INDOOR, OUTDOOR, AND PERSONAL AIR EXPOSURES TO PARTICLES, ELEMENTS, AND NICOTINE FOR 178 RESIDENTS OF RIVERSIDE, CALIFORNIA.

Lance A. Wallace[1], Halûk Özkaynak[2], John D. Spengler[2], Edo D. Pellizzari[3] and Peggy Jenkins[4]

[1] Atmospheric Research and Exposure Assessment Laboratory, Environmental Protection Agency, Warrenton, VA, USA
[2] Harvard University School of Public Health, Boston, MA, USA
[3] Research Triangle Institute, Research Triangle Park, NC, USA
[4] California Air Resources Board, Sacramento, CA, USA

ABSTRACT

Personal, indoor, and outdoor concentrations of inhalable particles and 15 elements were measured for a probability sample of 178 persons representing 139,000 nonsmoking residents of Riverside, California. Newly designed personal monitors were employed. Personal exposures often exceeded concurrent indoor and outdoor concentrations, both for particles and for 14 of 15 associated elements. The increase appears to be due to personal activities such as dusting, vacuuming, cooking, and sharing a home with a smoker. This suggests that reduction of dust levels in the home could decrease exposure to airborne particles.

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) joined with California's Air Resources Board to sponsor a study in the Los Angeles Basin. Small personal monitors were designed to measure inhalable particles (aerodynamic diameter less than 10 μm, or PM_{10}). The same monitors were used (with interchangeable sampling nozzles) to sample both inhalable (PM_{10}) and fine ($PM_{2.5}$) particles indoors and outdoors.

Following a 9-home study to test the measurement methods in the Azusa, CA area, a full-scale study was carried out in Riverside, CA in the fall of 1990. The main goal of the study was to estimate the frequency distribution of exposures to PM_{10} particles for all nonsmoking Riverside residents aged 10 and above, based on a probability sample of 178 residents. A second major objective was to estimate the frequency distribution of concentrations of PM_{10} and $PM_{2.5}$ in residences and nearby outdoor air (e.g., back yards). Other objectives included determining the effect of outdoor air on indoor concentrations, and the contribution of personal activities to exposure.

METHODS AND STUDY DESIGN

A personal exposure monitor (PEM) was designed to collect PM_{10} using a sharp-cut impactor with a circular set of holes each 1.9 mm in diameter. Particles are collected at a flow rate of 4 Lpm on a 37-mm Teflon filter mounted below a greased impactor plate. A backup filter coated with citric acid collected nicotine vapor. 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.

The same monitor, modified to operate with line current, was used to collect indoor and outdoor samples. This monitor is called the Stationary Ambient Monitor (SAM) when used outdoors and the Stationary Indoor Monitor (SIM) when used indoors. The sampling head can be replaced with one having holes 1.4 mm in diameter to collect fine particles ($PM_{2.5}$). Laboratory studies indicate that the PEM and the SAM_{10} have a sharp cutpoint at about 11 μm, while the $SAM_{2.5}$ has a sharp cutpoint at 2.5 μm.

A central site was selected to provide a record of temporal changes in outdoor particle levels for the duration of the study (48 days). The site had two high-volume samplers (Wedding & Assoc.) with 10-μm inlets (actual cutpoint about 9.0 μm), two dichotomous PM_{10} and $PM_{2.5}$ samplers (Sierra-Andersen) (actual cutpoint about 9.5 μm and 2.5 μm), one PEM and one SAM.

The city of Riverside, CA was selected for study because it is known to have highly variable outdoor PM_{10} concentrations (1). A wide range of outdoor concentrations offers the best chance of determining the contribution of outdoor levels to indoor levels and personal exposures. The fall season was selected since Santa Ana winds occur then; such winds can have strong effects on the outdoor concentrations of particles (1).

A three-stage probability sampling procedure was adopted. Thirty-six areas within Riverside were selected for study following socioeconomic stratification. Several homes from each area were sent letters explaining the study. Interviewers then collected information about each household and invited eligible residents to participate. Respondents represented 139,000 $\pm$ 16,000 (S.E.) nonsmoking Riverside residents aged 10 and above.

Smokers were excluded from participating, but nonsmoking members of their family were not. Employed persons were slightly oversampled, since employment was thought to be a possible risk factor for exposure to particles.

Each participant wore the PEM for two consecutive 12-hour periods. Concurrent PM_{10} and $PM_{2.5}$ samples were collected by the indoor SIM and outdoor SAM at each home. This resulted in 10 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-hour period, using perfluorotracers (1).

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

All filters were weighed on-site and then analyzed for elements by x-ray fluorescence (XRF). An additional set of about 600 citric-acid treated filters from personal and indoor samplers was analyzed for nicotine.

