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Exposure of High Risk Subpopulations to Particles:

Final Report (APM 21)

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by

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Abstract

This final report describes results of studies EPA conducted on the exposure of high-risk subpopulations to particles. The overall goal of these studies was to relate personal exposure to outdoor concentrations of particles, particularly fine particles less than 2.5 micrometers in diameter ($PM_{2.5}$). The studies were carried out by EPA's National Exposure Research Laboratory (NERL), and by two university consortia (Harvard University School of Public Health and the University of Washington Department of Environmental Studies). All studies included repeated measurements over 10-28 days of personal, indoor, and outdoor air $PM_{2.5}$, including indoor and outdoor air PM_{10} and associated co-pollutants. Nearly 2500 personal $PM_{2.5}$ filters and a much larger number of indoor and outdoor $PM_{2.5}$ and PM_{10} filters were collected from more than 200 participants in 5 cities. All participants filled out identical household questionnaires and time-activity diaries providing information on where they spent their time and what particle sources were active. Participants were chosen from several high-risk subpopulations: adults with chronic obstructive pulmonary disease (COPD), coronary heart disease (CHD), and hypertension, and children with asthma. Healthy cohorts were also included as controls. Some participants were in retirement homes but most lived in their own homes or apartments. All subjects were chosen on a non-probabilistic basis, and therefore ***all conclusions apply only to the subjects themselves; they must not be extrapolated to larger groups of people.***

Results indicated little difference in exposure between disease cohorts and healthy persons in the same geographic area. Across the cities, mean $PM_{2.5}$ exposures ranged between 9.3 and 23 $\mu g/m^3$, compared to mean indoor concentrations of 7.4-20 $\mu g/m^3$, and mean outdoor concentrations of 9.0-22 $\mu g/m^3$. Seasonal variations were important in some cities, unimportant in others. Median longitudinal correlations of personal $PM_{2.5}$ exposure with outdoor concentrations ranged from 0.1 to 0.65. However, all cohorts had some persons with high correlations with outdoor air and others with low correlations, indicating the importance of activities and household characteristics that either change over time or are basically repeatable from day to day. Different cities had different estimates of the mean infiltration factor, a measure of the influence of outdoor air particles on indoor concentrations. For private residences, calculated values of the infiltration factor for $PM_{2.5}$ ranged from 0.40 to 0.53 during the heating season and from 0.45 to 0.79 during the non-heating seasons.

Contents

Figures

Tables

Acknowledgements

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Grateful acknowledgement is made to the hundreds of participants and supportive family members who made these studies possible.

INTRODUCTION

In the 1990s, many studies around the world indicated that daily mortality increased with increasing outdoor air pollution. Suspicion mainly fell on particles, although associated co-pollutants (sulfates, SO_2, NO_2, CO, and ozone), which are often highly correlated with particles, continue to be considered possible contributors. The findings led the US EPA to propose a new National Ambient Air Quality Standard (NAAQS) for fine particles ($PM_{2.5}$). The United States Congress appropriated additional money to study the problem, and the National Academy of Sciences (NAS) was asked to outline a research program. The NAS report (NRC-NAS, 1998) envisioned a 13-year research "portfolio," the first three years of which would concentrate on measuring actual personal exposure, particularly for high-risk subpopulations, to fine particles and associated co-pollutants.

Even before the NAS report was published, the National Exposure Research Laboratory (NERL) at Research Triangle Park, NC had planned such a study and had already published a Request for Applications. The NAS report and Congressional funding increased the funds available for the study and made it possible to fund a series of studies instead of the one originally planned. Three of these were to be carried out by University consortia under co-operative agreements and the remainder by EPA. The studies began in 1999, and fieldwork has been completed in all but one of the studies.

Prior to these studies, which concentrated on persons living in their own homes and apartments, the NERL carried out two studies in retirement homes in Baltimore, MD and Fresno, CA. Last year, a report on the progress of the studies was published (US EPA 2002). That report provided the study designs and proposed measurement methods for all of the studies. Because the measurement methods were fully described in last year's report, these descriptions will not be repeated in this report. (An exception is the description of a new multipollutant monitor developed by the Harvard University School of Public Health. This description is found below in the Quality Assurance section.) Last year's report also included summaries of $PM_{2.5}$ and PM_{10} mass concentrations for the Baltimore and Fresno studies, and therefore these results will not be repeated in this report. (Again one exception is a table in the Discussion section comparing results from all cities.) This final report summarizes work on particles from studies in five cities: Research Triangle Park, NC; Atlanta, GA; Boston, MA; Los Angeles, CA; and Seattle, WA.

STUDY DESIGN

All studies had a basic study design involving personal, indoor, and outdoor monitoring of persons in certain high-risk subpopulations. All studies fielded after 1999 used the same questionnaire, which was approved by the Office of Management and Budget (OMB) in July of 1999. Within the basic design, each group was free to emphasize different aspects, so that some variation in pollutant monitoring existed among the groups and the various field studies. The major goal of all studies was to relate personal exposure to outdoor concentrations, a crucial need to support regulation of outdoor particles.

Because the epidemiology studies are time series studies, the relation of personal exposure to outdoor concentrations must be calculated over a number of days for each subject. Earlier studies had indicated that the minimum number of needed days would be greater than 3 and less than 7. Therefore all studies followed participants for a minimum of 7 days. All studies calculated longitudinal correlations of personal exposure to outdoor concentrations. Several hypotheses were developed that could be tested in the course of the studies. One hypothesis was that high-risk subpopulations would engage in fewer dust-generating activities and would therefore have lower indoor air concentrations and lower exposures than healthy cohorts. A second related hypothesis was that their personal exposures would have stronger correlations with outdoor air than healthy cohorts, due to fewer particle sources indoors. A brief description of the study design for each city follows.

Research Triangle Park, NC

Following two earlier studies of retirement home residents in Baltimore and Fresno, fully described in the first report in this series (US EPA 2002), the Research Triangle Institute International, a not-for-profit research institute, was contracted to collect environmental samples based upon a NERL study design (Ronald Williams, Project Officer). NERL scientists analyzed the resulting field and laboratory data. This group studied two cohorts in the Research Triangle Park, NC area: an African-American cohort with controlled hypertension living in a low-moderate socioeconomic status (SES) neighborhood, and persons with implanted cardiac defibrillators. Participants were non-smoking, 50+ years of age, and living in their own homes. The participants were monitored for 7 consecutive days during four consecutive calendar seasons. Besides personal particle monitors to measure mass, participants also carried a real-time light-scattering device, the MIE pDR (personal DataRAM®) to estimate short-term (1 minute average) particle exposures. Air exchange rates in each home were also measured. The study design is shown in Table 1.

Table 1. Study Design: Research Triangle Park

Study city	SE Raleigh	Chapel Hill	Monitor Types
Panel description	Low SES neighborhoods, minorities with controlled hypertension	Cardiac Defibrillators	--
Number of participants	27	8	--
Seasons (days/season)	4 (7)	4 (7)	--
PM 2.5 mass	P, I, O, A	P, I, O, A	PEM, HI, Dichot, FRM, TEOM,
PM 10 mass	I, O, A	I, O, A	PEM, Dichot
PM nephelometer	P, I	P, I	MIE pDR
PM number count and distribution	I, O select homes	I, O select homes	SMPS-APS
EC-OC	P, I, O, A	P, I, O, A	PEM
NO_2	I, A	I, A	Ogawa badge
O_3	P, A	P, A	Ogawa badge, TECO
CO	I, O, A	I, O, A	Draeger, TECO
Elements (SO_4)	P, I, O, A	P, I, O, A	PEM, HI, Dichot, FRM
Air Exchange	PFT	PFT	PFT tubes
Health Measures	PEF, FEV, pulse, 0_2 sat.	PEF, FEV, pulse, O_2 sat.	AirWatch, Nellcor N-20

P = personal, I = indoor residential, O = Outdoor residential, A= ambient, EC-OC = elemental and organic carbon, PFT = perfluorotracer method, PEF = Peak expiratory flow, FEV = Forced expiratory volume, O_2 sat= blood oxygen saturation. PEM = personal exposure monitor, HI = Harvard impactor, Dichot = dichotomous sampler, FRM = Federal Reference Method, pDR = personal Data Ram, SMPS-APS = Scanning Mobility Particle Sizer – Aerodynamic Particle Sizer. MIE, Ogawa, Airwatch, Draeger, TECO, TEOM, Nellcor = manufacturers or trade names.

Atlanta, Boston, and Los Angeles

The Harvard School of Public Health (Petros Koutrakis, Principle Investigator) studied subpopulations in Atlanta, Boston, and Los Angeles. In each city, personal, indoor and outdoor multi-pollutant samples were collected over seven-day periods in two seasons. The high-risk subpopulations monitored included those with chronic obstructive pulmonary disease (COPD) and those with heart disease, including myocardial infarctions (MI). A healthy cohort was also monitored. The pollutants monitored included personal, indoor, and outdoor $PM_{2.5}$, sulfate, ozone, SO_2, NO_2, elemental carbon and organic carbon (EC/OC). Personal PM_{10} was also monitored in Los Angeles only. The measurement methods included a multipollutant personal sampler developed at Harvard and capable of sampling $PM_{2.5}$, PM_{10}, SO_2 and sulfate (in Eastern cities), NO_2 and nitrate (in Western cities), ozone, and EC/OC. Indoor and outdoor particles were measured using Harvard Impactors (HIs) for both $PM_{2.5}$ and PM_{10}. The basic study design is provided in Table 2.

Table 2. Study Design—Atlanta, Boston, Los Angeles

		COHORT						
		Healthy		COPD		MI[†]		Target
		N	Days	N	Days	N	Days	Person-days
Atlanta	Fall 1999	--	--	15	7[a]	9	7	168
	Spring 2000	--	--	13	7[b]	9	7	154
Boston	Winter 1999-2000	8	7	4	7[c]	11	7	159
	Summer 2000	6	7	5	7	10	7	147
Los Angeles	Winter1999-2000	--	--	15	7[d]	--	--	103
	Summer 2000	--	--	14[e]	7	--	--	98
TOTAL		14		66		39		829

[†] Boston MI cohort includes individuals with conjunctive heart failure, history of by-pass surgery, and medication-treated angina

[a] One subject monitored for four days and another for six days.

[b] Three subjects monitored for only six days each.

[c] One subject monitored for 5 days.

[d] One subject monitored for 5 days.

[e] One subject was excluded, as the person was admitted to the hospital and spent little time at home during the sampling period.

Seattle

This city was studied by the University of Washington Dept. of Environmental and Occupational Studies (L-J. Sally Liu, Principle Investigator). Four different cohorts were studied: persons with chronic obstructive pulmonary disease (COPD), coronary heart disease (CHD), children with asthma, and healthy elderly persons (Table 3). Participants were monitored during two "seasons"—the heating season (October through February) and the non-heating season. The pollutants monitored are summarized in Table 4.

Table 3. Study Design: Seattle (Subpopulations)

Year	Starting Date	Asthmatics	CHD	COPD	Healthy	Total
1999	Oct 26	-	0	5	3	8
	Nov 8	-	0	5	4	9
	Nov 29	-	0	5	3	8
2000	Jan 10	-	0	3	6	9
	Feb 7	-	1	2	3	6
	Feb 21	-	0	3	3	6
	Mar 6	-	1	3	3	7
	Mar 27	-	0	4	4	8
	Apr 10	-	0	3	2	5
	May 1	-	0	5	3	8
	May 15	-	0	4	0	4
	Jul 10	-	1	4	2	7
	Jul 31	-	1	1	2	4
	Sep 25	-	2	4	-	6
	Oct 16	-	3	5	-	8
	Nov 6	-	6	-	-	6
	Nov 27	2	4	-	-	6
	Dec 25	4	1	-	-	5
2001	Jan 8	5	1	-	-	6
	Jan 22	3	3	-	-	6
	Feb 5	2	3	-	-	5
	Feb 26	4	4	-	-	8
	Mar 29	5	3	-	-	8
	Apr 16	3	2	-	-	5
	Apr 30	4	1	-	-	5
	May 14	1	3	-	-	4
		33	40	56	38	167

Number of subjects by cohort and session. A total of 108 subjects were monitored, about 50% were monitored twice.

Table 4. Study Design: Seattle (Measurement Methods)

Measurement	Personal	Indoor	Outdoor	Central Site
PM_{10}	-	HI (10 lpm)	HI (10 lpm)	HI (10 lpm) x 2
$PM_{2.5}$	HPEM (4 lpm)	HI (10 lpm)	HI (10 lpm)	HI (10 lpm) x 2
Pump	BGI	Medo	Gast	Gast
PM_1	pDR	Neph or pDR	Neph	Neph
WS/SVOC	PUF	PUF	PUF	PUF
EC/OC	HPEM	HI & IOGAPS	HI & IOGAPS	HI & IOGAPS
Biomarker	Urine sample	-	-	-
CO	Breath sample	Langan/bag	-	-
NO_2/SO_2	Ogawa	Ogawa	Ogawa	Ogawa
ACH	-	TelAir/PFT	TelAir	-
Continuous RH	-	Onset logger	-	-
Continuous T	-	Onset logger	-	-
Compliance	Motor on/off	-	-	-
Time/activity	Diary form	-	-	-
PEF/FEV_1[c]	Airwatch	-	-	-
Pulse rate / O_2 saturation	Pulse Oximeter	-	-	-
HRV and BP	Holter	-	-	-

HI = Harvard Impactor; HPEM = Harvard Personal Exposure Monitor; pDR = Thermo-MIE personal DataRam; Neph = Radiance Reflectance nephelometer; WS = wood smoke; SVOC = semi-volatile organic compounds; PUF = polyurethane foam; IOGAPS = indoor-outdoor gaseous air pollution sampler; Langan = Langan Instruments, Inc. T15 CO Measurer; BGI, Medo, Gast, TelAir and Ogawa = manufacturers; PFT = perfluorotracer method; PEF/FEV_1 = peak expiratory flow/forced expiratory flow (1 second); Airwatch = spirometer; HRV = heart rate variability; BP = blood pressure.

QUALITY ASSURANCE

Since the personal, indoor, and residential outdoor monitors employed in these studies are not EPA reference instruments, side-by-side comparisons with the EPA Federal Reference Method (FRM) or with instruments that have previously been compared with the FRM, such as the Harvard Impactor (HI) were carried out. The following text and figures illustrate that the precision and accuracy of these monitors compared well with EPA reference methods.

Research Triangle Park, NC

A major effort was made to compare all monitors used in this study and a journal article focusing on monitor performance was produced (Williams et al., 2000b). In general, precision was very good at levels of about 5%, and agreement with EPA reference methods was also good. The personal and indoor monitors (HI) were collocated with the EPA Federal Reference Method (FRM) monitors at the central site. The personal monitor showed a slightly positive bias with an intercept of 4 $\mu g/m^3$, a slope of 0.95, and an R^2 of 86% (Figure 1). The indoor monitor agreed better with a negligible intercept, a slope of 1.04 and an R^2 of 97%. The regression of the personal monitor vs. the HI had an intercept of 2.7 $\mu g/m^3$, a slope of 0.95, and an R^2 of 88%.

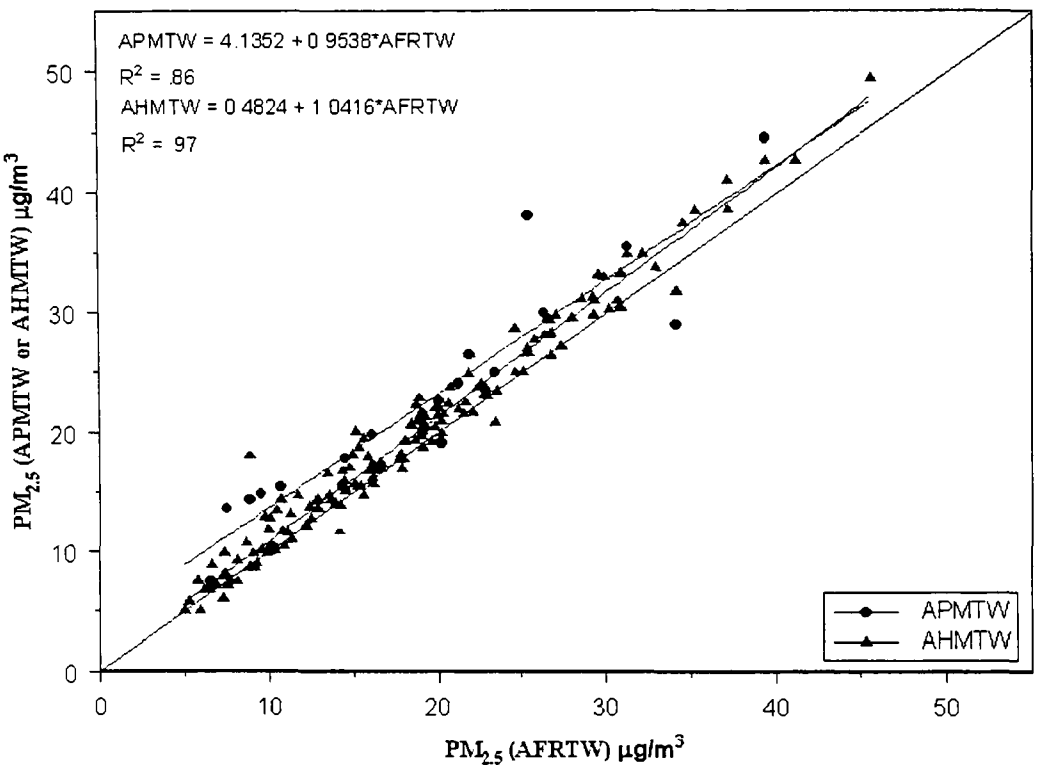

Figure 1. Collocated personal (APMTW) and indoor (AHMTW) monitors with EPA's Federal Reference Method (FRM) monitor at the Research Triangle Park central site. Regression lines are compared to the one-to-one line.

Atlanta

During the fall of 1999, the SKC PEM fine particle personal monitor was run side by side at the Atlanta central site against the HI, which has previously shown that it agrees well with the EPA FRM. Agreement was poor, with an intercept of 6 $\mu g/m^3$ and a low slope of 0.78 (Figure 2).

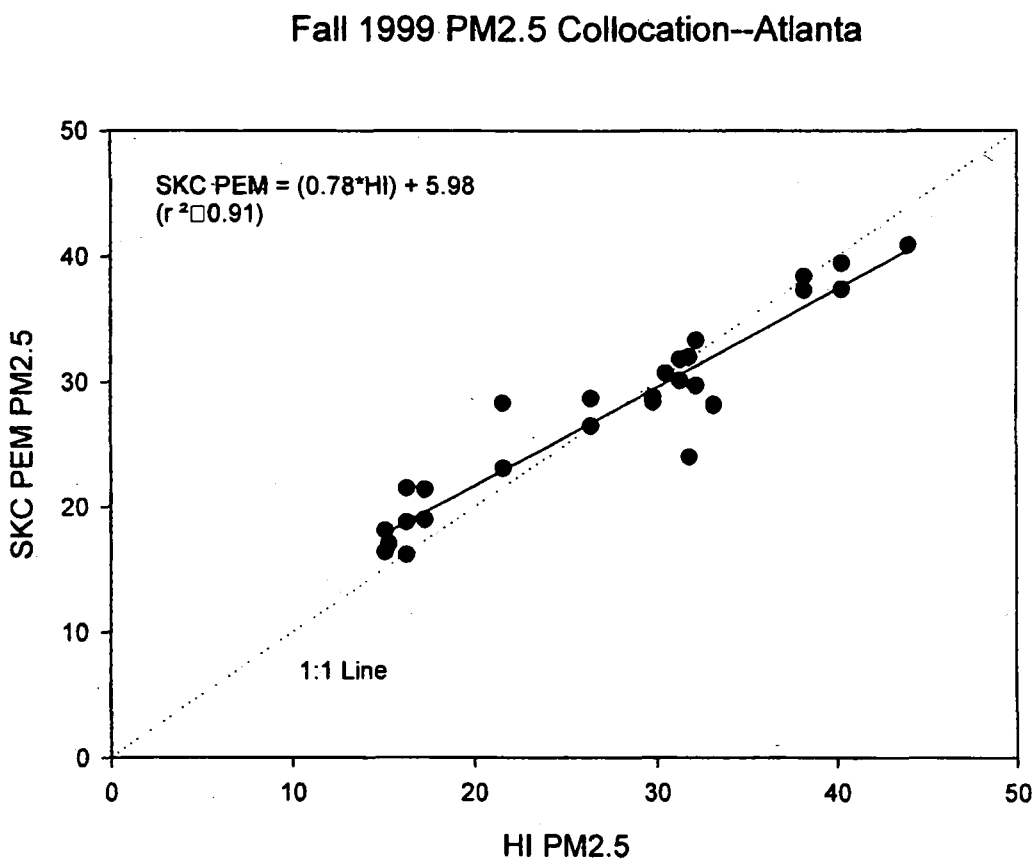

Figure 2: Wintertime comparison of the SKC Personal Exposure Monitor (PEM) with the Harvard Impactor (HI) at the Atlanta central site.

