%		mean	L95%	U95%	mean	L95%	U95%	mean	L95%	U95%
Acenaphthylene	0.40	-0.32	1.12	0.25	1.08	-1.48	3.63	3915	-5132	12962	2425	-1947	6797
Phenanthrene	0.91	0.61	1.21	0.43	0.38	0.03	0.73	10	-2108	2128	2777	1141	4413
Anthracene	0.81	0.49	1.12	0.40	0.88	0.06	1.70	246	-228	721	346	8	684
Fluoranthene	0.72	0.34	1.10	0.22	1.04	-0.10	2.18	161	-387	709	523	25	1021
Pyrene	1.00	0.23	1.77	0.64	1.65	0.58	2.71	122	19	226	32	2	62
Benzo(a)anthracene	1.00	0.53	1.47	0.80	0.76	0.33	1.20	192	82	301	13	-14	40
Chrysene	1.00	0.48	1.52	0.72	0.87	0.41	1.34	86	13	158	17	-5	38
Benzo(e)pyrene	1.00	0.31	1.69	0.74	1.29	0.64	1.94	264	101	426	48	9	87
Benzo(a)pyrene	1.00	0.13	1.87	0.44	0.68	-0.17	1.53	143	-156	441	65	-39	168
Indeno(1,2,3-cd)pyrene	1.00	0.51	1.49	0.71	0.82	0.39	1.24	244	-14	502	91	8	175
Benzo(g,h,i)perylene	0.83	0.47	1.19	0.71	0.52	0.17	0.87	245	44	447	70	6	134
Coronene	0.71	-0.25	1.24	0.24	1.46	0.07	2.85	10	-189872	189892	234632	73559	395704
Diethyl phthalate	0.92	-2.99	3.79	0.22	0.68	-0.11	1.47	10	-164691	164711	208921	72912	344930
Di-n-butyl phthalate													
Benzyl butyl phthalate													
Di-2-ethylhexyl phthalate													

r: correlation coefficients between predicted and observed concentrations.

Penetration factors are calculated using estimates of all four unknown parameters at same time.

Decay rate, smoking source strength and flux from other indoor sources are estimated assuming penetration factors are unity.



N = 61 Samples from 31 homes

N = 62 Samples from 33 homes

Figure 6. Sources of fine particles (top) and Inhalable particles (bottom) in 31 homes with smoking. Relative uncertainties in the estimates can be determined from the upper and lower confidence limits for the source strengths provided in Table 1.

Figure 7. Sources of fine particles (top) and inhalable particles (bottom) in homes with cooking. Relative uncertainties in the estimates can be determined from the upper and lower confidence limits for the source strengths provided in Table 1.

The measurements at the central site showed good agreement with the outdoor measurements at homes throughout the City of Riverside, indicating that a single central-site PM_{10} monitor can characterize a large urban area adequately. Although the correlations of indoor air concentrations with outdoor air are lower, there is evidence that outdoor air PM_{10} concentrations can affect indoor air concentrations.

The nonlinear least squares method of solving the mass-balance model improved on previous formulations in making fewer arbitrary assumptions and solving for all unknown parameters simultaneously. An interesting result from this effort was the

finding that the penetration factor was very close to one for nearly all particles, elements, PAHs and phthalate esters.

Conclusions

The personal and microenvironmental monitors designed especially for this study performed well. About 96% of all samples attempted were collected and median precision was 2% to 4%.

The major finding of the study was the 50% increase in daytime personal exposures to PM_{10} compared to indoor and outdoor concentrations. The increase appears to be due to personal activities such as dusting, vacuuming, cooking, and shar-

ing a home with a smoker. This suggests that reduction of dust levels in the home could decrease exposure to airborne particles.

A mass-balance model provided estimates for the source strengths of cigarettes and cooking for particles and elements in two size fractions. The model also provided estimates for the source strengths of cigarettes for a number of PAHs.

Elements in the Personal Cloud

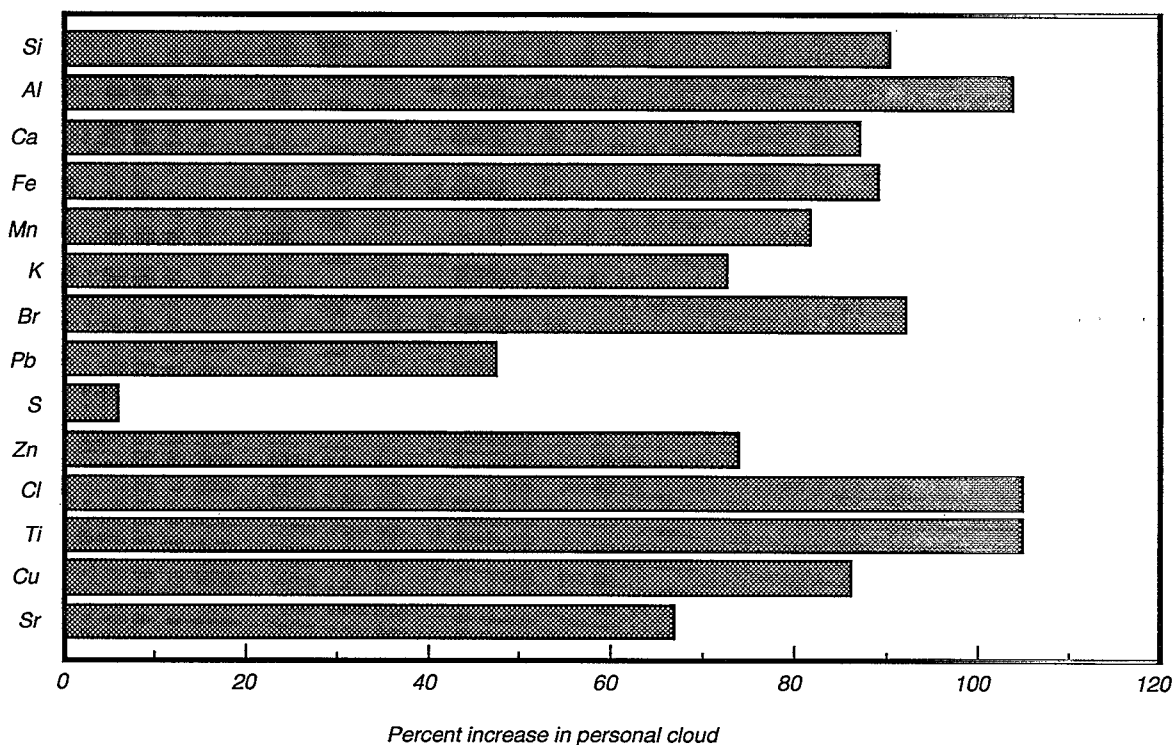


Figure 8. Increased concentrations of elements in the personal vs. the indoor samples.

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The complete report, entitled "The Particle Team (PTEAM) Study: Analysis of the Data—Final Report, Volume III," (Order No. PB97-102 495; Cost: \$85.00, subject to change) will be available only from:

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