Filters were weighed before use and again within 48 hours of collection at an on-site weighing facility with controlled temperature and humidity. Replicate weighings were required to be within 4 μg/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 samples were collected at the central site. Duplicate PEM samples (5% of the total) were also collected by EPA, RTI, and Harvard scientists while on site.

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.

Blank PEM and SIM/SAM filters (N = 51) showed consistent small increases in mass of 5-10 μg. XRF analyses indicated that the increase was not due to particles; possibilities include water vapor or electric charge, although stringent efforts were made to control humidity and static charge in the on-site weigh room. The effect of the increase is small (0.4-4 $\mu g/m^3$) and was corrected for by subtracting the mean blank value from all samples. Limits of detection (LODs), based on three times the standard deviation of the blanks, were on the order of 10 $\mu g/m^3$. 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 2-4%.

The collocated samplers at the central site showed good agreement, with correlations ranging from 0.96 to 0.99. As had been noted in the pilot study, the PEM and SAM collected about 12% more mass than the dichotomous samplers, perhaps due to their higher cutpoint (11 μm compared to 9.5 μm) or to a particle "bounce" effect, measured in the laboratory at less than 9%. The Wedding samplers collected about 13% less mass than the dichots at night, but about the same level during the day, reflecting a possible temperature dependency on the part of the Wedding. Although these small differences were significant, they do not affect the main conclusions.

All PEM, SIM, SAM, and dichotomous sampler filters (about 2500) were analyzed by x-ray fluorescence (XRF) for a suite of 42 elements (1). The analysis was carried out at EPA's Atmospheric Research and Exposure Assessment Laboratory in Research Triangle Park, NC. Some filters were analyzed twice under blind conditions. A subset of about 100 filters was analyzed by the Lawrence Berkeley Laboratory (LBL).

Background levels on laboratory and field blanks were very low for 19 of 20 elements. Blank levels for iron were slightly higher, but were 4 to 100 times lower than observed concentrations. 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 less than 15% for all 15 prevalent (more than 30% of samples with measurable quantities) elements. Median RSDs for duplicates analyzed by the two laboratories were less than 21% for all elements except manganese (76%) and copper (27%).

Outdoor 12-h PM-10 concentrations at the central site ranged from 20-200 $\mu g/m^3$, with the fine particles accounting for most of the variation. On the six windiest (16-20 knots) days, the coarse particles accounted for most of the PM_{10} mass.

Population-weighted daytime personal PM_{10} concentrations averaged about 150 $\mu g/m^3$, compared to concurrent indoor and outdoor mean concentrations of about 95 $\mu g/m^3$ (Table 1). The overnight personal PM_{10} mean was much lower (77 $\mu g/m^3$) and more similar to the indoor (63 $\mu g/m^3$) and outdoor (86 $\mu g/m^3$) means. About 25% of the population was estimated to have exceeded the 24-h National Ambient Air Quality Standard for PM_{10} of 150 $\mu g/m^3$. Over 90% of the population exceeded the California Ambient Air Quality Standard of 50 $\mu g/m^3$. Fine ($PM_{2.5}$) particles accounted for about 50% of the total PM_{10} mass both indoors and outdoors.

The measurements at the central site showed good agreement (r = 0.93 at night, 0.66 during the day) 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 (0.59 at night, 0.51 during the day), they provide some evidence that outdoor air PM_{10} concentrations can affect indoor air concentrations.

Air exchange rates varied from 0.15 to 4.74 h^{-1}, with mean (median) values of 1.24 (1.02) h^{-1} (daytime) and 1.09 (0.80) h^{-1} (overnight) in 174 homes. Nicotine levels ranged from 0.05 $\mu g/m^3$ to 14.9 $\mu g/m^3$, with a mean (median) value of 1.42 (0.45) $\mu g/m^3$. Nicotine levels were measurable (mean of 2.2 $\mu g/m^3$) in 35 of the 39 homes reporting smoking, and were below detectable levels (mean of .12 $\mu g/m^3$) in all but six homes without smokers. Personal nicotine mean levels were similar to indoor means (1.48 $\mu g/m^3$ vs. 1.39 $\mu g/m^3$).