During the spring of 2000, the SKC PEM was replaced by the Harvard PEM, and agreement with the HI at the central site was considerably improved, with a negligible intercept, a slope of 1.06 and an R^2 value of 84% (Figure 3).

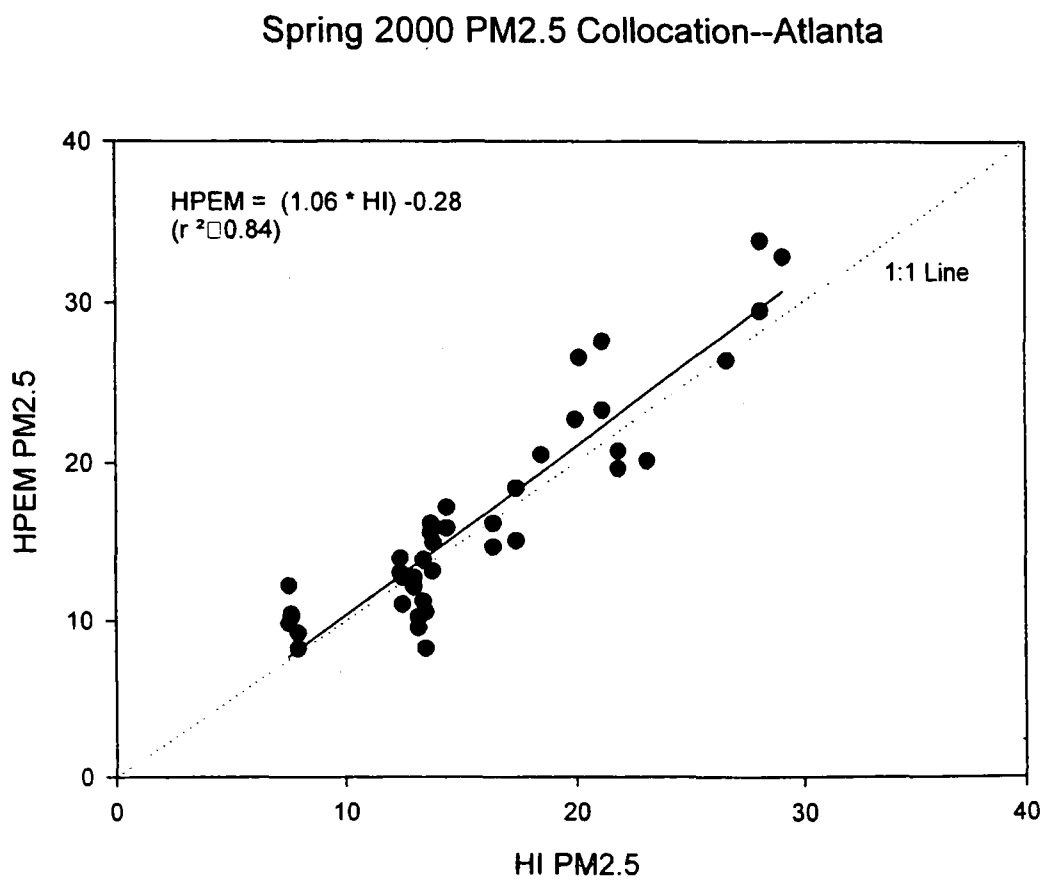

Figure 3. Springtime comparison of the Harvard Personal Exposure Monitor (HPEM) with the Harvard Impactor (HI) at the Atlanta central site.

Boston

The Harvard PEM (HPEM) fine particle personal monitor was run side by side at the Boston central site against the Harvard Impactor (HI). Agreement was good, with a slope of 1.13 and an R^2 value of 95% (Figure 4).

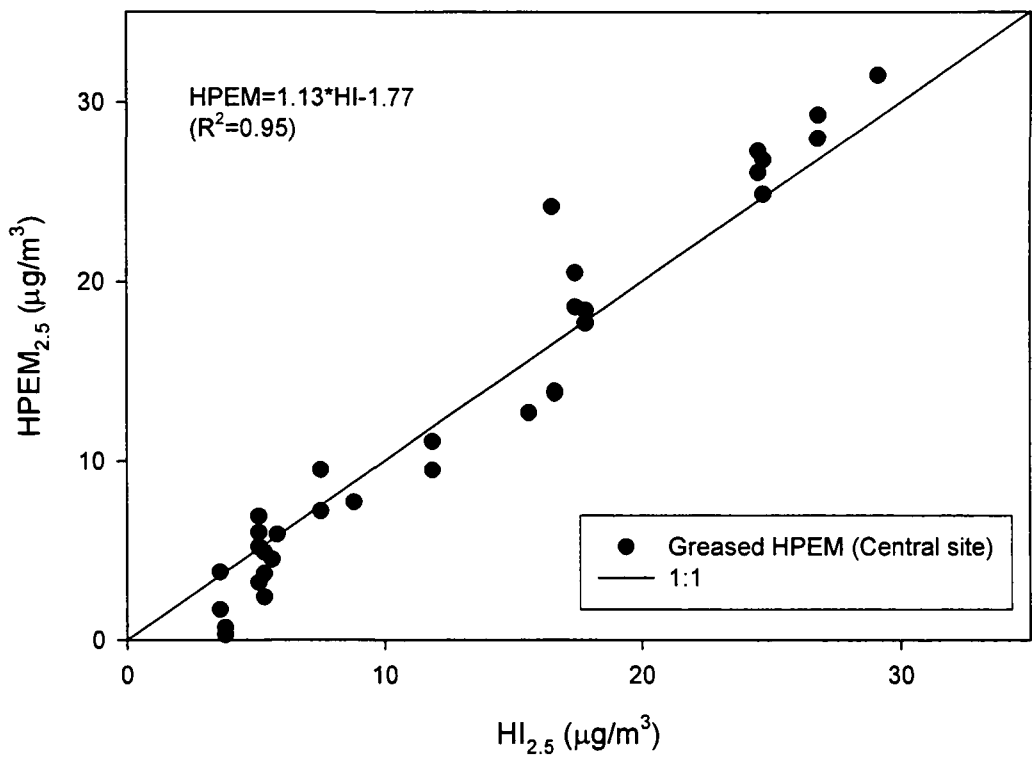

Figure 4. The Harvard PEM ($HPEM_{2.5}$) vs. the Harvard Impactor ($HI_{2.5}$) when collocated at the Boston central site.

Los Angeles

Following the Atlanta and Boston studies referenced above, the Harvard University School of Public Health developed an integrated multi-pollutant monitor to measure the simultaneous particulate and gaseous exposures for 22 COPD patients in Los Angeles. This sampler is essentially several individual samplers that have been joined together to form a simple, compact, and relatively lightweight personal monitor (Figure 5). The entire monitor (plus pump and battery pack) weighs approximately six pounds. Participants were able to wear the monitor successfully throughout the monitoring period. Participants were allowed to remove the monitor and place it nearby when they would be stationary for long periods of time, such as when they were sleeping or reading. For participants using oxygen, monitors were strapped to their oxygen tanks to ease the sampling burden.

For indoor, outdoor home, and central site monitoring, the monitors were placed on a tripod, with the inlets approximately one meter above the ground. Both indoor and outdoor monitors were placed away from any objects (e.g., trees, houses, vents) to minimize interference with pollutant measurements. Outdoor monitors were placed under a rain cap to protect the samplers from precipitation. For personal monitoring, the monitor was attached by Velcro to the shoulder strap of a padded backpack at breathing level. If the participant was mobility-restricted or otherwise hampered, the samplers were attached to fixed objects near the participant's body, with the inlet protruding into the breathing zone.

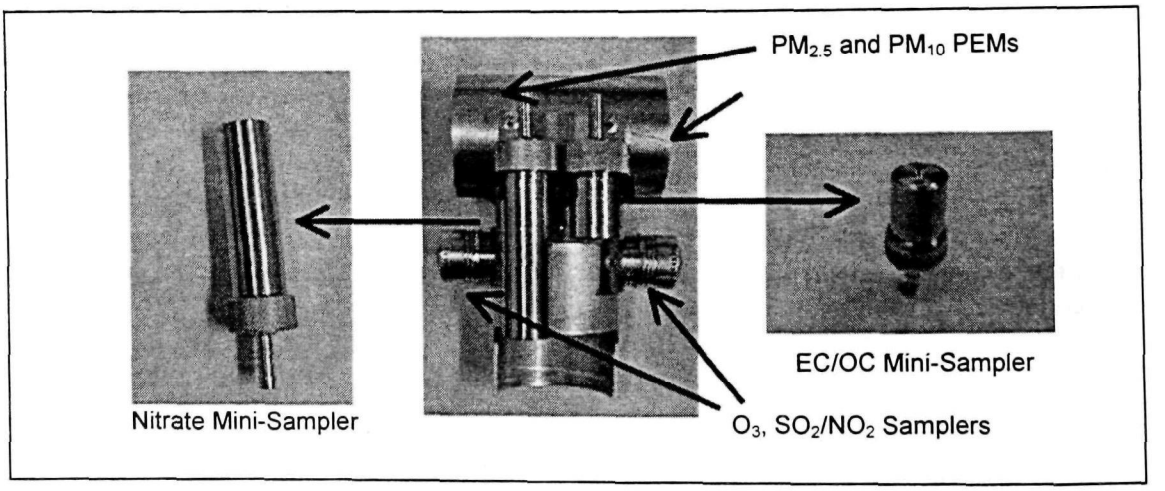

Figure 5. Multi-Pollutant Sampler developed by Harvard University School of Public Health.

The multi-pollutant sampler measured PM_{10} and $PM_{2.5}$ concentrations using PEMs, small inertial impactors designed specifically for personal and micro-environmental monitoring

(Marple *et al.*, 1987; Thomas *et al.*, 1993; Chang *et al.*, 1999; Demokritou *et al.*, 2001a). Impactor plates in all samplers were greased to minimize particle bounce (Demokritou *et al.*, 2001a; Demokritou *et al.*, 2001b). In both seasons, indoor and outdoor PM_{10} and $PM_{2.5}$ measurements were made using Harvard PEMs operated at flow rates of 4 LPM. In the winter, personal PM_{10} and $PM_{2.5}$ concentrations were measured using PEMs manufactured by SKC. Since these samplers were designed to operate at flow rates of 4 LPM, the SKC PEMs were modified to allow their use at flow rates of 1.6 and 2 LPM for PM_{10} and $PM_{2.5}$ sampling, respectively (Rojas-Bracho *et al.*, 2000). Because the cut-point of the impactors are a function of flow rate, the number of nozzle holes was reduced from ten to four for the PM_{10} PEM and to five for the $PM_{2.5}$ PEM to maintain the same size cut-offs as originally designed. In the summer, personal PM_{10} and $PM_{2.5}$ concentrations were measured using PEMs designed by Harvard to operate at 1.8 LPM, since these samplers were lighter, could be used without modification, and would be comparable to the Harvard PEM samplers used to sample indoors and outdoors.

Both the SKC and Harvard PEMs used Teflon filters as the particle collection media and included drain disk rings to prevent metal contamination for future ICP-MS analysis. The $PM_{2.5}$ and PM_{10} PEMs were attached to either side of the monitor using a 10 cm long elutriator (Figure 5). Nitrate and EC/OC mini-samplers were attached to the front of the elutriator using clips. The passive O_3 and SO_2/NO_2 badges were placed in the side of the elutriator, with their face exposed to the sample air stream to allow for constant sampler collection rates.

Collocated PEMs and HIs at the Los Angeles central site showed reasonable agreement with a slope of 0.95 and an R^2 value of 90% (Figure 6). However, the intercept of 3 $\mu g/m^3$ was a bit higher than could be hoped for. During the second season of the Los Angeles study, PM_{10} and $PM_{2.5}$ personal samples were collected simultaneously. The PM_{10} PEM was collocated at the L.A. central site with an HI_{10}, with reasonable agreement ($R^2 = 0.92$, slope = 1.22).

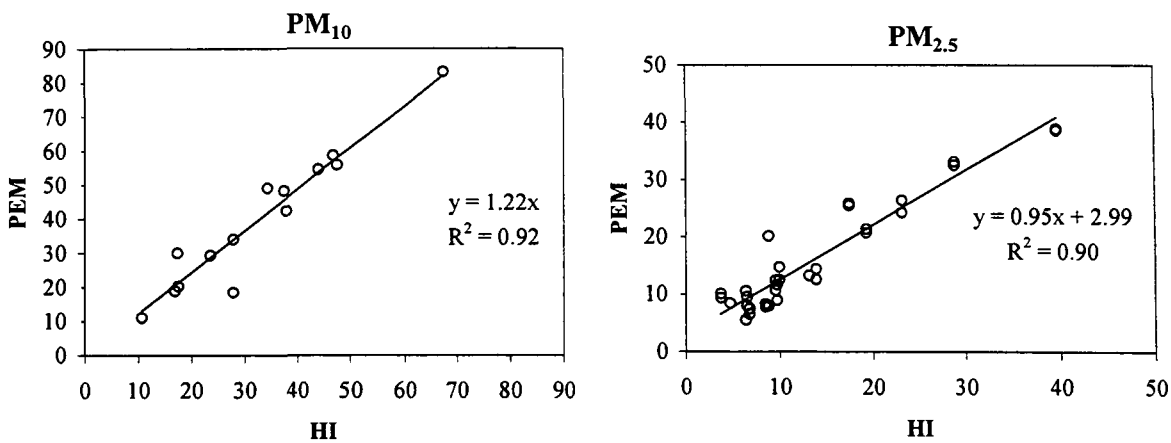

Figure 6. Collocated PEMs and HIs at the Los Angeles central site: winter PM_{10} and winter and summer $PM_{2.5}$ Measurements

Seattle

The limit of detection (LOD) for the 24-h integrated HI was 1 $\mu g/m^3$ and for the 24-h integrated $HPEM_{2.5}$ was 6.2 $\mu g/m^3$ for the first 4 sessions and was reduced to 4.5 $\mu g/m^3$ afterwards. The improvement was achieved by replacing the oiled porous impaction plate with vacuum grease to reduce contamination from silicon oil and by adding a drain disc downstream of the Teflon filter. All duplicates were highly correlated with each other, with a Pearson's *r* of 0.96 or higher. The mean difference between the duplicates was not significantly different from zero. The precision, calculated as the standard deviation of duplicate differences divided by $\sqrt{2}$, was 1.2 $\mu g/m^3$ for HI and 2.2 $\mu g/m^3$ for $HPEM_{2.5}$.

The accuracy of the $PM_{2.5}$ measurements was calculated by comparing with the collocated $FRM_{2.5}$ measurements at the central site. The investigators also collocated $HI_{2.5}$ and $HPEM_{2.5}$ whenever possible: 77 pairs at the stationary ambient monitoring sites and 17 pairs at subjects' homes (Figure 7). The Pearson's *r* between samplers was 0.93 or greater. There was a positive bias, 7.7 $\mu g/m^3$ ($p<0.001$), for $HPEM_{2.5}$ with an oiled impaction plate. For $HPEM_{2.5}$ with a greased impaction plate, the bias was negligible (0.4 $\mu g/m^3$, $p=0.08$). All HI and HPEM measurements were corrected for average blank values. The $HPEM_{2.5}$ measurements with oiled impaction plates during the first four monitoring sessions (N=269 out of 1347 personal filters) were removed from analysis due to the oil contamination problem.

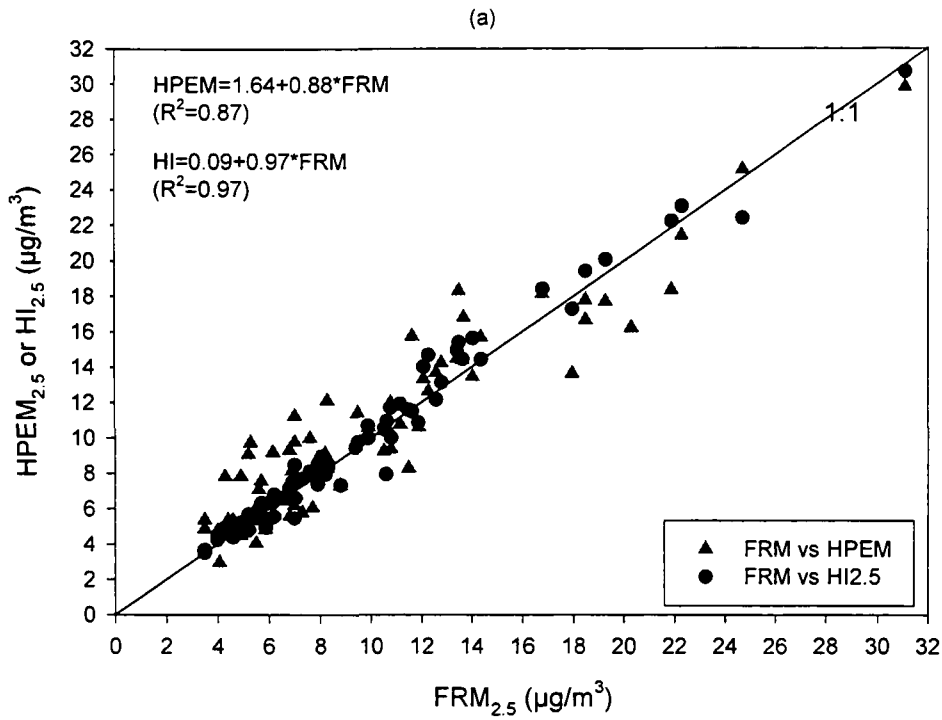

Figure 7. Comparison of Harvard impactor (HI), Harvard personal environmental monitor (HPEM), and federal reference method (FRM) measurements for $PM_{2.5}$: Seattle (central sites).

RESULTS

Research Triangle Park, NC

No significant differences in mean cohort exposures were observed between either of the two cohorts, their residences, locality or for the four seasons. Therefore all results were combined (Table 5). Mean personal $PM_{2.5}$ exposures (23.0 $\mu g/m^3$) were approximately 4 $\mu g/m^3$ greater than the comparable indoor and outdoor $PM_{2.5}$ concentrations. No differences were observed between the mean indoor, outdoor residence, and central site $PM_{2.5}$ concentrations. Mean outdoor PM_{10} (~30-31 $\mu g/m^3$) was slightly higher than the indoor PM_{10} concentrations (28 $\mu g/m^3$), and 8-12 $\mu g/m^3$ higher than the corresponding $PM_{2.5}$ concentrations.

Table 5. PM Mass Concentrations ($\mu g/m^3$): RTP

Variable	N	Geo Mean	Mean	Min	Max	RSD
[a]Personal $PM_{2.5}$	712	19.2	23.0	3.4	142.3	70.1
[b]Indoor $PM_{2.5}$	761	15.3	19.1	2.3	119.4	80.1
[b]Outdoor $PM_{2.5}$	761	17.5	19.3	5.0	51.6	43.7
[b]Central site $PM_{2.5}$	746	17.3	19.2	5.0	49.5	44.9
[a]Indoor PM_{10}	761	23.2	27.7	4.4	155.7	70.6
[a]Outdoor PM_{10}	761	27.5	30.4	7.9	105.1	46.4
[a]Central site PM_{10}	752	27.9	31.4	4.8	105.0	51.5
[c]Indoor $PM_{10-2.5}$	761	6.3	8.6	-2.8	116.6	111.8
[c]Outdoor $PM_{10-2.5}$	761	8.5	11.1	-2.8	82.2	86.9
[c]Central site $PM_{10-2.5}$	210	8.6	10.0	2.6	32.1	62.3

[a]Measured using PEMs, [b]measured using HI samplers, [c]measured by difference in PEM PM_{10} monitor and collocated HI $PM_{2.5}$ mass concentrations. Negative values associated with the minimum $PM_{10-2.5}$ measurements are suspected to be the result of methodological differences between the two monitoring methods relative to semi-volatile retention and particle cut-off parameters.

The cumulative distributions for the four types of $PM_{2.5}$ samples (personal, indoor, residential outdoor, and central site) are displayed by season and for all seasons combined in Figure 8. The plotted points are the 1st, 5th, 10th, 25th, 50th, 75th, 90th, 95th, and 99th percentiles.

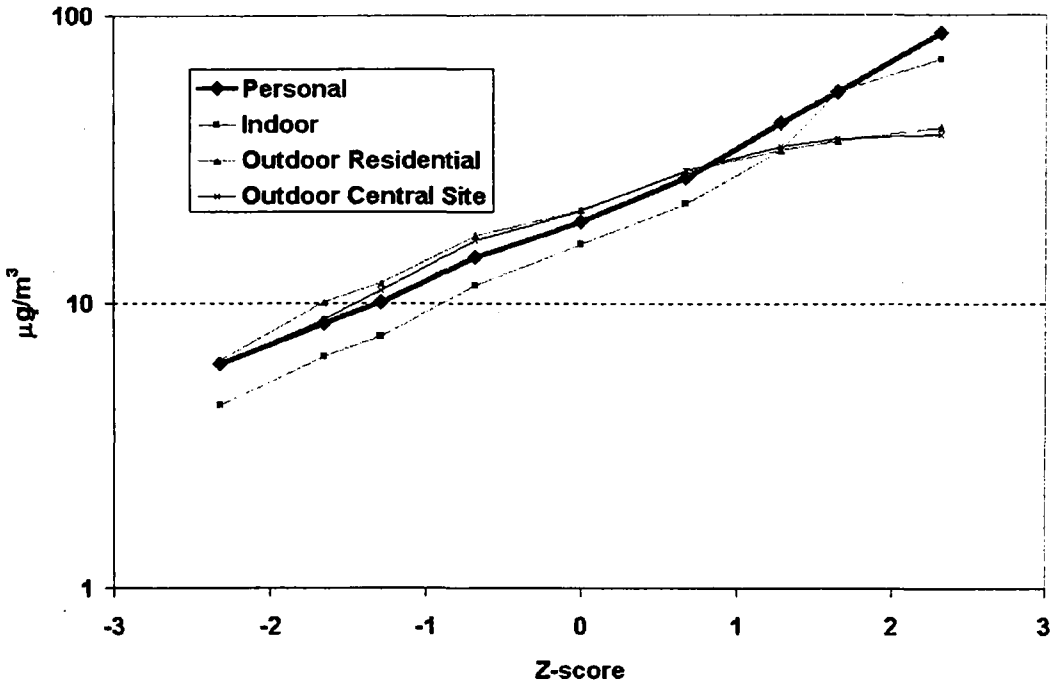

Fig. 8a. Cumulative $PM_{2.5}$ distributions: RTP Summer 2000 (N = 206-224)

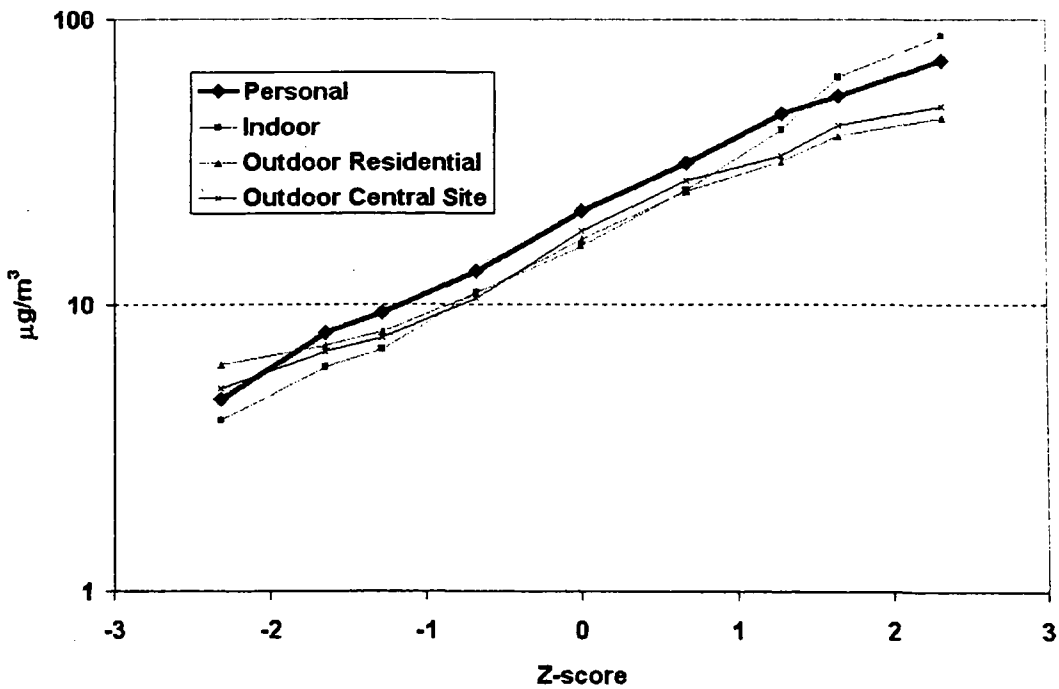

Fig. 8b. Cumulative $PM_{2.5}$ distributions: RTP Fall 2000 (N = 204-210).

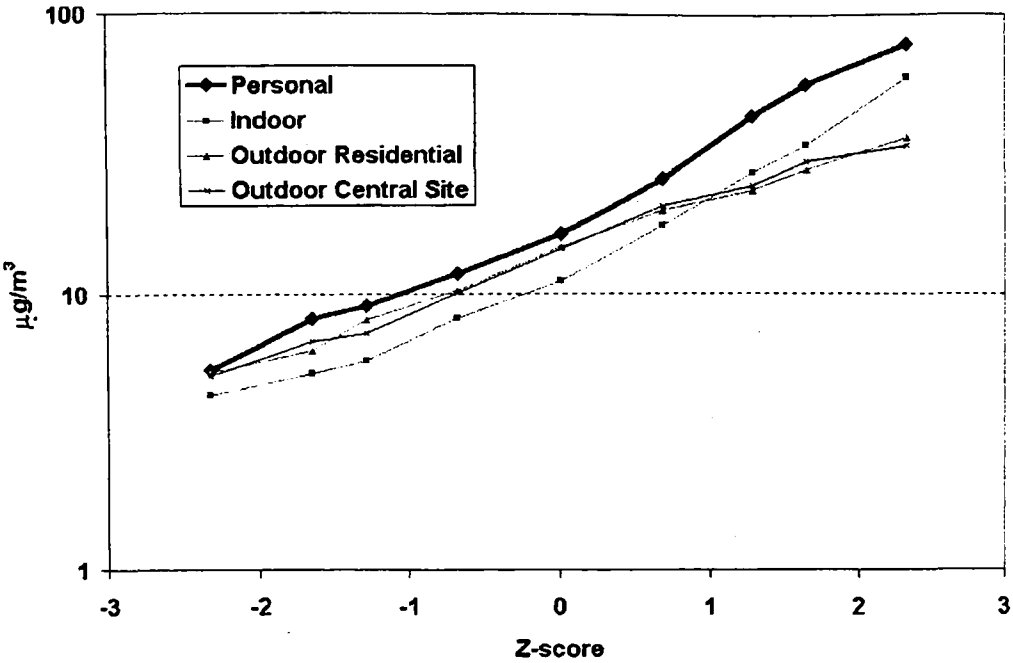

Fig. 8c. Cumulative $PM_{2.5}$ distributions: RTP Winter 2001 (N = 182).

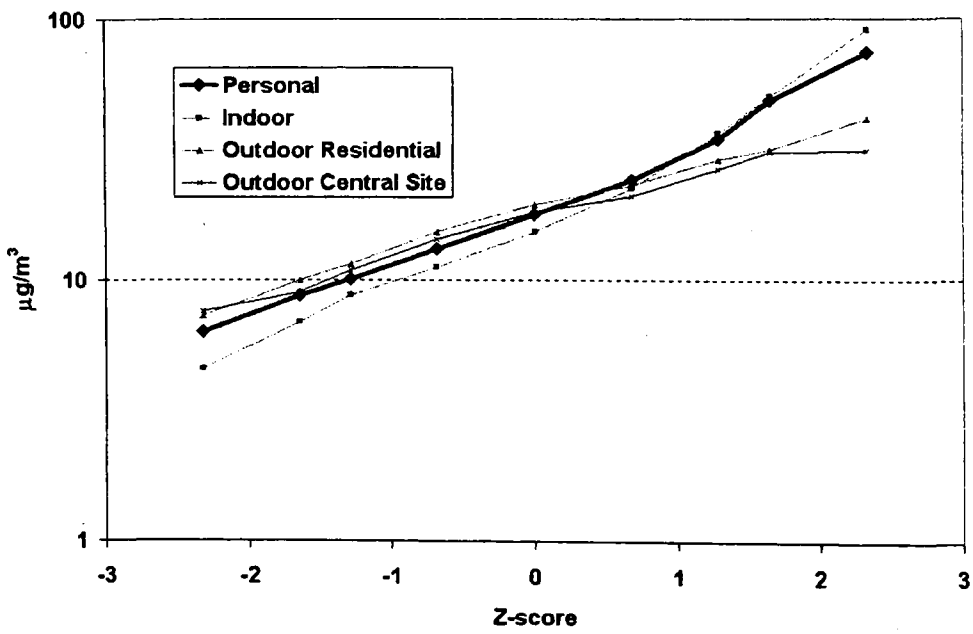

Fig. 8d. Cumulative $PM_{2.5}$ distributions: RTP Spring 2001 (N = 132-147).

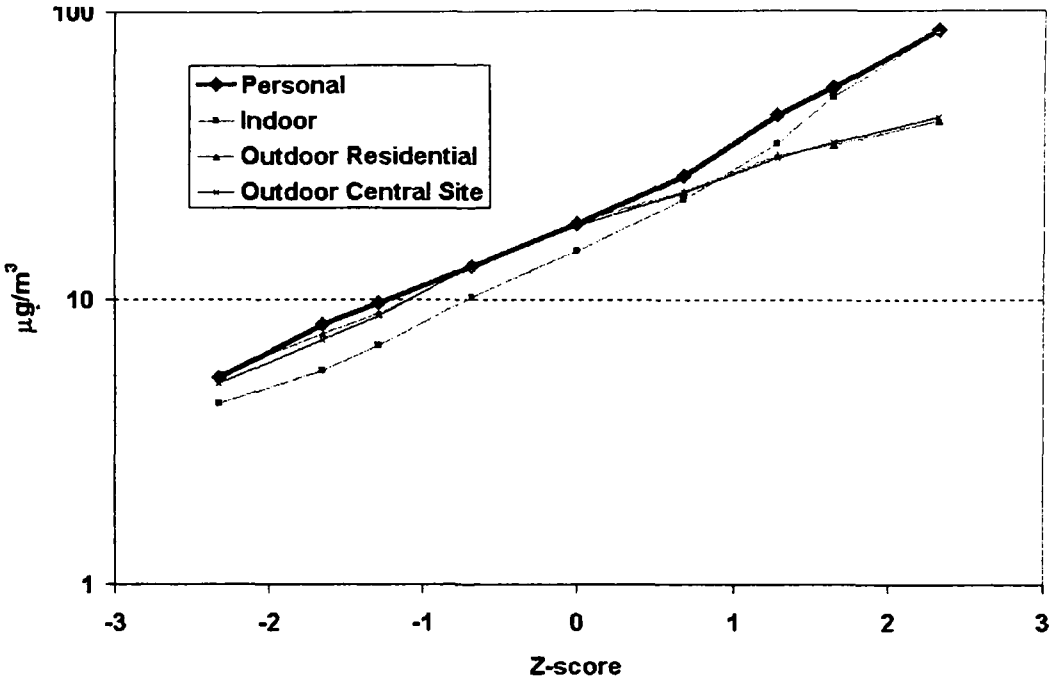

Fig. 8e. Cumulative $PM_{2.5}$ distributions: RTP All Seasons (N = 735-763).

The cumulative distributions for the three types of PM_{10} samples (indoor, residential outdoor, central site) are compared to their counterpart $PM_{2.5}$ samples in Figure 9.

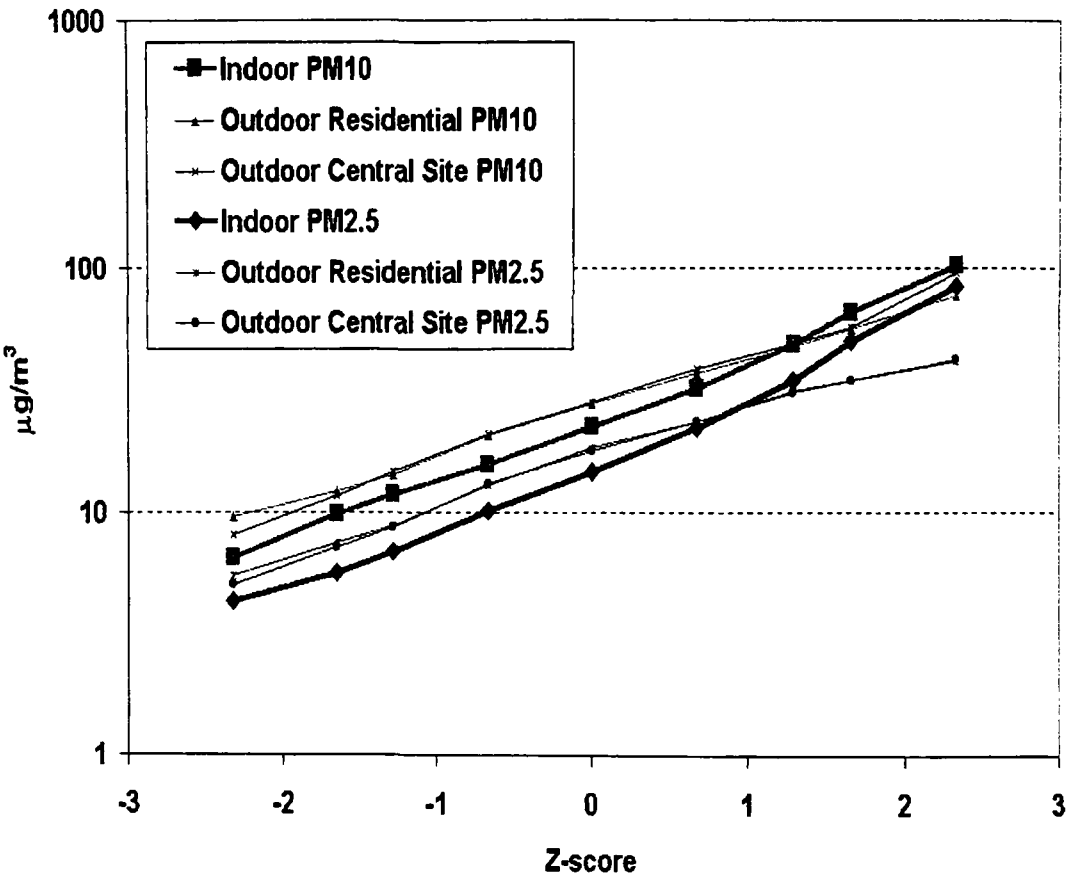

Fig. 9. Comparison of PM10 and PM2.5 combined over all seasons: RTP.

Outdoor temperatures are important influences on behavior patterns that can influence air exchange rates and infiltration factors. Figure 10 displays the daily temperature variations over the course of the RTP study.

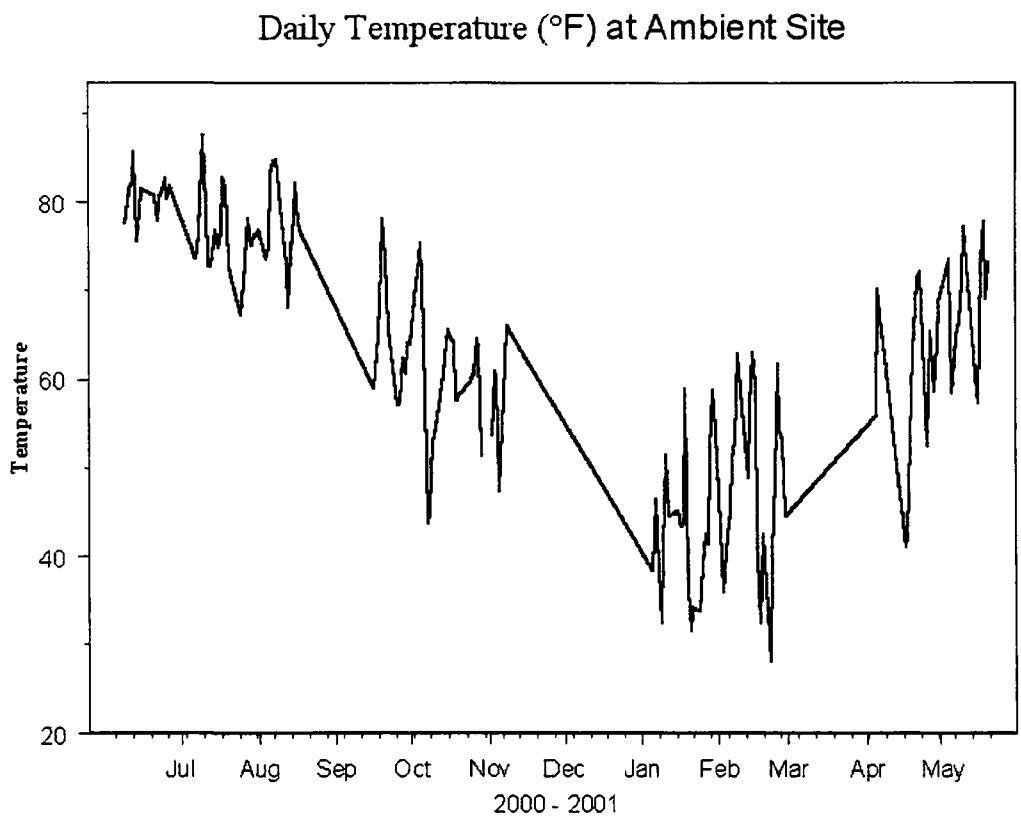

Figure 10. Daily average temperatures (°F) during field study: RTP.

Air exchange rates were surprisingly high during the winter and low during the summer, the reverse of what has been observed in other studies in temperate zone cities (Table 6). The unusually mild winter, with few daily average temperatures below freezing, may have contributed to the high and highly variable air exchange rates that season.

Table 6. Air exchange rates by season: RTP

Season	N	Mean (h^{-1})	SD (h^{-1})
Summer 2000	34	0.48	0.51
Fall 2000	33	0.63	0.36
Winter 2001	28	1.07	0.81
Spring 2001	34	0.70	0.40

Particle concentrations showed little seasonal variation (Figure 11). Indoor concentrations, although their mean values were lower than outdoor for both size fractions, had a larger number of high values in every season.

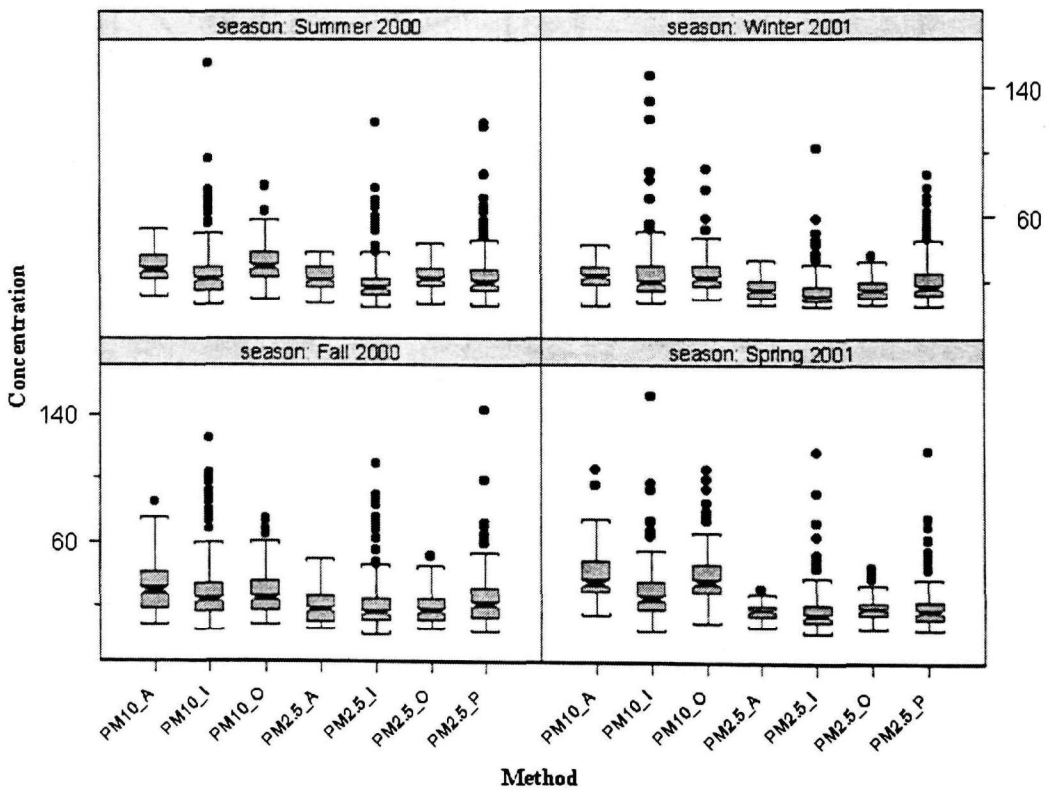

Figure 11. Boxplots of the PM_{10} and $PM_{2.5}$ concentrations ($\mu g/m^3$): RTP. A=Ambient (central site); I = indoor; O = Outdoor (backyard); P = Personal.

Infiltration factors were calculated for each home (Figure 12). Again no significant seasonal difference was noted. The number of unphysical values (<0 or >1) occurring in every season suggests difficulties in the mass balance model employed.

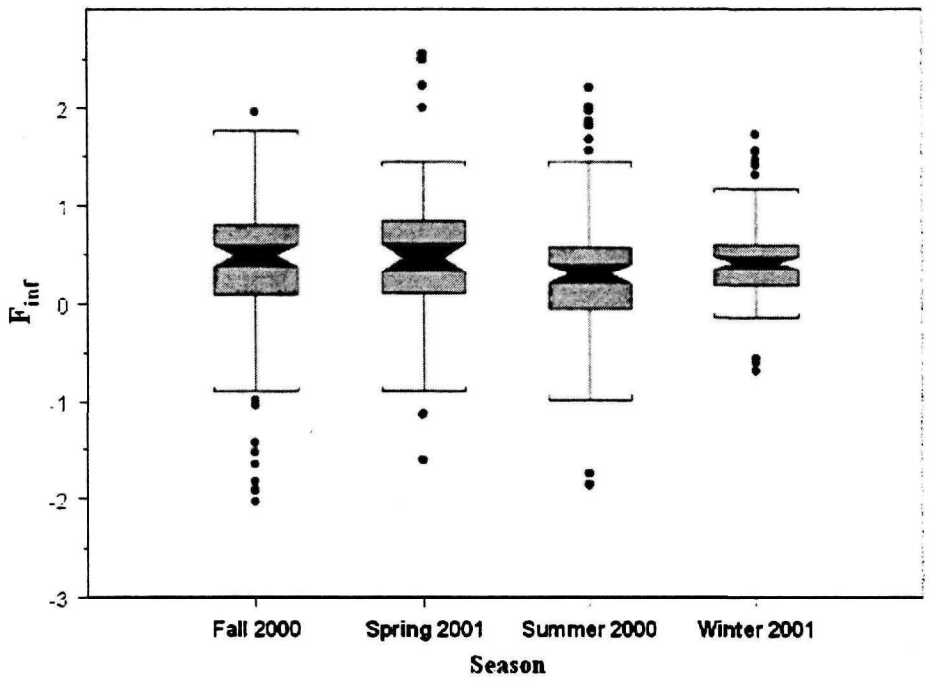

Figure 12. Boxplots of the PM2.5 infiltration factor by season: RTP.

Several calculations using linear regression were performed to estimate the contribution of outdoor particles to indoor concentrations and personal exposures (Table 7). All approaches agreed in finding an infiltration factor F_{inf} ranging between 0.40 and 0.45. The personal exposure attenuation factor F_{pex} ranged between 0.54 and 0.58 in all calculations except one, in which it was 0.47. The average $PM_{2.5}$ indoor air concentration due to indoor sources C_{ig} (indoor-generated) ranged between 9 and 10 ug/m^3, while the average personal exposure due to indoor sources and personal activities E_{na} (non-ambient) ranged between 12 and 15 ug/m3. Since the mean indoor $PM_{2.5}$ concentration was 19 ug/m^3, the contribution of indoor sources (9-10 ug/m^3) was close to half the total. The contribution of indoor sources and personal activities to the mean personal exposure of 23 ug/m^3 was slightly more than half the total.

Table 7. Maximum Likelihood Estimates of Indoor and Outdoor Source Contributions to Indoor and Personal $PM_{2.5}$: RTP

Mixed Model Specifications	Indoor [a](± SE)	Personal[b](± SE)
1. Single fixed intercept Single fixed slope	C_{ig} = 10.34(±1.32) F_{inf} = 0.40(± 0.06)	E_{na} = 11.82(±1.52) F_{pex}= 0.58(±0.07)
2. Multiple fixed intercepts Single fixed slope	9.02(±1.24) 0.43(± 0.06)	13.59(±1.62) 0.54(± 0.07)
3. Random intercepts Single fixed slope	9.18(±1.59) 0.45(±0.06)	12.64(± 1.78) 0.56(± 0.07)
4. Single fixed intercept Multiple fixed slopes	9.32(±1.16) 0.45(±0.06)	12.47(±1.35) 0.59(±0.08)
5. Multiple fixed intercepts Multiple fixed slopes	10.19(±1.64) 0.42(± 0.06)	15.16(±2.05) 0.47(±0.07)
6. Single fixed intercept Random slopes	9.73(± 1.22) 0.42(± 0.08)	12.27(± 1.40) 0.56(±0 .08)
7. Random intercepts Random slopes	9.18(± 1.68) 0.45(± 0.06)	12.59(±1.64) 0.56(±0.07)

[a] Subject specific indoor model: Indoor $PM_{2.5}$ = C_{ig} + F_{inf}* Outdoor $PM_{2.5}$ + Residual, where Indoor $PM_{2.5}$ = daily indoor HI measurements ($\mu g/m^3$), and Outdoor $PM_{2.5}$ = outdoor backyard HI measurements ($\mu g/m^3$).

[b] Subject specific personal model: Personal $PM_{2.5}$ = E_{na} + F_{pex}* Outdoor $PM_{2.5}$ + Residual, where Personal $PM_{2.5}$ = personal Marple sampler measurements ($\mu g/m^3$).

The house-by-house estimates of the infiltration factor F_{inf}, indoor-generated concentration C_{ig}, and personal exposure attenuation factor F_{pex} are provided in Table 8

Table 8. Least squares estimate of indoor filtration and personal exposure factors

Subject	C_{ig}	F_{inf}	R^2	E_{ig}	F_{pex}	R^2
1	8.7	0.38	0.14	8.3	0.92	0.24
2	8.5	0.33	0.07	4.7	0.65	0.22
3	7.0	0.6	0.24	21.3	0.62	0.05
4	20.0	0.87	0.06	19.6	0.85	0.07
5	4.2	0.74	0.28	8.0	1.1	0.11
6	9.3	0.43	0.19	7.6	0.74	0.07
7	-3.2	0.99	0.59	-5.4	1.27	0.43
8	23.9	-0.22	0.15	29.2	-0.22	0.07
9	10.2	0.24	0.09	16.2	0.18	0.03
10	2.7	0.42	0.74	8.2	0.29	0.16
11	35.3	-0.26	0.02	44.0	-0.37	0.02
12	9.3	0.22	0.13	10.2	0.24	0.17
13	1.8	0.51	0.46	19.1	-0.42	0.06
14	6.1	0.31	0.11	6.7	0.34	0.15
15	17.9	0.02	0	15.6	0.27	0.02
16	2.5	0.47	0.78	9.4	0.51	0.22
17	-0.8	0.81	0.29	10.8	0.62	0.06
18	6.0	0.51	0.33	39.4	-0.36	0.02
19	6.8	0.64	0.59	6.0	0.69	0.51
20	2.2	0.45	0.89	6.3	0.51	0.41
21	23.5	0.45	0.21	37.1	0.28	0.07
22	4.9	0.97	0.15	13.0	0.78	0.1
23	36.7	-0.55	0.04	21.5	0.2	0.02
24	36.0	0.17	0	25.3	0.71	0.06
26	8.5	0.53	0.29	23.0	0.47	0.14
27	30.9	0.21	0	14.8	0.47	0.1
28	4.8	0.36	0.28	6.1	0.63	0.25
29	-0.8	1.62	0.6	17.1	0.08	0.01
31	4.0	0.69	0.78	4.7	0.78	0.62
32	10.1	0.2	0.04	11.4	0.3	0.04
33	6.0	0.34	0.12	8.1	0.47	0.13
34	7.3	0.43	0.15	8.1	0.62	0.12
35	17.6	0.22	0.02	10.2	0.79	0.2
36	6.4	0.11	0.25	13.0	0.21	0.1
37	6.0	0.32	0.28	19.4	0	0
38	0.9	0.53	0.89	4.7	0.78	0.53
Mean	10.6	0.42	0.28	14.5	0.44	0.16
SD	10.7	0.38	0.27	10.5	0.40	0.16

The first model is defined by indoor $PM_{2.5} = C_{ig} + F_{inf}$ (outdoor $PM_{2.5}$), where C_{ig} = concentration of indoor generated $PM_{2.5}$ (regression intercept), and F_{inf} = infiltration of ambient $PM_{2.5}$ (regression slope). C_{ig} and E_{ig} have units of $\mu g\ m^{-3}$. R^2 = coefficient of determination (square of correlation between indoor and outdoor $PM_{2.5}$). The second model is defined by personal $PM_{2.5} = E_{ig} + F_{pex}$ (outdoor $PM_{2.5}$), where E_{ig} = exposure to indoor generated $PM_{2.5}$ (regression intercept), and F_{pex} = ambient $PM_{2.5}$ contribution to personal exposure (regression slope). R^2 = coefficient of determination (square of correlation between personal and outdoor $PM_{2.5}$).

A nonlinear mass-balance model was applied to the 24-h average gravimetric data collected by the indoor and outdoor Harvard impactors. The model is (Ozkaynak et al., 1996):

$$C_{in} = PaC_{out}/(a+k) + f_{cook}S_{cook}/[(a+k)V] + f_{other}S_{other}/[(a+k)V]$$

where

C_{in} = indoor concentration ($\mu g/m^3$)
C_{out} = outdoor concentration ($\mu g/m^3$)
P = penetration coefficient (dimensionless)
a = air change rate (h^{-1})
k = deposition rate (h^{-1})
f_{cook} = fraction of time cooking (dimensionless)
f_{other} = fraction of time not cooking (dimensionless)
V = volume of house (m^3)
S_{cook} = source strength (mass flux) from cooking ($\mu g\ h^{-1}$)
S_{other} = source strength (mass flux) from all other indoor sources ($\mu g\ h^{-1}$)

Measured quantities include *Cin, Cout, a,* and *V*, while the time spent cooking was obtained from the time-activity diaries. The four unknowns (P, k, S_{cook}, S_{other}) were obtained from the SAS NLIN procedure. P was constrained to be between 0 and 1; k, S_{cook}, S_{other} were constrained to be ≥ 0.

Thirty-six homes of 38 measured had sufficient data (up to 28 days) to carry out the calculations. The results are shown in Table 9. In this table, C_{cook}, the average concentration produced by cooking during the time of cooking, is obtained from the 2nd term on the right-hand side of the equation above (the ratio $f_{cook}S_{cook}/[(a+k)V]$, and C_{other}, the average concentration due to all other indoor sources, is obtained from the third term in the equation above, the ratio $f_{other}S_{other}/[(a+k)V]$.

Table 9 shows considerable scatter. For example, six values of k are at the lower limit of 0, and 14 values of P are at the upper limit of 1. This indicates a high degree of error, either measurement error, errors in entering the cooking times on the time-activity diary, or errors in the assumption of the model. However, many of the individual estimates for the homes appear to be in reasonable agreement with what is known or guessed to be the case. Few previous studies have been able to arrive at individual estimates for the penetration coefficient and deposition rates in individual homes.

When all data are combined, the overall nonlinear estimate for the penetration coefficient P is 0.99. This is likely to be an underestimate, since P was constrained from above by 1; values greater than 1 would have increased the average value. In fact, the regression was run again with P unconstrained and this resulted in a value of P averaged across the 36 homes of 1.17. Even so, this value of P is in good agreement with the value of P obtained from the University of Washington approach (0.97) and the PTEAM result (1). However,

if the average of the estimated penetration coefficient is calculated for the homes individually, the estimate of P drops to 0.76. This is an underestimate, since 14 values of P are equal to 1 and some would have been greater than 1 if unconstrained.

The estimates of the concentration due to cooking (while cooking) range from zero to 16.7 $\mu g/m^3$, with an average of 6.94 $\mu g/m^3$. However, since the fraction of time cooking was only 4%, the average concentration over 24 hours due to cooking was only 0.3 $\mu g/m^3$. The estimates of the concentration due to other indoor activities range from zero to 16.5 $\mu g/m^3$, with an average of 5.56 $\mu g/m^3$, somewhat less than the PTEAM estimate. This leads to an estimate of the fraction of indoor air concentrations due to outdoor air of 0.81. (The PTEAM estimate was 0.71). Finally, the estimates for the deposition rate k vary from 0 to 1, with an average rate of 0.25 h^{-1}. This average is less than the estimated PTEAM average of 0.39 h^{-1}. Also, it is likely to be an overestimate, since k was constrained from below by zero. However, some theoretical estimates of deposition rates support a lower value (Lai and Nazaroff, 2001).

Table 9. Nonlinear Model Estimates of the Deposition Rate *k*, Penetration Coefficient *P*, and Indoor Source Terms

Subject	Days	F_{inf}	C_out	k	P	a	C_cook	C_other	residual	C_PRED	C_in	f_{cook}	f_{other}
1	21	0.36	17.67	0.00	0.36	0.84	6.85	6.85	0.81	15.55	16.35	0.05	0.95
2	21	0.56	22.88	0.62	1.00	0.91	0.00	5.30	0.51	16.27	16.78	0.05	0.95
3	22	0.81	24.90	0.19	1.00	0.97	10.67	0.68	0.47	21.04	21.50	0.08	0.92
4	20	0.78	22.77	0.07	1.00	0.27	7.10	7.10	2.54	39.30	41.83	0.07	0.93
5	20	0.67	19.93	0.14	0.88	0.59	7.81	3.84	0.04	19.84	19.88	0.02	0.98
6	26	0.39	19.15	0.00	0.39	0.56	11.36	5.38	-0.03	17.75	17.72	0.04	0.96
7	25	0.88	23.96	0.05	0.92	1.31	0.00	0.00	-0.38	21.10	20.73	0.03	0.97
8	6	0.17	27.58	0.01	0.18	0.25	7.16	3.00	0.60	17.15	17.75	0.04	0.96
9	21	0.23	19.78	0.48	0.46	0.54	10.52	10.52	0.19	15.49	15.68	0.01	0.99
10	21	0.45	17.30	0.18	0.61	0.59	8.39	1.37	0.02	10.11	10.13	0.04	0.96
11	9	0.63	20.92	0.18	0.80	1.71	0.00	12.40	2.14	27.75	29.89	0.02	0.98
12	27	0.08	18.66	0.46	0.17	0.45	11.18	10.45	-0.01	13.51	13.50	0.02	0.98
13	7	0.44	13.35	0.28	1.00	0.23	1.03	1.22	0.00	8.53	8.53	0.00	1.00
14	26	0.36	16.59	0.18	0.55	0.36	10.63	2.55	0.02	11.27	11.28	0.03	0.97
15	25	0.57	15.88	0.18	0.77	0.53	5.10	5.10	1.23	16.37	17.60	0.04	0.96
16	28	0.42	16.44	0.18	0.52	0.89	4.03	3.02	-0.05	10.26	10.21	0.06	0.94
17	18	0.39	18.19	0.00	0.39	0.20	7.61	0.33	-0.11	11.89	11.78	0.06	0.94
18	19	0.65	20.01	0.34	1.00	1.00	11.57	2.52	0.38	15.97	16.35	0.05	0.95
19	21	0.69	20.41	0.47	0.98	1.23	10.56	9.31	0.09	19.77	19.86	0.07	0.93
20	20	0.43	17.98	0.05	0.50	0.36	1.17	0.88	-0.02	10.32	10.30	0.04	0.96
21	24	0.69	21.72	0.21	1.00	0.59	10.87	10.87	3.00	30.29	33.29	0.02	0.98
22	14	1.00	19.45	0.00	1.00	0.39	11.12	0.19	1.66	21.99	23.65	0.07	0.93
23	13	0.68	21.01	0.34	1.00	0.79	5.38	5.38	6.08	18.94	25.02	0.06	0.94
24	27	0.79	18.13	0.11	1.00	0.45	11.16	11.16	3.62	35.45	39.06	0.06	0.94
26	17	0.48	26.23	0.80	1.00	0.86	16.67	16.48	0.63	22.94	23.57	0.04	0.96
27	26	1.00	19.15	0.00	1.00	1.41	7.83	7.83	7.11	27.22	34.32	0.01	0.99
28	24	0.40	18.83	0.18	0.56	0.52	8.73	2.52	0.16	11.63	11.79	0.02	0.98
31	14	0.66	25.60	0.30	0.78	2.18	0.00	10.42	0.13	21.39	21.52	0.04	0.96

Table 9. Continued

Subject	Days	F_{inf}	C_out	k	P	a	C_cook	C_other	residual	C_PRED	C_in	f_{cook}	f_{other}
32	26	0.26	16.48	0.72	0.95	0.28	0.00	9.21	0.02	13.33	13.35	0.01	0.99
33	20	0.41	17.53	0.30	0.70	0.54	10.42	0.94	0.14	9.82	9.97	0.09	0.91
34	12	0.52	25.88	0.37	1.00	0.41	8.01	3.20	0.07	18.15	18.22	0.07	0.93
35	24	0.47	17.14	0.53	1.00	0.50	13.59	13.59	0.87	21.49	22.35	0.10	0.90
36	5	0.16	26.59	1.00	0.79	0.26	1.02	6.57	0.02	9.52	9.54	0.00	1.00
37	19	0.31	18.18	0.41	0.62	0.46	4.94	5.21	0.04	11.88	11.91	0.05	0.95
38	14	0.53	12.73	0.04	0.61	0.39	3.40	0.31	-0.01	7.69	7.68	0.00	1.00
Mean		0.54	19.91	0.26	0.76	0.68	6.94	5.56	0.90	18.04	18.94	0.04	0.96
SD		0.24	3.66	0.25	0.26	0.44	4.50	4.37	1.68	7.62	8.68	0.03	0.03

The nonlinear model provided estimates of the infiltration factor F_{inf}, the deposition rate k, the penetration coefficient P and the fraction of indoor air particle concentrations produced by outdoor air infiltration (*frac out*). Linear regression provided estimates of *Finf* and *frac out*. Parameters P and k were estimated with an iterative procedure by first allowing values of P and k each to increment between 0.1 and 1.0 by 0.1. All possible combinations of P and k were then combined with each 24-h measurement of a to produce a calculated infiltration factor F_{calc}. Calculated infiltration factors were then matched with weekly estimates of F_{inf} obtained from the mixed-model where multiple slopes and intercepts were estimated for each subject by season and assuming an autoregressive variance-covariance structure. Values of P, k and a were retained for calculated values F_{calc} that differed from estimates of F_{inf} by no more than ± 0.01.

The nonlinear model was run for each house separately with P unbounded and also with P bounded from above at 1. The model was also run by combining all data (N = 708 days). The linear model, as mentioned above, was run for each house separately and also for all data combined. The results (Table 10) indicate that the two models disagree. In particular, the linear model predicts a much smaller effect of outdoor air (44-46% of the total indoor concentration) than the nonlinear model (63-81%). Similar disagreements between models have been observed by collaborating researchers at the University of Washington and Harvard School of Public Health on data from their respective studies. At present, the reasons for the discrepancies are unknown.

Table 10. Comparison of Linear and Nonlinear Model Estimates of Parameters in the Mass Balance Equation: RTP

Model	Site	N	F_{inf}	P	k	frac out
NonLinear; P≤1	RTP	36	0.54	0.76	0.26	0.63
NL; P unbounded	RTP	36	0.73	1.17	0.36	0.68
NL, overall	RTP	708	0.73	0.99	0.18	0.81
Linear; by house	RTP	36	0.44	0.72	0.42	0.44
Linear; overall	RTP	761	0.40	0.75	0.51	0.46

For the RTP cohorts, regression and correlation analyses indicated that, although there were significant associations between ambient concentrations of $PM_{2.5}$ and the gaseous co-pollutants, personal $PM_{2.5}$ exposures were only significantly related to ambient O_3. Personal exposures and indoor concentrations of the pollutants were not associated with their corresponding ambient concentrations, except for $PM_{2.5}$.

X-ray fluorescence (XRF) studies of the personal, indoor, and outdoor $PM_{2.5}$ samples collected on Marple PEMs indicated that sulfates made up the bulk of the mass (35-46%). Measurements on co-located indoor and outdoor monitors with quartz filters followed by thermal quantitation of elemental and organic (EC/OC) indicated that total carbon made up the bulk of the remainder of the outdoor mass (30-34%). A small amount of crustal material, nitrate, and sodium chloride

accounted for the remainder of the mass. However, a positive artifact was observed for the indoor samples and data uncertainty for the OC component has to be considered.

Finally, the ultimate goal of these studies was to determine the longitudinal correlation between personal exposures and central site measurements. Because no particular seasonal component was noticed for the 37 subjects, their four 7-day sampling periods were combined into a single 28-day sequence and Pearson correlation coefficients between exposure and ambient concentration were determined for each subject. Because the cohorts showed some differences, they are kept separate in the summary boxplot of correlations (Figure 13). Median correlation coefficients are quite low at 0.45 and 0.3, suggesting that for these persons, only 10-20% of the variance in personal exposures could be explained by ambient concentrations. For some persons, correlations were quite high, but for others they were not only very low, but even negative.

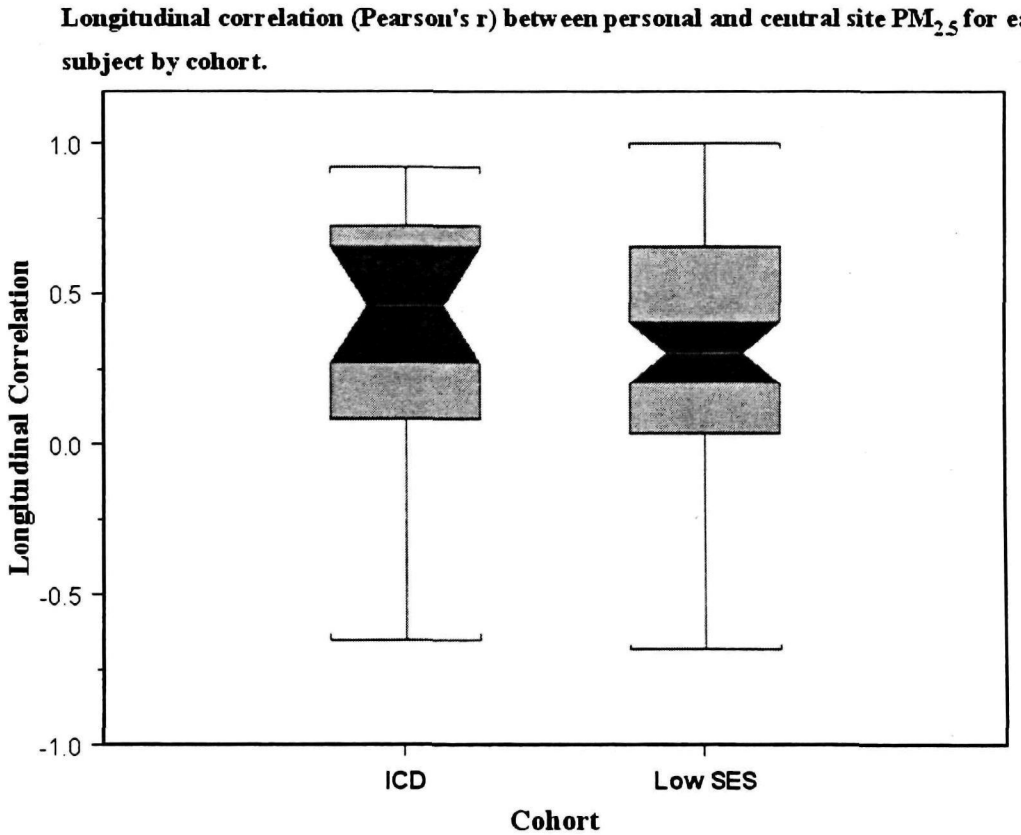

Figure 13: Boxplots of longitudinal correlation coefficients (Pearson) between personal exposure and ambient concentration of PM2.5: RTP, NC.

Atlanta

Summary statistics for personal, indoor, and outdoor $PM_{2.5}$, elemental carbon (EC), and particulate sulfate (SO_4^{2-}) stratified by season and sample type are presented in Table 11. For all pollutant measures, outdoor concentrations tended to be higher than corresponding indoor and personal levels, with differences generally most pronounced for SO_4^{2-}, reflecting the fact that its major sources are located outdoors. Indoor and outdoor concentrations were higher in the spring as compared to fall for each of the measured pollutants. In contrast, spring and fall personal exposures tended to be comparable, with no consistent pattern observed across the three measured pollutants (Figure 14).

Table 11. Descriptive Statistics for Outdoor, Indoor, and Personal Pollutant Concentrations in Atlanta: Particles (in $\mu g/m^3$).

Pollutant/ Season/Sample	N	Mean + Std. Dev.	Median	Max
Fall $PM_{2.5}$				
Outdoor	141	14.7 ± 7.7	13.1	46.7
Indoor	143	14.3 ± 10.8	12.2	89.3
Personal	141	16.6 ± 9.8	14.3	77.2
Spring_$PM_{2.5}$				
Outdoor	118	22.0 ± 10.8	20.8	53.8
Indoor	127	19.9 ± 14.2	16.0	99.1
Personal	142	15.0 ± 7.5	14.2	45.8
Fall EC				
Outdoor	141	1.5 ± 1.0	1.2	5.9
Indoor	152	1.0 ± 0.7	0.9	6.5
Personal	146	1.5 ± 0.8	1.3	4.6
Spring EC				
Outdoor	134	1.8 ± 0.8	1.7	5.2
Indoor	139	1.7 ± 0.7	1.5	5.1
Personal	138	1.7 ± 0.6	1.7	4.2
Fall SO_4^{2-}				
Outdoor	102	4.0 ± 2.2	3.9	10.5
Indoor	112	2.5 ± 1.4	2.2	8.7
Personal	131	2.7 ± 1.6	2.5	9.3
Spring SO_4^{2-}				
Outdoor	137	5.4 ± 2.9	4.8	14.1
Indoor	144	3.4 ± 1.9	3.0	10.7
Personal	135	2.8 ± 1.6	2.5	9.3

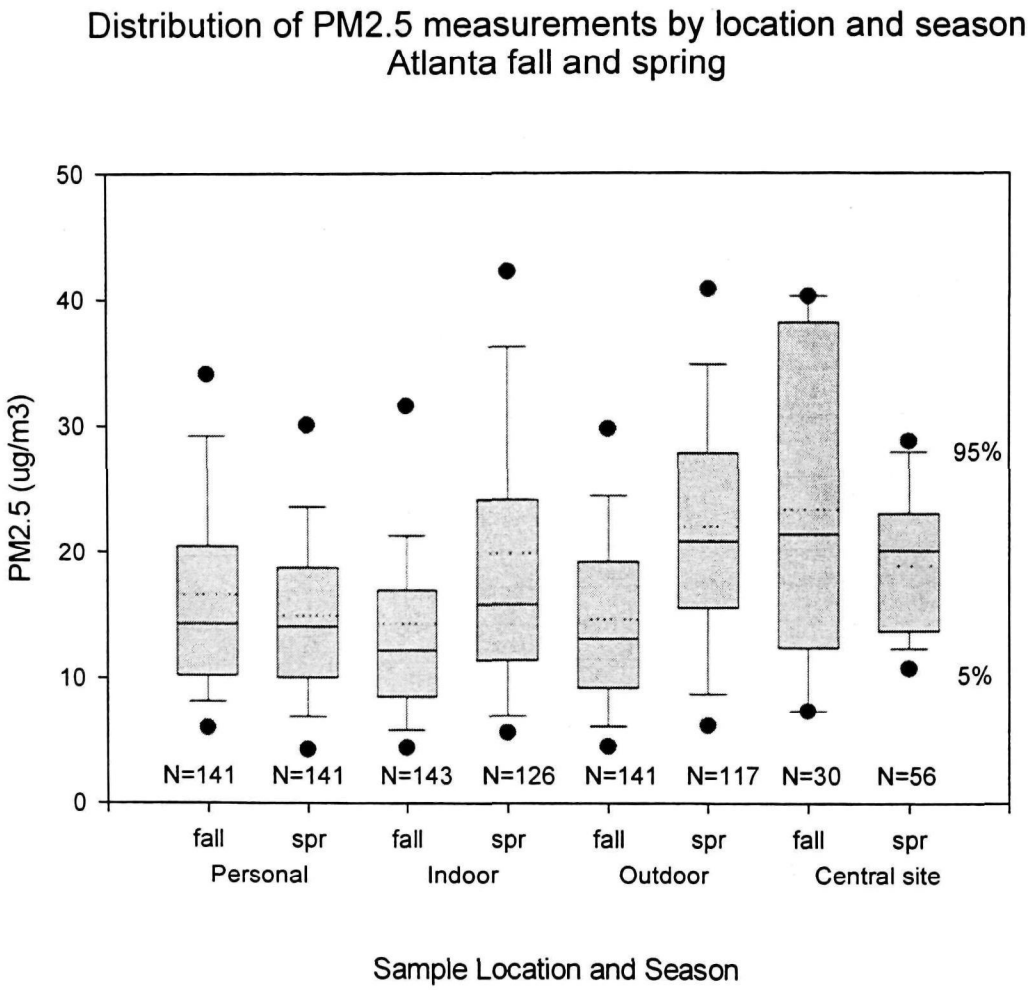

Figure 14. $PM_{2.5}$ concentrations by location and season--Atlanta. Boxplots indicate 5th, 25th, 50th (median), 75th and 95th percentiles by horizontal lines, with the mean indicated by the dashed line and outliers indicated by dots.

The cumulative $PM_{2.5}$ distributions over all seasons are compared in a lognormal probability graph in Figure 15. On such graphs, lognormal distributions appear as a straight line. Personal, indoor, and outdoor residential measurements are very similar from the 1st to the 95th percentiles. The outdoor central site records higher concentrations than the outdoor residential sites for the lower half of the distribution, suggesting that it is impacted by sources more strongly.

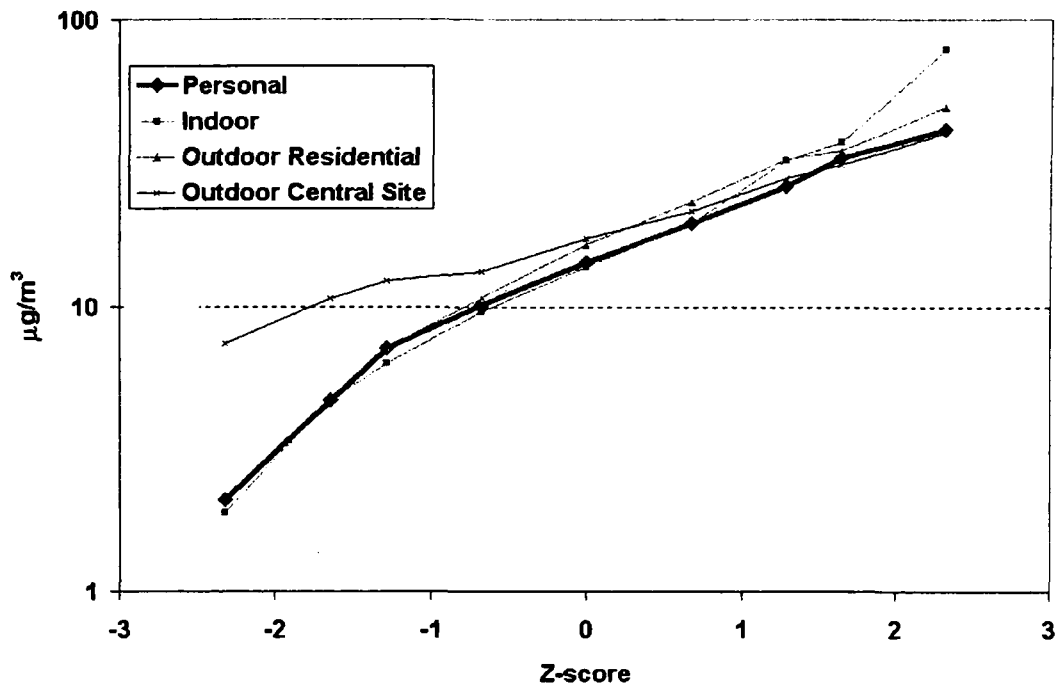

Figure 15. Atlanta. The four types of $PM_{2.5}$ measurements plotted on lognormal probability coordinates. Plotted points are the 1st, 5th, 10th, 25th, 50th, 75th, 90th, 95th, and 99th percentiles. N = 258-282.

Concentrations of elemental carbon (EC), an indicator of diesel traffic, were increased in the spring compared to the fall for all types of measurements (personal, indoor, outdoor) (Figure 16). Sulfate concentrations, on the other hand, were elevated in the spring for the indoor and outdoor measurements, but were nearly unaffected by season for the personal measurements (Figure 17).

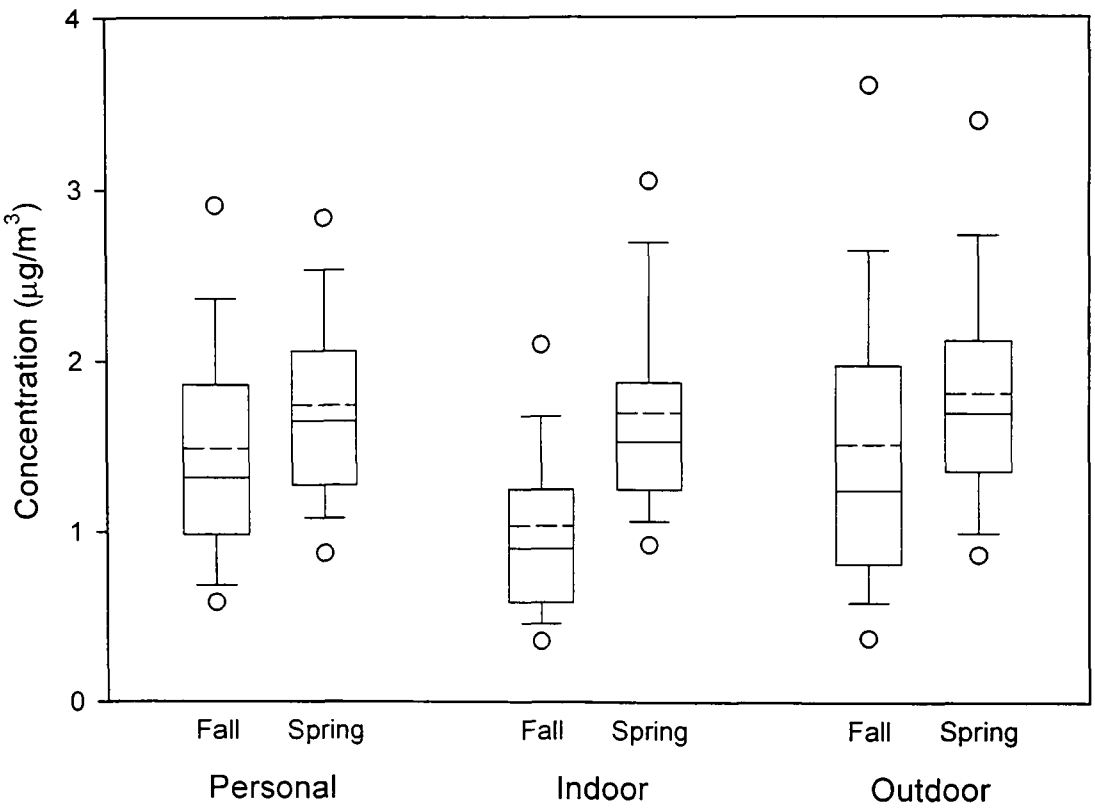

Figure 16. EC Concentrations by location and season: Atlanta.

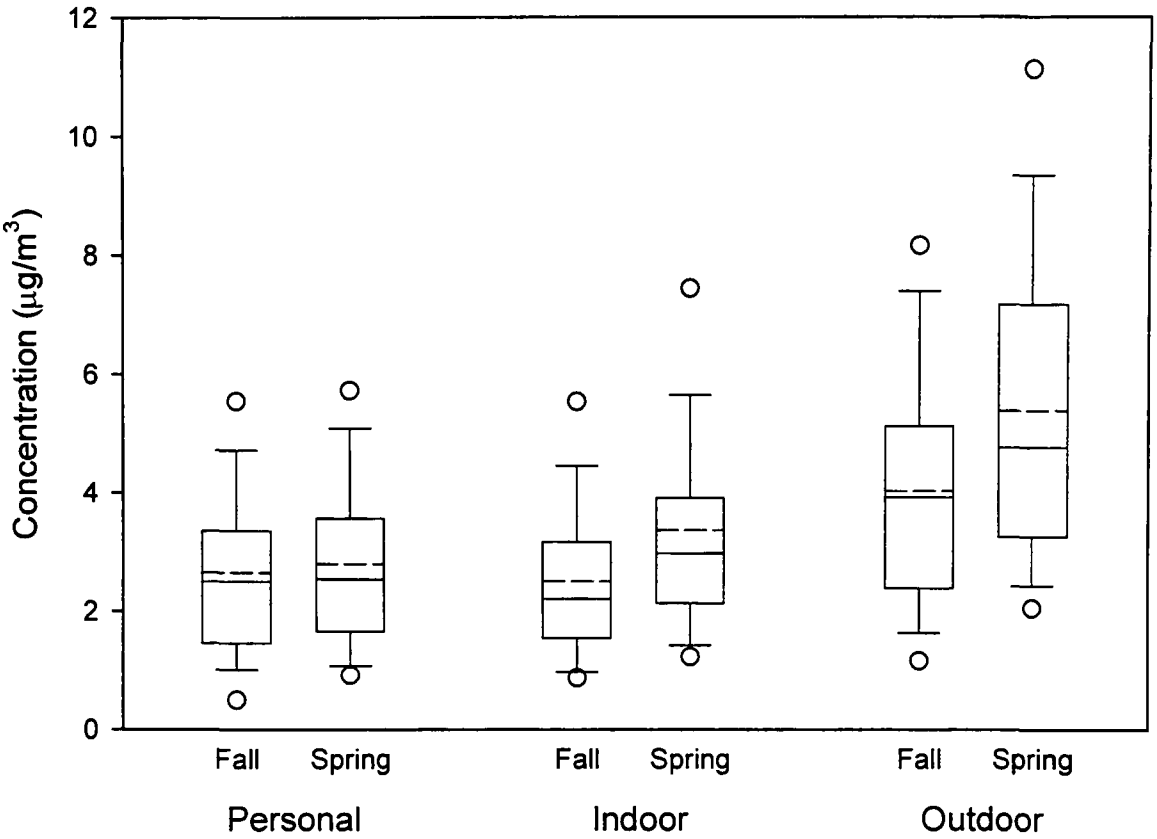

Figure 17. SO_4^{2-} Concentrations by location and season: Atlanta

Summary statistics for personal, indoor, and outdoor ozone (O_3), nitrogen dioxide (NO_2), and sulfur dioxide (SO_2) stratified by season and sample type are presented in Table 12. During both seasons, outdoor concentrations for each of the gases tended to be higher than their respective indoor concentrations and personal exposures. Outdoor gaseous pollutant concentrations were comparable during both sampling seasons.

Table 12. Descriptive Statistics for Outdoor, Indoor, and Personal Pollutant Concentrations in Atlanta: Gases (in ppb).

Pollutant					
Season/Sample Type	N	Mean + Std. Dev	Median	Max	Min
Fall O_3					
Outdoor	150	20.5 ± 11.2	18.5	57.7	-2.4
Indoor	151	2.7 ± 5.3	1.1	48.0	-2.3
Personal	146	3.6 ± 5.0	2.1	36.8	-2.4
Spring O_3					
Outdoor	146	26.7 ± 10.4	25.6	60.9	8.1
Indoor	145	1.3 ± 2.4	0.7	13.6	-2.9
Personal	144	2.1 ± 2.8	1.3	12.8	-3.1
Fall NO_2					
Outdoor	143	13.9 ± 8.3	11.0	40.8	-0.3
Indoor	142	9.2 ± 12.6	6.1	123.3	1.4
Personal	138	9.5 ± 7.8	7.3	49.7	1.6
Spring NO_2					
Outdoor	143	13.5 ± 9.6	11.0	55.0	-0.6
Indoor	145	11.6 ± 15.8	6.4	85.8	0.2
Personal	143	12.1 ± 15.2	7.7	128.2	0.6
Fall SO_2					
Outdoor	143	3.2 ± 3.6	2.6	19.0	-5.3
Indoor	142	1.4 ± 6.2	0.4	54.6	-5.6
Personal	138	-0.4 ± 4.5	-0.6	24.4	-5.8
Spring SO_2					
Outdoor	143	4.8 ± 7.0	3.7	48.0	-8.5
Indoor	145	-1.3 ± 9.8	-1.0	105.2	-8.6
Personal	143	-1.2 ± 3.8	-0.5	9.8	-10.1

Longitudinal correlations (Spearman) between personal exposures and outdoor concentrations of $PM_{2.5}$ were calculated for the Atlanta participants (Figure 18). Median correlations were about 0.65 for the COPD cohort and about 0.55 for those with myocardial infarctions.

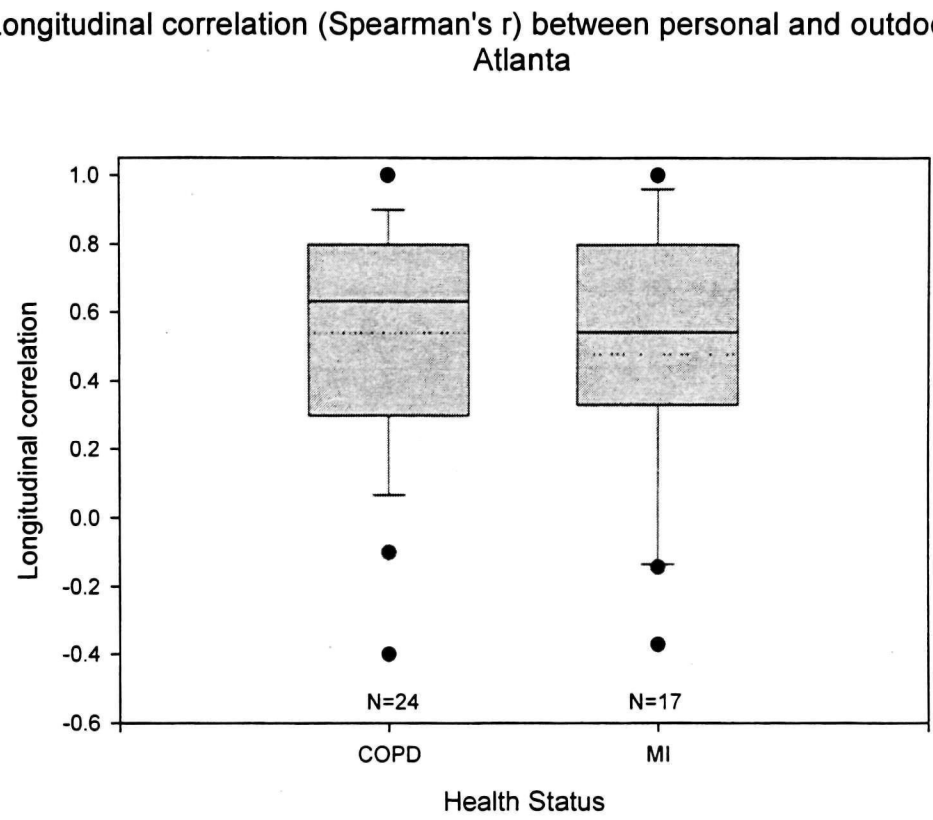

Figure 18. Longitudinal correlations between personal exposures and ambient concentrations of $PM_{2.5}$: Atlanta

Boston

Summary statistics for personal, indoor, and outdoor $PM_{2.5}$, EC and sulfate stratified by season and sample location (personal, indoor, and outdoor) are presented in Table 13. For all pollutant measures, concentrations tended to be similar among the microenvironments. This result is not surprising for sulfate and EC, which have few indoor sources; however, previous studies have shown higher personal exposures than indoor or outdoor concentrations. While median $PM_{2.5}$ exposures were higher than outdoor, the concentrations differed by less than 1 ug/m^3.

: 13. Descriptive Statistics for Outdoor, Indoor, and Personal Pollutant entrations in Boston: Particles (in μg/m^3).

Pollutant Season/Sample Type	N	Mean + Std. Dev.	Median	Max
Winter $PM_{2.5}$				
Outdoor	75	11.4 ± 7.7	9.9	46.3
Indoor	72	10.4 ± 9.4	7.8	48.9
Personal	55	17.6 ± 26.9	10.6	155.9
Second Personal	47	12.5 ± 17.3	9.9	122.8
Summer $PM_{2.5}$				
Outdoor	44	12.8 ± 7.7	11.9	39.8
Indoor	86	12.7 ± 8.7	10.5	49.8
Personal	63	9.4 ± 5.1	8.3	26.1
Second Personal	31	12.0 ± 8.5	11.0	37.0
Summer sulfate				
Outdoor	97	4.0 ± 2.7	3.3	11.6
Indoor	105	3.1 ± 2.2	2.6	10.6
Personal	103	3.1 ± 2.1	2.6	8.9
Second Personal	39	2.9 ± 2.4	1.8	10.9
Winter EC				
Outdoor	94	2.3 ± 1.4	1.9	6.7
Indoor	102	2.0 ± 1.5	1.5	10.8
Personal	94	1.5 ± 1.6	1.1	11.6
Second Personal	54	1.5 ± 1.7	1.2	11.5
Summer EC				
Outdoor	95	1.5 ± 0.7	1.4	4.7
Indoor	100	1.5 ± 0.6	1.4	3.3
Personal	101	1.6 ± 0.6	1.5	4.9
Second Personal	38	1.5 ± 0.6	1.3	3.8

The cumulative $PM_{2.5}$ distributions observed in Boston are plotted on lognormal probability coordinates in Figure 19. Between the 5th and 95th percentiles, all four types of measurements appear to be very similar. The personal measurements show the greatest variation, with both the lowest and highest concentrations of any of the four types of measurements. The similarity of the outdoor residential concentrations to the outdoor central site suggests both the spatial homogeneity of the fine particles across the city and the representativeness of the central site in estimating residential outdoor concentrations.

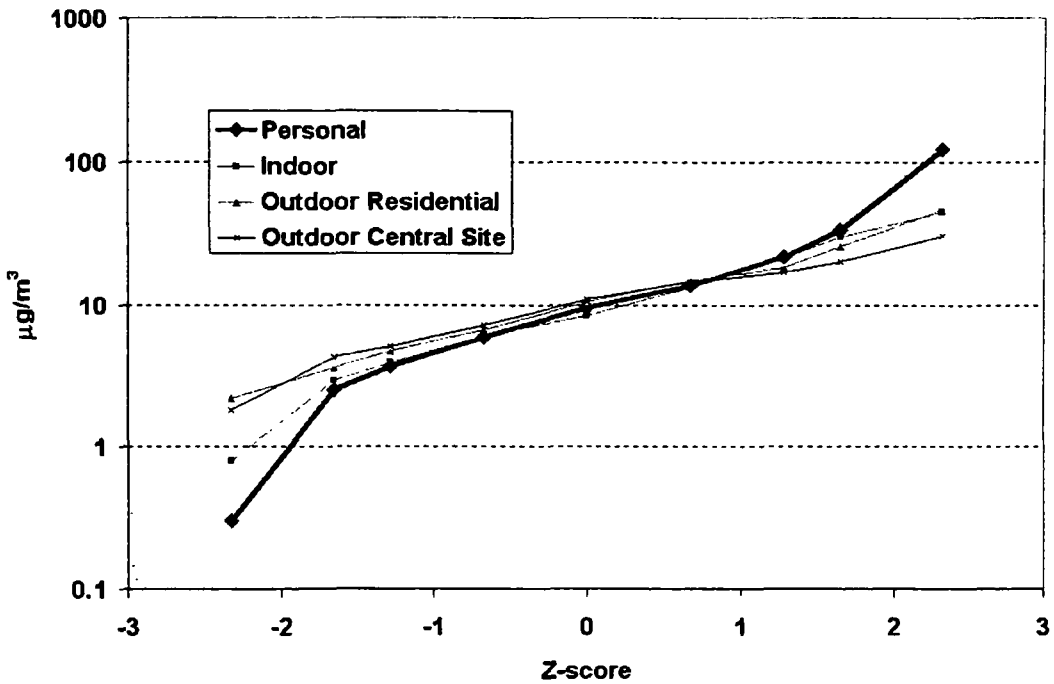

Figure 19. Boston. Cumulative $PM_{2.5}$ distributions of personal, indoor, residential outdoor and central-site concentrations. N = 266-301

Longitudinal correlation coefficients between personal and ambient concentrations were calculated for each subject in the three cohorts: cardiovascular heart disease, spouses of patients, and persons with COPD (Figure 20). As at RTP and Atlanta, median coefficients were relatively low (range of 0.55-0.65). Mean coefficients (dashed line) were even lower due to some strongly negative coefficients.

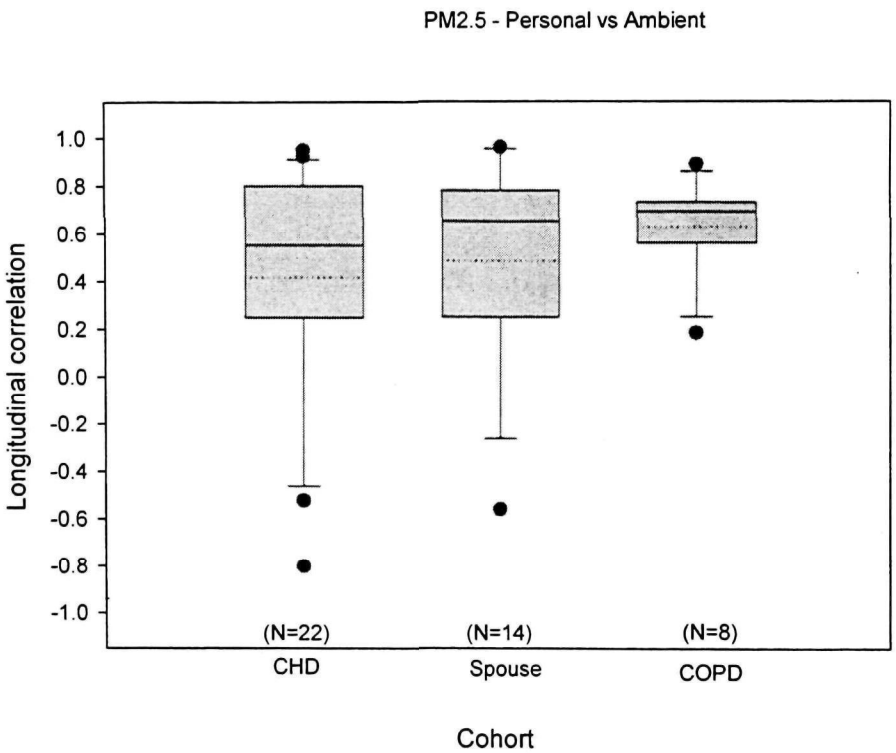

Figure 20. Longitudinal correlations between personal exposures and ambient concentrations of $PM_{2.5}$: Boston.

Los Angeles

Summary statistics for personal, indoor, and outdoor $PM_{2.5}$, elemental carbon (EC), and particulate sulfate (SO_4^{2-}) stratified by season and sample type are presented in Table 14.

Table 14. Descriptive Statistics for Outdoor, Indoor, and Personal Pollutant Concentrations in Los Angeles: Particles (in μg/m³).

Pollutant				
Season/Sample Type	**N**	**Mean ± Std. Dev.**	**Median**	**Max**
Winter $PM_{2.5}$				
Outdoor	92	13.5 ± 8.5	11.2	56.5
Indoor	92	16.9 ± 11.7	12.8	49.5
Personal	87	19.6 ± 14.5	14.4	63.5
Summer $PM_{2.5}$				
Outdoor	96	19.3 ± 9.0	17.4	53.5
Indoor	97	18.1 ± 11.1	17.0	94.8
Personal	92	25.1 ± 20.8	18.8	137.8
Winter NO_3^-				
Outdoor	92	3.1 ± 2.6	2.2	11.8
Indoor	94	1.1 ± 1.0	0.9	4.7
Personal	98	1.2 ± 1.1	0.8	6.4
Summer NO_3^-				
Outdoor	95	2.8 ± 1.5	2.5	7.1
Indoor	96	1.7 ± 0.8	1.5	4.2
Personal	97	1.6 ± 0.9	1.4	5.0
Winter EC				
Outdoor	94	1.9 ± 1.1	1.7	5.5
Indoor	90	1.6 ± 0.9	1.4	5.2
Personal	91	1.9 ± 1.0	1.7	4.9
Summer EC				
Outdoor	95	0.1 ± 0.7	0.0	2.7
Indoor	95	0.2 ± 0.7	0.2	2.1
Personal	85	0.3 ± 0.8	0.2	3.3

Cumulative $PM_{2.5}$ distributions for Los Angeles are plotted on lognormal probability coordinates in Figure 21. The most striking aspect of this plot, unlike those for Atlanta and Boston, is the evidence of a "personal cloud", with personal exposures consistently higher than either indoor or outdoor concentrations from the 5th to the 99th percentiles.

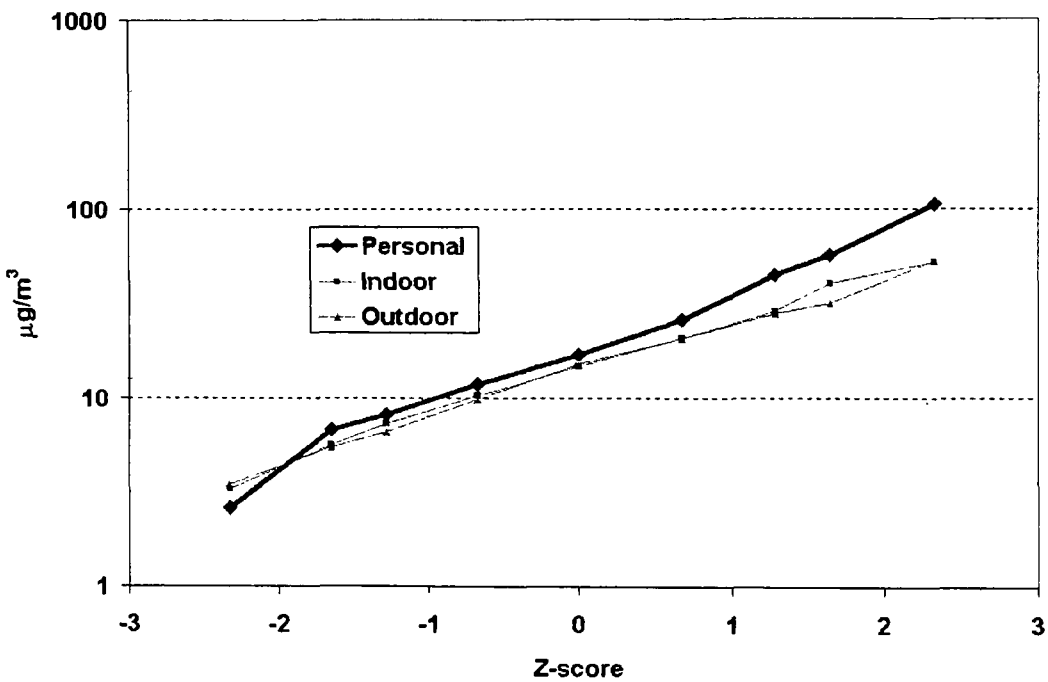

Figure 21. Los Angeles. Cumulative $PM_{2.5}$ distributions of personal, indoor, and residential outdoor concentrations. N = 179-189

Longitudinal correlations (Spearman) between personal exposure and ambient concentrations of $PM_{2.5}$ for the summer and winter seasons were calculated for the respondents in Los Angeles (Figure 22). The median personal-outdoor correlation was quite low in summer at 0.29 and only slightly higher in winter at 0.49. Surprisingly, personal-indoor correlations were very little higher. Mean values were again lower than the medians due to several negative relationships.

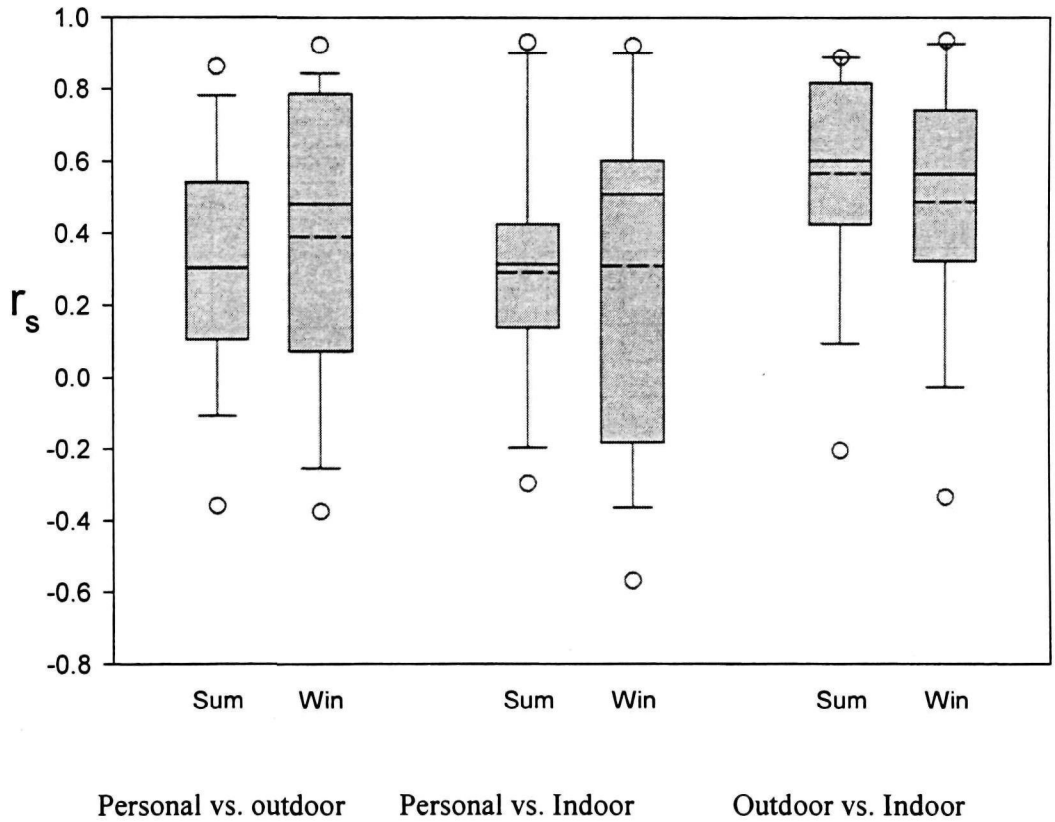

Figure 22. Longitudinal correlations between personal exposures and indoor and outdoor concentrations of $PM_{2.5}$: Los Angeles.

Table 15 provides a more detailed look at the individual longitudinal correlations between personal and outdoor exposure to $PM_{2.5}$. The table shows that only 4 of the 28 week-long monitoring efforts resulted in significant relations between personal exposures and outdoor concentrations.

Table 15. Individual-Specific Spearman Correlation Coefficients: $PM_{2.5}$ in Los Angeles

SEASON	SUBJECT	Indoor vs. Outdoor			Personal vs. Indoor			Personal vs. Outdoor		
		n	R	p-value	n	r	p-value	n	r	p-value
Summer	LPD-16	7	0.61	0.15	5	0.90	0.04	5	0.70	0.19
	LPD-17	7	0.61	0.15	7	0.32	0.48	7	-0.07	0.88
	LPD-18	7	0.89	0.01	6	0.94	0.00	6	0.89	0.02
	LPD-19	6	0.14	0.79	7	0.29	0.53	6	0.54	0.27
	LPD-21	6	0.71	0.11	6	0.14	0.79	7	0.39	0.38
	LPD-22	7	0.82	0.02	6	0.14	0.79	6	0.43	0.40
	LPD-23	7	-0.29	0.53	7	0.32	0.48	7	0.11	0.82
	LPD-24	7	0.43	0.34	6	0.31	0.54	6	0.77	0.07
	LPD-25	7	0.89	0.01	6	-0.03	0.96	6	0.14	0.79
	LPD-26	7	0.61	0.15	7	-0.32	0.48	7	0.29	0.53
	LPD-27	7	0.79	0.04	7	0.43	0.34	7	0.00	1.00
	LPD-28	6	0.54	0.27	7	0.54	0.22	6	0.20	0.70
	LPD-29	7	0.43	0.34	7	0.32	0.48	7	0.32	0.48
	LPD-30	7	0.82	0.02	7	-0.18	0.70	7	-0.43	0.34
Winter	LPD-01A	1	--	--	2	--	--	2	--	--
	LPD-02	7	0.07	0.88	7	0.61	0.15	7	-0.21	0.64
	LPD-03	6	0.49	0.33	3	0.50	0.67	4	0.80	0.20
	LPD-04	7	0.57	0.18	6	0.60	0.21	6	0.49	0.33
	LPD-05	5	0.20	0.75	5	-0.10	0.87	7	0.43	0.34
	LPD-06	3	--	--	3	--	--	2	--	--
	LPD-07	6	0.89	0.02	6	-0.26	0.62	7	0.36	0.43
	LPD-08	6	0.37	0.47	6	0.31	0.54	6	0.94	0.00
	LPD-09	6	0.94	0.00	6	-0.26	0.62	5	0.10	0.87
	LPD-10	5	0.70	0.19	4	-0.60	0.40	6	0.49	0.33
	LPD-11	4	-0.40	0.60	6	0.43	0.40	4	-0.40	0.60
	LPD-12	6	0.49	0.33	5	0.60	0.28	5	0.00	1.00
	LPD-13	7	0.61	0.15	7	0.93	0.00	7	0.50	0.25
	LPD-14	7	0.93	0.00	7	0.89	0.01	7	0.82	0.02
	LPD-15	7	0.57	0.18	7	0.61	0.15	7	0.79	0.04

A mixed-effects multiple regression model was run to determine the factors affecting indoor $PM_{2.5}$ concentrations. The model estimate for the infiltration factor was 0.42 in winter and 0.70 in summer, consistent with increased air exchange rates in summer.

Seattle

PM mass results for all four cohorts are presented in Table 16. Mean personal $PM_{2.5}$ concentrations ranged from 9.3-10.8 $\mu g/m^3$ for adults but were significantly higher (13.3 $\mu g/m^3$) for asthmatic children. Indoor means were slightly lower at 7.4-9.5 $\mu g/m^3$, and outdoor means slightly higher at 9.0-12.6 $\mu g/m^3$. Considering the difference between personal and indoor concentrations as an estimator for the personal cloud, mean values were only about 1-2 $\mu g/m^3$ for the three adult cohorts but 4 $\mu g/m^3$ for the asthmatic children.

Table 16. Summary of PM Concentrations Between Oct 1999 and May 2001 by Health Group: Seattle

Location	Pollutant ($\mu g/m^3$)	Group	N	Mean	Std Dev	GM	GSD	Min	Max
Personal	$PM_{2.5}$	COPD	307	10.5	7.2	8.6	1.9	0.8	45.6
		Healthy	183	9.3	8.4	7.7	1.8	0.8	96.2
		Asthmatic	263	13.3	8.2	11.1	1.9	1.0	49.4
		CHD	325	10.8	8.4	8.8	1.9	1.4	66.6
Indoor	$PM_{2.5}$	COPD	443	8.5	5.1	7.3	1.7	1.0	49.9
		Healthy	193	7.4	4.8	6.1	1.9	0.4	38.0
		Asthmatic	276	9.2	6.0	7.9	1.7	2.2	36.3
		CHD	329	9.5	6.8	8.0	1.8	1.6	65.3
	PM_{10}	COPD	437	14.1	6.6	12.7	1.6	2.5	40.1
		Healthy	206	12.6	7.8	10.6	1.9	0.6	62.2
		Asthmatic	274	19.4	11.1	16.8	1.7	2.2	107.7
		CHD	324	16.2	11.3	13.6	1.8	0.6	110.6
Outdoor	$PM_{2.5}$	COPD	437	9.2	5.1	8.0	1.7	-0.2	28.9
		Healthy	194	9.0	4.6	7.9	1.7	0.7	24.5
		Asthmatic	272	11.3	6.4	9.8	1.7	2.8	40.4
		CHD	323	12.6	7.9	10.6	1.8	1.3	41.5
	PM_{10}	COPD	435	14.3	6.8	12.8	1.6	2.9	41.4
		Healthy	200	14.5	7.0	13.0	1.6	2.9	54.9
		Asthmatic	269	16.4	7.4	14.7	1.6	1.2	47.3
		CHD	324	18.0	9.0	16.1	1.6	3.3	54.3
Central Site	$PM_{2.5}$	All	222	10.1	5.7	8.6	1.8	1.0	29.5
	PM_{10}	All	221	17.3	9.1	14.9	1.8	0.4	49.9

Correlations between the particulate samples are presented for both size fractions in Table 17. Correlations were high for the residential outdoor and central site $PM_{2.5}$ measurements, and slightly lower for the PM_{10} measurements. Personal exposures correlated fairly well with indoor concentrations (0.57) but not with outdoor levels (0.34). The complete $PM_{2.5}$ distributions, summed across all cohorts, are compared in Figure 23. The indoor distribution was substantially lower than the personal or outdoor distributions.

Table 17. Correlations Between Personal, Indoor, Outdoor, and Central Site Monitors for $PM_{2.5}$ and PM_{10}: Seattle

		Personal $PM_{2.5}$	Indoor $PM_{2.5}$	Outdoor $PM_{2.5}$	Central $PM_{2.5}$	Indoor PM_{10}	Outdoor PM_{10}	Central PM_{10}
Personal $PM_{2.5}$	corr	1						
	N	1078						
Indoor $PM_{2.5}$	corr	**0.57**	1					
	N	996	1500					
Outdoor $PM_{2.5}$	corr	**0.34**	0.49	1				
	N	1010	1426	1498				
Central $PM_{2.5}$	corr	**0.29**	0.40	0.83	1			
	N	974	1293	1298	1408			
Indoor PM_{10}	corr	0.43	0.75	0.33	0.30	1		
	N	1008	1455	1441	1304	1515		
Outdoor PM_{10}	corr	0.31	0.46	0.89	0.79	0.31	1	
	N	1003	1428	1447	1294	1448	1497	
Central PM_{10}	corr	0.25	0.34	0.67	0.87	0.31	0.73	1
	N	965	1288	1290	1368	1298	1288	1398

Note: All p-values < 0.0001.

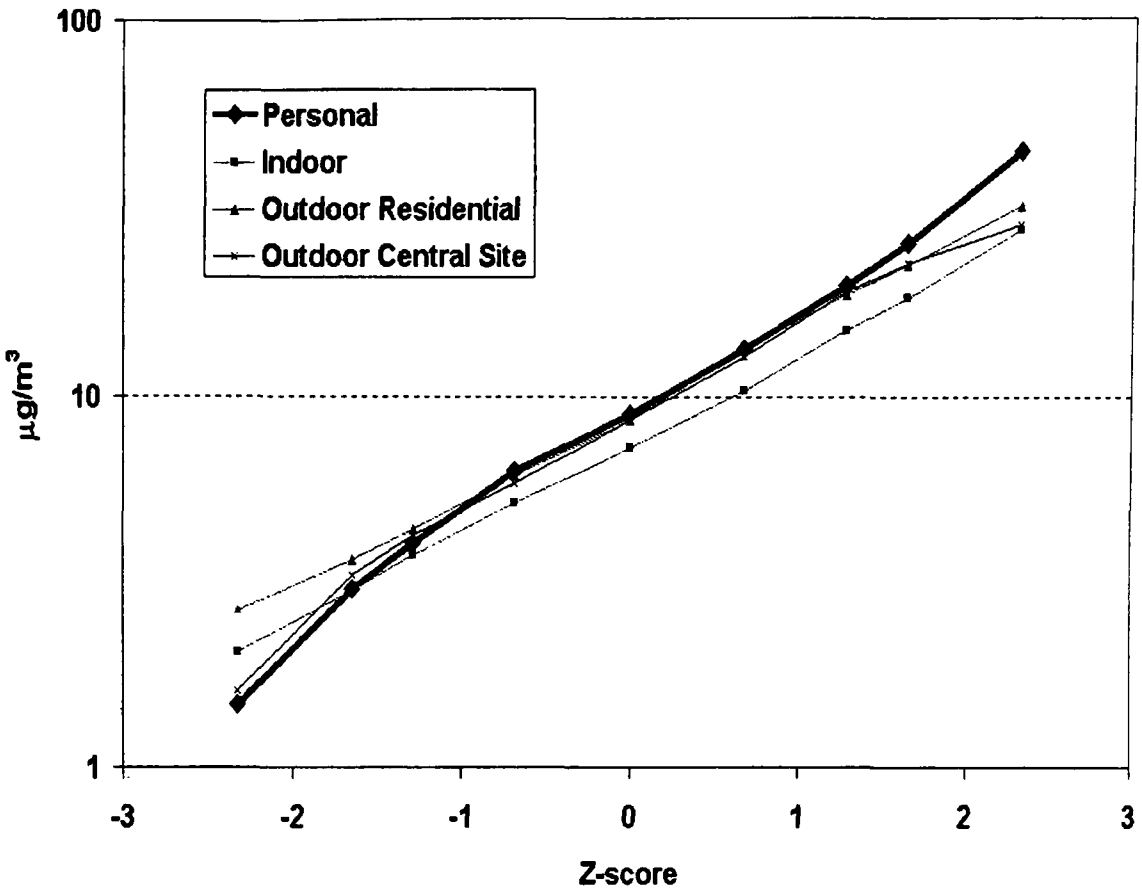

Figure 23. Cumulative distribution functions for personal, indoor, outdoor, and central site $PM_{2.5}$. Seattle. N = 1078-1500.

At equilibrium, as mentioned above, the infiltration factor $F_{inf} = Pa/(a+k)$, where P is the penetration factor (dimensionless), a is the air exchange rate (h^{-1}) and k is the deposition rate (h^{-1}). The magnitudes of these parameters were estimated using the recursive mass balance model based on the MIE pDR measurements (Allen et al., in press). An algorithm was developed to identify all indoor peaks. These peaks were removed, leaving only the concentrations due to penetration of outdoor particles. The 10-minute average measured concentrations from the indoor and outdoor pDRs were combined to form one-hour concentrations. Using a one-hour time step, the recursive mass balance model was run to provide separate estimates of P, a, k, and F_{inf} for each home. Summary statistics from this effort are provided in Table 18 and Boxplots showing the range of values for each parameter across the 65 homes are shown in Figure 24. The penetration factor P was close to 1, as was also the case in the earlier PTEAM Study (Pellizzari et al., 1992; Özkaynak et al., 1996a.b; Clayton et al., 1993). The deposition constant k was 0.15 ($\pm$ 0.19) h^{-1}, smaller than the value of 0.39 h^{-1} observed in PTEAM.

Table 18. Summary of Estimated Particle Penetration (P), Air Exchange Rate (a), Particle Decay Rate (k), and the Ambient PM Infiltration Efficiency (F_{inf}) Using the Recursive and Nonlinear Regression Models: Seattle

Variable	N	Mean	Median	Std Dev
P	65	0.97	1.00	0.08
a	65	0.59	0.73	0.26
k	65	0.15	0.07	0.19
F_{inf}	65	0.78	0.81	0.19

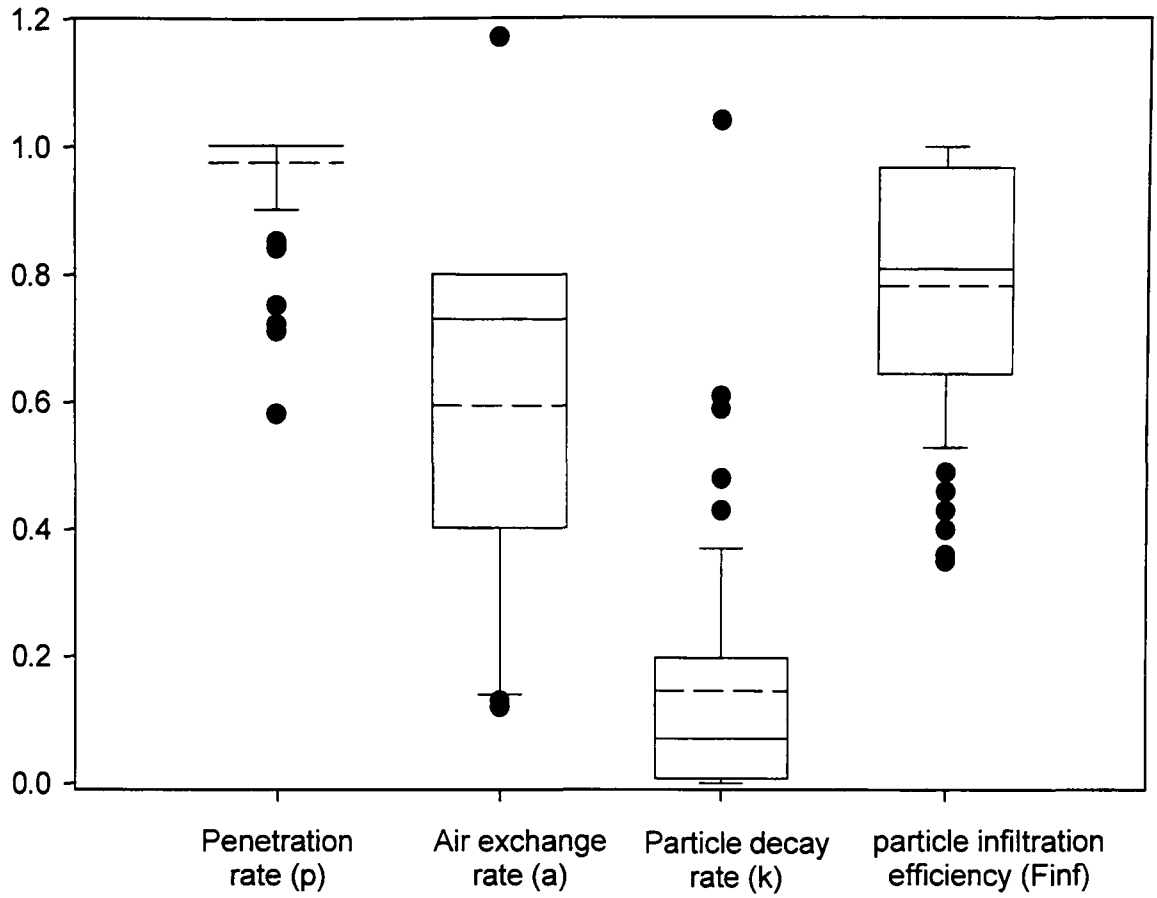

Figure 24. Distribution of P, a, k, and F_{inf} among residences: Seattle.

Longitudinal correlations between personal and outdoor $PM_{2.5}$ were calculated for each subject (Figure 25). Correlations covered a wide range from negative to nearly 1, as has been observed in other studies, with the median correlation between 0.3 and 0.4 for all four cohorts. The hypothesis that the sick persons would have higher longitudinal correlations with outdoor air than healthy persons was not confirmed.

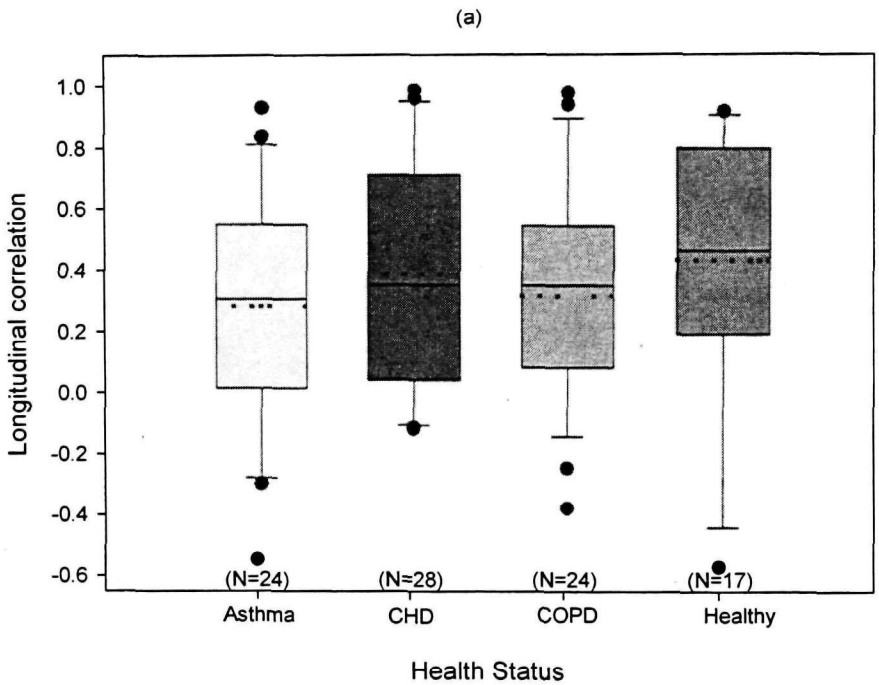

Figure 25. Longitudinal correlation between personal and central site $PM_{2.5}$ for each subject by health status (N≥6 for each subject): Seattle

Elderly high-risk subpopulations may spend more of their time in the home and less in transit than their healthy counterparts. This hypothesis was tested in all studies by administering a questionnaire asking for time spent in several microenvironments. In the Seattle study, the hypothesis was confirmed (Table 19), although the differences were relatively small.

Table 19. Percentage of Time Spent in Microenvironments by Health Group: Seattle.

		Percentage of time spent in each microenvironment			
Group	**Microenvironment**	**Mean**	**Std Dev**	**Min**	**Max**
Asthmatic Kids (N=33)	Home	66.4	5.7	55.5	80.0
	Yard	1.7	2.6	0.0	8.2
	In Transit	4.4	1.7	1.3	8.2
	Work	1.1	3.5	0.0	16.5
	Outdoors	4.7	3.5	0.1	17.5
	Indoors away from home	21.0	6.4	4.5	33.2
	Cooking, self	0.1	0.1	0.0	0.5
	Cooking, others	0.7	0.5	0.0	1.9
CHD, Adults (N=38)	Home	85.5	7.8	65.0	96.5
	Yard	1.0	1.4	0.0	6.0
	In Transit	3.6	2.3	0.1	9.2
	Work	0.3	1.7	0.0	10.6
	Outdoors	0.9	1.2	0.0	4.8
	Indoors away from home	6.9	5.1	0.1	20.9
	Cooking, self	1.7	1.6	0.0	5.8
	Cooking, others	0.2	0.3	0.0	1.5
COPD, Adults (N=56)	Home	87.6	6.9	71.4	100.0
	Yard	0.8	1.0	0.0	4.3
	In Transit	3.2	1.9	0.0	7.3
	Work	0.1	0.6	0.0	3.1
	Outdoors	1.0	1.9	0.0	11.6
	Indoors away from home	6.1	4.6	0.0	21.3
	Cooking, self	1.0	1.3	0.0	5.6
	Cooking, others	0.2	0.6	0.0	2.7
Healthy, Adult (N=39)	Home	82.7	8.3	66.8	99.2
	Yard	1.2	1.6	0.0	6.7
	In Transit	4.0	2.5	0.5	9.3
	Work	1.0	2.8	0.0	12.4
	Outdoors	1.7	1.8	0.0	7.9
	Indoors away from home	8.0	5.3	0.1	19.4
	Cooking, self	1.0	1.0	0.0	4.4
	Cooking, others	0.3	0.6	0.0	2.7

DISCUSSION

The mean concentrations of $PM_{2.5}$ are compared across all studies in Table 20. (Results from two earlier studies of retirement homes by NERL are included for comparison.) $PM_{2.5}$ exposures were lower in Seattle than in the other cities. Seasonal differences were not strong in Boston or Raleigh, but were higher in spring than in fall in Atlanta, higher in summer than in winter in Los Angeles, and (not shown) higher in the fall-winter heating season in Seattle.

Indoor $PM_{2.5}$ concentrations were often quite similar to outdoor concentrations in all cities. The only exceptions were the two retirement homes in Baltimore and Fresno, where recirculation and filtering of outdoor air was provided on a constant basis by the HVAC systems. Personal exposures were often but not always higher than indoor concentrations, but only by a few (1-4) $\mu g/m^3$.

Table 20. Arithmetic Mean $PM_{2.5}$ Concentrations ($\mu g/m^3$)—All Studies.

Site	N (Personal)	Personal	Indoor	Outdoor
Raleigh/Chapel Hill	712	23.0	19.1	19.3
Atlanta--Fall	141	16.6	14.3	14.7
Atlanta--Spring	142	15.0	19.8	22.0
Boston--Winter	55/47[a]	17.6/12.5[a]	10.4	11.4
Boston--Summer	63/31	9.4/12.0	12.7	12.8
Los Angeles--Winter	87	19.6	16.9	13.5
Los Angeles--Summer	92	25.1	18.1	19.3
Seattle--COPD	307	10.5	8.5	9.2
Seattle--healthy	183	9.3	7.4	9.0
Seattle--CHD	325	10.8	9.5	12.6
Seattle—asthmatic children	263	13.3	9.2	11.3
Baltimore retirement home	325	13.0	10.0	22.0
Fresno retirement home	120	13.3	9.7	20.5

[a] Second personal sample from same household.

Over 2100 indoor and outdoor PM_{10} samples were collected in three cities (Table 21). Once again Seattle had the lowest outdoor concentrations. Indoor and outdoor PM_{10} concentrations were quite comparable in all areas, again with the exception of the two retirement homes, which were able to reduce the outdoor levels penetrating indoors by substantial amounts.

Table 21. Arithmetic Mean PM_{10} Concentrations ($\mu g/m^3$)—All Studies.

Site	N (Indoor)	Indoor	Outdoor
Raleigh/Chapel Hill	761	23.2	27.2
Seattle--COPD	437	14.1	14.3
Seattle--healthy	206	12.6	14.5
Seattle--CHD	324	16.2	18.0
Seattle—asthmatic children	274	19.4	16.5
Baltimore retirement home	28	11.0	30.0
Fresno retirement home	24	15.1	28.2

A fundamental goal of all these studies was to relate personal exposure to outdoor concentrations. For different cohorts, median longitudinal correlation coefficients (either Pearson or Spearman) ranged between 0.10 and 0.65. These values suggest that for the median person in each of the high-risk sensitive subpopulations, outdoor air explained as little as 1% up to a maximum of about 40% of the variation in personal exposure. All studies included some persons with very high correlations with ambient air and others with very low or even negative correlations. These correlations are lower than those found for healthy adults and children (Ebelt et al., 2000; Janssen 1998; Janssen et al., 1998; Williams et al., 2000a).

An important parameter governing the effect of outdoor air particles on indoor concentrations is the infiltration factor:

$$F_{inf} = Pa/(a+k)$$

The infiltration factor is expected to be lower when the house is closed and higher when windows are open. Therefore F_{inf} was calculated separately for the heating and non-heating seasons in each city. During the heating season, the infiltration factor varied over a tight range of 0.40 to 0.53. In the non-heating seasons, the range was much wider, from 0.40 to 0.79 (Table 22). Every city except for RTP showed an increase in F_{inf} between the heating and non-heating seasons. The increase was quite large in Los Angeles, Boston, and Seattle, and relatively small in Atlanta.

Table 22. Variation of $PM_{2.5}$ Infiltration Factor by Season—All Studies

City	N	Heating	SE	N	Non-heating	SE
RTP: linear model	29	0.46	0.05	25	0.40	0.04
Los Angeles	15	0.42	0.08	15	0.70	0.11
Boston	14	0.40	0.13	15	0.67	0.10
Boston sulfate	N/A	N/A	N/A	15	0.75	0.03
Atlanta	24	0.43	0.10	22	0.49	0.14
Atlanta sulfate	24	0.40	0.04	22	0.45	0.04
Seattle	55	0.53	0.16	55	0.79	0.18

A summary of the longitudinal correlations between personal and outdoor air for participants in all the panel studies is provided in Table 23. The three studies all succeeded in collecting valid personal and outdoor data for at least four consecutive days on about 100 separate occasions. All studies agreed with each other, and also with previous studies, in having roughly half the participants with correlations below 0.5. These values indicate that for the median person in each of the studies, outdoor air explained between 18 and 25% of the variance in personal exposure. The studies also agreed in showing less than one fourth of the participants (14-21%) with significant personal-outdoor correlations.

Table 23. Longitudinal Correlations Between Personal and Outdoor Air for Participants—All Studies

Organization	Cities	N persons[1]	N with $p<0.05$	Fraction with $p<0.05$	N with $r>0.5$	Fraction with $r>0.5$
NERL-RTI	RTP	112	16	0.14	48	0.43
Univ. Wash.	Seattle	98	16	0.16	49	0.50
Harvard	Boston-Atlanta-L.A.	105	22	0.21	49	0.47

[1] Number with at least four consecutive days of personal and residential outdoor measurements within one season. Persons monitored in two or more seasons are counted separately for each season.

CONCLUSIONS

A strong effort has been made to gather information about the actual exposures of those persons at high risk from pollution. Over 200 persons were monitored in five cities and over 2500 personal, indoor, and outdoor $PM_{2.5}$ samples were collected, with another 2000 indoor and outdoor PM_{10} samples collected. Associated co-pollutants and elements were also sampled, many with personal as well as indoor and outdoor measurements. Thousands of questionnaires were administered, creating a rich database on activity patterns, time budgets, and particle-generating activities. These data are a valuable resource for current and future analyses, and it is expected that a large number of journal articles will be produced in the next few years investigating different aspects of these studies.

The main goal of the studies, to document how the exposure of high-risk subpopulations to fine particles is related to ambient concentrations, both cross-sectionally and longitudinally, has been amply fulfilled. All major identified high-risk subpopulations were included in the study efforts. Good geographic distribution across the country was incorporated into the study designs, and highly precise measurements of both exposure and outdoor concentrations performed. The second goal of the studies, to calculate the contribution of ambient concentrations to total exposure, was more difficult. While estimates have been achieved in some of the studies, additional work in this area will depend on elemental data still being analyzed and on which of several statistical approaches might be adopted. However, the basic data that will ultimately lead to considerable advances in fully completing this goal have been collected and will form a major focus of future journal articles bearing on this question.

The organizations performing the work gained valuable experience in field studies involving extensive personal and indoor monitoring. As a result, new improved personal monitors were developed and the Harvard Multipollutant Personal Sampler, developed as part of this study, is now available commercially.

Hundreds of volunteers took part in the studies and in most cases successfully completed them, indicating that the burden of carrying the monitoring equipment, making room for the indoor monitors, keeping activity diaries, and filling out questionnaires each day had been successfully alleviated by good planning and study design.

Two hypotheses were advanced at the beginning of these studies. One was that high-risk subpopulations would engage in fewer dust-generating activities and would therefore have lower indoor air concentrations and lower personal exposures than healthy cohorts. This hypothesis was not confirmed. In Seattle, for instance, the healthy cohort had the lowest personal exposures of the four cohorts studied. In Boston, the spouses of the high-risk persons had lower concentrations in one season and higher concentrations in the other, but with no overall difference.

A second related hypothesis was that the personal exposures of the high-risk cohorts would have stronger correlations with outdoor air than healthy cohorts, due to fewer particle sources indoors. Again the hypothesis was not confirmed, with the healthy cohort in Seattle actually showing higher mean correlations with outdoor air than the three high-risk cohorts.

The following conclusions may be ventured. However, it should be carefully noted that all subjects were chosen on a non-probabilistic basis, and therefore ***all conclusions apply only to the subjects themselves; they must not be extrapolated to larger groups of people.***

- Personal $PM_{2.5}$ monitors were fully evaluated in side-by-side comparisons with EPA reference or equivalent instruments at all sites and generally agreed well.
- Mean personal $PM_{2.5}$ exposures for the different cohorts ranged from 9 to 25 $\mu g/m^3$. The Seattle cohorts were at the low end and the Los Angeles and Raleigh/Chapel Hill cohorts at the high end.
- Mean indoor and outdoor $PM_{2.5}$ concentrations were similar for all cohorts living in private homes in all cities, and ranged from 7-20 $\mu g/m^3$ indoors and from 9-22 $\mu g/m3$ outdoors.
- Personal clouds on the order of 1-4 $\mu g/m^3$ were observed for $PM_{2.5}$ for most cohorts.
- Mean indoor and personal $PM_{2.5}$ concentrations were much lower than outdoor concentrations for cohorts living in retirement homes, due possibly to extensive filtering and recirculation of outdoor air by the HVAC systems.
- Personal $PM_{2.5}$ exposures were similar for the healthy and sick cohorts. For these subjects, there was no indication that the sensitive persons were reducing their exposure.
- Longitudinal correlations of personal $PM_{2.5}$ exposure with outdoor air concentrations were low for some individuals in each cohort and high for others, suggesting the importance of individual activity patterns and household characteristics in affecting the personal-outdoor relationship. In all three studies, fewer than half of these correlation coefficients exceeded 0.5, and fewer than one fourth were significant at the $p<0.05$ level.
- Calculations of the infiltration factor, which determines the contribution of outdoor air to indoor concentrations, are difficult and have resulted in divergent estimates between studies and even between different applications of the mass-balance model (linear vs. nonlinear) in the same study. It is not clear if this is fully explained by differences among the cities in climate, house construction practices, use of air conditioning, or dependence on the assumptions of the mass balance model. This is a problem that will require further analyses of the collected data.

FUTURE WORK

Although much has been learned, there remain some important questions requiring further research, development, and field studies. Presently our estimates of the infiltration factor, the penetration coefficient, and the $PM_{2.5}$ deposition rate appear to be conflicting. The assumptions underlying the use of the mass balance model need to be carefully examined.

A continuing challenge is to understand the organic chemical (OC) loading on particles, which provides a substantial portion of the total mass. Because of both positive and negative artifacts on quartz fiber filters, the standard thermo-optical method of determining OC is usually unable to arrive at a trustworthy estimate of the actual OC mass on the particles in their normal state in the atmosphere. This problem is being further studied at EPA-NERL.

Because of increasing interest in air toxics, future studies may combine studies of particles with studies of polar and non-polar volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs). EPA-NERL is presently planning studies that will add these air toxics to their continuing PM studies. The importance of ultrafine particles in contributing to mortality and morbidity continues to be of interest, but a lack of personal monitors capable of measuring ultrafine particles has limited our knowledge of personal exposure to ultrafines, and data is sparse on indoor concentrations. Additional work in the area of low-burden, low-cost personal exposure monitors and the adaptation of survey instruments (activity diaries and questionnaires) to better characterize potential personal exposures to PM of ambient sources in the general population is needed.

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APPENDIX

Journal Articles Published or in Preparation from the Panel Studies

Journal Articles Published or in Preparation from the Panel Studies

Research Triangle Park, NC

Conner, T., Norris, G., Landis, M., and Williams, R. (2001). Individual particle analysis of indoor, outdoor, and personal samples from the 1998 Baltimore retirement home study. *Atmospheric Environment*, 35:3935-3946.

Landis, M.S., Norris, GA., Williams, R.W., and Weinstein, J.P. (2001) Personal exposures to $PM_{2.5}$ mass and trace elements in Baltimore, Maryland. *Atmospheric Environment* 35: 6511-6524.

Rea, A., Zufall, M., Williams, R., Reed, C., and Sheldon, L. (2001). The influence of human activity patterns on personal PM exposure: a comparative analysis of filter-based and continuous particle measurements. *Journal of Air and Waste Management Association*, 51:1271-1279.

Williams, R., Suggs, J., Rea A., Leovic, K., Vette, A., Sheldon, L., Rodes C., and Thornburg J. The Research Triangle Park particulate matter panel study: modeling ambient source contribution to personal and residential PM mass concentrations. *Atmospheric Environment*, in press.

Williams, R., Suggs, J., Rea, A., Leovic, K., Vette, A., Croghan, C., Sheldon, L., Rodes, R., Thornburg, J., Ejire, A., Herbst, M., and Sanders, W. The Research Triangle Park particulate matter panel study: PM mass concentration relationships. *Atmospheric Environment*, in press.

Rodes C, Lawless P., Thornburg J., Williams, R., Evans G., Zweidinger R., Norris, G., McDow S. The potential influence of face velocity on the loss of volatile species collected on Teflon filters. Submitted to *Journal of Air and Waste Management Association.*

Thornburg J., Rodes, C.E., Williams, R. Relationship between HVAC system operation, air exchange rate, and indoor-outdoor particulate matter ratios. Submitted to *Atmospheric Environment.*

Lawless, P., Rea, A., Williams, R. Personal monitoring compliance observed in the NERL Research Triangle Park particulate matter panel study. Planned journal submission.

Rea A., Croghan C., Thornburg J., Rodes, E., Williams R. PM concentrations associated with personal activities based on real-time personal nephelometry data from the NERL RTP PM panel study. Submitted to *Atmospheric Environment.*

Boston

Brown et al. Assessing exposures to particulate and gaseous pollutant for senior adults, individuals with COPD and MI patients in Boston, MA. Planned journal submission.

Sarnat et al. Examining the impact of ambient fine particulate matter sources on personal exposures: the effect of cohort, city and season. Planned journal submission.

Atlanta

Chang et al. The relationship between outdoor, indoor, and personal exposures to $PM_{2.5}$ and Its components for two sensitive cohorts. Planned journal submission.

Reid et al. Factors affecting the relationship between indoor and outdoor concentrations for ozone, NO_2, and SO_2. Planned journal submission.

Wheeler et al. associations between cardiovascular health and particulate exposures for two sensitive cohorts. Planned journal submission.

Los Angeles

Chang et al. Characterization of $PM_{2.5}$, EC and NO_3^- Exposures for the Metropolitan Los Angeles Area. Planned journal submission.

Lau et al. Potential for confounding by gaseous pollutants: results from Los Angeles. Planned journal submission.

Seattle

Liu, L.-J. S., Slaughter, C., Larson, T. (2002). Comparison of light scattering devices and impactors for particulate measurements in indoor, outdoor, and personal environments. *Environmental Science & Technology*, 36, 2977-2986.

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Allen, R., Box, M., Larson, T., Liu, L.-J. S. (2001). A cost-effective weighing chamber for particulate matter filters. *Journal of Air & Waste Management Association* 51: 1659-1653.

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Pang, Y., Gundel, L. A., Larson, T., Finn, D., Liu, L.-J. S., Claiborn, C (2002). Development and evaluation of a novel personal particulate organic and mass sampler (PPOMS). *Environmental Science & Technology* 36:5205-5210.

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Liu et al., Outdoor contribution to personal $PM_{2.5}$ (planned submission in Fall 2003).

Claiborn et al. Indoor OC artifact (planned submission in Fall 2003).

Larson et al. Source apportionment for indoor and outdoor PM exposures (planned submission in Spring 2003).

Seattle (Cont.)

Johns et al. Comparisons of breath, personal, indoor, and ambient CO (planned submission in Fall 2003).

Claiborne et al. Characterization of indoor and outdoor EC/OC in Seattle (planned submission in Fall 2003).

Wu et al. Use of real-time CO_2 monitors and nephelometer measurements to estimate and characterize air exchange rates (planned submission in Winter 2004).

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