Table 1. Population-Weighted[a] Concentrations (± SE) ($\mu g/m^3$)

Sample type	N	Median	Arith. Mean	Percentile 90%	98%
Daytime PM_{10}					
Personal	171	130 ± 8	150 ± 9	260 ± 12	380
Indoor	169	82 ± 8	95 ± 6	180 ± 11	240
Outdoor	165	83 ± 5	94 ± 6	160 ± 7	240
Overnight PM_{10}					
Personal	168	66 ± 4	77 ± 4	140 ± 10	190
Indoor	163	52 ± 4	63 ± 3	120 ± 5	160
Outdoor	162	74 ± 4	87 ± 4	170 ± 5	210
Daytime $PM_{2.5}$					
Indoor	173	34 ± 4	48 ± 4	100 ± 7	170
Outdoor	167	36 ± 4	49 ± 3	100 ± 6	170
Overnight $PM_{2.5}$					
Indoor	166	26 ± 2	36 ± 2	83 ± 6	120
Outdoor	161	35 ± 2	51 ± 4	120 ± 5	160

[a] Personal samples weighted to represent nonsmoking population of 139,000 Riverside residents aged 10 or above. Indoor-outdoor samples weighted to represent 61,500 homes with at least one nonsmoker aged 10 or above.

Population-weighted mean elemental concentrations for 15 prevalent elements were calculated. As with the particle mass, daytime personal exposures were consistently higher than either indoor or outdoor concentrations of all the elements save sulfur. At night, levels were similar in all three types of samples. The personal and indoor PM_{10} samples are depleted in the crustal elements (Si, Al, Fe) compared to the outdoor samples, by amounts ranging from 15 to 25%. The indoor $PM_{2.5}$ samples show no depletion in any elements, and may be slightly enriched in Ca, K, Cl, and (night only) S.

Questionnaires were analyzed to detect activities associated with increased exposure. Housework (dusting, vacuuming, cooking) was associated with significantly increased personal exposures and indoor air concentrations during the day. Sharing a home with one or more smokers also led to increased personal exposures and indoor air concentrations during the night. Persons who commuted to work had significantly lower exposures than those who stayed at home.

DISCUSSION

The personal and microenvironmental monitors designed especially for this study performed well. About 96% of all samples attempted were collected and median precision was 2-4%. A positive bias of about 12% was noted with respect to the reference dichotomous sampler method.

The source of the roughly 50% increase in daytime personal exposure compared to the indoor and outdoor air concentrations appears to be generation or reentrainment of particles during personal activities such as cooking, dusting, and vacuuming. Other possible sources have been found to be unlikely. These include 1) different sampling characteristics of the monitors (no differences noted in laboratory tsts); 2) skin flakes or clothes fibers accumulating on the personal monitor (does not account for the observed higher elemental concentrations); and 3) increased exposures encountered while participants are out of the house (not supported by multivariate analyses).

House dust is a mixture of airborne outdoor aerosols, tracked-in soil and road dust, and aerosols produced by indoor sources such as combustion. 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-hour overnight monitoring period, and were thus not engaging in these particle-generating or reentraining activities. There remains the problem of sulfur, which showed no increase in personal samples compared to indoor or outdoor samples. This may be due to the fact that sulfate ions have a much smaller mass median diameter and a lower deposition velocity than other ionic constituents of fine particles. Thus sulfur would not tend to accumulate in house dust as much as other elements. Also, smaller particles may be harder to dislodge from surfaces, due to electrostatic or other forces.

REFERENCES

1. Pellizzari, E.D., Thomas, K.W., Clayton, C.A., Whitmore, R.W., Shores, R.C., Zelon, H.S., and Perritt, R.L. Particle Total Exposure Assessment Methodology (PTEAM): Riverside, California Pilot Study. Vol. 1. US EPA. Research Triangle Park, NC. 1992.

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use. This research was funded in part by the California Air Resources Board.

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before cc

1. REPORT NO.	2.	
EPA/600/A-93/144		

4. TITLE AND SUBTITLE	5. REPORT DATE
Indoor, Outdoor, and Personal Air Exposures to Particles, Elements, and Nicotine for 178 Residents of Riverside, California	7/93
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) L.A. Wallace, H. Ozkaynak, J. Spengler, E. Pellizzari, P. Jenkins	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Research Triangle Institute Research Triangle Park, NC 27709	10. PROGRAM ELEMENT NO.
	11. CONTRACT/GRANT NO. 68-02-4544
12. SPONSORING AGENCY NAME AND ADDRESS US EPA Research Triangle Park, NC 27711	13. TYPE OF REPORT AND PERIOD COVERED
	14. SPONSORING AGENCY CODE EPA/600/09

15. SUPPLEMENTARY NOTES

16. ABSTRACT

Personal, indoor, and outdoor concentrations of inhalable particles and 15 elements were measured for a probability sample of 178 persons representing 139,000 nonsmoking residents of Riverside, California. Newly designed personal monitors were employed. Personal exposures often exceeded concurrent indoor and outdoor concentrations, both for particles and for 14 of 15 associated elements. The increase appears to be due to personal activities such as dusting, vacuuming, cooking, and sharing a home with a smoker. This suggests that reduction of dust levels in the home could decrease exposure to airborne particles.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES 7
	20. SECURITY CLASS (This page)	22. PRICE

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE