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Estimating Contributions of Outdoor Fine Particles to Indoor Concentrations and Personal Exposures:

Effects of Household Characteristics and Personal Activities

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Estimating Contributions of Outdoor Fine Particles to Indoor Concentrations and Personal Exposures:

Effects of Household Characteristics and Personal Activities

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Notice

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Abstract

A longitudinal study of personal, indoor, and outdoor exposures to $PM_{2.5}$ and associated elements was carried out involving 37 residents of the Research Triangle Park area in North Carolina. Participant exposures were monitored for 7 consecutive days in each of four seasons. A main goal of the study was to estimate the contribution of outdoor $PM_{2.5}$ to indoor concentrations and personal exposures This contribution depends on the infiltration factor (the fraction of outdoor $PM_{2.5}$ remaining airborne after penetrating indoors), which can be estimated using sulfur as a marker for particles of outdoor origin. The annual average infiltration factors ranged from 0.26 to 0.89, and depended strongly on air exchange rates. The outdoor contributions to personal exposure were then regressed longitudinally on outdoor concentrations measured at a central monitoring station, with a range of R^2 values from 0.19 to 0.88. Variables significantly affecting indoor air $PM_{2.5}$ concentrations included smoking and cooking, the number of persons in the household, burned food, use of a kitchen exhaust fan, and duration of candle use. These findings might have important implications for epidemiological studies

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Chapter 1
Introduction

Many studies worldwide in the last decade have documented an association between health effects and particle concentrations measured at central monitoring sites (Schwartz et al., 1996). Since the health effects are presumably related to personal exposures, an important research need, identified by the National Academy of Sciences in 1995, is to determine how personal exposures correlate with these outdoor concentrations (NRC-NAS 1998). A number of studies (Abt 2000a,b; Allen et al., 2003; Clayton et al., 1993; Ebelt et al., 2000; Evans et al., 2000; Hopke et al., 2003; Howard-Reed et al., 2000; Janssen et al., 1997, 1998, 1999, 2000; Keeler et al., 2002; Landis et al., 2001; Liu et al., 2003; Long et al., 2000, 2001, Özkaynak et al., 1996a,b; Pellizzari et al., 1992; Rea et al., 2001; Rojas-Bracho et al., 2000; Sarnat et al., 2000, 2001; Thomas et al., 1993; US EPA 2002, 2003; Vette et al., 2001; Wallace et al., 2003a; Williams et al., 2000a,b, 2003a,b) have measured personal exposure directly using personal monitors, and the correlations of personal exposure with outdoor concentrations are straightforward to determine (Wallace, 2000). However, the correlation that interests epidemiologists is not that between total personal exposure and outdoor concentrations, but the correlation between that component of personal exposure due to outdoor particles and the outdoor concentrations. This requires the ability to estimate the contribution to personal exposures from particles originating outdoors. Only a few studies have reported making this estimate (Ebelt et al., 2000; Allen et al., 2003, 2004). The goal of this report is to estimate the contribution of outdoor particles to personal exposure for a group of 37 persons monitored one week per season over four seasons in 2000-2001 (US EPA 2002, 2003; Williams et al., 2003a,b). The correlation between this portion of personal exposure and $PM_{2.5}$ outdoor concentrations will then be calculated for each person. Many of the main findings of this report appear in Wallace and Williams (2005).

Since people spend on average 89% of their time indoors, the contribution of outdoor particles to indoor concentrations will also be explored. For many people, the indoor-outdoor relationship may be the major determinant of the personal-outdoor relationship.

The contribution of outdoor particles to indoor concentrations is described by the mass balance equation. The full mass balance equation includes such phenomena as coagulation, condensation, and gas-to-particle conversion (Nazaroff and Cass, 1989). We will consider here a simplified version involving only infiltration, exfiltration, deposition, and indoor sources. The differential form of this simplified mass balance equation is

$$dC_{in}/dt = PaC_{out} - (a+k)\,C_{in} - S/V \qquad (1\text{-}1)$$

where C_{in} = indoor number or mass concentration (cm^{-3} or $\mu g/m^3$),
C_{out} = outdoor number or mass concentration
P = penetration coefficient across building envelope
a = air exchange rate (h^{-1})
V = volume of building (cm^3 or m^3)
S = source strength (h^{-1} or $\mu g\ h^{-1}$)
k = deposition rate of particles (h^{-1})

The equation is assumed to be applicable to all particle sizes, with all terms except air exchange rate and building volume considered to be functions of particle size. We assumed that the entire house is a single well-mixed zone, with instantaneous mixing of particles throughout the house, and that the measured air exchange rate in one room applies to the entire house. This assumption is probably violated by most homes, which are likely to have different zones on each floor or even on the same floor. We also assume that the deposition rate is constant over the period of integration. This assumption will not hold if persons open windows, turn on fans, run the furnace or the air conditioner, or otherwise make changes in the household operating characteristics that will affect particle deposition during the integration period. Finally, we assume that the averaging time over which the equation is to be evaluated is sufficiently long that transient terms due to short-term changes in the outdoor concentration are negligible compared to the long-term average concentrations. Since we are dealing with 24-hour averages, this assumption is probably a good one. Under these assumptions, the solution to the mass balance equation is

$$C_{in} = [Pa/(a+k)] * C_{out} + S/[V(a+k)] \qquad (1\text{-}2)$$

The coefficient of the outdoor concentration is sometimes called the infiltration factor F_{inf}:

$$F_{inf} = Pa/(a+k) \qquad (1\text{-}3)$$

The infiltration factor has a major effect on the indoor-outdoor and the personal-outdoor relationships. This is expected to vary by household and resident characteristics. For example, a tightly built house may have a lower penetration coefficient than a drafty house, although almost no data are available to support this claim. A house with a large surface area (e.g., many carpets, rugs, or fibrous wall hangings) may have higher deposition rates (Lai and Nazaroff 2000). Use of fans or filters may also increase particle deposition rates (Howard-Reed et al., 2003; Riley et al.,

2002; Thatcher et al., 2002; Thatcher and Layton, 1995; Wallace et al., 2004a). Open windows will increase F_{inf} by increasing the air exchange rate (Howard-Reed et al., 2002; Wallace and Howard-Reed, 2002) and possibly by redirecting infiltrating particles through the open window (P = 1) rather than through the rest of the building envelope (P < 1) (Liu and Nazaroff, 2001; Mosley et al., 2001; Thornburg et al., 2001). Use of air conditioning has been shown to lower the infiltration factor, either because of reducing air exchange rates by shutting windows or increasing deposition rates by recirculating indoor air through ductwork (Howard-Reed et al., 2003; Sarnat et al., 2000; Lai et al., 1999; Thornburg, 2004; Wallace et al., 2002).

Despite these clear indications that exposure to outdoor air particles indoors depends heavily on household and behavioral characteristics, studies capable of estimating the infiltration factor reliably for individual homes are rather few. Even more rare are studies capable of estimating the values of P and k for individual homes (Allen et al., 2004). In this study we attempt to estimate both F_{inf} and its parameters P and k for individual homes, together with an estimate of the uncertainties involved.

Several investigators have noted that sulfur has few indoor sources (Ebelt et al., 2000; Sarnat et al., 2002). If that is the case, the source term in Equation 1-2 above may be ignored, and the equation takes the very simple form

$$S_{in}/S_{out} = F_{inf} \qquad (1\text{-}4)$$

where S_{in} and S_{out} are the sulfur concentrations indoors and outdoors.

Indoor-outdoor comparisons of sulfur concentrations thus provide a direct way to estimate F_{inf} for each individual home. Strictly speaking, the sulfur data provides only information for particles with similar behavior to that of sulfur with respect to penetration, deposition, and reactivity. Sulfur particles are smaller than most other fine particles. Since the particles in the 1-2.5 μm range may have higher deposition velocities than sulfur, the estimates for the sulfur deposition rate k_S would be slight underestimates for the typical aerosol mixture in the $PM_{2.5}$ category. To avoid extra notation, we shall use P and k throughout rather than P_S and k_S but the reader should remember that these are values appropriate only for particles in the size range of sulfur particles. Later we provide evidence that this "sulfur-related" infiltration factor is actually a very good estimate of the $PM_{2.5}$ infiltration factor.

Chapter 2
Description of Study Methods and Database

A full description of the measurement methods used in this study has been provided in the two preceding reports in this series (US EPA 2002, 2003) and also in two published articles (Williams et al., 2003a,b). The following abbreviated description includes only the methods discussed in this report.

In this study, 37 persons in 36 homes in the Research Triangle Park, NC area were monitored for up to four seasons, 7 days per season. (Thirteen of these were monitored for only one, two, or three seasons; see Appendix A.) Two gravimetric monitor types were employed for $PM_{2.5}$ measurements: the Harvard Impactor (HI), operating at 20 Lpm, and a Personal Exposure Monitor (PEM), operating at 2 Lpm for a nominal 24-h period. The PEM was used for personal samples and the HI for indoor-outdoor samples, the latter measured just outside the home. An HI, a PEM and a Federal Reference Method (FRM) monitor were also operated every day of the study at a central site. There were also fixed PEM_{10} monitors co-located with the $HI_{2.5}$ monitors—filters from these instruments were analyzed for a suite of elements using x-ray fluorescence. Since sulfur particles are expected to be in the size range <0.5 μm, the PEM_{10} filters should have the same amount of sulfur as the $PEM_{2.5}$ filters.)

The HI mass measurements had a precision of about 5%; the PEM measurements had a precision of about 8% (Williams et al., 2003a). The precision of the sulfur measurements was calculated to be about 8% (Kellogg, R, personal communication).

The data analysis concentrated on sulfur as a tracer of outdoor-generated particles. Indoor-outdoor ratios were used to estimate the infiltration factor for $PM_{2.5}$; personal-outdoor ratios were used to estimate the portion of personal exposure due to outdoor-generated particles. The estimates of exposure due to outdoor-generated particles were then regressed on outdoor measurements, both at the home and at a central site, to determine the relationship between central site measurements and exposure to particles of outdoor origin.

Participants filled out activity logs each day, identifying cooking, cleaning, and other activities that might affect particle exposures. A questionnaire on household characteristics was also completed for each house. The full set of questionnaire variables and their definitions and units is provided in Appendix A (Table A-4). These variables were then included in multivariate regressions to determine their influence on air exchange rates, the infiltration factor, and the observed $PM_{2.5}$ and sulfur concentrations. A detailed description of this analysis is provided in the section on multivariate regressions.

Chapter 3
Results

A total of 876 person-days had at least one personal, indoor, or outdoor $PM_{2.5}$ measurement. About 868 days had at least one filter analyzed by XRF. Samples were flagged if they failed any of a number of quality control criteria. For example, flow rates were required to be within 10% of the target values, filters were discarded if seen to be torn or pierced. All outdoor $PM_{2.5}$ filters collected between April 11 and April 17 were discarded due to excessive contamination with pollen. An additional requirement for the sulfur measurements was that indoor values not be more than 1.08 times outdoor values (the 1.08 factor was chosen because the estimated uncertainty of the sulfur measurements was 8%). Even if an indoor/outdoor ratio exceeding 1.08 were correct, it would indicate an indoor source of sulfur, and therefore should not be used in analyses to determine the infiltration factor. Six samples were flagged for exceeding a 1.08 indoor/outdoor ratio. Another 21 samples were flagged when regressions suggested an indoor source of sulfur, and it was discovered that the participant was using a humidifier during three seasons (7 days per season). Since different combinations of variables will have different numbers of flags, the number of valid data points sometimes varies in the following tables.

Calculation of F_{inf} Using the Indoor/Outdoor Sulfur Ratio

Table 3-1 lists the distributional characteristics of all paired indoor-outdoor samples with validated $PM_{2.5}$ and sulfur measurements. Also included in Table 3-1 are estimates of the indoor/outdoor sulfur ratio. The average indoor/outdoor sulfur ratio was 0.59 (0.16 SD).

Assuming all the sulfur is in the form of ammonium sulfate, we can calculate the mass by multiplying by the ratio of the molecular weights (3.5). The result is an estimate of 8.0 $\mu g/m^3$ of ammonium sulfate, or about 41% of the total $PM_{2.5}$. Several Eastern cities included in EPA's speciation network ranged from 26-31% in their sulfate/$PM_{2.5}$ values (AQCD 2003).

All the validated indoor and outdoor sulfur concentrations (N = 775) are displayed in Figure 3-1.

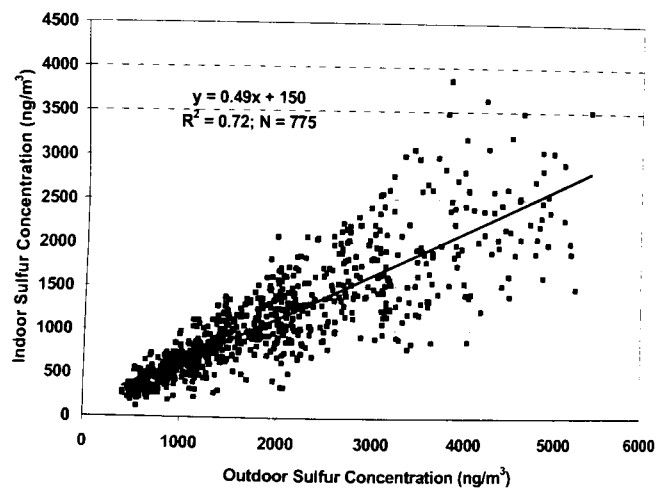

Figure 3-1. Valid pairs of indoor and outdoor 24-h average sulfur measurements (ng/m^3).

The indoor and outdoor sulfur measurements averaged over all visits to each home are displayed in Table 3-2, together with their standard deviations and standard errors. Home 29 had two participants, 29 and 35. It was visited twice in the first season but only once thereafter. Thus it had a maximum of 35 visits compared to 28 for most of the other homes. Several homes were monitored for 8 days instead of 7 in the first season so that their maximum number of visits was 29.

The ratios of the mean indoor sulfur concentration to the mean outdoor sulfur concentration averaged over all visits are shown by house in Table 3-3. Since the ratio involves two measurements, each with associated error, the standard deviation and standard error of the ratio are calculated by propagating the proportional errors:

$$SD_{calc} = [(\sigma_{in}/\mu_{in})^2 + (\sigma_{out}/\mu_{out})^2]^{1/2} * \mu_{in}/\mu_{out} \qquad (3\text{-}1)$$

Table 3-1. $PM_{2.5}$ ($\mu g/m^3$) and Sulfur Concentrations (ng/m^3) Observed in Matched Indoor-Outdoor Samples in the RTP Study

	N	Mean	SD	Min	10th	25th	Median	75th	90th	Max
$PM_{2.5}$ In	774	19.4	16	2	7	10	15	22	36	119
$PM_{2.5}$ Out	774	19.5	9	5	9	13	19	24	32	52
Sulfur In	775	1116	653	123	426	615	974	1441	2034	3852
Sulfur Out	775	1964	1123	404	765	1061	1759	2667	3610	5406
S In/Out	775	0.59	0.16	0.17	0.39	0.48	0.59	0.69	0.79	1.06

Table 3-2. Indoor and Outdoor 24-hour Sulfur Measurements (ng/m^3) Averaged Over All Visits to Each Home

House	N	S_{in}	SD	SE	S_{out}	SD	SE
1	27	955	372	72	1532	612	118
2	26	1069	477	94	1877	1030	202
3	29	1516	534	99	2078	756	140
4	23	882	359	75	2106	1056	220
5	27	1199	723	139	1892	1034	199
6	28	1293	651	123	2116	1008	191
7	28	1476	901	170	1947	1261	238
8	6	1270	408	167	3664	1376	562
9	24	1087	688	141	1968	1310	267
10	27	1049	558	107	1945	1202	231
11	8	1661	825	292	2446	1266	447
12	28	944	449	85	1945	1149	217
13	7	553	224	85	1534	661	250
14	27	701	342	66	1579	774	149
15	27	928	449	86	1322	588	113
16	28	1040	554	105	1639	952	180
17	26	923	467	92	1889	1140	224
18	6	1328	436	178	2625	996	407
19	27	1568	769	148	2198	1218	234
20	27	1139	641	123	2272	1229	237
21	27	1240	704	135	2152	1322	254
22	13	1231	607	168	2338	1175	326
23	13	1175	601	167	2558	1508	418
24	27	909	498	96	1795	1153	222
25	8	1827	679	240	2712	1292	457
26	24	1829	1009	206	2620	1457	297
27	28	1335	704	133	2113	1276	241
28	25	1059	502	100	1938	1147	229
29	34	991	446	76	2001	1269	218
31	14	1678	722	193	1920	839	224
32	28	607	287	54	1730	769	145
33	23	522	322	67	1493	978	204
34	12	1746	808	233	2811	1139	329
36	6	658	300	123	2525	1130	461
37	20	1079	327	73	1836	698	156
38	17	534	137	33	974	299	72
Sum/Mean	775	1139	541	126	2058	1058	252

Table 3-3. Ratios of the Mean Indoor Sulfur Concentrations to the Mean Outdoor Sulfur Concentrations

House	S_{in}/S_{out}	SD_{calc}[b]	SE_{calc}[b]
1	0.62	0.35	0.07
2	0.57	0.40	0.08
3	0.73	0.37	0.07
4	0.42	0.27	0.06
5	0.63	0.52	0.10
6	0.61	0.42	0.08
7	0.76	0.67	0.13
8	0.35	0.17	0.07
9	0.55	0.51	0.10
10	0.54	0.44	0.08
11	0.68	0.49	0.17
12	0.49	0.37	0.07
13	0.36	0.21	0.08
14	0.44	0.31	0.06
15	0.70	0.46	0.09
16	0.63	0.50	0.09
17	0.49	0.38	0.08
18	0.51	0.25	0.10
19	0.71	0.53	0.10
20	0.50	0.39	0.08
21	0.58	0.48	0.09
22	0.53	0.37	0.10
23	0.46	0.36	0.10
24	0.51	0.43	0.08
25	0.67	0.41	0.14
26	0.70	0.55	0.11
27	0.63	0.51	0.10
28	0.55	0.41	0.08
29	0.50	0.38	0.07
31	0.87	0.54	0.14
32	0.35	0.23	0.04
33	0.35	0.31	0.07
34	0.62	0.38	0.11
36	0.26	0.17	0.07
37	0.59	0.29	0.06
38	0.55	0.22	0.05
Mean	0.56[a]	0.39	0.09

[a] This value obtained by dividing the mean indoor sulfur concentration for all homes by the mean outdoor sulfur concentration. The arithmetic mean of the ratios was 0.59.
[b] Values of the standard deviation and standard error are calculated by propagation of error.

where SD_{calc} = the calculated standard deviation of the ratio for agiven house,
σ_{in} = the standard deviation of the indoor sulfur measurements,
μ_{in} = the mean of the indoor sulfur measurements,
σ_{out} = the standard deviation of the outdoor sulfur measurements,
μ_{out} = the mean of the outdoor sulfur measurements

The calculated standard error is $SD_{calc}/N^{1/2}$.

The range of indoor/outdoor sulfur ratios averaged over all visits to a home is shown in Figure 3-2. Although the interquartile range of 0.49 to 0.66 stays within 20% of the ratio of the mean indoor to outdoor sulfur concentrations of 0.56, the complete range spans a greater than threefold variation, from 0.26 to 0.87. The lowest values are homes that participated only in the summer season, when apparently air conditioning caused lower infiltration generally across most homes.

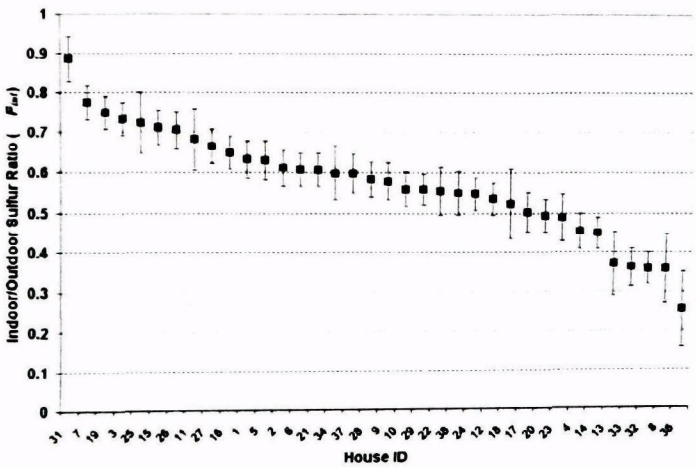

Figure 3-2. Indoor/outdoor sulfur ratios (F_{inf}) by home averaged across all seasons. Error bars are standard errors calculated by propagation of error.

Figure 3-2 shows the indoor/outdoor sulfur ratios as they were averaged over all seasons. However, the ratios should vary as a function of air exchange rate, and the air exchange rate varies by season. The highest air exchange rates were in the winter (1.07 h^{-1}) and the lowest in the summer (0.48 h^{-1}) (Table 3-4), a reversal of the normal pattern in many northern U.S. cities (Abt et al., 2000b; Ebelt et al, 2000; Long et al., 2001), where windows are opened more often in summer. The lower indoor/outdoor ratios in the summer (average 0.50) are presumably due to the use of air conditioning systems, which reduce air exchange rates by requiring closed windows and reduce sulfur concentrations by increasing the residence time in the house as well as forcing recirculation of indoor air through ductwork and filters. On the other hand, the higher air exchange rates in winter were not accompanied by comparably higher indoor/outdoor sulfur ratios, which were virtually identical (0.62-0.63) for all three seasons.

A plot of the indoor/outdoor sulfur ratios by season shows that the seasonal difference in the summer sulfur ratios was shared by nearly all of the homes (Figure 3-3).

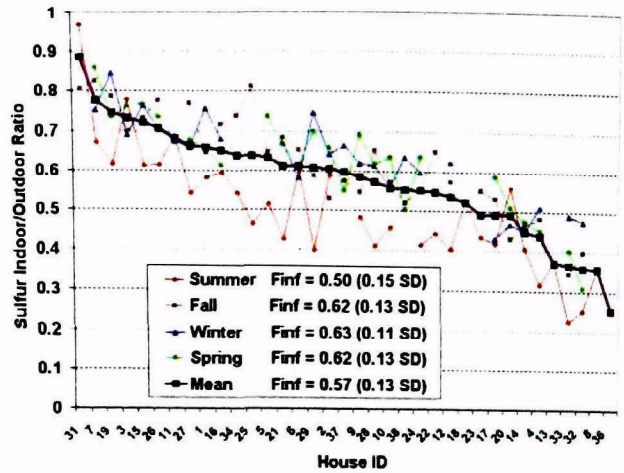

Figure 3-3. Indoor/outdoor sulfur ratios by home and by season.

Calculation of F_{inf} by Regressing Indoor Sulfur on Outdoor Sulfur

A second way to calculate F_{inf} is to regress indoor sulfur concentrations on outdoor values. Although this calculation is obviously related to the simple calculation of the mean indoor/outdoor ratio, the value of the intercept adds to our understanding. That is, a nonzero intercept might indicate possible indoor sources. In fact, the intercept for home 18 was very high (about 1000 ng/m^3) and investigation revealed that this subject was using a humidifier in his home for three of the four seasons. It is suspected that the subject was using tap water in the humidifier and that the dissolved solids in the tap water included sulfates or sulfides (Highsmith et al., 1988). Therefore the indoor sulfur values for the three seasons not including summer (21 person-days) were not used in this analysis.

Table 3-5 shows generally high R^2 values for each individual home: 14 values of 0.90 or higher and only one value below 0.50. The slope of the regression was significantly different from zero ($p<0.05$) for every home. (The p-values for the slopes are suppressed in the table to save space.) 22 of 36 homes had intercepts not significantly different from zero, indicating no apparent source of sulfur in the home. The intercepts that are different from zero (shown in boldface in the intercept column) tend to be relatively small compared to the average indoor S value of >100 ng/m^3 and thus may be due to scatter rather than to indoor sources of sulfur or sulfates. The scatter is due both to variations in F_{inf} with changing seasons and air exchange rates

Table 3-4. Air Exchange Rates (h^{-1}) and Sulfur Indoor/Outdoor Ratios by Season

Season	N	airex	SD	S_{in}/S_{out}	SD[a]
Summer	223	0.49	0.57	0.50	0.16
Fall	187	0.61	0.40	0.63	0.14
Winter	179	1.01	0.73	0.63	0.13
Spring	171	0.68	0.49	0.62	0.16

[a] Observed standard deviation; SD calculated by propagation of error would be larger.

Table 3-5. Results of Regressions of Indoor Sulfur on Outdoor Sulfur by Home, Compared to Average Values of the Indoor/Outdoor Sulfur Ratio

House	N	S_{in}/S_{out}	SE	Slope	SE	Inter.	SE	p	R^2
1	27	0.63	0.02	0.57	0.04	86	72	0.24	0.86
2	26	0.60	0.02	0.45	0.02	**231**	53	0.00	0.93
3	29	0.74	0.02	0.68	0.04	103	81	0.21	0.93
4	23	0.46	0.02	0.28	0.04	**287**	97	0.01	0.67
5	27	0.65	0.02	0.60	0.07	70	156	0.66	0.72
6	28	0.61	0.02	0.63	0.03	-46	60	0.45	0.96
7	28	0.77	0.02	0.69	0.04	132	83	0.12	0.93
8	6	0.36	0.05	0.27	0.06	291	249	0.31	0.77
9	24	0.58	0.02	0.49	0.04	115	90	0.21	0.88
10	27	0.57	0.02	0.42	0.04	**227**	88	0.02	0.82
11	8	0.68	0.04	0.63	0.07	116	180	0.54	0.93
12	28	0.54	0.02	0.36	0.03	**238**	63	0.00	0.86
13	7	0.37	0.04	0.32	0.05	60	78	0.48	0.88
14	27	0.45	0.02	0.41	0.03	55	59	0.36	0.85
15	27	0.69	0.02	0.72	0.05	-23	74	0.76	0.88
16	28	0.65	0.02	0.57	0.03	**111**	50	0.04	0.94
17	26	0.52	0.02	0.36	0.04	**238**	85	0.01	0.78
18	6	0.52	0.05	0.42	0.06	226	173	0.26	0.90
19	27	0.75	0.02	0.60	0.04	**239**	91	0.01	0.91
20	27	0.49	0.02	0.50	0.03	-8	66	0.90	0.94
21	27	0.61	0.02	0.47	0.05	232	127	0.08	0.77
22	13	0.55	0.03	0.44	0.08	211	217	0.35	0.69
23	13	0.49	0.03	0.39	0.03	**182**	79	0.04	0.95
24	27	0.55	0.02	0.40	0.03	**190**	69	0.01	0.85
25	8	0.72	0.04	0.38	0.15	793	438	0.12	0.45
26	24	0.70	0.02	0.67	0.04	83	121	0.50	0.92
27	28	0.66	0.02	0.53	0.03	**215**	74	0.01	0.92
28	25	0.59	0.02	0.38	0.04	**315**	99	0.00	0.76
29	34	0.57	0.02	0.30	0.03	**398**	79	0.00	0.70
31	14	0.89	0.03	0.84	0.06	74	124	0.56	0.94
32	28	0.36	0.02	0.30	0.04	90	83	0.28	0.63
33	23	0.39	0.02	0.27	0.04	119	73	0.12	0.66
34	12	0.62	0.03	0.64	0.09	-65	283	0.82	0.81
36	6	0.26	0.05	0.26	0.03	10	94	0.92	0.92
37	20	0.60	0.02	0.41	0.05	**326**	104	0.01	0.75
38	17	0.56	0.03	0.40	0.06	**140**	57	0.03	0.76

and measurement error. These have the well-known effect of lower slopes and higher intercepts than the true values. Therefore the calculated slopes are likely to be underestimates of the true infiltration factor. Only 4 of the 36 homes had slopes higher than the mean indoor/outdoor sulfur ratio. The overall average indoor/outdoor sulfur ratio is 0.56, compared to the overall average slope of 0.49.

Comparison of Methods for Calculating F_{inf}

The methods for calculating F_{inf} (regression vs. the simple indoor/outdoor ratio) are compared in Figure 3-4. The comparison differentiates between the 22 homes with intercepts not different from zero and the 14 homes with intercepts significantly different from zero. The latter set of homes cannot be said with complete confidence to have no indoor sources of sulfur, although random measurement errors could also cause nonzero intercepts. The slopes for these homes are generally lower, as would be expected if measurement errors are affecting the regressions. Home number 25 is an outlier in this graph (large red diamond), having the highest intercept of all homes, but with high uncertainty, due to a small number of samples (N = 8). Without home 25, the 21 remaining homes with intercepts not different from zero have slopes that are related to the mean indoor/outdoor ratios with a high R^2 of 0.91.

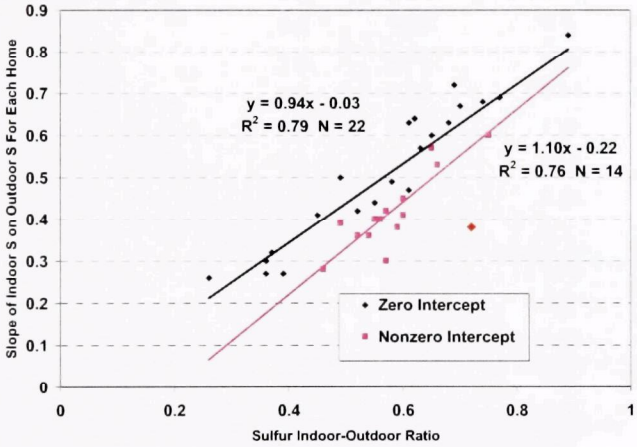

Figure 3-4. Comparison of the results of regressing indoor sulfur on outdoor sulfur with the simple indoor/outdoor ratio averaged over all visits to a home.

Estimating Indoor and Outdoor Contributions to Indoor $PM_{2.5}$

The indoor/outdoor sulfur ratio may be multiplied by the outdoor air concentration to estimate the contribution of outdoor air particles to the total indoor particle level for each home (Table 3-6). The indoor-generated particles are then the difference between the total indoor $PM_{2.5}$ and the outdoor contribution to indoor $PM_{2.5}$. The fractional contributions of outdoor air particles to indoor concentrations, averaged over all visits to each home, are shown in Figure 3-5. Fourteen homes had more than 80% of their indoor $PM_{2.5}$ concentrations supplied by outdoor air, and 7 homes had less than 40% supplied by outdoor air.

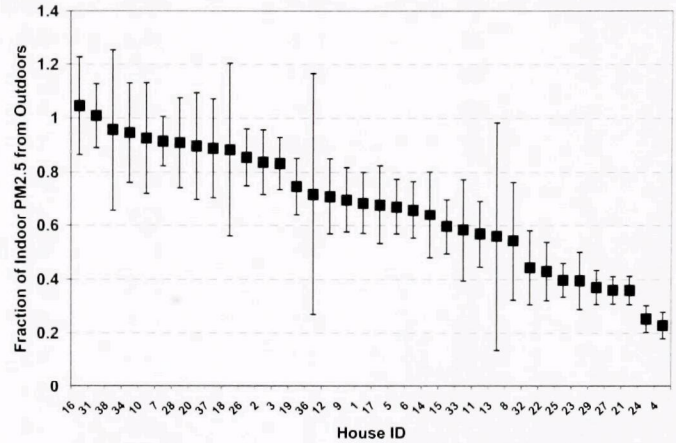

Figure 3-5. Estimates of the fractional contribution of outdoor particles to total indoor $PM_{2.5}$ concentrations, averaged over all home visits. Error bars are standard errors calculated by propagation of error.

The outdoor and indoor-generated contributions to total indoor $PM_{2.5}$, averaged over all visits to each house, are shown in Figure 3-6. The relative importance of these two sources varies widely, with some homes having essentially no indoor contribution and others having more than 50% indoor contributions.

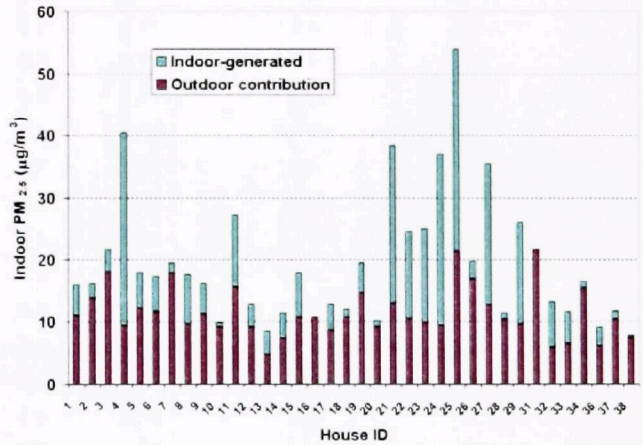

Figure 3-6. Comparison of average outdoor-generated and indoor-generated particles based on indoor/outdoor sulfur ratios.

The 95% confidence limits are displayed separately for outdoor and indoor contributions in Figures 3-7 and 3-8. (These values are taken from Table 3-6.) The indoor contributions cover a wider range than the outdoor contributions. For example, three homes had indoor sources producing an average concentration over the four seasons > 25 $\mu g/m^3$, compared to no homes with outdoor contributions that high. On the other hand, 20 homes had indoor contributions of 5 $\mu g/m^3$ or lower compared to only

Table 3-6. Estimated Annual Average Contributions of Outdoor and Indoor-Generated Particles to Total Indoor $PM_{2.5}$ Concentrations by House ($\mu g/m^3$)

House	N	Outdoor contribution to indoor particles	SD_{calc}	SE_{calc}	Indoor-generated particles	SD_{calc}	SE_{calc}
1	27	11.0	12.9	2.5	5.0	15.9	3.1
2	26	13.8	18.8	3.7	2.3	21.3	4.2
3	29	19.4	16.6	3.1	1.6	20.0	3.7
4	23	9.4	16.8	3.5	31.0	37.0	7.7
5	27	12.2	17.4	3.4	5.9	20.9	4.0
6	28	11.7	15.0	2.8	5.8	16.5	3.1
7	28	18.0	22.6	4.3	1.6	25.5	4.8
8	6	9.6	15.8	6.5	8.1	16.5	6.7
9	24	11.3	20.0	4.1	4.8	22.0	4.5
10	27	10.9	18.9	3.6	0.2	19.3	3.7
11	8	15.5	17.8	6.3	11.8	22.2	7.8
12	28	9.1	16.1	3.1	3.7	16.9	3.2
13	7	4.8	9.6	3.6	3.8	10.4	3.9
14	27	7.4	13.3	2.6	4.1	14.8	2.8
15	27	10.8	11.4	2.2	7.2	20.3	3.9
16	28	10.7	15.1	2.9	-0.5	15.6	3.0
17	26	8.7	15.4	3.0	4.1	18.6	3.6
18	6	10.7	11.9	4.8	1.4	12.2	5.0
19	27	17.2	20.4	3.9	3.3	21.5	4.1
20	27	10.8	19.8	3.8	-0.3	20.2	3.9
21	27	13.0	20.6	4.0	25.4	29.6	5.7
22	13	10.6	15.6	4.3	14.0	23.5	6.5
23	13	9.9	19.0	5.3	15.1	33.7	9.4
24	27	9.4	16.6	3.2	27.6	29.1	5.6
25	8	21.4	19.1	6.7	32.6	29.0	10.3
26	24	17.6	22.3	4.5	2.2	24.2	4.9
27	28	12.7	18.1	3.4	22.7	32.1	6.1
28	25	10.4	15.8	3.2	1.0	16.6	3.3
29	34	9.7	16.8	2.9	16.3	22.7	3.9
31	14	21.7	20.7	5.5	-0.2	23.2	6.2
32	28	5.8	12.3	2.3	7.3	13.6	2.6
33	23	6.4	18.0	3.7	5.1	20.0	4.2
34	12	15.4	16.7	4.8	1.3	17.3	5.0
36	6	6.2	17.4	7.1	3.0	17.5	7.2
37	20	10.4	10.4	2.3	1.3	10.9	2.4
38	17	7.4	8.3	2.0	0.3	8.8	2.1
Sum/ Mean	775	11.7	16.5	3.9	7.8	20.5	4.8

one home with an outdoor contribution that low. This reflects much more variability in indoor particle-generating activities compared with outdoor particle concentration variability.

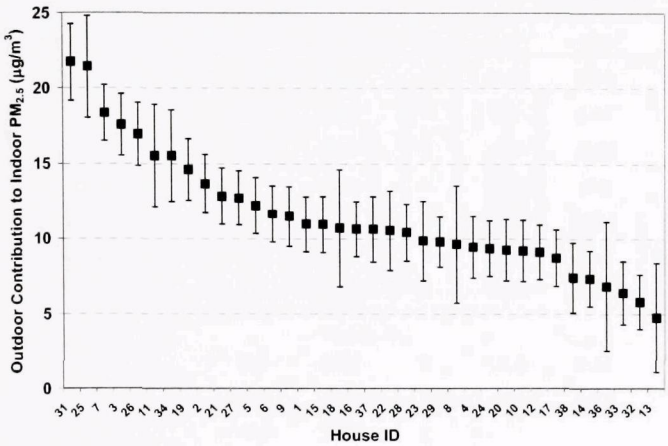

Figure 3-7. Estimates of average outdoor contribution to indoor $PM_{2.5}$. Error bars are standard errors calculated by propagation of error techniques applied to the three measurements required to estimate the outdoor contribution.

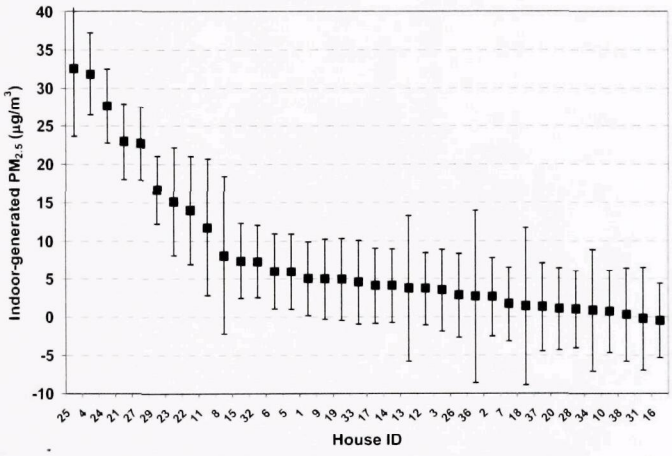

Figure 3-8. Estimates of average indoor-generated $PM_{2.5}$. Error bars are standard errors calculated by propagation of error techniques applied to the four measurements required to estimate indoor-generated $PM_{2.5}$.

We saw in Figure 3-3 that the indoor/outdoor sulfur ratio varied by season, and was much lower in summer than the other three seasons. We next look at the estimates of indoor and outdoor contributions by season (Table 3-7; Figures 3-9 through 3-12). Table 3-7 shows that indoor-generated particle concentrations were 45-46% of the total indoor particle concentrations averaged across all homes in the summer and fall seasons, but only 31% in the winter and spring seasons. This was due mostly to a reduction in indoor sources in the latter two seasons, since the outdoor contribution did not change greatly over the four seasons. Because of the small number of measurements in each house and each season (N = 1-7), the uncertainty, particularly in the estimates of indoor-generated concentrations, is much larger than for the equivalent concentrations for each home over all seasons (as in Table 3-6). The average standard error was 15% for the estimates of the outdoor contributions, but 66% for the indoor estimates.

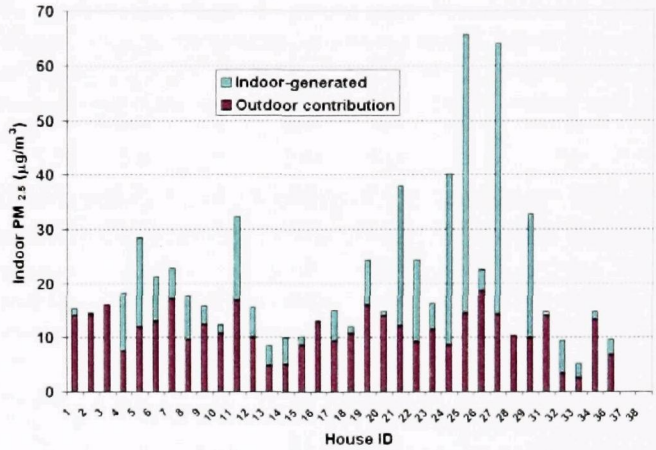

Figure 3-9. Indoor-outdoor average contributions to indoor $PM_{2.5}$. Summer 2000.

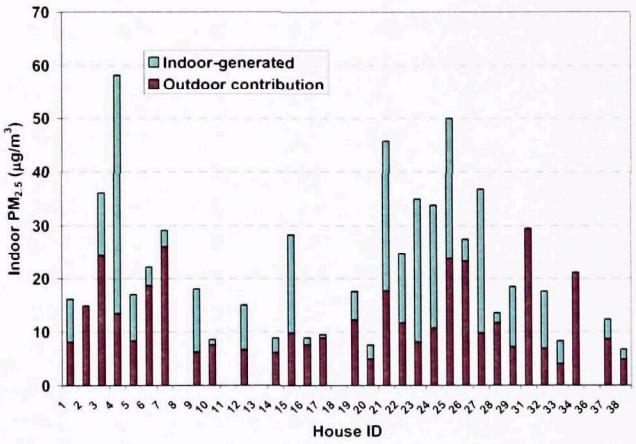

Figure 3-10. Indoor-outdoor average contributions to indoor $PM_{2.5}$. Fall 2000.

Table 3-7. Estimated Average Contributions of Outdoor Particles and Indoor-Generated Particles to Total Indoor Concentrations (μg/m^3) by House and by Season

	Summer 2000			Fall 2000			Winter 2001			Spring 2001		
House	N	Out	In	N	Out	In	N	Out	In	N	Out	In
1	8	14.0	1.6	6	8.1	8.1	6	10.8	12.5	2	8.7	0.7
2	6	14.2	0.5	4	14.9	-1.8	6	14.3	-1.9	7	12.0	11.3
3	7	16.2	-0.1	6	24.3	11.8	7	23.6	-6.8	7	12.9	2.0
4	2	7.5	10.9	7	13.5	44.8	7	8.0	43.9	4	6.9	11.7
5	6	12.0	16.5	6	8.3	8.7	7	16.6	-1.2			
6	7	12.9	8.5	5	18.7	3.5	7	9.3	8.2	7	7.8	3.2
7	7	17.3	5.6	7	26.0	3.1	6	15.0	-0.1	5	14.7	-1.8
8	6	9.6	8.1									
9	6	12.4	3.5	5	6.2	11.8	5	15.6	8.8	5	12.0	-1.3
10	6	10.7	1.8	7	7.5	1.1	7	17.3	-1.4	5	7.5	0.2
11	6	17.0	15.4							2	11.2	0.9
12	7	10.0	5.7	7	6.7	8.3	7	10.4	1.5	7	9.4	-0.7
13	7	4.8	3.8									
14	7	5.0	4.9	6	6.1	2.7	7	10.4	7.7	7	7.7	0.9
15	6	8.7	1.5	7	9.8	18.5	7	13.1	1.4	3	12.6	3.4
16	6	13.0	-0.4	7	7.6	1.3	7	11.4	-1.7	7	10.9	-1.0
17	7	9.4	5.6	5	8.9	0.6	6	9.7	1.2	1	2.8	2.5
18	6	10.7	1.4									
19	6	16.1	8.4	7	12.3	5.3	7	25.2	-2.4	7	15.2	2.8
20	7	13.9	1.0	6	5.0	2.6	7	16.0	-4.9	7	7.5	0.5
21	7	12.1	25.8	6	17.7	28.1	7	13.9	18.6	6	7.7	19.5
22	6	9.2	15.3	7	11.7	13.0						
23	7	11.4	5.1	6	8.1	26.9						
24	7	8.6	31.4	7	10.7	23.1	6	11.9	27.9	7	6.4	28.1
25	2	14.4	51.5	6	23.8	26.2						
26	6	18.6	4.1	6	23.3	4.2	5	21.5	-2.4	5	9.2	4.1
27	6	14.2	49.9	7	9.8	27.0	7	15.2	12.0	7	11.0	3.9
28	5	10.5	0.0	7	11.7	1.8	6	11.9	3.3	6	8.3	-0.9
29	13	9.8	23.0	6	7.2	11.3	7	12.9	18.1	5	8.9	9.7
31	7	13.9	0.8	7	29.5	-1.2						
32	7	3.3	6.1	7	6.9	10.7	7	6.7	6.4	7	6.4	6.1
33	5	2.6	2.7	5	4.0	4.3	6	10.1	3.0	4	9.1	4.2
34	7	13.1	1.6	3	21.1	-0.7						
36	5	6.9	2.7									
37				5	8.7	3.7	7	12.2	0.7	7	10.5	0.3
38				3	4.9	1.9	7	8.0	0.5	5	7.4	-0.1
Sum/Mean	216	11.3	9.5	186	12.4	10.0	171	13.5	5.9	142	9.4	4.2

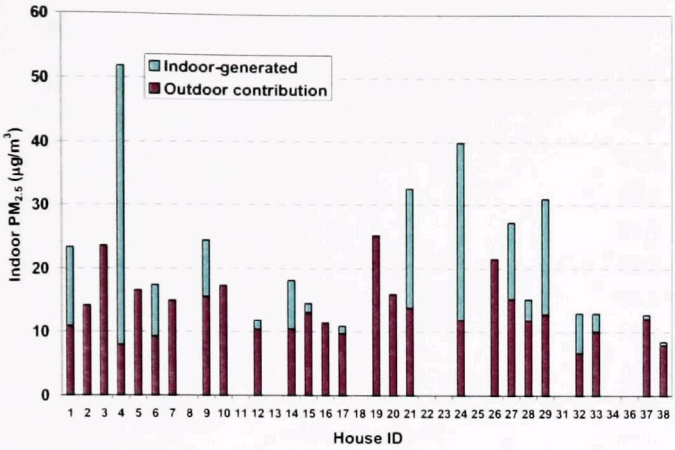

Figure 3-11. Indoor-outdoor average contributions to indoor $PM_{2.5}$. Winter 2001.

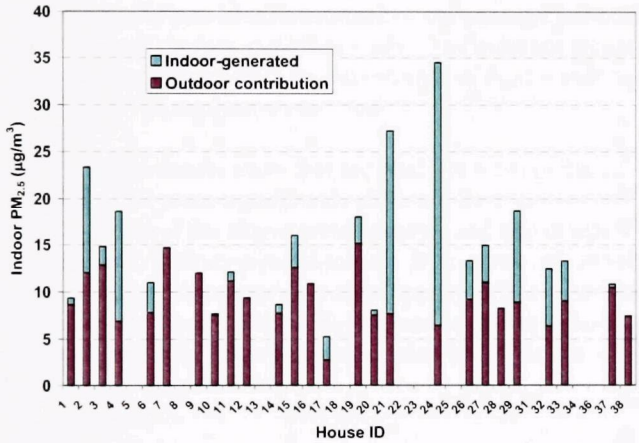

Figure 3-12. Indoor-outdoor average contributions to indoor $PM_{2.5}$. Spring 2001.

Relationship Between Outdoor Particles and Indoor Particles of Outdoor Origin

As described in the Introduction, a quantity of interest to epidemiologists is the relationship between outdoor particles and indoor particles of outdoor origin. Since epidemiologists work with measurements made at a central site, we examined how this relationship changes as we go from the particles measured just outside the home to those measured at the central site, and as we go from particles measured by the same type of equipment to those measured by different monitors. We first performed regressions for each home of the estimated contributions to indoor $PM_{2.5}$ made by particles of outdoor origin vs. the measured concentrations just outside the home (Table 3-8). These individual regressions by home have high R^2 values in most cases (median = 0.77; range 0.39-0.95). For all 752 valid daily measurements, the regression has a slope of 0.56 (0.01 SE) and a non-significant intercept of 0.31 (0.31) (Table 3-8). The Pearson correlation coefficient was 0.82, with an R^2 (adjusted) value of 0.665. (A Spearman rank correlation was also 0.82, indicating that the Pearson coefficient was not affected by outliers.) When the regression was run against the HI at the central site, the R^2 value was reduced to 0.58 (N = 736). Against the FRM at the central site, it was reduced to 0.49 (N = 775). We caution that these R^2 values must be considered upper bounds, because the assumptions of our model, which force the infiltrated particles of outdoor origin to be proportional to outdoor concentrations, virtually ensure that R^2 values will be high.

Estimating Contributions of Outdoor Air to Indoor Concentrations Using the RCS Model

Another way of estimating an average infiltration factor, this time using the $PM_{2.5}$ mass data, is the Random Component Superposition (RCS) model developed by Ott and co-workers (Ott et al., 2000). The RCS model simply assumes that a regression of all the measured indoor vs. outdoor mass concentrations will provide an overall average infiltration factor together with an estimate of the indoor source strength averaged across all homes (that being the constant term in the regression). The overall average infiltration factor estimated by this approach is 0.60 (0.06 SE, N = 774) (Figure 3-13), compared with the average indoor/outdoor sulfur ratio of 0.589 (0.006 SE, N = 775) from Table 3-1. The close agreement of these two values for infiltration factor, one based on particle concentrations alone and the other based on sulfur concentrations alone, is important evidence that using the sulfur results to estimate fine particle parameters is justified, at least for these overall averages. It should be noted that the RCS model determines only one average value of F_{inf} for all homes, whereas the methods discussed above provide estimates for individual homes. The RCS approach as applied to PM_{10} values in Riverside, CA, Philipsburg, NJ, and Toronto, Canada found similar values of 0.55, 0.60 and 0.61 for the infiltration factor (Ott et al., 2000) compared to our values using $PM_{2.5}$ sulfur and mass data. Further evidence for the applicability of the sulfur results to the $PM_{2.5}$ fraction is the close agreement between the RCS estimate of 7.7 µg/m^3 for the indoor-generated $PM_{2.5}$ compared to 7.8 µg/m^3 estimated using the sulfur indoor-outdoor ratios (Table 3-6).

Table 3-8. Regressions of Indoor $PM_{2.5}$ of Outdoor Origin on Outdoor $PM_{2.5}$ Measured at the Home

House	N	Slope	SE	Intercept	SE	p	R^2
1	27	0.53	0.03	1.47	0.65	0.03	0.91
2	26	0.45	0.05	3.06	1.16	0.01	0.79
3	24	0.62	0.04	2.34	0.99	0.03	0.93
4	23	0.35	0.05	1.71	1.18	0.16	0.69
5	27	0.67	0.09	-0.56	1.81	0.76	0.69
6	28	0.63	0.03	-0.35	0.68	0.61	0.93
7	28	0.81	0.05	-0.73	1.25	0.56	0.91
8	6	0.22	0.08	3.55	2.14	0.17	0.61
9	24	0.52	0.06	1.05	1.31	0.43	0.76
10	22	0.37	0.04	2.81	0.86	0.00	0.77
11	8	0.67	0.10	0.26	2.46	0.92	0.85
12	28	0.32	0.04	3.26	0.71	0.00	0.75
13	7	0.31	0.04	0.67	0.64	0.34	0.88
14	27	0.41	0.04	0.47	0.68	0.50	0.82
15	27	0.66	0.07	0.55	1.15	0.64	0.77
16	28	0.60	0.03	0.74	0.52	0.17	0.94
17	26	0.41	0.06	1.69	1.18	0.16	0.62
18	6	0.40	0.10	2.32	2.15	0.34	0.75
19	22	0.57	0.07	3.13	1.40	0.04	0.78
20	22	0.52	0.04	-0.30	0.75	0.69	0.90
21	27	0.58	0.07	0.47	1.58	0.77	0.74
22	13	0.38	0.11	2.95	2.20	0.21	0.51
23	13	0.38	0.04	1.88	0.90	0.06	0.89
24	27	0.47	0.06	1.03	1.21	0.40	0.68
25	8	0.78	0.33	-1.59	9.98	0.88	0.39
26	21	0.69	0.05	0.26	1.36	0.85	0.90
27	28	0.52	0.04	2.49	0.85	0.01	0.86
28	25	0.37	0.06	3.58	1.24	0.01	0.59
29	34	0.27	0.05	4.62	1.10	0.00	0.43
31	14	0.74	0.05	2.86	1.40	0.06	0.95
32	28	0.31	0.05	0.72	0.92	0.44	0.56
33	23	0.38	0.06	-0.07	1.13	0.95	0.66
34	12	0.59	0.15	0.53	3.80	0.89	0.58
36	6	0.26	0.05	-0.14	1.23	0.91	0.85
37	20	0.43	0.06	2.86	1.16	0.02	0.71
38	17	0.52	0.05	0.45	0.67	0.51	0.89
Sum/Mean	752	0.56	0.01	0.31	0.31	0.32	0.67

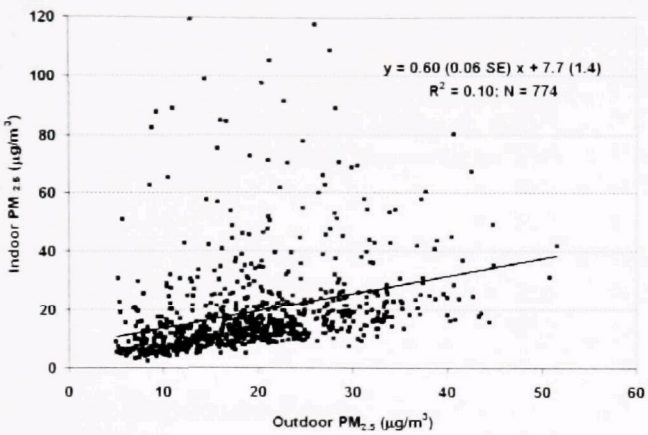

Figure 3-13. Regression of indoor PM2.5 on residential outdoor PM2.5 data.

Many studies have $PM_{2.5}$ measurements indoors and outdoors but do not have sulfur or sulfate measurements to determine F_{inf}. The RCS model provides an estimate of the *average* infiltration factor across all homes in a given study. The question arises: Is it possible to apply the RCS model estimate of F_{inf} for *individual* homes from PM measurements alone?

The regression results show that only half (N = 18) of the homes had slopes that were significantly different from zero (Table 3-9). Also, three of the slopes were negative and two others were greater than 1, both unphysical results. By contrast, all 36 sulfur regressions had slopes that were significantly different from zero Comparing the slopes of the $PM_{2.5}$ regressions to the estimates of F_{inf} from the sulfur indoor/outdoor ratios resulted in a low R^2 value of 0.11 (Figure 3-14).

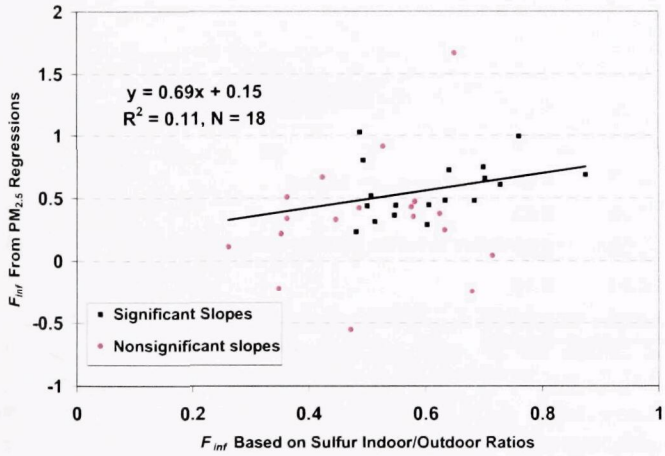

Figure 3-14. Estimates of the infiltration factor F_{inf} from sulfur indoor/outdoor ratios compared to regressions of indoor vs. outdoor fine particles. The regression line shown is for the 18 cases with slopes significantly different from zero.

Estimates of the Contribution of Outdoor Air Particles to Personal Exposure

If total personal exposure E to fine particles is the sum of exposures while indoors and exposures while outdoors, then the following equation holds:

$$E = f_{in}\, C_{in} + f_{out}\, C_{out} \qquad (3\text{-}2)$$

where f_{in} and f_{out} are the fractions of time spent indoors and outdoors.

Exposure can also be expressed as the sum of outdoor and non-outdoor sources:

$$E = E_o + E_{no} \qquad (3\text{-}3)$$

where E_o is the exposure due to outdoor sources and E_{no} is the exposure due to non-outdoor sources. The exposure due to outdoor sources may be written as

$$E_o = f_{in}\, F_{inf}\, C_{out} + f_{out}\, C_{out} \qquad (3\text{-}4)$$

Now we assume that all of the time spent indoors was spent indoors at home where a measurement of outdoor concentration is available, and all of the time spent both outdoors and in transit is considered to involve exposure to the outdoor concentration. This outdoor concentration may be that measured at the home or at the central site if that is available. In this case, the fractions f_{out} and f_{in} add up to 1. We now define a factor F_{pex} such that

$$E = F_{pex}\, C_{out} + E_{no} \qquad (3\text{-}5)$$

The factor F_{pex} plays a role with respect to personal exposure that F_{inf} played with respect to indoor concentrations. Some investigators have labeled F_{pex} as an "attenuation factor," marking the decrease in the effect, or attenuation, of outdoor concentrations on personal exposure. However, this would imply that attenuation increases as F_{pex} increases, whereas the reverse is true. We shall call F_{pex} the "outdoor exposure factor," indexing the contribution of outdoor particles to personal exposure. From Equations 3-3 and 3-4, we have a relationship for the outdoor exposure factor F_{pex}:

$$F_{pex} = f_{in} F_{inf} + f_{out} \qquad (3\text{-}6)$$

Rewriting the equation as

$$F_{pex} = F_{inf} + f_{out}\,(1 - F_{inf}) \qquad (3\text{-}7)$$

we see that F_{pex} is always larger than F_{inf}, although the difference is small if the fraction f_{out} is small, as it usually is.

Table 3-9. Regressions of Indoor on Outdoor $PM_{2.5}$ by House

House	N	Slope	SE	p-level	Intercept	SE	p-level	R^2
1	27	0.37	0.21	0.09	9.4	4.1	0.03	0.08
2	26	0.43	0.22	0.07	6.0	5.6	0.30	0.10
3	24	0.61	0.23	0.01	6.4	6.2	0.31	0.21
4	23	0.67	0.76	0.39	26.0	17.8	0.16	0.00
5	27	0.72	0.24	0.01	4.5	5.0	0.38	0.23
6	28	0.44	0.17	0.02	9.0	3.5	0.02	0.18
7	28	0.99	0.16	0.00	-3.4	3.9	0.39	0.59
8	6	-0.22	0.26	0.45	23.9	7.5	0.03	0.00
9	27	0.35	0.19	0.08	9.4	4.0	0.03	0.08
10	22	0.31	0.05	0.00	4.5	0.9	0.00	0.67
11	9	-0.25	0.68	0.73	34.4	15.6	0.06	0.00
12	28	0.23	0.11	0.04	8.6	2.1	0.00	0.12
13	7	0.51	0.25	0.09	1.8	3.5	0.64	0.35
14	27	0.33	0.18	0.08	6.0	3.3	0.08	0.08
15	28	0.04	0.51	0.94	17.5	8.9	0.06	0.00
16	28	0.48	0.05	0.00	2.1	0.9	0.03	0.76
17	27	0.80	0.24	0.00	-1.1	4.4	0.81	0.29
18	21	0.52	0.17	0.01	5.9	3.5	0.11	0.30
19	22	0.66	0.12	0.00	6.4	2.6	0.02	0.58
20	22	0.44	0.04	0.00	2.3	0.8	0.01	0.85
21	28	0.47	0.38	0.23	28.2	8.9	0.00	0.02
22	13	0.91	0.69	0.22	6.6	14.5	0.66	0.06
23	13	-0.55	0.85	0.53	36.6	19.6	0.09	0.00
24	27	0.42	0.62	0.51	29.6	11.9	0.02	0.00
25	8	1.66	1.18	0.21	4.6	35.8	0.90	0.12
26	21	0.75	0.14	0.00	1.8	3.7	0.64	0.58
27	28	0.24	0.60	0.69	30.6	13.0	0.03	0.00
28	25	0.36	0.12	0.01	4.8	2.4	0.06	0.24
29	34	1.03	0.27	0.00	6.8	5.5	0.22	0.29
31	14	0.69	0.11	0.00	4.0	3.0	0.22	0.76
32	28	0.22	0.18	0.24	9.6	3.2	0.01	0.02
33	23	0.34	0.19	0.08	5.8	3.6	0.12	0.09
34	12	0.48	0.16	0.01	4.7	4.1	0.28	0.43
36	6	0.11	0.10	0.31	6.4	2.4	0.06	0.07
37	20	0.29	0.12	0.03	6.6	2.3	0.01	0.19
38	17	0.44	0.06	0.00	1.8	0.9	0.05	0.78

Several articles have discussed this idea that personal exposure to particles of outdoor origin may be greater than indoor concentrations of such particles (Wilson and Suh, 1997; Wilson, Mage, and Grant 1998). The central idea is that persons spend some time outdoors or in vehicles where they are exposed to the full outdoor concentrations, thus adding to their exposures indoors where they are "protected" somewhat by the infiltration factor F_{inf}.

As an example, suppose f_{in} = 0.89, as found by the NHAPS study, and f_{out} = {fraction of time outdoors (6%) plus fraction of time in vehicles (5%)} = 0.11. Setting F_{inf} = 0.60 as found by the RCS model, we find that F_{pex} would be 0.64, an increase of 7%.

Outdoor Exposure Factor F_{pex} Estimated Using PM Measurements

A regression of personal $PM_{2.5}$ on outdoor $PM_{2.5}$ concentrations provides an estimate of F_{pex} averaged across all participants: 0.64 $\pm$ 0.01 SE (Figure 3-15). However, the fit is poor (R^2 = 0.09). The intercept in this case (11.6 $\pm$ 1.4 μg/m^3) is an estimate of E_{no}, the average exposure due to non-outdoor sources. The difference between the indoor source contributions (estimated at 8.0 μg/m^3 in Table 3-1) and the non-outdoor source contribution is sometimes attributed to the "personal cloud," which in this case would equal 3.6 $\pm$ 2.0 μg/m^3.

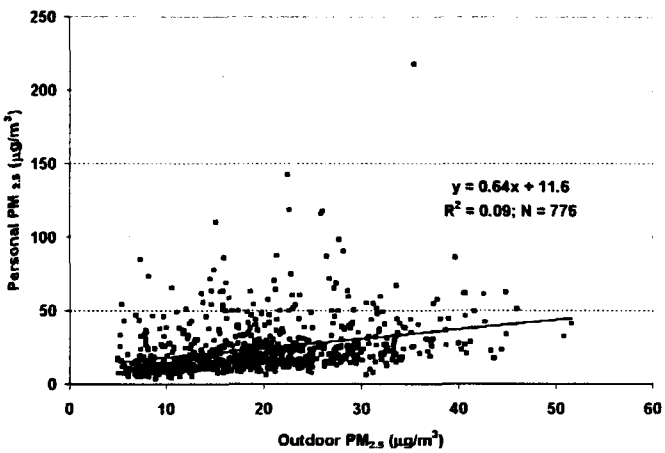

Figure 3-15. 24-h average fine particle personal exposures vs. outdoor air concentrations.

One outlier in Figure 3-15 (personal $PM_{2.5}$ > 200 μg/m^3) has a strong influence on the slope of the line. If the outlier is removed, the slope decreases from 0.64 to 0.59 (Figure 3-16), while the intercept increases from 11.6 to 12.3. This would increase the personal cloud from 3.6 to 4.3 μg/m^3. The similarity of the slopes for the personal vs. outdoor and indoor vs. outdoor regressions (0.59-0.64 compared to 0.61) suggests that the time spent indoors drives the relationship between personal exposure and outdoor concentrations. That is, the infiltration factor F_{inf}, which governs the reduction of particle concentrations as they enter a house, is very similar to the outdoor exposure factor F_{pex}, which governs the reduction in outdoor concentrations contributing to personal exposure.

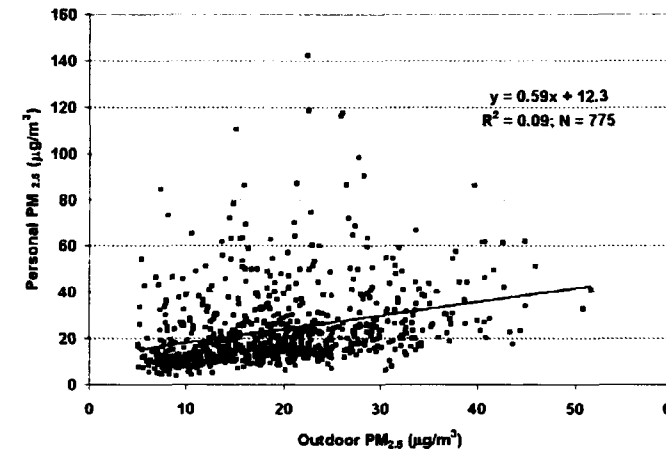

Figure 3-16. Personal vs. outdoor $PM_{2.5}$; one outlier removed.

Estimating the Outdoor Exposure Factor F_{pex} Using Sulfur Measurements

Another estimate of the outdoor exposure factor is given by the regression of personal sulfur exposures vs. outdoor sulfur concentrations (Figure 3-17). The slope is 0.49, identical to the slope of the indoor/outdoor sulfur regression (Figure 3-1), while the intercept of 90 ng/m^3 is less than the indoor/outdoor intercept of 150 ng/m^3. The estimate using sulfur (R^2=0.78) is preferred to the estimate using $PM_{2.5}$ (R^2=0.09).

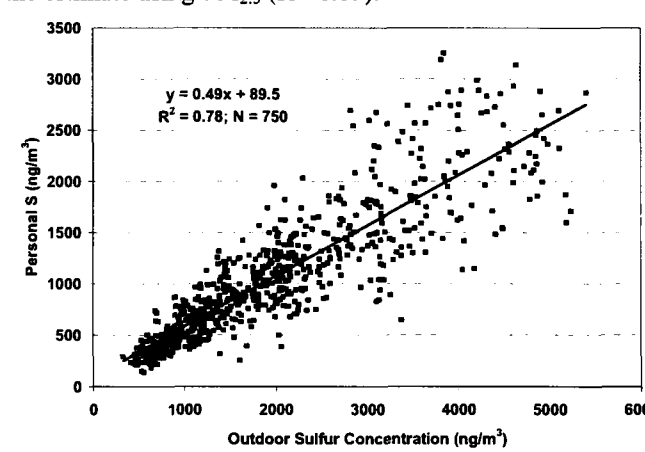

Figure 3-17. Personal vs. outdoor sulfur.

Comparison of F_{pex} and F_{inf}

Both the $PM_{2.5}$ and sulfur regressions fail to support the argument based on Equation 3-7 that F_{pex} is larger than F_{inf}. One possibility is that time spent in office buildings, which usually recirculate a portion of the air through filters, will involve lower exposures to sulfur than time spent in homes without such recirculation and refiltration. Some evidence for this can be found in the PTEAM study, where persons who worked away

from home had lower particle exposures than those who stayed home (Ozkaynak et al., 1996). In that case, the proper equation for total exposure would have to take account of the different concentrations in homes and offices:

$$E = f_{inhome}\, C_{inhome} + f_{out}\, C_{out} + f_{inoffice}\, C_{inoffice} \qquad (3\text{-}8)$$

where the relationship $C_{inoffice} < C_{inhome}$ would hold. This will reduce the magnitude of the outdoor factor.

Comparison of Indoor-Outdoor Sulfur and Personal Sulfur Measurements

Since the personal samples were collected using the 2 Lpm PEM, but the indoor and outdoor samples were collected using the 20 Lpm HI, we need to address the question of how the two samplers agree. During the summer season, 166 co-located indoor and outdoor samples were collected. As shown in Figures 3-18 and 3-19, the agreement was excellent ($R^2 = 0.97$ for both), with the slopes near 1 for both. This agreement between different monitor types is justification for comparing the indoor/outdoor sulfur ratio (the infiltration factor F_{inf}) with the personal/outdoor sulfur ratio (the outdoor exposure factor F_{pex}).

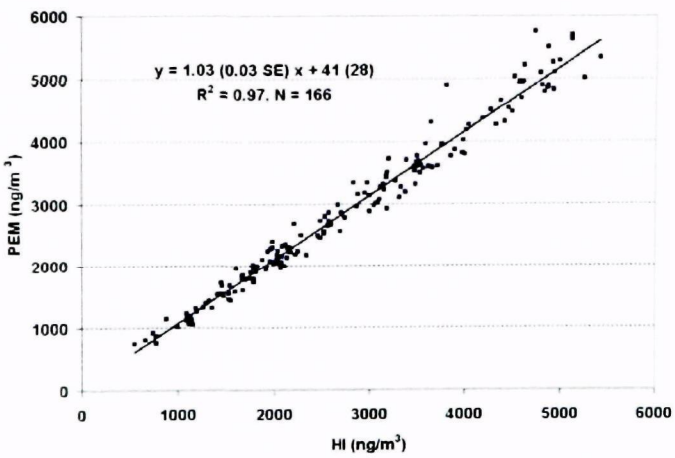

Figure 3-18. Co-located $PEM_{2.5}$ and $HI_{2.5}$ sulfur concentrations outdoors. Summer 2000.

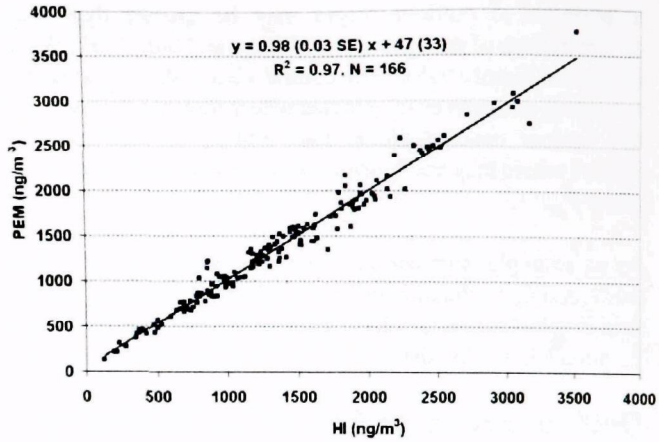

Figure 3-19. Co-located $PEM_{2.5}$ and $HI_{2.5}$ sulfur concentrations indoors. Summer 2000.

The overall comparison shows that personal sulfur exposures were somewhat smaller than indoor concentrations at home (Table 3-10). When compared on a home-by-home basis, only 33% of the participants had sulfur exposures higher than indoor concentrations (Table 3-11; Figure 3-20). The ratio of the mean personal sulfur concentration averaged over all visits to a home to the mean outdoor sulfur concentration (F_{pex}) is 0.54 (Table 3-12).

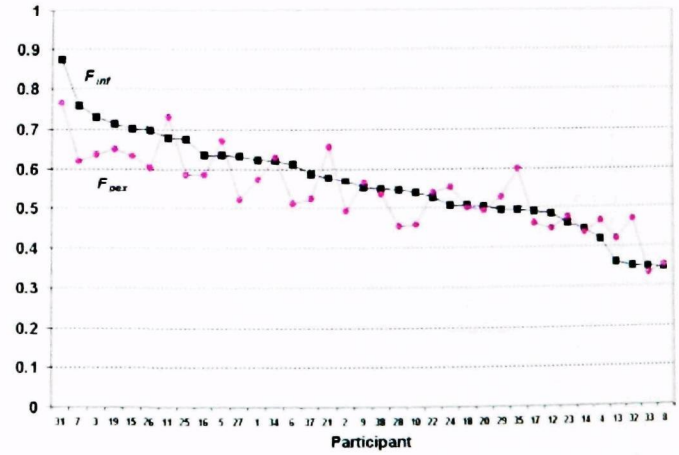

Figure 3-20. Comparison of infiltration factor F_{inf} and outdoor exposure factor F_{pex} by participant.

Finally, only 32% of 117 cases compared by home and season had higher personal sulfur exposures than indoor sulfur levels (Table 3-13). Only in the summer season was the average exposure higher than the average indoor concentration, but only by 1%. 18 of 35 homes had higher exposures than indoor concentrations in the summer, compared to only 8, 4, and 8 cases in the other seasons. In most cases, F_{pex} was less than F_{inf}, contrary to the assumption that personal exposures to particles of outdoor origin would be larger than indoor concentrations due to the time spent outdoors. This supports the idea that indoor concentrations in buildings, where our participants spent some of

Table 3-10. Sulfur Concentrations (ng/m^3) and Ratios in Matched Personal, Indoor, and Outdoor Samples

Concentrations/ratios	N	Mean	SD	Min	10th	25th	Median	75th	90th	Max
Personal	780	1062	632	137	393	556	915	1419	1975	3254
Indoor	780	1116	655	123	423	621	957	1442	2039	3852
Outdoor	750	1944	1140	323	755	1037	1671	2592	3647	5406
Indoor/outdoor	745	0.59	0.17	0.17	0.38	0.48	0.58	0.69	0.80	1.08
Personal/outdoor	750	0.55	0.14	0.16	0.38	0.46	0.54	0.63	0.73	1.08
Personal/indoor	780	0.97	0.23	0.28	0.75	0.83	0.92	1.03	1.21	2.64

Table 3-11. Personal and Indoor Sulfur Concentrations (ng/m^3) by Subject

Subject	N	Personal	SE	Indoor	SE	Pers/In	SE
1	27	854	71	932	74	0.91	0.01
2	24	910	86	1058	86	0.84	0.02
3	26	1338	108	1508	108	0.88	0.02
4	25	934	105	853	72	1.08	0.07
5	26	1254	141	1196	145	1.07	0.04
6	27	1082	117	1281	127	0.85	0.02
7	27	1210	155	1488	176	0.79	0.02
8	7	1256	165	1234	145	1.01	0.04
9	28	1124	141	1130	128	0.97	0.04
10	27	940	105	1029	106	0.90	0.02
11	10	1671	264	1542	251	1.08	0.06
12	27	892	85	959	87	0.93	0.02
13	7	643	112	553	85	1.16	0.10
14	26	707	69	720	66	0.97	0.02
15	27	841	80	950	87	0.88	0.02
16	27	951	108	1028	108	0.91	0.01
17	26	878	93	976	103	0.91	0.04
18	7	1398	189	1421	177	0.97	0.04
19	28	1464	139	1597	146	0.91	0.02
20	28	1104	128	1120	120	0.97	0.02
21	28	1369	149	1211	134	1.12	0.04
22	14	1329	189	1285	165	1.01	0.04
23	13	1209	213	1175	167	0.99	0.04
24	25	1012	119	880	98	1.15	0.06
25	8	1686	246	1827	240	0.91	0.03
26	24	1562	175	1833	206	0.85	0.02
27	28	1101	106	1335	133	0.84	0.02
28	27	896	88	1079	97	0.82	0.02
29	28	996	109	989	88	0.99	0.06
31	14	1469	161	1678	193	0.88	0.02
32	28	811	65	607	54	1.43	0.08
33	25	485	54	524	65	0.97	0.04
34	12	1692	224	1746	233	0.97	0.02
35	6	1056	170	997	154	1.05	0.03
36	6	1143	249	658	123	1.77	0.17
37	20	940	82	1079	73	0.86	0.04
38	17	511	41	534	33	0.96	0.04
Mean	780	1101	132	1135	126	0.99	0.04

Table 3-12. Ratios of the Mean Personal Sulfur Exposure to the Mean Outdoor Sulfur Concentration (F_{pex}) by Subject

Subject	N	S_{pers}/S_{out}	SD[b]	SE[b]
1	25	0.57	0.34	0.07
2	23	0.49	0.33	0.07
3	21	0.64	0.37	0.08
4	23	0.46	0.37	0.08
5	26	0.67	0.54	0.11
6	27	0.51	0.38	0.07
7	27	0.62	0.58	0.11
8	6	0.35	0.18	0.08
9	25	0.57	0.54	0.11
10	21	0.46	0.44	0.10
11	8	0.73	0.53	0.19
12	27	0.45	0.34	0.06
13	7	0.42	0.26	0.10
14	26	0.44	0.30	0.06
15	27	0.63	0.42	0.08
16	27	0.59	0.49	0.09
17	25	0.46	0.38	0.08
18	6	0.50	0.27	0.11
19	22	0.65	0.55	0.12
20	22	0.49	0.45	0.10
21	28	0.66	0.57	0.11
22	13	0.54	0.40	0.11
23	14	0.47	0.44	0.12
24	23	0.55	0.47	0.10
25	14	0.59	0.36	0.10
26	21	0.60	0.50	0.11
27	28	0.52	0.41	0.08
28	25	0.45	0.36	0.07
29	27	0.53	0.47	0.09
31	14	0.77	0.46	0.12
32	28	0.47	0.29	0.05
33	22	0.33	0.29	0.06
34	13	0.63	0.37	0.10
35	27	0.60	0.42	0.08
36	6	0.45	0.32	0.13
37	21	0.52	0.29	0.06
38	20	0.53	0.33	0.07
Sum/Mean	765	0.54[a]	0.40	0.09

[a] Ratio of the mean personal sulfur concentration to the mean outdoor sulfur concentration. The mean of the ratios is also 0.54.
[b] Standard Deviations (SD) and Standard Errors (SE) Calculated by Error Propagation.

Table 3-13. Comparison of Personal/Outdoor, Indoor/Outdoor, and Personal/Indoor Sulfur Ratios by House and by Season

	Summer				Fall				Winter				Spring			
House	N	P/O[a]	I/O[b]	P/I[c]	N	P/O	I/O	P/I	N	P/O	I/O	P/I	N	P/O	I/O	P/I
1	6	0.50	0.57	0.88	6	0.58	0.63	0.93	7	0.60	0.67	0.89	6	0.63	0.64	0.99
2	6	0.46	0.57	0.81	6	0.48	0.51	0.93	6	0.48	0.63	0.76	5	0.59	0.66	0.90
3	5	0.72	0.83	0.86	7	0.64	0.70	0.91	7	0.59	0.67	0.88	2	0.59	0.74	0.79
4	2	0.41	0.28	1.45	7	0.46	0.47	0.98	7	0.48	0.48	1.00	7	0.48	0.39	1.25
5	6	0.57	0.50	1.13	6	0.66	0.66	1.01	7	0.66	0.72	0.92	7	0.80	0.74	1.09
6	6	0.43	0.57	0.75	7	0.57	0.63	0.89	7	0.50	0.59	0.85	7	0.53	0.61	0.87
7	6	0.54	0.67	0.81	7	0.70	0.84	0.82	7	0.58	0.76	0.77	7	0.71	0.85	0.84
8	6	0.35	0.35	1.02												
9	6	0.47	0.48	0.97	7	0.58	0.69	0.84	7	0.57	0.60	0.94	5	0.70	0.63	1.12
10	5	0.45	0.45	1.00	7	0.44	0.57	0.77	7	0.50	0.59	0.86	2	0.47	0.50	0.93
11	6	0.74	0.68	1.09					2	0.60	0.67	0.90				
12	7	0.39	0.40	0.98	7	0.50	0.56	0.89	7	0.56	0.62	0.91	6	0.43	0.46	0.93
13	7	0.42	0.36	1.16												
14	7	0.39	0.39	1.02	6	0.44	0.46	0.96	6	0.45	0.47	0.95	7	0.46	0.46	0.99
15	6	0.60	0.58	1.04	7	0.63	0.74	0.85	7	0.64	0.77	0.82	7	0.66	0.78	0.85
16	6	0.57	0.57	0.99	7	0.65	0.72	0.91	7	0.55	0.67	0.82	7	0.57	0.62	0.92
17	6	0.45	0.42	1.08	6	0.38	0.48	0.80	7	0.47	0.56	0.85	6	0.57	0.58	0.98
18	6	0.50	0.51	0.99												
19	6	0.59	0.62	0.96	7	0.71	0.79	0.90	7	0.71	0.82	0.86	2	0.66	0.68	0.97
20	7	0.54	0.53	1.01	6	0.44	0.43	1.01	7	0.41	0.47	0.86	2	0.52	0.51	1.02
21	7	0.55	0.43	1.27	7	0.74	0.70	1.05	7	0.71	0.68	1.05	7	0.75	0.67	1.13
22	6	0.46	0.43	1.08	7	0.63	0.64	0.98								
23	7	0.47	0.43	1.10	7	0.48	0.58	0.83								
24	7	0.54	0.41	1.32	6	0.52	0.51	1.02	6	0.60	0.57	1.06	4	0.64	0.62	1.04
25	7	0.51	0.56	0.92	7	0.71	0.80	0.89								
26	6	0.52	0.62	0.84	6	0.69	0.78	0.88	7	0.56	0.68	0.81	2	0.74	0.78	0.95
27	7	0.45	0.54	0.83	7	0.61	0.77	0.80	7	0.58	0.66	0.88	7	0.54	0.67	0.81
28	5	0.39	0.40	0.97	7	0.51	0.66	0.77	7	0.49	0.61	0.80	6	0.47	0.60	0.78
29	14	0.42	0.39	1.07	6	0.62	0.59	1.06	7	0.56	0.68	0.82	7	0.62	0.69	0.90
31	7	0.91	0.96	0.95	7	0.67	0.82	0.82								
32	7	0.49	0.25	1.95	7	0.51	0.40	1.29	7	0.56	0.48	1.18	7	0.37	0.30	1.22
33	4	0.25	0.20	1.27	6	0.39	0.38	1.01	5	0.47	0.50	0.95	7	0.29	0.37	0.80
34	7	0.52	0.54	0.97	6	0.75	0.84	0.89								
36	6	0.45	0.26	1.74												
37					7	0.52	0.65	0.80	7	0.54	0.67	0.81	7	0.51	0.53	0.96
38					6	0.54	0.54	1.00	7	0.56	0.63	0.90	7	0.51	0.50	1.03
Sum/Mean	215	0.50	0.49	1.01	205	0.58	0.64	0.91	179	0.55	0.62	0.89	146	0.56	0.59	0.95

[a] Personal/outdoor sulfur concentration ratio (outdoor exposure factor)
[b] Indoor/outdoor sulfur concentration ratio (infiltration factor)
[c] Personal/indoor sulfur concentration ratio

their time, may be lower than in homes, due to recirculation and filtration of outdoor air. The fact that the summer season gave similar estimates for personal exposure and indoor concentrations of sulfur is additional evidence for this hypothesis, since air conditioning was in general use in summer and would provide extra filtration for the indoor sulfur particles.

Use of the Outdoor Exposure Factor F_{pex} to Calculate the Contribution to Personal Exposure Made by Particles of Outdoor Origin

We next used our estimated outdoor exposure factors F_{pex} for each subject (based on the sulfur personal/outdoor ratio) to calculate the contribution of particles of outdoor origin to personal exposure (Table 3-14; Figures 3-21 through 3-23). On average, particles of outdoor origin contributed 10.9 μg/m^3 (47%) of the total personal exposure compared with 12.5 μg/m^3 (53%) for the particles of non-outdoor origin. The range of the non-outdoor-generated particle contributions (5-33 μg/m^3) was much greater than the range of the outdoor-generated contributions (6-19 μg/m^3).

The contribution of particles of non-outdoor origin to personal exposure can be subdivided into particles generated indoors while the person is at home and particles from all other sources.

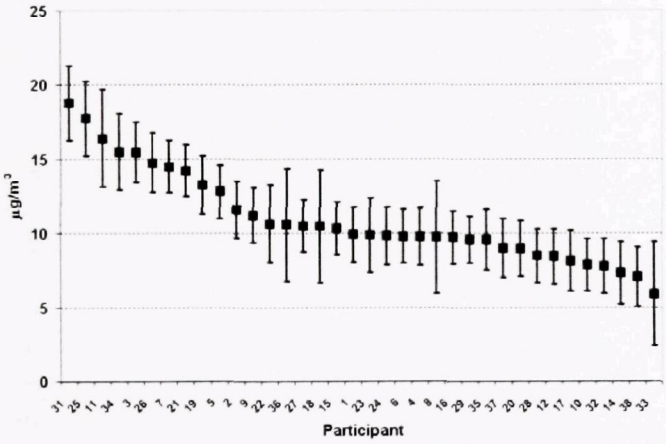

Figure 3-21. Outdoor contributions to personal exposure. Error bars are standard errors calculated by propagation of error.

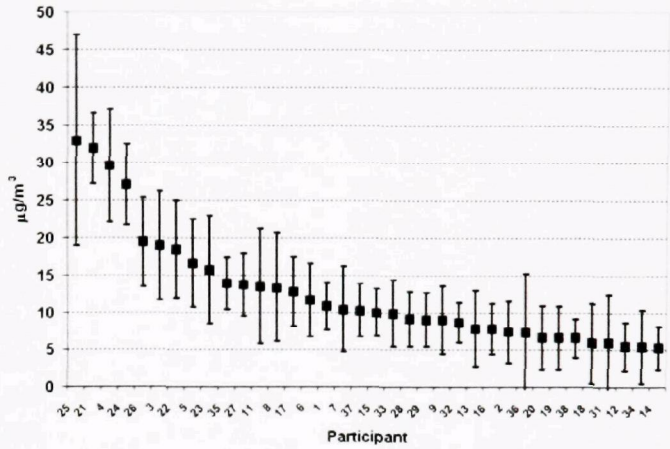

Figure 3-22. Non-outdoor contributions to personal exposure. Error bars are standard errors calculated by propagation of error.

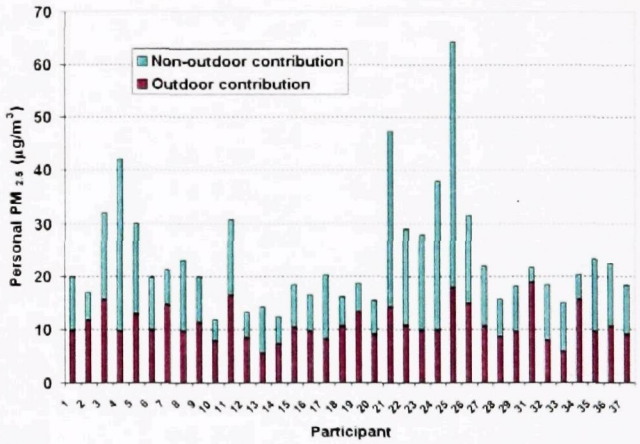

Figure 3-23. Outdoor and non-outdoor contributions to personal $PM_{2.5}$ exposure.

The particles from other sources include particles generated while the person is indoors at other locations or in a vehicle, as well as particles from the "personal cloud" that may be generated throughout the day at all locations (Yakovleva et al., 1999). The indoor-generated particle concentrations estimated from the F_{inf} values can be multiplied by the fraction of time spent indoors at home (from the activity logs) to calculate the contribution of indoor-generated particles to personal exposure. The outdoor contribution is calculated by multiplying the outdoor concentration times the fraction of time spent outdoors. The contribution of indoor-generated particles at all other indoor locations as well as the particles from the personal cloud is then found by subtracting the sum of the particles of outdoor origin and the indoor-generated particles while at home from the total personal exposures (Table 3-15; Figure 3-24).

Table 3-14. Estimated Contribution of Outdoor Particles to Personal Exposure (μg/m^3). Standard Deviations and Standard Errors Calculated by Propagation of Error

Subject	N	Outdoor contribution to personal exposure	SD	SE	Nonoutdoor contribution to personal exposure	SD	SE
1	25	9.9	13.3	2.7	10.9	16.2	3.2
2	23	11.6	17.9	3.7	7.4	20.3	4.2
3	21	15.5	17.4	3.8	19.0	33.1	7.2
4	23	9.8	19.4	4.0	29.6	36.1	7.5
5	26	12.8	17.2	3.4	16.6	29.8	5.8
6	27	9.8	15.9	3.1	11.7	25.6	4.9
7	27	14.5	23.8	4.6	10.4	29.7	5.7
8	6	9.7	16.5	6.7	13.4	17.8	7.3
9	25	11.2	20.5	4.1	9.0	22.8	4.6
10	21	7.9	18.9	4.1	5.1	19.7	4.3
11	8	16.4	17.9	6.3	13.5	21.9	7.8
12	27	8.4	16.3	3.1	5.5	16.9	3.2
13	7	5.6	10.0	3.8	7.9	13.7	5.2
14	26	7.3	13.3	2.6	5.2	14.6	2.9
15	27	10.3	12.6	2.4	10.0	16.7	3.2
16	27	9.7	15.8	3.0	7.8	17.8	3.4
17	25	8.2	16.1	3.2	12.8	23.5	4.7
18	6	10.5	12.4	5.1	5.9	13.2	5.4
19	22	13.3	18.7	4.0	6.6	20.1	4.3
20	22	9.0	19.1	4.1	6.7	20.4	4.3
21	28	14.2	20.8	3.9	31.9	24.9	4.7
22	13	10.6	16.3	4.5	18.4	23.5	6.5
23	14	9.9	21.2	5.7	15.7	26.9	7.2
24	23	9.8	16.8	3.5	27.1	26.2	5.5
25	14	17.8	20.1	5.4	33.0	52.7	14.1
26	21	14.8	22.6	4.9	19.5	27.1	5.9
27	28	10.5	17.9	3.4	13.7	22.0	4.1
28	25	8.5	16.3	3.3	9.1	18.7	3.7
29	27	9.6	17.6	3.4	9.0	19.0	3.7
31	14	18.8	20.5	5.5	5.9	24.5	6.5
32	28	7.8	11.8	2.2	8.7	14.3	2.7
33	22	5.9	17.7	3.8	9.9	21.4	4.6
34	13	15.5	16.0	4.4	5.4	17.9	5.0
35	28	9.6	13.7	2.6	13.9	10.4	3.4
36	6	10.6	18.6	7.6	7.3	19.4	7.9
37	21	9.0	11.2	2.5	10.3	16.5	3.6
38	20	7.1	10.1	2.3	6.6	11.6	2.6
Sum/Mean	766	10.9	16.9	4.0	12.5	21.8	5.2

Table 3-15. Contributions to Personal Exposure from $PM_{2.5}$ Particles of Outdoor and Non-Outdoor Origin

Subject	Fraction of time at home	Outdoor contribution to personal exposure[a]	Home contribution[b]	Other[c]	$PM_{2.5}$ Personal exposure	f_{out}[d]	f_{in}[e]	f_{other}[f]
1	0.90	9.9	5.0	5.9	20.8	0.48	0.24	0.28
2	0.77	11.6	2.1	5.3	19.0	0.61	0.11	0.28
3	0.89	15.5	3.7	15.2	34.5	0.45	0.11	0.44
4	0.83	10.0	26.4	3.2	39.6	0.25	0.67	0.08
5	0.78	12.8	4.7	12.0	29.5	0.43	0.16	0.41
6	0.88	9.8	5.2	6.5	21.5	0.46	0.24	0.30
7	0.84	14.5	1.3	9.1	24.9	0.58	0.05	0.37
8	0.75	9.7	6.1	7.3	23.1	0.42	0.26	0.32
9	0.73	11.1	3.5	5.5	20.2	0.55	0.17	0.27
10	0.90	7.9	0.6	4.5	13.0	0.61	0.05	0.35
11	0.85	16.4	10.0	3.6	29.9	0.55	0.33	0.12
12	0.84	8.4	3.3	2.3	14.0	0.60	0.24	0.16
13	0.89	5.6	3.4	4.5	13.5	0.41	0.25	0.33
14	0.86	7.3	3.6	1.7	12.6	0.58	0.29	0.13
15	0.92	9.8	6.8	3.2	19.8	0.49	0.34	0.16
16	0.79	9.7	-0.4	8.2	17.5	0.55	-0.02	0.47
17	0.88	8.2	3.7	9.1	21.0	0.39	0.18	0.43
18	0.84	10.5	1.2	4.7	16.4	0.64	0.07	0.29
19	0.87	13.3	4.3	2.4	19.9	0.67	0.22	0.12
20	0.71	9.0	0.8	5.9	15.6	0.58	0.05	0.38
21	0.61	14.6	15.5	16.5	46.5	0.31	0.33	0.35
22	0.88	10.6	12.4	6.0	29.0	0.37	0.43	0.21
23	0.70	10.0	10.7	5.0	25.7	0.39	0.42	0.19
24	0.81	9.8	21.5	5.6	36.9	0.27	0.58	0.15
25	0.69	19.3	22.6	10.5	52.4	0.37	0.43	0.20
26	0.83	14.6	2.2	17.3	34.1	0.43	0.06	0.51
27	0.87	10.5	19.7	-6.0	24.2	0.43	0.81	-0.25
28	0.84	8.5	0.9	8.3	17.6	0.48	0.05	0.47
29	0.91	9.6	16.1	-7.1	18.6	0.52	0.87	-0.38
31	0.87	18.8	-0.2	6.0	24.6	0.76	-0.01	0.24
32	0.67	7.8	4.9	3.8	16.5	0.47	0.30	0.23
33	0.95	5.9	5.0	4.9	15.8	0.37	0.32	0.31
34	0.83	14.9	1.0	4.4	20.3	0.73	0.05	0.22
35	0.97	9.5	11.6	2.3	23.4	0.41	0.50	0.10
36	0.74	10.6	2.2	5.1	17.9	0.59	0.12	0.28
37	0.87	9.1	1.1	9.2	19.4	0.47	0.06	0.47
38	0.68	7.1	0.2	6.4	13.6	0.52	0.01	0.47
Mean	0.82	10.9	6.6	5.9	23.3	0.49	0.25	0.26
SD	0.08	3.3	7.0	4.8	9.3	0.12	0.22	0.18

[a] product of F_{pex} and the mean residential outdoor concentration
[b] product of the indoor-generated concentration and the fraction of time at home
[c] sum of particle concentrations indoors at other locations times the fraction of time spent there plus contributions from the personal cloud; obtained by subtraction of outdoor and home contributions from total personal exposure
[d] Proportion of personal exposure contributed by outdoor-generated particles
[e] Proportion of personal exposure contributed by indoor-generated particles while at home
[f] Proportion of personal exposure contributed by indoor-generated particles while indoors other than at home plus personal activities throughout the day

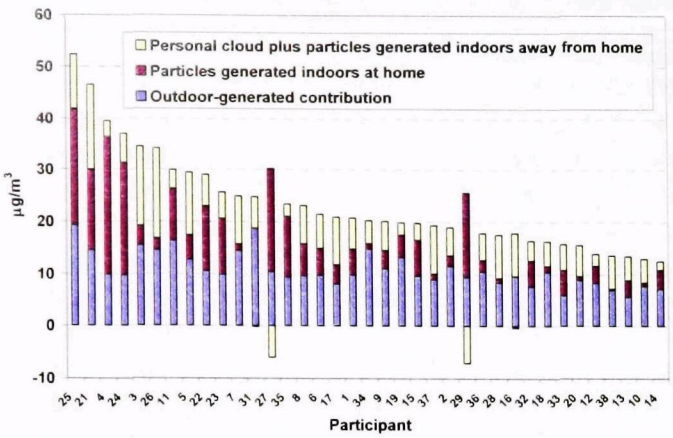

Figure 3-24. Outdoor and non-outdoor contributions to personal $PM_{2.5}$ exposure. The non-outdoor contribution is divided into indoor-generated $PM_{2.5}$ encountered while at home and the sum of indoor-generated $PM_{2.5}$ while away from home and $PM_{2.5}$ due to the personal cloud.

From Table 3-15, the contribution of at-home indoor-generated particles to personal exposure varied widely, from about 0 to 26 µg/m^3 (mean 6.6 µg/m^3). The contribution of particles generated while the person was away from home (either from exposures indoors at other locations or from personal activities throughout the day) ranged from –7 to 17 µg/m^3 (mean 5.9 µg/m^3). Two of the 37 participants had negative values for this variable due to uncertainties in estimating the outdoor and home contributions. On average, the home contribution plus the contribution of the personal cloud and exposures in locations other than the home added up to a bit more than the contribution from outdoor-generated particles (12.5 µg/m^3 vs. 10.9 µg/m^3). On an individual basis, however, some participants have virtually all of their exposures contributed from outdoor particles, while others have most of their exposure from indoor-generated or personal cloud particles (Figure 3-24).

Relationship Between Outdoor Concentrations and the Contribution to Personal Exposure of Particles of Outdoor Origin

As described in the Introduction, our ultimate aim is to determine the relationship between personal exposure to particles of outdoor origin and ambient measurements. We regressed our estimate of the outdoor contribution to personal exposure on the concentrations just outside each home (Table 3-16). Adjusted R^2 values ranged from 0.42-0.93 (median of 0.81). The overall R^2 was 0.71 (N = 750). When the regression was run against the HI at the central site, the overall R^2 was reduced to 0.64 (N = 735). The regressions of exposure to particles of outdoor origin against central site FRM concentrations is the one that interests epidemiologists, who use values at the central site to estimate personal exposure. For this set of regressions, the mean R^2 was 0.60 (N = 743), with a range from 0.19 to 0.88 (median = 0.73). Again, we caution that our assumptions force high R^2 values, and these should be considered upper bounds for the true R^2 values.

Tables 3-16 and 3-17 provide clear evidence of some degradation in the relationship between personal exposure to particles of outdoor origin and the outdoor concentrations as we consider measurements made further from the home. 30 of the 37 participants showed higher correlations with outdoor concentrations measured just outside the home than with concentrations measured at the central site by the FRM. The range in R^2 differences was –0.24 to +0.52, with an average difference of 0.11.

Would these regressions show improved results if we considered each season separately? There would be an advantage perhaps in that the infiltration factor would probably be less variable within a season than across seasons. However, a drawback would be the limitation to no more than 7 observations per household per season. After testing this on the seasonal values, we found that the drawback of fewer observations outweighed the advantage of reducing cross-season variability. That is, adjusted R^2 values by season included many that were well below the minimum of 0.19 observed for the full set of year-round observations. The adjusted R^2 results for seasonal regressions with the residential outdoor and central-site outdoor monitors are shown in Figures A-1 to A-3 in the Appendix.

Use of Reported Time in Indoor and Outdoor Microenvironments to Predict the Outdoor Exposure Factor F_{pex} from the Infiltration Factor F_{inf}

In some studies, personal exposure is not measured; therefore, a general theory has been developed using time-activity data and measurements of indoor and outdoor concentrations as a way of estimating personal exposure. Equation 3-6 provides a way to predict the outdoor exposure factor F_{pex} from the fractions of time indoors and outdoors and the measured infiltration factor F_{inf}. Since we also have a measured value of F_{pex}, using the sulfur exposure/outdoor ratio, we can compare values predicted by Equation 3-6 to measured values.

From the daily time-activity questionnaires, we can find the time spent in various activities (Table 3-18). We define the fraction of time outdoors f_{out} as the time either outdoors or in travel, with a mean value of 10%. The fraction of time indoors f_{in} (mean 90%) includes indoors at home and all other locations. For each participant and each day we can calculate F_{pex} from Equation 3-6. Table 3-18 and Figure 3-25 show that the predicted value of F_{pex} exceeds the measured value at nearly all points of the distribution. Although the R^2 is 0.57, we can almost match it

Table 3-16. Regression of Personal Exposure to Particles of Outdoor Origin on Outdoor Concentrations Measured Near Residence by Harvard Impactor (HI)

Person	N	Slope	SE	Inter	SE	p	R^2
1	25	0.48	0.04	1.47	0.83	0.09	0.84
2	24	0.44	0.05	1.36	1.20	0.27	0.77
3	21	0.50	0.05	3.12	1.29	0.03	0.84
4	23	0.44	0.05	0.46	1.21	0.71	0.76
5	27	0.68	0.06	-0.08	1.28	0.95	0.82
6	27	0.56	0.05	-0.93	0.98	0.35	0.84
7	27	0.65	0.07	-0.81	1.66	0.63	0.79
8	6	0.31	0.04	1.10	1.24	0.42	0.91
9	25	0.53	0.08	0.76	1.76	0.67	0.63
10	21	0.35	0.03	1.83	0.66	0.01	0.84
11	9	0.32	0.03	2.45	0.69	0.00	0.80
12	27	0.42	0.10	0.00	1.39	1.00	0.77
13	7	0.72	0.12	-0.11	3.11	0.97	0.74
14	27	0.44	0.04	-0.12	0.70	0.87	0.83
15	27	0.67	0.05	-0.90	0.97	0.36	0.85
16	27	0.55	0.03	0.59	0.55	0.30	0.93
17	25	0.40	0.07	1.29	1.25	0.31	0.59
18	6	0.45	0.09	1.02	2.03	0.64	0.81
19	22	0.54	0.04	2.43	0.95	0.02	0.88
20	22	0.55	0.04	-1.10	0.73	0.15	0.92
21	28	0.55	0.04	-1.10	0.73	0.15	0.85
22	13	0.48	0.10	1.14	2.19	0.61	0.63
23	14	0.48	0.06	0.02	1.36	0.99	0.83
24	23	0.52	0.04	0.68	0.83	0.42	0.87
25	14	0.69	0.11	-2.20	3.21	0.51	0.76
26	21	0.60	0.06	0.20	1.57	0.90	0.83
27	28	0.40	0.03	2.48	0.61	0.00	0.88
28	25	0.36	0.04	1.90	0.75	0.02	0.78
29	27	0.40	0.06	2.38	1.19	0.06	0.62
31	14	0.58	0.06	4.00	1.69	0.04	0.88
32	28	0.30	0.07	2.77	1.17	0.03	0.42
33	22	0.27	0.04	1.39	0.78	0.09	0.67
34	13	0.55	0.17	1.77	4.34	0.69	0.44
35	27	0.40	0.06	2.56	1.22	0.05	0.60
36	6	0.46	0.12	-0.46	3.01	0.89	0.74
37	21	0.43	0.07	1.61	1.28	0.23	0.64
38	21	0.50	0.05	0.46	0.76	0.55	0.81
Sum/Mean	750	0.53	0.01	0.26	0.26	0.40	0.71

Table 3-17. Regression of Personal Exposure to Particles of Outdoor Origin on Outdoor Concentrations Measured at Central Site by Federal Reference Method (FRM)

Subject	N	Slope	SE	p (slope)	Intercept	SE	p (Int)	R^2
1	25	0.58	0.06	0.00	0.95	1.08	0.39	0.77
2	24	0.45	0.06	0.00	2.71	1.16	0.03	0.74
3	21	0.48	0.06	0.00	5.81	1.35	0.00	0.76
4	23	0.40	0.05	0.00	1.49	1.19	0.22	0.73
5	27	0.76	0.10	0.00	0.24	1.75	0.89	0.69
6	27	0.50	0.04	0.00	0.39	0.87	0.65	0.84
7	27	0.57	0.07	0.00	2.07	1.77	0.25	0.69
8	6	0.36	0.06	0.00	0.49	1.54	0.77	0.88
9	25	0.55	0.09	0.00	1.11	1.82	0.55	0.59
10	21	0.46	0.06	0.00	1.05	0.97	0.29	0.74
11	9	0.62	0.18	0.01	3.08	4.32	0.50	0.57
12	27	0.35	0.04	0.00	2.58	0.67	0.00	0.77
13	7	0.54	0.15	0.01	-0.92	1.88	0.64	0.67
14	27	0.48	0.05	0.00	-0.11	0.77	0.88	0.8
15	27	0.71	0.05	0.00	0.25	0.83	0.76	0.87
16	27	0.58	0.07	0.00	0.85	1.11	0.45	0.74
17	25	0.35	0.10	0.00	2.09	1.77	0.25	0.34
18	6	0.52	0.22	0.08	0.08	4.54	0.99	0.47
19	22	0.47	0.06	0.00	5.18	1.22	0.00	0.71
20	22	0.58	0.07	0.00	-1.47	1.32	0.28	0.77
21	28	0.51	0.14	0.00	4.08	2.93	0.17	0.33
22	13	0.43	0.12	0.00	1.64	2.64	0.55	0.5
23	14	0.45	0.06	0.00	1.39	1.20	0.27	0.83
24	23	0.41	0.09	0.00	2.48	1.68	0.15	0.49
25	14	0.68	0.10	0.00	-1.56	3.12	0.63	0.76
26	21	0.60	0.07	0.00	1.56	1.70	0.37	0.78
27	28	0.41	0.04	0.00	3.01	0.73	0.00	0.82
28	25	0.34	0.04	0.00	2.23	0.83	0.01	0.73
29	27	0.37	0.06	0.00	3.61	1.14	0.00	0.56
31	14	0.60	0.09	0.00	5.77	2.30	0.03	0.76
32	28	0.31	0.06	0.00	2.66	1.09	0.02	0.47
33	22	0.33	0.06	0.00	0.85	1.00	0.41	0.59
34	13	0.57	0.11	0.00	0.93	2.95	0.76	0.68
35	27	0.23	0.08	0.01	6.06	1.40	0.00	0.19
36	6	0.56	0.09	0.00	-2.39	2.20	0.34	0.88
37	21	0.46	0.06	0.00	1.99	1.02	0.07	0.73
38	21	0.53	0.08	0.00	0.38	1.10	0.73	0.67
Sum/ Mean	750	0.52	0.02	0.00	1.29	0.31	0.00	0.60

Table 3-18. Time (in Minutes) Spent in Various Activities/Locations

Activity/Location	N	Mean	SD	Min	10th	25th	Median	75th	90th	Max
Indoors at home	727	1179	181	495	915	1065	1215	1320	1395	1440
Cooking	727	98	72	0	15	45	75	135	195	480
Cleaning	727	39	45	0	0	0	30	60	105	210
Grooming	727	57	47	0	15	30	45	75	120	480
Other indoor locations	727	90	82	0	0	30	75	120	200	540
Travel	727	78	61	0	0	30	60	105	165	345
Outdoors	727	67	63	0	0	15	45	105	150	390
Unknown	727	11	22	0	0	0	0	15	30	135
Exposed to smoke	727	14	44	0	0	0	0	0	45	375
Outdoors + travel	727	145	103	0	30	60	120	210	285	600
Indoors	727	1295	103	840	1155	1230	1320	1380	1410	1440
f_{out} (fraction of time outdoors or in travel)	727	0.10	0.07	0.00	0.02	0.04	0.08	0.15	0.20	0.42
f_{in} (fraction of time indoors)	727	0.90	0.07	0.58	0.80	0.85	0.92	0.96	0.98	1.00
Infiltration factor F_{inf} (indoor/outdoor S)	727	0.59	0.16	0.17	0.38	0.47	0.58	0.68	0.79	1.06
Measured outdoor factor F_{pex} (personal/outdoor S)	727	0.55	0.14	0.16	0.38	0.45	0.54	0.63	0.73	1.08
Predicted outdoor factor F_{pex} from Equation 3-6	727	0.63	0.15	0.19	0.44	0.53	0.63	0.72	0.82	1.05

by using only F_{inf} (not F_{pex}) to estimate personal exposure (Figure 3-26). The latter regression has a higher slope (0.85 vs. 0.80), a lower intercept (0.12 vs. 0.19) and an R^2 of 0.49, indicating that the extra information from the time-activity diary was not sufficient or was not precise enough to produce a better result than simply using the indoor/outdoor sulfur ratio alone.

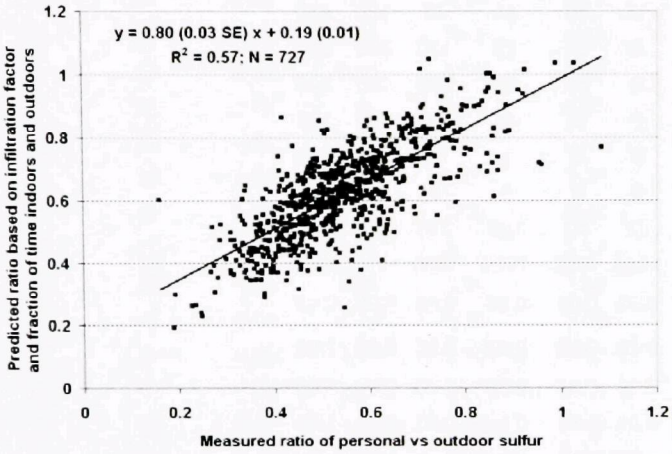

Figure 3-25. Predicted value of the outdoor exposure factor F_{pex} using Equation 3-6 compared to the measured value using the personal/outdoor sulfur ratio.

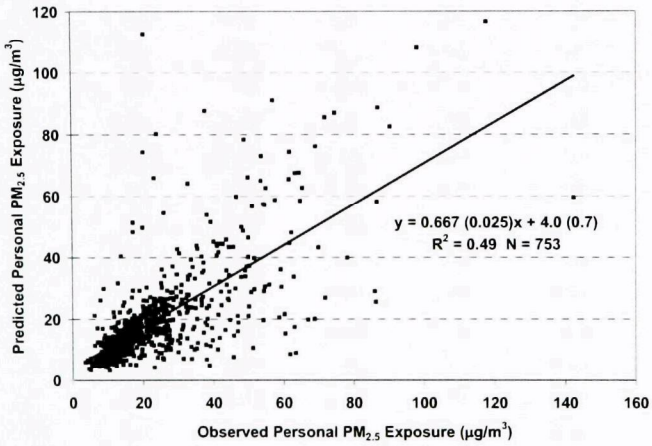

Figure 3-26. Predicted personal exposure to $PM_{2.5}$ using only F_{inf}.

Equation 3-2 relates personal exposure to the time-weighted averages of the indoor and outdoor microenvironments. We have measurements of fine particle concentrations both indoors and outdoors, but they are only 24-h averages and are not necessarily the actual concentrations experienced by the persons at the time they were in those microenvironments. Also, fine particle concentrations were not measured in some locations such as the car and workplace. If we assign the measured residential outdoor value to all the outdoor and transport microenvironments, and the measured indoor concentrations in the home to all the other indoor locations, we can test how well Equation 3-2 does in predicting personal exposure by comparing with the measured personal exposure. Results (Figure 3-26) show a moderate ability of Equation 3-2 to predict exposure, with an adjusted R^2 of 0.49 (N=753). However, the average personal exposure predicted from Equation 3-2 was only 19.4 µg/m³ compared to the observed average exposure of 23.1 µg/m³. The difference of 3.7 µg/m³ provides an independent estimate of the magnitude of the personal cloud, and this value agrees well with the earlier estimates above. The inability of Equation 3-2 to estimate personal exposure has been shown previously (Özkaynak et al., 1996; Pellizzari et al., 1999).

Estimating *P* and *k*

Calculating Average Values of P and k

Equation 1-3 for the infiltration factor is nonlinear in the air exchange rate a. Therefore if we plot our measured 24-h average indoor/outdoor sulfur ratios versus the measured air exchange rate, we can solve for the overall average P and k by minimizing some appropriate function. We tried minimizing the squares of the differences between the measured and modeled concentrations (ordinary Gaussian least squares approach) and also tried minimizing the absolute differences (a procedure giving less weight to outliers). Both approaches gave almost identical results, so we report only the results from the ordinary least squares approach. Figure 3-27 gives the results for all the individual 24-h measurements (N = 720). The estimate for the penetration coefficient P averaged across all measurements is 0.85 (0.02 SE). The estimate for the average deposition rate k is 0.22 h^{-1} (0.01 SE). The R^2 value was moderately high at 0.45.

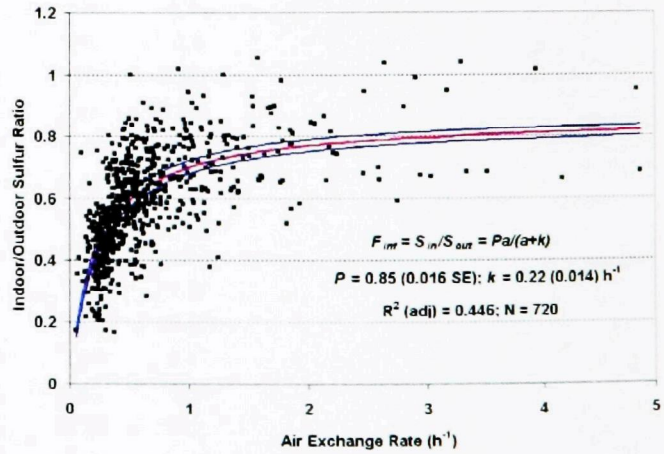

Figure 3-27. Nonlinear least-squares fit to the indoor/outdoor sulfur ratio vs. the air exchange rate. Bounding curves are ± 1 SE.

A second (linear) approach to calculating average values of P and k follows from the simple time-averaged mass balance equation for sulfur with no indoor sources:

$$S_{in}/S_{out} = Pa/(a+k) \tag{3-9}$$

This equation is not linear for one of the unknowns (k), and it also mixes the values of P and k together in one factor. We can partially separate P from k by inverting the equation (Long et al., 2001):

$$S_{out}/S_{in} = k/(Pa) + 1/P \quad (3\text{-}10)$$

Note that writing the equation this way isolates P in a single term, the intercept of the regression. This value can then be used to calculate k from the other term. Since the inverse of the air exchange rate a is the residence time τ, we can put this equation into the form of a linear regression on τ:

$$S_{out}/S_{in} = (k/P)\tau + 1/P \quad (3\text{-}11)$$

The intercept of the regression will be our best estimate of P and the slope will lead to an estimate of k (based on our value for P). One problem with this approach is that the intercept is always an extension of a line determined by points that may be far from the intercept. For example, participants 4 and 6 in the following graphs have no values of τ lower than 2 h or 1 h, respectively, so that the extrapolation of the line to a value of 0 goes well beyond the domain where all the points lie. This will lead to more variation and a greater uncertainty in our estimates of P.

As an initial check on whether the sulfate data provides useful data for determining P and k we can run the regression on the full data set (Figure 3-28).

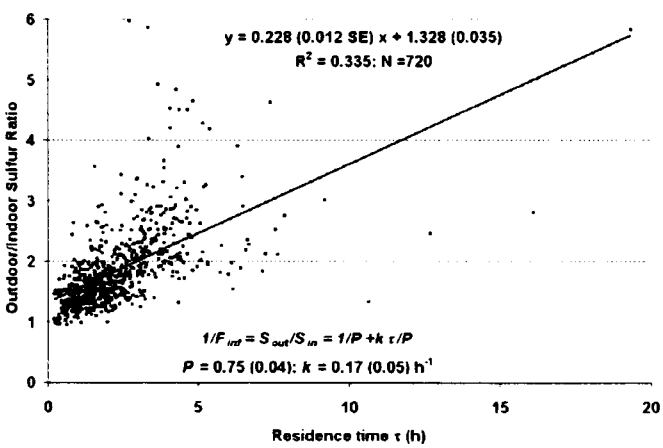

Figure 3-28. Regression of the outdoor/indoor sulfur ratio vs. residence time.

However, there are four very influential outliers at unusually high residence times (air exchange rates < 0.1 h^{-1}). These are unlikely values given other measurements in the same house. If we rerun the regression without these outliers we obtain somewhat larger values for P and k, as well as an improved R^2 (Figure 3-29). The overall average deposition rate $k = 0.23$ (0.01 SE) h^{-1} while the overall average penetration coefficient $P =$

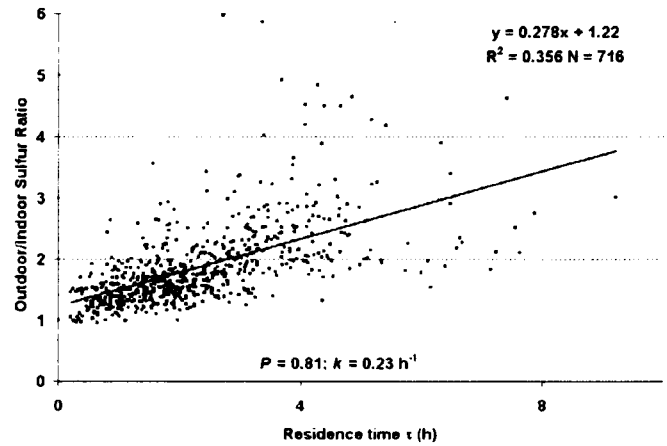

Figure 3-29. Same regression as in Figure 3-28 without four outliers.

0.81 (0.03 SE). Since measurement error generally leads to lower slopes and higher intercepts, it may be that these estimates are lower bounds for the actual average k and P. These linear estimates agree well with the nonlinear estimates of 0.22 h^{-1} for k and 0.85 for P.

Calculating Individual Home Values of P and k

In general the different ways of analyzing the data do not disagree violently, and therefore we can go ahead and try to calculate k and P for the individual homes. We first assume that P and k do not change greatly across seasons for any home. This is unlikely to be the case if certain household characteristics such as window opening, use of fans and filters, changes by season. Nonetheless, by making this assumption, we can make use of all measurements for each house in a single regression. We can employ a nonlinear fit to the observed values of the indoor/outdoor sulfur ratio (see Equations 1-3 and 1-4), calculating the best value of P and k for each home averaged across all seasons (Table 3-19). The results are mixed, with R^2 values ranging from zero to 0.94. 32 of 37 estimates for P were significantly different from zero, but only 16 estimates of k were significantly different from zero (significant results shown in boldface). Of the 32 significant estimates for P, three were considerably greater than 1, a physical impossibility. The range of significant P estimates was from 0.52 to 1.40, but the interquartile range was more tightly clustered between 0.66 and 0.98.

There are 5 values for P (3 of them significant) that are well above 1, ranging from 1.16 to 2.02. We can rerun the regressions for these 5 cases bounding P from above at 1 (Table 3-20). All five of the new estimates of k are significantly different from zero, compared to three when P was unbounded. The R^2 values are decreased slightly compared to the unbounded case, but remain quite high, from 0.31 to 0.81.

Table 3-19. Estimates of *P* and *k* for Individual Homes Using Nonlinear Fit to the Indoor/Outdoor Sulfur Ratio

House	N	*P*	SE	p (*P*)	*k*	SE	p (*k*)	R^2
1	22	**0.85**	0.10	0.000	**0.26**	0.13	0.049	0.29
2	23	**0.61**	0.04	0.000	-0.01	0.04	0.861	0.00
3	28	**0.83**	0.04	0.000	**0.12**	0.04	0.012	0.27
4	21	**0.61**	0.15	0.001	0.09	0.09	0.326	0.17
5	20	**1.16**	0.15	0 000	**0.41**	0.12	0.003	0.53
6	26	**0.67**	0.07	0.000	0.06	0.06	0.377	0.04
7	25	**0.88**	0.04	0.000	**0.14**	0.05	0.013	0.32
8	6	0.44	0.23	0.134	0.06	0.16	0.746	0.05
9	21	**0.80**	0.09	0.000	**0.17**	0.08	0.036	0.35
10	25	**0.71**	0.08	0.000	0.14	0.07	0.065	0.23
11	8	**0.67**	0.05	0.000	-0.01	0.03	0.725	0.02
12	28	**0.91**	0.12	0.000	**0.29**	0.10	0.006	0.51
13	7	**0.30**	0.08	0.013	-0.04	0.04	0.358	0.11
14	27	**0.45**	0.05	0.000	0.00	0.04	0.994	0.00
15	24	**1.03**	0.18	0.000	0.23	0.13	0.097	0.25
16	28	**0.67**	0.03	0.000	0.02	0.03	0.557	0.02
17	19	**0.60**	0.07	0.000	0.03	0.02	0.192	0.15
18	6	1.17	0.86	0.246	0.37	0.50	0.498	0.40
19	27	**1.00**	0.07	0.000	**0.35**	0.10	0.002	0.51
20	27	**0.52**	0.04	0.000	0.02	0.03	0.505	0.02
21	26	**0.88**	0.08	0.000	**0.21**	0.07	0.004	0.53
22	13	**0.97**	0.23	0.001	0.28	0.16	0.106	0.48
23	13	**0.82**	0.10	0.000	**0.50**	0.15	0.007	0.74
24	27	**0.77**	0.10	0.000	**0.18**	0.08	0.033	0.28
25	8	**1.37**	0.15	0.000	**0.48**	0.12	0.007	0.94
26	22	**0.83**	0.07	0.000	0.13	0.07	0.071	0.22
27	27	**0.77**	0.04	0.000	**0.15**	0.06	0.021	0.27
28	24	**0.75**	0.08	0.000	**0.13**	0.06	0.046	0 29
29	31	**1.46**	0.38	0.001	**0.69**	0.31	0.033	0.53
32	28	**0.60**	0.20	0.006	0.18	0.15	0.246	0.15
33	20	**0.55**	0.09	0.000	0.22	0.12	0.086	0.37
34	10	2.02	2.05	0.352	0.93	1.36	0.511	0.39
37	19	**0.79**	0.06	0.000	**0.12**	0.04	0.004	0.55
38	15	**0.68**	0.06	0.000	**0.07**	0.03	0.048	0.37
31	14			Did not converge				
36	5			Did not converge				
35	0			Same as house 29				

Table 3-20. Values for *k* when *P* is Bound from Above by 1

Subject	N	R^2	*k*	SE
25	8	0.81	0.21	0.03
29	31	0.48	0.33	0.03
34	10	0.31	0.26	0.03
18	6	0.40	0.28	0.02
5	20	0.50	0.28	0.03

A similar approach to the above nonlinear use of the indoor/outdoor sulfur ratio is to use the linear equation involving the inverse of that ratio, as well as the inverse of the air exchange rate (Table 3-21). 24 homes had both slope and intercept significantly different from zero (shown in boldface).

Five values of P were well above 1, so the regressions were rerun bounding P from above by 1 (Table 3-22). All 5 new estimates of k are significantly different from zero, compared to none when P was unbounded. The R^2 values are decreased slightly compared to the unbounded case, but remain quite high, from 0.32 to 0.82.

The estimates of P and k from the nonlinear approach (Table 3-19) are compared to the estimates from the linear approach (Table 3-21) in Figures 3-30 and 3-31. Only values significantly different from zero are included. Figure 3-30 suggests that the nonlinear approach gave consistently smaller estimates of P at the high end of the range. In fact, of the five estimates exceeding unity due to the linear approach, all five were lower, and two were less than one, in the nonlinear approach.

The median value for P was 0.81 (interquartile range 0.66 to 0.90). The median for k was 0.24 h^{-1} (interquartile range 0.12 to 0.35 h^{-1}).

Having calculated P and k for each home from the linear regression of the outdoor/indoor sulfur ratio vs. the residence time, the estimated average infiltration factor for each home can be calculated from the equation

$$F_{inf} = Pa/(a+k) \qquad (3\text{-}12)$$

where a in this case is the arithmetic, geometric, or harmonic average of the air exchange rates for each home.

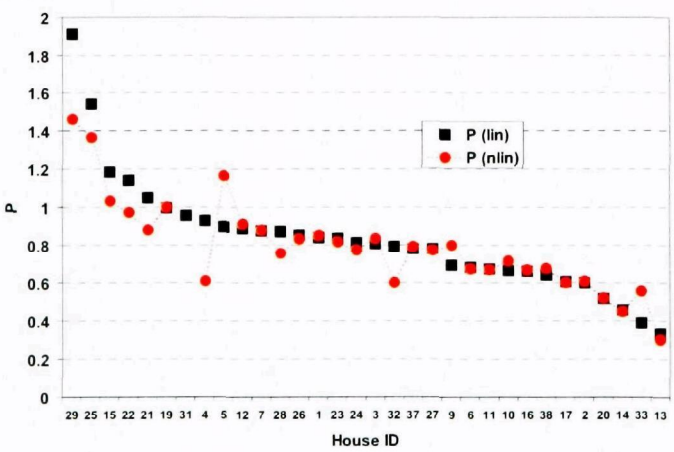

Figure 3-30. Comparison of estimates of P from the linear and nonlinear approaches described in the text. Only values significantly different from zero are plotted (N = 32 homes).

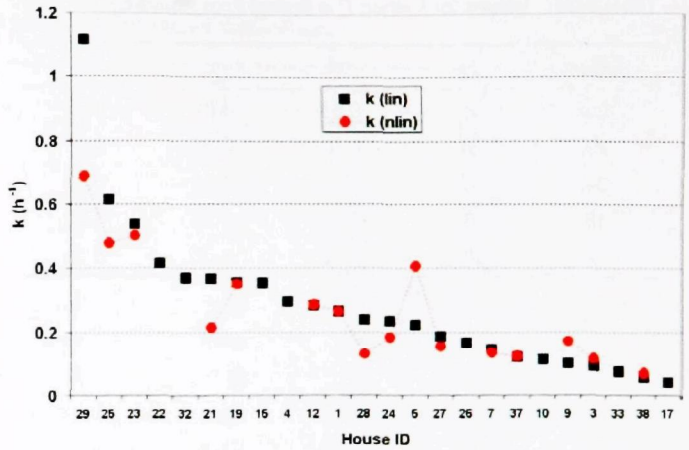

Figure 3-31. Comparison of the estimates of k from the linear and nonlinear approaches described in the text. Only values significantly different from zero are plotted (N = 24 homes).

Estimating F_{inf} from Individual Values of P and k

These estimated values of P and k from the two approaches (linear and nonlinear) can be used to estimate F_{inf} for each home. These estimates are compared to the observed indoor/outdoor sulfur ratio in Figure 3-32. Although the excellent agreement (R^2 = 0.96 for the nonlinear estimate and 0.99 for the linear estimate) of the F_{inf} estimates may be an artifact due to the related nature of the three calculations, at least this agreement suggests that the P and k values used to estimate F_{inf} from both the linear and nonlinear regressions (which both involve the air exchange rate or its inverse) are at least consistent with the measured indoor/outdoor sulfur ratios (which do not involve the air exchange rate). However, the many cases in which neither the linear nor the nonlinear approach gave values for k significantly different from zero, and the several cases in both approaches in which values of P were greater than unity, suggests that the assumption of constant values for k and P across seasons for each home is violated.

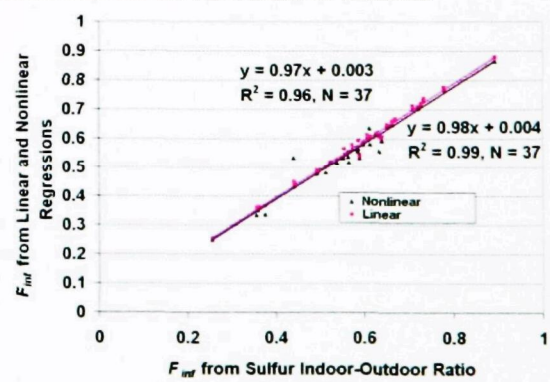

Figure 3-32. Comparison of the infiltration factor (F_{inf}) estimates from the simple ratio of indoor sulfur to outdoor sulfur by home vs. the nonlinear regression of the same ratio using the measured air exchange rates and the linear regression of the inverse ratio (outdoor/indoor) against the residence time.

Table 3-21. Results of Linear Regressions of the Outdoor/Indoor Sulfur Ratio on Residence Time for 36 Homes

House	N	1/*P*	SE	*P*	SE	*k*/*P*	SE	*k*	R^2
1	22	1.19	0.13	**0.84**	0.09	0.31	0.09	**0.26**	0.34
2	23	1.66	0.11	0.60	0.04	0.00	0.07	0.00	0.00
3	28	1.24	0.06	**0.81**	0.04	0.11	0.04	**0.09**	0.20
4	21	1.08	0.45	**0.93**	0.39	0.32	0.11	**0.29**	0.28
5	20	1.12	0.21	**0.90**	0.17	0.25	0.09	**0.22**	0.25
6	26	1.47	0.15	0.68	0.07	0.10	0.08	0.07	0.02
7	25	1.14	0.05	**0.88**	0.04	0.16	0.05	**0.14**	0.33
8	6	2.25	1.36	0.44	0.27	0.14	0.33	0.06	0.00
9	21	1.45	0.13	**0.69**	0.06	0.14	0.04	**0.10**	0.32
10	25	1.51	0.15	**0.66**	0.06	0.17	0.06	**0.11**	0.20
11	8	1.49	0.10	0.67	0.05	-0.01	0.04	-0.01	0.00
12	28	1.13	0.16	**0.89**	0.13	0.32	0.06	**0.28**	0.52
13	7	3.03	0.79	0.33	0.09	-0.05	0.17	-0.02	0.00
14	27	2.20	0.24	0.45	0.05	0.04	0.07	0.02	0.00
15	24	0.85	0.18	**1.18**	0.26	0.30	0.09	**0.35**	0.30
16	28	1.51	0.07	0.66	0.03	0.03	0.05	0.02	0.00
17	19	1.66	0.20	**0.60**	0.07	0.07	0.03	**0.04**	0.22
18	6	0.99	0.65	1.01	0.67	0.29	0.19	0.29	0.19
19	27	1.00	0.06	**1.00**	0.06	0.36	0.06	**0.35**	0.60
20	27	1.93	0.14	0.52	0.04	0.04	0.04	0.02	0.00
21	26	0.96	0.12	**1.05**	0.13	0.35	0.05	**0.37**	0.66
22	13	0.88	0.29	**1.14**	0.37	0.36	0.10	**0.41**	0.52
23	13	1.19	0.17	**0.84**	0.12	0.64	0.12	**0.54**	0.72
24	27	1.23	0.21	**0.81**	0.14	0.29	0.08	**0.23**	0.29
25	8	0.65	0.07	**1.54**	0.16	0.40	0.03	**0.61**	0.96
26	22	1.17	0.10	**0.85**	0.07	0.19	0.07	**0.16**	0.25
27	27	1.28	0.08	**0.78**	0.05	0.24	0.07	**0.18**	0.31
28	24	1.15	0.17	**0.87**	0.13	0.27	0.07	**0.24**	0.40
29	31	0.52	0.25	**1.91**	0.91	0.58	0.09	**1.11**	0.55
31	14	1.05	0.08	0.96	0.07	0.16	0.11	0.15	0.07
32	28	1.26	0.62	**0.79**	0.39	0.47	0.16	**0.37**	0.21
33	20	2.59	0.35	**0.39**	0.05	0.19	0.07	**0.08**	0.27
34	10	0.58	0.54	1.73	1.64	0.44	0.21	0.77	0.28
36	5	3.46	1.78	0.29	0.15	0.14	0.44	0.04	0.00
37	19	1.28	0.10	**0.78**	0.06	0.15	0.03	**0.12**	0.53
38	15	1.56	0.11	**0.64**	0.04	0.09	0.03	**0.05**	0.39
Sum/Mean	720	1.33	0.03	0.75	0.00	0.23	0.01	0.17	0.34

Table 3-22. Values for *k* when *P* is Bound from Above by 1

Subject	N	*k*	SE	R^2
15	24	0.22	0.02	0.32
22	13	0.32	0.03	0.55
25	8	0.26	0.03	0.82
29	31	0.28	0.03	0.55
34	10	0.44	0.05	0.34

Seasonal Analysis

Up to this point, the analyses have been carried out using values averaged across all visits during the year. However, some seasonal variation was noted in the values for the infiltration factor F_{inf} and the personal exposure factor F_{pex}. In this section, we test whether we can obtain useful results on an individual home or person on a seasonal basis by regressing personal and indoor sulfur on outdoor sulfur concentrations by season. The advantage to this is that the seasonal variations, if any, will be observed; the disadvantage is that limiting the regression to a maximum of 7 values is likely to lead to wider variance and more uncertainty in estimating the two quantities of interest.

Since air exchange rates are the only measured variable contributing to the infiltration factor (the others being the unmeasured P and k parameters), we compare the (harmonic) average air exchange by house and by season to the estimate of F_{inf} obtained by taking the average indoor/outdoor sulfur ratio by house and by season (Appendix Table A-1). Then we compare the estimate of F_{inf} from the indoor/outdoor sulfur ratio to the corresponding estimate obtained by regressing indoor on outdoor sulfur and taking the slope of the regression as an estimate of F_{inf} for each house and season (Appendix Table A-2). The estimates of F_{inf} from the ratio are compared to the estimates from the slope in Figure 3-33. The R^2 value is 0.62, and the estimates from the regression slopes vary more widely (from 0 to >1) than the estimates from the indoor/outdoor ratios (0.2 to <1).

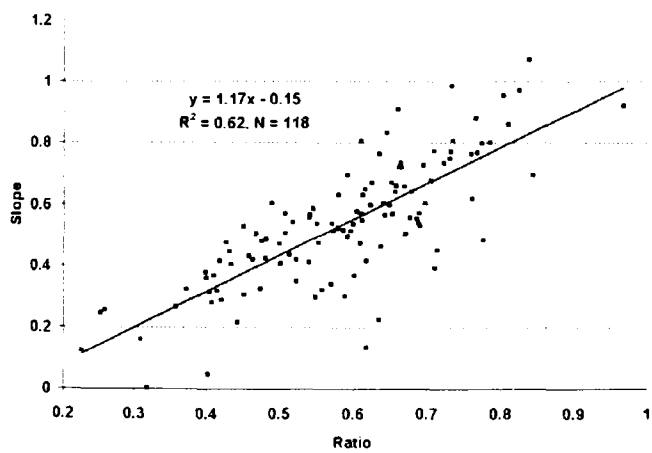

Figure 3-33. Estimates for each home by season of the infiltration factor F_{inf} from regressing indoor sulfur on outdoor sulfur (Slope) compared to estimates from the simple ratio of indoor sulfur to outdoor sulfur averaged over all visits in a season.

Another way of comparing the estimates using the regressions to the estimates using the ratios is provided by Figures 3-34 and 3-35. Figure 3-34 presents the homes ordered by season and by the value of F_{inf} as determined from the ratio; Figure 3-35 presents the comparable results from the regressions. Comparing the two figures shows many cases in which the regression approach predicts low values for the infiltration factor (< 0.4), as well as a smaller number of cases in which the approach predicts very high values, including one impossible result (>1). This analysis confirms that the ratio provides the more stable estimate of the infiltration factor.

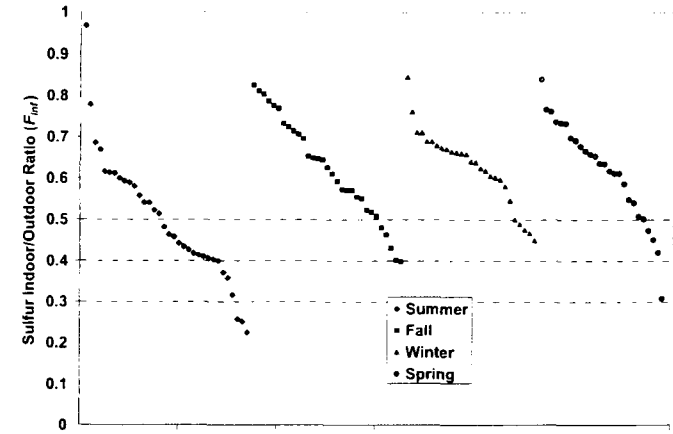

Figure 3-34. Estimates of F_{inf} by home from the indoor/outdoor sulfur ratio.

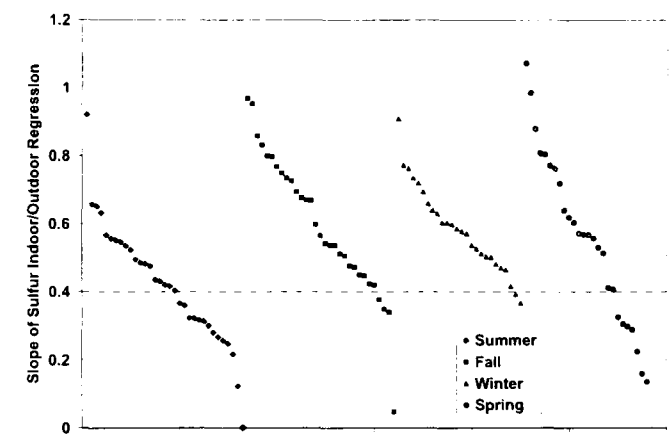

Figure 3-35. Estimates of F_{inf} by home from regressions of indoor on outdoor sulfur.

Multivariate Regressions

All participants filled out a questionnaire on their household characteristics and daily activities. A total of 54 questionnaire variables were included in the dataset (see Table A-4 in the Appendix for the variable names and definitions). We carried out a series of multiple regressions on the personal, indoor, and outdoor fine particle and sulfur concentrations vs. these questionnaire responses to try to identify sources of increased exposure.

Our first priority was to investigate the 54 independent variables for possible collinearity. This procedure is explained fully in the book *Regression Diagnostics* by Belsley, Kuh, and Welsch (1980). A factor matrix is prepared of the 54 variables and a "condition number" is calculated for each of the eigenvalues of the matrix. Belsley, Kuh, and Welsch (1980) recommend that if

a condition number exceeds 30, then variables that have a heavy weight on that eigenvalue should be inspected and either combined or a way found to drop one of the variables. Four eigenvalues did in fact exceed a condition number of 30. The pairs of variables with the heaviest loadings on each eigenvalue were ROOMS/AREA, DRYER/DRYER_VENT, DSTFACTORavg/C_FUEL, and TEMPC/TEMPDELTA. Since AREA seemed the more precise factor, we dropped ROOMS. Since DRYER seemed the more fundamental variable, we dropped DRYER_VENT. And since TEMPDELTA was a calculated variable including a number of imputed values, we dropped it. The choice between DSTFACTORavg and C_FUEL was more difficult. The dust factor as estimated by the technician has previously (in the PTEAM Study, for example) been one of the strongest predictors of airborne particles. However, the cooking fuel (electric or gas) is the more fundamental variable, so we reluctantly dropped DSTFACTORavg from the list of variables. The final list contained 50 variables dealing with household characteristics, personal activities, and two measured quantities (air exchange and outdoor temperature).

To include 50 variables in the regression and achieve an overall significance of $p<0.05$, the Bonferroni criterion was applied by dividing the chosen significance level by the number of variables. This gives us a p-value of 0.001 as the value required to achieve an overall significance of $p<0.05$ for the final model. In all of the following tables dealing with multiple regression results, the variables that meet the Bonferroni criterion for significance are listed in boldface type.

The first step in the multiple regression approach was to carry out a stepwise regression (combined forward selection and backward elimination) on all 50 household characteristics and personal activities as well as one or two measured particulate matter or sulfur variables using Statistica 6.1 software. A second regression was run including only those variables at the $p < 0.05$ level. It is important to run the second regression on the smaller set of variables since this will include more cases (typically, for our data set, 20-70 additional records that were dropped because of the casewise elimination employed for all 50 independent variables.) To check our results, all of the initial 50-variable regressions were rerun on SAS statistical software using a standard backward elimination stepwise regression. Normally, SAS and Statistica identified the same significant variables, with minor differences in the slopes, intercepts, and R^2 values (the latter were always within 0-3% of each other). When a variable differed, both final versions were run one more time in Statistica, and the run with the higher R^2 value was selected.

Although major collinearities were avoided, less strong ones remain. This could cause one or two of a collinear pair of variables to fail to register as significant in the first run involving 50 variables; they will then have no chance to be considered in the final run picking out only the significant variables from the first run. In some situations, we inserted a variable that we suspected might be relevant after the "final" run and discovered that it too was significant, but never succeeded in changing the final R^2 value by more than 1% when adding individual variables, indicating that the most important variables in each regression were identified.

In carrying out analyses of how household characteristics and personal activities affect indoor concentrations of particles, we need to carefully examine how the study design may have created unequal conditions among the homes. First, not all homes were visited for an equal number of seasons. Only 24 homes were visited in all four seasons. Second, even within a season, only 3-6 homes were visited each week. Therefore different homes within any given season encountered different outdoor conditions. The homes were scrambled each season so that the same homes were not visited together in a given week over two seasons, so this should have evened out the outdoor conditions encountered to some extent, at least for the 24 homes visited all four seasons. However, for the entire group of 36 homes, there was extensive variation in the outdoor $PM_{2.5}$ concentrations, whether at the central site or outside the homes, ranging over a factor of two from the highest to the lowest (Figure 3-36).

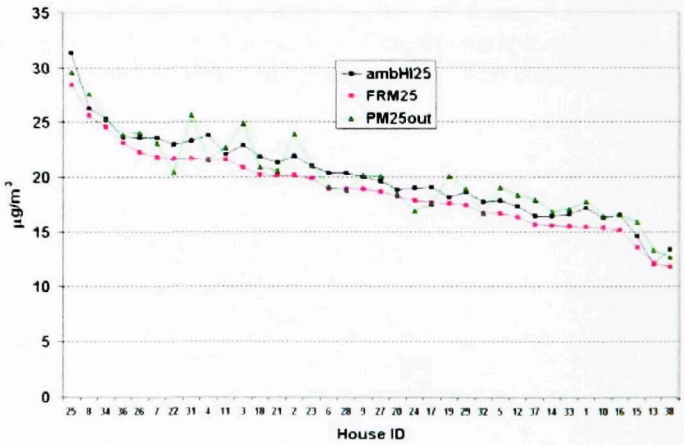

Figure 3-36. Central-site and residential outdoor concentrations averaged over all visits to a home.

Clearly, if homes at the high end of the outdoor concentrations happen by chance to have some characteristics substantially different from those at the low end, a simple regression of outdoor concentrations at the central site on these household characteristics may show significant results, even though the relationship cannot be causal (household characteristics cannot affect outdoor concentrations). In fact, some of these "impossible" relationships are observed. For example, a regression of the two central site monitors and the outdoor residential monitor on various household characteristics showed a number of artifactual relationships (Table 3-23). The building age and air exchange rates were among the strongest variables in

Table 3-23. Multiple Regression of Outdoor Concentrations on Household Characteristics and Personal Activities

Variable	Slope	Std.Err.	p-level	N	R^2(adj.)
PM25out				720	0.12
Intercept	13.78	1.13	0.000000		
AGE	0.13	0.02	0.000000		
Windopen	3.94	0.65	0.000000		
airex	-3.18	0.57	0.000000		
outdoor	0.01	0.01	0.003668		
DRYER	1.84	0.87	0.034377		
ambHI25				743	0.10
Intercept	10.58	1.82	0.000000		
airex	-2.82	0.52	0.000000		
AGE	0.11	0.02	0.000004		
C_FUEL	2.95	0.72	0.000042		
Windopen	2.45	0.66	0.000202		
FAN	-3.66	1.00	0.000264		
MILDEWavg	-2.77	1.03	0.007241		
spacehtr	-4.63	1.96	0.018484		
VAC	2.00	1.02	0.049494		
FRM				772	0.10
Intercept	10.60	1.68	0.000000		
airex	-2.56	0.49	0.000000		
AGE	0.10	0.02	0.000002		
FAN	-3.73	0.93	0.000067		
C_FUEL	2.46	0.65	0.000168		
Windopen	1.97	0.60	0.001016		
MILDEWavg	-2.74	0.96	0.004431		
spacehtr	-4.27	1.87	0.022320		
VAC	1.95	0.95	0.039863		

all three cases, along with open windows in two of the three cases, but it is likely that these variables simply represent differential outdoor air conditions encountered at the different times that the homes were monitored. The variables "explain" only 10-12% of the variance of the outdoor monitors, but they represent a caution nonetheless in our interpretation of "significant" variables in all other of the multiple regressions we will consider. In particular, those regressions that include both an outdoor measurement and one or more of the variables *AGE* and *airex* (and *windopen*) will be putting the variables into "double jeopardy", examining their effect both explicitly and implicitly as part of the outdoor particle measurement. Since the signs of *airex* and *windopen* are opposite, one might think that because they are correlated they end up with opposite signs, a common occurrence in regressions with correlated variables. Although it might seem that air exchange rates should be fairly well correlated with open windows, the Spearman correlation of *airex* and *windopen* was only 0.24. Nonetheless, the first regression in Table 3-23 was rerun twice, dropping *airex* from the first run and then restoring it and dropping *windopen* from the second run, to test whether some kind of cross-term relationship was occurring. However, in both cases the variable left in continued to be significant with the sign in the same direction as before, and the adjusted R^2 dropped in both cases, once to 0.10 and once to 0.08. Therefore, the "best" model for the residential outdoor $PM_{2.5}$ measurements continues to be the one listed first in Table 3-23. Again, for the definitions of the variables appearing in the next five tables, see Table A-4 in the Appendix.

The indoor model is

$$PM25in = \alpha + \beta_{out}*PM25out + \beta_i*x_i \qquad (3\text{-}13)$$

where the x_i are the 50 appropriate continuous/categorical variables.

The model above can be repeated using FRM25 as the independent variable:

$$PM25in = \alpha + \beta_{out}*FRM25 + \beta_i*x_i \qquad (3\text{-}14)$$

The point of using the FRM25 as the outdoor variable is that epidemiological studies are often limited to the central-site monitor.

The results of these model runs are provided in Table 3-24. The first regression is a simple regression of indoor $PM_{2.5}$ on outdoor $PM_{2.5}$. As can be seen, the relationship is particularly poor, with an R^2 of only 0.09. Since epidemiologists are often restricted to use of a fixed-site urban monitor, the regression is rerun using the central-site FRM monitor. The R^2 remains low at 0.11. These results are consistent with other studies showing low cross-sectional relationships between indoor and outdoor PM levels.

The R^2 value is increased to above 0.40 for multiple regressions including the questionnaire responses and either the residential outdoor monitor or the central-site monitor. In each case, the outdoor PM concentration has the greatest influence on the indoor PM (as judged by the p-value). Estimates for the outdoor contribution to indoor PM ranged from 48% (using residential outdoor PM) to 57% (using central site FRM). The difference in these estimates can be attributed to the consistently lower values returned by the FRM. Investigators have noted that the FRM may underestimate PM levels due to loss of volatile species such as nitrates.

Burned food added about 12 $\mu g/m^3$ to the indoor concentration, while a nearby dirt road added 8.4 to 8.9 $\mu g/m^3$, and use of an exhaust fan added about 5 $\mu g/m^3$. It is likely that the use of the exhaust fan because of cooking or burned food added to the indoor concentration.

Homes with persons who reported being near cigarette smoke also had higher indoor fine particle concentrations. The number of smokers increased indoor PM concentrations by 4-6 $\mu g/m^3$, while time spent near smokers increased concentrations by 0.04 $\mu g/m^3$ per minute exposed.

Cooking increased $PM_{2.5}$ concentrations by about 0.03 $\mu g/m^3$ per minute. Homes with electric stoves had $PM_{2.5}$ concentrations 6.6-6.8 $\mu g/m^3$ higher than homes with gas stoves, but it should be remembered that the homes with electric stoves also had higher outdoor $PM_{2.5}$ levels (see Table 3-23). Each additional person living in the household added 1.3 $\mu g/m^3$ to the daily average $PM_{2.5}$ concentrations. Vacuuming appeared capable of increasing daily average $PM_{2.5}$ concentrations by about 4 $\mu g\ m^3$, although the variable was only marginally significant (using the Bonferroni criterion) in one of the two regressions. Finally, homes with a clothes dryer were associated with a decrease of 4.7 $\mu g/m^3$ in their daily average concentrations. The presence of a clothes dryer (nearly all of which were vented outdoors) increases air exchange since almost the same volume of air must enter the house to replace the heated air vented outdoors.

The next series of two multiple regressions takes advantage of our ability to split indoor concentrations into indoor-generated and outdoor-generated fine particles (Table 3-25).

The model for indoor-generated fine particles is the following:

$$Incontrib = \alpha + \beta_i*x_i \qquad (3\text{-}15)$$

"Incontrib" is the contribution of indoor sources after subtracting the outdoor contribution determined by the indoor/outdoor sulfur ratio; therefore, this model does not include the outdoor $PM_{2.5}$ variable. The model was initially run with this variable included

Table 3-24. Dependence of Indoor Fine Particle Concentrations on Household Characteristics and Personal Activities

Variable	Slope	Std.Err.	p-level	N	R^2(adj.)
PM25in				775	0.09
Intercept	8.47	1.37	0.000000		
PM25out	0.54	0.06	0.000000		
PM25in				760	0.11
Intercept	7.21	1.36	0.000000		
FRM25	0.67	0.07	0.000000		
PM25in				762	0.42
Intercept	-10.50	2.91	0.000325		
PM25out	0.48	0.05	0.000000		
DIRT_RD	8.89	1.42	0.000000		
Burning	11.80	1.93	0.000000		
C_FUEL	6.77	1.36	0.000001		
Numpeopl	1.34	0.29	0.000003		
Exhstfan	5.04	1.11	0.000007		
cooking	0.03	0.01	0.000099		
numsmok	6.33	1.71	0.000225		
DRYER	-4.66	1.40	0.000912		
vacuum	3.89	1.20	0.001220		
smoke	0.04	0.01	0.001506		
Other_indoor	0.09	0.03	0.002624		
Unknown	0.06	0.02	0.012012		
outdoor	-0.02	0.01	0.016669		
PM25in				747	0.42
Intercept	-11.15	2.91	0.000138		
FRM25	0.57	0.06	0.000000		
Burning	11.94	1.91	0.000000		
DIRT_RD	8.42	1.40	0.000000		
Exhstfan	5.34	1.11	0.000002		
C_FUEL	6.57	1.36	0.000002		
Numpeopl	1.30	0.28	0.000006		
cooking	0.03	0.01	0.000045		
vacuum	4.04	1.19	0.000750		
DRYER	-4.65	1.40	0.000909		
Other_indoor	0.10	0.03	0.001069		
Unknown	0.06	0.02	0.004758		
smoke	0.04	0.01	0.006304		
numsmok	4.55	1.76	0.009757		
outdoor	-0.02	0.01	0.021123		

Table 3-25. Dependence of Indoor-Generated and Outdoor-Generated Particles on Household Characteristics and Personal Activities

Variable	Slope	Std.Err.	p-level	N	R^2(adj.)
Incontrib				709	0.37
Intercept	-12.03	2.88	0.000033		
Burning	12.06	1.97	0.000000		
C_FUEL	8.70	1.45	0.000000		
DIRT_RD	8.01	1.45	0.000000		
airex	-4.42	0.83	0.000000		
Numpeopl	1.43	0.30	0.000003		
Exhstfan	4.89	1.15	0.000024		
S_WIN	-4.56	1.12	0.000055		
cooking	0.03	0.01	0.000064		
smoke	0.05	0.01	0.000068		
Other_indoor	0.10	0.03	0.001016		
outdoor	-0.03	0.01	0.002000		
Cleaning_1	2.60	1.00	0.009668		
DRYER	-3.64	1.45	0.012452		
Unknown	0.05	0.02	0.024292		
Outcontin				775	0.69
Intercept	0.04	0.31	0.889273		
PM25out	0.58	0.01	0.000000		
Outcontin				686	0.82
Intercept	-0.64	0.54	0.239662		
PM25out	0.59	0.01	0.000000		
TempC	-0.15	0.02	0.000000		
AGE	0.06	0.01	0.000000		
Windopen	1.68	0.23	0.000000		
cigsmokd	0 35	0.06	0.000000		
A_C	0.39	0.07	0.000000		
windowall	0.0022	0.0006	0.000069		
Numpeopl	-0 18	0.06	0.002316		
FLRCOVav	-0.02	0.01	0.002557		
vacuum	0.64	0.25	0.011674		
airex	0 55	0.23	0.014491		
fry	-0.47	0.21	0.025024		

as an independent variable to confirm that it had been fully accounted for. Results showed that it was not significant and could be omitted from the final model.

The indoor-generated contribution (*Incontrib*) depends on many of the variables appearing in the Equation 3-13 and 3-14 regressions for indoor $PM_{2.5}$ with about the same coefficient values, but does not depend on the outdoor concentration. The value of R^2 is 0.37 and again, burned food, cooking, use of electric stoves, nearness to a dirt road, use of the exhaust fan, the number of people living in the home, and exposure to ETS turn up as significant variables affecting indoor-generated particles. A new variable, the air exchange rate (*airex*), reduces indoor-generated concentrations as *airex* increases.

The model for the outdoor contribution to indoor concentrations is the following:

$$Outcontin = \alpha + \beta_{out}*PM25out + \beta_i*x_i \qquad (3\text{-}16)$$

"Outcontin" is the contribution of outdoor sources to indoor $PM_{2.5}$ levels determined by the indoor/outdoor sulfur ratio.

Results (Table 3-25) show that air exchange (*airex*, *windopen*, *windowall*) again significantly influences the results, this time increasing the outdoor contribution as their values increase. The additional variables increase R^2 to 0.82, compared with an R^2 of 0.69 for outdoor PM alone. The fact that air exchange variables appear as highly significant in these two regressions is important confirmation of our assumptions in using sulfur to identify indoor-generated and outdoor-generated PM. That is, if the assumption of no indoor sulfur source were violated in some homes, we would not be able to separate indoor-generated PM from outdoor-generated PM in those homes, and the relationship with air exchange would be weakened by the amount of the misclassification. Air exchange variables are not selected as significant in regressions of *total* indoor PM, which lumps both indoor-generated and outdoor-generated PM, because the influence of air exchange can work in two directions depending on whether indoor air PM is greater than or less than outdoor PM. But increased air exchange must increase outdoor-generated PM and it must decrease indoor-generated PM; therefore our finding of a strong effect in the expected directions when we analyze the indoor- and outdoor-generated concentrations separately must reflect a successful separation of the two sources.

A model for indoor sulfur concentration is the following:

$$S_{in} = \alpha + \beta_{out}*S_{out} + \beta_i*x_i \qquad (3\text{-}17)$$

Once again we first examine whether the outdoor concentration shows artifactual dependence on household variables by looking at outdoor sulfur (S_{out}) alone (Table 3-26). Several artifacts appeared—air exchange, storm windows, time spent outdoors. However, these variables increased R^2 from 0.59 using only $PM_{2.5}$ outdoors as the independent variable, to 0.65 using all significant variables, a relatively small increase.

The results for indoor sulfur (S_{in}) suggest that there may be more sources of indoor sulfur than we have assumed above. For example, the number of pilot lights (*pilot*) was a strongly significant variable, adding 133 ng/m^3 to the indoor sulfur concentration per each additional pilot light. Natural gas sometimes contains some sulfur, although if the level exceeds a certain amount it must be scrubbed. Smoking was also significant (*cigsmokd*), possibly due to the use of matches. The effect of open windows was also clear and expected. Homes with at least one open window (*windopen*) had increased indoor sulfur concentrations of 120 ng/m^3. A quantitative measure (*windowall*) was an increase of 0.32 ng/m^3 for every inch-hour a window was open. Thus a window opened 6 inches wide for a day would result in an increase of 48 ng/m^3 S. Building age (*AGE*) was shown to be artifactually associated with outdoor PM; therefore, its appearance in this multiple regression may also be artifactual. Adding these variables to the simple regression of indoor on outdoor sulfur increased the R^2 from 0.70 to 0.83.

We can estimate personal exposure from (1) outdoor concentrations alone; (2) indoor concentrations alone; (3) outdoor and indoor concentrations together; (4) outdoor concentrations and indoor concentrations together with household characteristics; and (5) outdoor concentrations alone together with household characteristics. These choices result in the following models of personal exposure:

$$PM25pers = \alpha + \beta_{out}*PM25out \qquad (3\text{-}18)$$

$$PM25pers = \alpha + \beta_{out}*FRM25 \qquad (3\text{-}19)$$

$$PM25pers = \alpha + \beta_{in}*PM25in \qquad (3\text{-}20)$$

$$PM25pers = \alpha + \beta_{out}*PM25out + \beta_{in}*PM25in \qquad (3\text{-}21)$$

$$PM25pers = \alpha + \beta_{out}*PM25out + \beta_{in}*PM25in + \beta_i*x_i \qquad (3\text{-}22)$$

$$PM25pers = \alpha + \beta_{out}*PM25out + \beta_i*x_i \qquad (3\text{-}23)$$

$$PM25pers = \alpha + \beta_{out}*FRM25 + \beta_i*x_i \qquad (3\text{-}24)$$

The results of the regressions are provided in Table 3-27. Outdoor $PM_{2.5}$ was a poor predictor of personal exposure, accounting for only 11-12% of the observed variance. Indoor $PM_{2.5}$ was a much better predictor, with an R^2 of 0.45. Adding the outdoor $PM_{2.5}$ variable to the indoor variable provided little improvement (R^2 = 0.47). Only one personal activity (grilling) was independently significant, increasing the R^2 to 0.49.

Table 3-26. Dependence of Outdoor Sulfur on Outdoor $PM_{2.5}$ and of Indoor Sulfur on Outdoor Sulfur and Household Characteristics and Personal Activities

Variable	Slope	Std.Err.	p-level	N	R^2(adj.)
S_{out}				720	0.59
Intercept	47.09	66.44	0.478737		
PM25out	97.14	3.01	0.000000		
S_{out}				720	0.65
Intercept	594.47	85.83	0.000000		
PM25out	96.98	2.85	0.000000		
airex	-354.67	43.81	0.000000		
S_WIN	-216.48	54.80	0.000086		
outdoor	-1.50	0.41	0.000240		
numsmok	-209.72	82.71	0.011441		
A_C	-40.72	18.05	0.024394		
S_{in}				720	0.70
Intercept	151.44	27.27	0.000000		
S_{out}	0.49	0.01	0.000000		
S_{in}				741	0.83
Intercept	-19.76	48.56	0.684261		
S_{out}	0.51	0.01	0.000000		
AGE	7.91	0.73	0.000000		
PILOT	133.01	13.43	0.000000		
windowall	0.32	0.05	0.000000		
TempC	-9.74	1.59	0.000000		
Windopen	120.11	23.85	0.000001		
cigsmokd	28.21	5.66	0.000001		
FAN	-140.52	30.52	0.000005		
A_C	33.49	7.29	0.000005		
FLRCOVav	-1.98	0.51	0.000104		
Numpeopl	-20 14	6.13	0.001074		
fry	-60.97	20.94	0.003705		
Unknown	1.21	0.47	0.009623		
DIRT_RD	81.66	32.60	0.012471		
vacuum	61.71	25.90	0.017422		

Table 3-27. Regressions of Personal Exposures to $PM_{2.5}$ on Household Characteristics and Personal Activities

Variable	Slope	Std.Err.	p-level	N	R^2(adj.)
PM25pers				750	0.10
Intercept	10.78	1.56	0.000000		
PM25out	0.66	0.07	0.000000		
PM25pers				734	0.11
Intercept	10.44	1.53	0.000000		
FRM25	0.73	0.08	0.000000		
PM25pers					
Intercept	9.98	0.69	0.000000	727	0.45
PM25in	0.67	0.03	0.000000		
PM25pers				727	0.47
Intercept	5.66	1.12	0.000001		
PM25in	0.63	0.03	0.000000		
PM25out	0.27	0.05	0.000001		
PM25pers				727	0.49
Intercept	4.32	1.21	0.000370		
PM25in	0.60	0.03	0.000000		
PM25out	0.26	0.05	0.000001		
grill	11.54	3.22	0.000359		
cooking	0.02	0.01	0.003309		
PM25pers				737	0.37
Intercept	-11.15	3.63	0.002239		
numsmok	13.76	1.54	0.000000		
PM25out	0.55	0.06	0.000000		
CANDLDUR	0.05	0.01	0.000000		
Burning	11.19	2.23	0.000001		
C_FUEL	7.42	1.56	0.000003		
DIRT_RD	6.05	1.69	0.000363		
Other_indoor	0.11	0.03	0.000800		
A_C	1.32	0.40	0.001039		
Exhstfan	4.29	1.36	0.001652		
Cleaning_1	3.46	1.11	0.001968		
cooking	0.02	0.01	0.009918		
outdoor	-0.02	0.01	0.013482		
PM25pers				721	0.39
Intercept	-11.81	3.69	0.001449		
FRM25	0.60	0.07	0.000000		
numsmok	13.31	1.61	0.000000		

Table 3-27. Continued

Variable	Slope	Std.Err.	p-level	N	R^2(adj.)
CANDLDUR	0.05	0.01	0.000000		
Burning	11.71	2.25	0.000000		
C_FUEL	7.44	1.59	0.000003		
A_C	1.42	0.41	0.000492		
Other_indoor	0.11	0.03	0.000553		
Exhstfan	4.49	1.37	0.001106		
DIRT_RD	5.45	1.70	0.001407		
Cleaning_1	3.52	1.13	0.001927		
cooking	0.02	0.01	0.010664		
outdoor	-0.02	0.01	0.015074		

When only outdoor $PM_{2.5}$ and the household characteristics were included in the regression, the most significant variable other than outdoor $PM_{2.5}$ was the number of smokers, adding between 13.3 and 13.8 μg/m^3 to the daily average personal exposure. The use of candles was highly significant, adding 0.05 μg/m^3 per minute burned to daily average personal exposure. Burned food added between 11.2 and 11.7 μg/m^3 to personal exposure. Electric stoves increased particle concentrations by 7.4 μg/m^3. Although these household characteristics and personal activities were useful in improving the R^2 for personal exposure, collectively they were not able to improve it as much as simply adding in the measured indoor $PM_{2.5}$ concentration (R^2 = 0.37-0.39 for the outdoor + household characteristics variables compared to 0.47 for the outdoor + indoor concentrations).

By assuming that no sulfur is created indoors, we can estimate the contribution of personal activities to personal exposure (*Perscontrib*) by multiplying the outdoor $PM_{2.5}$ by the ratio of the personal sulfur concentration to the outdoor sulfur concentration, and subtracting that product from the observed personal exposure. As with the similarly defined indoor-generated particle contribution to total indoor concentrations, we can regress *Perscontrib* on the household characteristics and personal activities variables without including the outdoor concentration as an input, since we used it to determine the values of *Perscontrib*. Again, we checked to confirm that, when included, the outdoor concentration was not significant. The model is

$$Perscontrib = \alpha + \beta_i * x_i \qquad (3\text{-}25)$$

The focus on the non-outdoor part of personal exposure again showed that air exchange was an important variable, tending to decrease the personal contribution by 4.1 μg/m^3 per unit change in the air exchange variable (Table 3-28). Smoking, burned food, and cooking fuel variables significantly influence personal exposure concentration. Candle burning increased personal exposure by 0.05 μg/m^3 per minute burned.

The factors affecting the outdoor contribution to PM2.5 personal exposure were investigated using multiple regression (Table 3-29). Here the model is

$$Outcontribpers = \alpha + \beta_{out} * pm25out + \beta_i * x_i \qquad (3\text{-}26)$$

The most influential factor, as expected, was the outdoor PM2.5 concentration. This is so much stronger than all the rest of the factors put together (t-value of 45 compared to values <7 for the other variables) that it can be considered the single variable driving personal exposure to particles of outdoor origin. The next strongest factor (AGE) may be an artifact due to the unbalanced design of the study, in which not all homes were monitored on the same day. The next three factors have to do with air exchange, which depends on window opening behavior (*windopen*, *windowall*) and on indoor-outdoor temperature differences (roughly approximated by *TempC*). The increased air exchange rates due to increases in these variables brings more outdoor particles indoors, and thus affects personal exposure to the extent the person is at home. Note that the R^2 of 0.82 for this model of the influence of outdoor particles on personal exposure precisely matches that for the influence of outdoor particles on indoor particle concentrations (last part of Table 3-25).

Finally, personal exposure to sulfur was regressed against outdoor sulfur and the household characteristics, using the following set of models:

$$Spers = \alpha + \beta_{in} * Sin \qquad (3\text{-}27)$$

$$Spers = \alpha + \beta_{out} * Sout + \beta_{in} * Sin \qquad (3\text{-}28)$$

$$Spers = \alpha + \beta_{out} * Sout \qquad (3\text{-}29)$$

$$Spers = \alpha + \beta_{out} * Sout + \beta_{in} * Sin + b_i * x_i \qquad (3\text{-}30)$$

$$Spers = \alpha + \beta_{out} * Sout + \beta_i * x_i \qquad (3\text{-}31)$$

These correspond to having greater or less information about the outdoor and indoor sulfur concentrations and the questionnaire variables.

In general, very high values of R^2 are achieved (Table 3-30). The worst case is having only the outdoor sulfur concentration, and that alone explains 0.78 of the variance in personal exposure to sulfur. If the indoor sulfur were known alone, the R^2 could be improved to 0.88. Using both measured indoor and outdoor concentrations to estimate personal exposure increases R^2 to 0.91. Other influential variables included time spent near a smoker and the number of pilot lights in the house, both possible sources of sulfur as mentioned above.

Variables Affecting Air Exchange and the Infiltration Factor

Our primary focus has been on documenting the influence of outdoor fine particles on indoor concentrations and personal exposures. Two of the most influential factors are air exchange and the infiltration factor.

Variables Affecting Air Exchange

Previous work (Howard-Reed et al., 2002; Wallace et al., 2002) identified window opening and the absolute indoor-outdoor temperature difference as two main variables affecting air exchange. To create a variable approximating the absolute indoor-outdoor temperature difference, we took the absolute value of the difference between the outdoor temperature (*TempC*) and a typical indoor temperature of 72 F (22.2 C). The new variable was called *AbsTempDif*.

Table 3-28. Regression of the Non-ambient-related Contribution (*Perscontrib*) to Personal $PM_{2.5}$ Exposure

Variable	Slope	Std.Err.	p-level	N	R^2(adj.)
Perscontrib				677	0.30
Intercept	-9.68	3.55	0.006489		
numsmok	11.45	1.57	0.000000		
CANDLDUR	0.05	0.01	0.000000		
C_FUEL	8.44	1.62	0.000000		
Burning	10.83	2.25	0.000002		
airex	-4.12	0.97	0.000024		
Cleaning_1	4.26	1.14	0.000194		
DIRT_RD	5.85	1.69	0.000569		
Exhstfan	4.61	1.38	0.000850		
outdoor	-0.03	0.01	0.001324		
A_C	1.31	0.41	0.001585		
Other_indoor	0.09	0.03	0.005808		
cooking	0.02	0.01	0.022302		

Table 3-29. Regression of the Ambient-Related Contribution to Personal $PM_{2.5}$ Exposure

	B	SE	t(691)	p-level	N	R^2(adj.)
Intercept	-0.952	0.588	-1.6	0.105780	710	0.82
PM25out	0.521	0.012	45.2	0.000000		
AGE	0.044	0.007	6.7	0.000000		
windowall	0.003	0.000	6.4	0.000000		
Windopen	1.078	0.211	5.1	0.000000		
TempC	-0.062	0.012	-5.0	0.000001		
cigsmokd	0.221	0.056	3.9	0.000101		
MILDEWavg	1.113	0.296	3.8	0.000187		
smoke	0.009	0.002	3.6	0.000288		
cooking	0.005	0.001	3.6	0.000311		
A_C	0.234	0.068	3.5	0.000593		
DIRT_RD	0.959	0.295	3.3	0.001196		
Travel	0.004	0.002	2.9	0.004144		
Pets	0.754	0.298	2.5	0.011484		
dust	-0.688	0.271	-2.5	0.011302		
FLRCOVav	-0.012	0.004	-2.8	0.005730		
Grooming	-0.006	0.002	-3.0	0.002672		
AREA	-0.001	0.000	-3.2	0.001409		

Table 3-30. Regressions of Personal Exposure to Sulfur on Indoor and Outdoor Concentrations and Questionnaire Variables

Variable	Slope	Std.Err.	p-level	N	R^2(adj.)
Spers				727	0.88
Intercept	42.50	15.67	0.006860		
Sin	0.91	0.01	0.000000		
Spers				727	0.91
Intercept	-12.71	14.32	0.374879		
Sin	0.66	0.02	0.000000		
Sout	0.17	0.01	0.000000		
Spers				750	0.78
Intercept	89.52	21.54	0.000036		
Sout	0.49	0.01	0.000000		
Spers				722	0.93
Intercept	-1.59	31.71	0.959998		
Sin	0.65	0.02	0.000000		
Sout	0.17	0.01	0.000000		
DIRT_RD	117.68	18.99	0 000000		
smoke	0.87	0.16	0.000000		
windowall	0 14	0.03	0.000006		
MILDEWavg	81 04	20.08	0 000060		
C_FUEL	-55.33	15.33	0.000327		
Grooming	-0.45	0.14	0.001283		
outdoor	0.34	0.11	0.002197		
Other_loc	0.22	0.08	0.010413		
TempC	2.46	1.07	0.022455		
Spers				715	0.85
Intercept	28.25	58.70	0.630470		
Sout	0.50	0.01	0 000000		
PILOT	123.65	13.27	0.000000		
windowall	0 36	0.05	0.000000		
AGE	3.63	0 74	0.000001		
AREA	-0.10	0.02	0.000040		
smoke	1.03	0.26	0.000105		
DIRT_RD	111 72	31.23	0.000372		
FLRCOVav	-1.62	0.48	0.000862		
Windopen	72.43	22.40	0.001280		
TempC	-4 59	1 51	0.002480		
MILDEWavg	92.35	30.90	0.002903		
Travel	0.47	0.16	0 003808		
BUSY_RD	70.43	24.83	0 004686		
FAN	-76.58	29.86	0.010533		
cooking	0.33	0.14	0.017800		
cigsmokd	13 98	6 01	0.020369		
sweep	-45.29	21.61	0.036424		

Because we saw increased air exchange rates in summer, we considered using *Season* as a separate independent variable. The reasoning here is that perhaps people would tend to keep the windows closed and the air conditioner running in Summer even on a relatively cool day, whereas on a day with the identical temperature in another season they might not bother turning on the air conditioner. Thus, both temperature and season could have separate effects. First, *Season* was coded by average temperature, using a 3-point scale (Winter = 1, Fall and Spring = 2, Summer = 3); however, a regression including either season or TempC showed that TempC produced slightly higher R^2. It was then thought that a 3-point scale might be too restrictive, so a 5-point scale was created by month (e.g., Jan-Feb = 1, July-August =5). There were only 10 months, with March and December missing. The new variable was called *MonthbyTemp*. This time when the regression was run putting in either *TempC* or *MonthbyTemp*, the R^2 using the *MonthbyTemp* variable was slightly higher than that using *TempC* (Table 3-31).

The two strongest variables are in fact those that we already know from previous studies: the window opening width and the absolute indoor-outdoor temperature difference. The latter has a coefficient of 0.04 ach/°C, which agrees well with the estimate of 0.02 ach/°C in Wallace et al. (2002). The next strongest variable is the number of persons in the home. This variable, as well as the *pets* variable, was included on the questionnaire because of results from past studies (e.g., Thatcher and Layton, 1995) indicating that people and pets can increase particle levels due to resuspension and going outdoors more often, which will bring in outdoor particles when the door is open. The *pets* variable also appears, although just missing significance. The next strongest variable is age of the home—previous studies have noted that older homes are constructed more loosely. The use of the exhaust fan tends to increase air exchange, as noted in a previous study (Wallace et al., 2002). Another variable is the *MonthbyTemp* variable, reflecting the increased use of air conditioners (and therefore closed windows) in the summer. The presence of a clothes dryer (nearly all of which were vented outdoors) increases air exchange since almost the same volume of air must enter the house to replace the heated air vented outdoors. We have no explanation for the appearance here of the VAC and FLRCOVav variables, although their effect on the total R^2 value of the model is very small.

Variables Affecting the Infiltration Factor

The multiple regression on the indoor/outdoor sulfur ratio was run on all variables, including *airex* (Table 3-32). However, the five strongest variables contributing to this ratio can all be seen to be variables already contributing to the air exchange rate. Therefore these variables are, in a sense, being double-counted. They are obscuring and weakening the actual relationship with the air exchange rate. Therefore the regression was run again including all variables except those already found to contribute to the air exchange rate (Table 3-33). The final model shows that the air exchange rate (together with the windows open variable) is the strongest variable affecting the sulfur indoor/outdoor ratio. The air exchange rate is the only one of our variables that explicitly appears in the equation for the infiltration factor: $(Pa/(a+k))$. The three remaining variables provide very little contribution (about 3%) to the total R^2 of the model. One of these, A/C, appears to have the wrong sign, since presence of an air conditioner would be expected to reduce the infiltration factor. However, this variable ranges from 1 to 5 air conditioners and therefore is measuring the falloff in efficiency as one goes from central air conditioning to multiple window air conditioning.

Table 3-31. Variables Affecting Air Exchange Rate

Variable	Slope	Std.Err.	t(711)	p-level	N	R^2 (adj.)
airex					744	0.51
Intercept	0.5156	0.1355	3.8	0.00015		
windowall	0.0010	0.0001	13.2	0.000000000000		
ABSTempDIF	0.0434	0.0040	10.8	0.000000000000		
Numpeopl	0.0693	0.0094	7.3	0.000000000001		
AGE	0.0079	0.0013	6.3	0.00000000062		
VAC	-0.3079	0.0496	-6.2	0.00000000093		
FLRCOVav	-0.0043	0.0008	-5.1	0.00000044		
DRYER	0.2071	0.0458	4.5	0.0000073		
MILDEWavg	-0.2406	0.0544	-4.4	0.000011		
Monthbytemp	-0.0673	0.0153	-4.4	0.000012		
Mealsckd	-0.1488	0.0390	-3.8	0.00014		
Exhstfan	0.1375	0.0410	3.4	0.00085		
Unknown	0.0025	0.0008	3.3	0.0012		
Pets	0.1586	0.0523	3.0	0.0025		

Table3- 32. Variables Affecting Indoor/Outdoor Sulfur Ratio

Variable	Slope	Std.Err.	t(638)	p-level	N	R^2 (adj.)
Sinout					659	0.46
Intercept	0.68439	0.02916	23.5	0.00000000000000		
Monthbytemp	-0.03656	0.00343	-10.7	0.00000000000000		
AGE	0.00299	0.00037	8.1	0.00000000000000		
airex	0.06455	0.00925	7.0	0.000000000008		
Windopen	0.06168	0.01027	6.0	0.0000000031		
windowall	0.00014	0.00002	5.9	0.0000000062		
C_FUEL	-0.06117	0.01047	-5.8	0.0000000083		
DIRT_RD	0.06170	0.01362	4.5	0.0000070		
VAC	-0.05484	0.01323	-4.1	0.000039		
DUSTY_RD	-0.04057	0.01203	-3.4	0.00079		

Table 3-33. Variables Affecting Indoor/Outdoor Sulfur Ratio: Reduced Model

Variable	Slope	Std.Err.	t(699)	p-level	N	R^2 (adj.)
Sinout					704	0.43
Intercept	0.409	0.010	40.3	0.000000000000		
airex	0.121	0.008	15.1	0.000000000000		
Windopen	0.090	0.010	9.2	0.000000000000		
A_C	0.015	0.003	4.3	0.000018		
numsmok	0.055	0.015	3.7	0.00021		
BUSY_RD	0.036	0.011	3.2	0.0013		

Chapter 4
Discussion

To reach our primary goal of estimating the contribution of outdoor $PM_{2.5}$ to personal exposures, we set an intermediate goal of estimating the contribution of outdoor particle concentrations to indoor particle concentrations. Since personal exposures depend heavily on indoor concentrations, this may be a good first approximation. We calculated the contribution of outdoor particles by multiplying the outdoor concentration by the ratio of the indoor to outdoor sulfur concentrations:

$$C_{ino} = S_{in}/S_{out}\, C_{out} \qquad (4\text{-}1)$$

where C_{ino} is the concentration indoors of particles infiltrating from outdoors, and where the S_{in}/S_{out} term is an estimate of F_{inf}. We also calculated F_{inf} using a combination of the indoor/outdoor sulfur ratios and the air exchange measurements and found excellent agreement (R^2 = 0.96-0.99), indicating that the estimates of F_{inf} for individual homes were quite stable. This is one of the first studies to calculate infiltration factors and outdoor exposure factors for individual homes and persons, respectively, and is also one of the first to carry out the regressions of that portion of personal exposure due to particles of outdoor origin on outdoor concentrations.

Limitations of the Regressions and Influence of Assumptions

When we run a regression of C_{ino} on C_{out}, we are in fact regressing a term on a portion of itself:

$$F_{inf}\, C_{out} = A\, C_{out} + B \qquad (4\text{-}2)$$

The infiltration factor does not have a high degree of day-to-day variability: F_{inf} = 0.59 (0.16 SD) for all homes, and an even smaller standard deviation within homes. Therefore our regression is not very different from regressing a term on a *constant fraction* of itself, which will result in an R^2 of 1. This is a result that is forced by our basic assumptions no indoor sources of sulfur, no coagulation, condensation, particle-gas conversion, or indoor chemistry, negligible transient terms, instantaneous perfect mixing, penetration and deposition characteristics of $PM_{2.5}$ identical to those of sulfur - leading to a simple linear relation between the outdoor concentration and the fraction of outdoor air particles infiltrating the house. The regression on outdoor air measured at all the homes resulted in an R^2 of 0.71, a value that is certainly higher than reality due to our assumptions. The individual regressions on 36 homes gave even higher values of R^2 (median 0.77). When the regressions were run on the outdoor $PM_{2.5}$ concentrations measured by the FRM at the central site, the overall R^2 value was lowered to 0.60 (median 0.73), but with the same caveat that our assumptions force a high correlation and therefore these estimates are not be considered best estimates, but rather upper bounds.

A regression of personal $PM_{2.5}$ on outdoor $PM_{2.5}$ concentrations provides an estimate of F_{pex} of 0.59 $\pm$ 0.01 SE, with one influential outlier removed. With the outlier included, the estimate was 0.64 $\pm$ 0.02 SE. The intercept with the outlier excluded (12.3 $\pm$ 1.4 $\mu g/m^3$) is an estimate of E_{no}, the exposure due to non-outdoor sources. The difference between the indoor source contributions estimated at 7.7 $\mu g/m^3$ in Table 3-5 and the non-outdoor source contribution is sometimes attributed to the "personal cloud," which in this case equals 4.6 $\pm$ 2.0 $\mu g/m^3$.

The similarity of the slopes for the personal vs. outdoor and indoor vs. outdoor $PM_{2.5}$ regressions (0.59 compared to 0.60) suggests that the time spent indoors drives the relationship between personal exposure and outdoor concentrations. That is, the infiltration factor F_{inf}, which governs the reduction of particle concentrations as they enter a house, is very similar to the outdoor exposure factor F_{pex}, which governs the reduction in outdoor concentrations contributing to personal exposure.

A major assumption has been that fine particles in general will behave like sulfur in terms of penetration and deposition. How good is this assumption? Sulfur is found primarily in the fine fraction, and within that fraction it typically has diameters <0.5

μm, smaller than most other elements. From theoretical and experimental studies, deposition velocities appear to reach a minimum around diameters of 0.1-0.2 μm (Lai and Nazaroff, 2000). Deposition rates for sulfur and other elements were estimated in the PTEAM study (Ozkaynak et al, 1996). The rate for sulfur was 0.16 h^{-1} (0.02 SE). Rates for other elements were generally higher (e.g., iron, 0.70 h^{-1} (0.30 SE)). Therefore sulfur will stay elevated indoors longer than most other elements, and possibly longer than most of the remainder of the fine particle mass. This leads to overestimates of the contribution of outdoor particles to indoor $PM_{2.5}$. Evidence for the amount of the overestimate is provided by the results for iron concentrations. The indoor/outdoor ratio for iron was 0.38 (0.18 SD; N = 766). This was the lowest ratio of all the elements. For Si it was 0.58 (N = 766); for Mn it was 0.52 (N = 409). These were elements that appeared to have few indoor sources, judging from their high indoor-outdoor product-moment correlations (0.81 for Fe, 0.74 for Si, N = 762 samples for each; 0.58 for Mn, N = 460 samples; these correlations were almost as high as the value of 0.85 for sulfur). For the other elements, the indoor/outdoor correlations were lower and the indoor/outdoor ratios higher due to indoor sources, and cannot be used to help estimate the amount of the overestimate due to using the sulfur ratios. Iron itself makes up only a small fraction of $PM_{2.5}$ mass, but it could be a marker for the behavior of a larger fraction.

Sulfur accounted for 20% of indoor $PM_{2.5}$, assuming it is in the form of ammonium sulfate. If the remaining 80% of the indoor particle mass behaved more like iron, outdoor air particles would account for only 42% of indoor air $PM_{2.5}$, rather than 58% as was found using sulfur alone. However, it is unlikely that the remainder of the indoor air particles behave more like iron, because the RCS model regression of indoor $PM_{2.5}$ on outdoor $PM_{2.5}$ (Figure 3-13) resulted in a slope very close to the slope predicted by the sulfur indoor-outdoor ratio. These considerations indicate that the overestimate of the outdoor contribution due to relying on the sulfur ratios is not a major one and does not affect the relative order of the homes in terms of the fraction of indoor $PM_{2.5}$ produced by outdoor particles.

Based on previous studies, we attempted to use the particle measurements to estimate an infiltration factor for each home; however, we found poor agreement with the estimates in this study using the sulfur ratio. Most other studies of personal exposure and/or indoor concentrations of fine particles have fewer measurements per home than this study. Therefore our results suggest that particle mass measurements alone cannot provide reliable estimates of F_{inf}.

We estimated the outdoor exposure factor F_{pex} using two methods: personal outdoor sulfur ratios and a model using time indoors and outdoors. The model consistently overestimated the outdoor factor. We speculate that this is because people spend time in unmonitored environments, and our assumption that exposures are the same in those environments as in the monitored home environment may be incorrect. We found that estimating personal exposure using only the indoor/outdoor sulfur ratio (F_{inf}) gave better results than estimates using the modeled outdoor exposure factor F_{pex} based on time spent indoors and outdoors.

Although several different methods were employed in making these estimates, the methods ultimately depended on the sulfur measurements only. Because of the way P and k are related in the equation for F_{inf}, an infinite number of solutions are available for any given infiltration factor. The solution surface for P and k is very flat (meaning a very wide error associated with all point estimates); therefore a slight error in any measurement can lead to a very large error in the estimation of P and k. For example, each of the slightly different methods produced four or five physically impossible estimates for P Although bounds can be established to prevent P from exceeding unity, the existence of these nonphysical solutions suggests that measurement errors or violations of our assumptions may have caused large errors in estimating these parameters. Since we have no independent methods of estimating these parameters, we are unable to validate our estimates of P and k for individual homes. The estimates of P and k that were significantly different from zero included about 24 of the 36 homes. The median value for the penetration coefficient P was 0.81 (interquartile range 0.66 to 0.90). The median for the deposition rate k was 0.24 h^{-1} (interquartile range 0.12 to 0.35 h^{-1}). These estimates are similar to those obtained in some other studies (Liu et al., 2003; Wallace et al., 2002; Thatcher et al., 2003).

Because not all homes were done in the same time periods, there were certain unavoidable differences in the outdoor particle concentrations encountered at the time they were monitored. We attempted to identify possible artifacts (significant associations with no possible causal explanations) by regressing central-site and residential outdoor concentrations vs. the questionnaire variables. We did find some artifacts, and therefore caution that some apparently significant relationships may be due to these temporal variations rather than a true causal relationship. This may be particularly true for certain unvarying household characteristics such as building age, location near a road, and type of cooking fuel (gas vs. electric) because these unvarying characteristics enter the regressions as a block of entries repeated up to 28 times per house. Therefore their number of degrees of freedom is greatly reduced and any variation due to temporal heterogeneity will be multiplied by this repeated appearance. This caution extends to the variables AGE, DIRT ROAD, C_FUEL (cooking fuel), S_WIN (storm windows), VAC, and AREA among others.

Nonetheless, regressions of indoor concentrations and personal exposures showed greatly improved R^2 results when questionnaire variables were added to the outdoor concentrations. The improvement was from R^2 about 0.1 to R^2 about 0.4, for both indoor and personal estimates. Among the

significant contributors to indoor and personal exposure were smokers in the household, cooking, and number of people in a household, all variables that have previously been found to contribute to indoor PM concentrations (Özkaynak et al., 1996b; Wallace et al., 2004b). Some variables not previously identified as contributing to indoor or personal exposures were burning food, duration of candle use, number of pilot lights, use of an exhaust fan, proximity to a dirt road, and presence of electric stoves (compared to gas stoves). The latter two variables are to be treated with caution since they are unvarying household characteristics and therefore may have been subject to temporal unevenness of sampling. Vacuuming was another variable that achieved significance at times. The presence of a clothes dryer was significantly associated with reduced indoor concentrations and personal exposure.

An important result was the appearance of the air exchange variables (including windows open or closed) in the regressions on estimated concentrations of indoor-generated and outdoor-generated particles. The fact that these variables appeared in the expected directions whereas they did not appear when regressing on the mixture of both indoor-generated and outdoor-generated particles is confirmation that our division into the two components using the sulfur ratios was successful.

We found several variables that were important in affecting air exchange rates. Opening windows was the single most important variable, as has been shown in previous studies (Howard-Reed et al., 2002). The indoor-outdoor temperature difference (which affects the indoor-outdoor pressure difference and therefore the driving force for air exchange) was the next most powerful variable. The coefficient for this variable (0.04 ach/°C) was of similar magnitude to that found by Wallace et al. (2002). The number of people in the home and the presence of pets, both significant variables in this study but not often found in other studies, contribute to increased air exchange through the increased number of times going in and out of the house. The use of a kitchen exhaust fan was associated with an increase of 0.14 ach. This also compares well with the finding of a 0.8 ach increase associated with use of an attic exhaust fan (Wallace et al., 2002), given the much longer periods that an attic exhaust fan may run compared to a kitchen exhaust fan.

The single strongest variable affecting the infiltration factor F_{inf} is the air exchange rate, as can be seen from the equation for F_{inf} and the observed relationship shown in Figure 3-27. All other variables entering our analysis are either highly correlated with air exchange rate or appear to be artifacts arising from the unequal monitoring of homes on different days.

Chapter 5
Conclusions

The sulfur measurements showed excellent agreement between the personal samplers and the fixed indoor-outdoor Harvard Impactors. More than 160 co-located measurements both indoors and outdoors resulted in slopes insignificantly different from 1 and intercepts insignificantly different from zero, with R^2 values of 0.97 for both indoor and outdoor locations. Uncertainties of individual sulfur measurements were estimated at 8%.

The data appear to be consistent with the hypothesis that there are few indoor sources of sulfur. Only about 1% of measurements showed higher levels indoors than out. Although nearly all regressions of indoor sulfur vs. outdoor sulfur gave positive intercepts, these intercepts were often relatively small and could be due to measurement error. Therefore we accepted the indoor/outdoor sulfur ratio as our best estimate of F_{inf} for $PM_{2.5}$.

Estimates of F_{inf} averaged over all seasons varied over a large range by household (0.26-0.87), but half of the homes were within 20% of the median value of 0.59. The infiltration factor was significantly lower for many homes in the summer season, presumably due to the closed windows and increased recirculation and filtration of air associated with the use of air conditioners. Evidence from measurements of iron and silicon suggested that the estimates of the infiltration factor using the sulfur indoor/outdoor ratio are likely to overestimate the influence of outdoor air due to the low deposition velocity of sulfur compared to other likely constituents of $PM_{2.5}$. However, other considerations lead us to think that the overestimate is not large; also, it would not affect the relative ranking of the homes with respect to the outdoor contributions to indoor $PM_{2.5}$.

In general, the outdoor exposure factor F_{pex} was very similar to F_{inf}. This is expected, since persons spend most of their time indoors, but the very close agreement is also an indication that the sulfur measurements were reproducible and agreed well even though the personal monitor collected 10 times less material than the indoor/outdoor monitors.

F_{pex} was usually smaller by a few percent than F_{inf}, although a simplistic application of time-activity budgets indicates that it should be larger by about 6 or 7% for persons spending a typical amount of time outdoors and in vehicles. This may be due to time spent in unmonitored indoor locations (e.g., office buildings, department stores) that have mechanical ventilation, recirculation, and filtration, thus lowering exposure to sulfur while in those locations. The fact that F_{pex} was slightly larger than F_{inf} during the summer season, when many homes were closed and using recirculated and filtered air, supports this hypothesis. We conclude that the model advocated in earlier publications for determining F_{pex} by using time-activity budgets is not useful for studies that do not measure indoor concentrations in schools, workplaces, shopping malls, and other locations where persons spend substantial amounts of time. The preferred way to estimate F_{pex} is by using personal sulfur measurements, but lacking those, it may be just as useful to use F_{inf} as the best estimate of F_{pex}.

Regressions of indoor air concentrations due to particles of outdoor origin vs. outdoor concentrations had generally high R^2 values, as did regressions of personal exposure to particles of outdoor origin vs. outdoor concentrations. However, this result is partially due to the many assumptions in our approach leading to a simple linear relation between indoor concentrations of particles of outdoor origin and outdoor concentrations. Therefore these estimates should not be treated as best estimates, but rather as upper bounds to the actual amount of variance in personal exposure to $PM_{2.5}$ of outdoor origin that can be explained by outdoor measurements at the central site.

We investigated whether particle measurements alone could be used to estimate the infiltration factor. However, comparisons with the sulfur ratio method indicated that particle measurements

alone cannot be used to give reliable estimates of F_{inf}.

The two unmeasured parameters contributing to F_{inf} (P and k) were estimated using both linear and nonlinear approaches. The median value for the penetration coefficient P was 0.81 (interquartile range 0.66 to 0.90). The median for the deposition rate k was 0.24 h^{-1} (interquartile range 0.12 to 0.35 h^{-1}). Recall that these values of k and P are for particles in the same size range as sulfur particles. However, we conclude that the unphysical values obtained for P in some cases casts doubt on all the estimates of P and k for individual homes; we are unable to validate these estimates.

Air exchange rates were found to depend primarily on opening windows and on the absolute indoor-outdoor temperature difference. Other contributors included use of a kitchen exhaust fan, presence of a vented clothes dryer, and number of persons in a household. The infiltration factor was primarily dependent on air exchange, as expected from the basic equilibrium equation.

Regressions of indoor concentrations and personal exposures showed considerably improved R^2 estimates by consideration of questionnaire variables. In particular, smoking, cooking, number of persons in a household, and burned food were important contributors. Air exchange rates were also very important variables, but only after the total indoor particle concentration had been split into indoor-generated and outdoor-generated portions. The strong effect of air exchange (increasing outdoor-generated particle concentrations and decreasing indoor-generated particle concentrations) was an important confirmation of the success of our efforts to resolve these two contributors to total indoor particle levels.

Chapter 6
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Appendix

In this Appendix we document the seasonal variation of the infiltration factor F_{inf} and the ambient exposure factor F_{pex}. Table A-1 presents the seasonal average of F_{inf} and the seasonal harmonic average of the air exchange rate for each home.

Table A-2 compares the seasonal average indoor/outdoor ratio for sulfur to the slopes obtained when regressing indoor on outdoor sulfur. The slope is usually smaller than the ratio, averaging 91% of its value, and also has a wider range, with a standard deviation almost twice that of the ratio (0.26 compared to 0.14). This is to be expected, since measurement error causes lower slopes in regressions, and the regressions are based on a maximum of 7 values (with a few exceptions), and usually only 5, 6, or 7; this leads to more variability than is desirable.

Table A-3 compares the personal/outdoor ratios (F_{pex}) averaged over one season with the slopes determined from regressions of personal on outdoor sulfur. As with the F_{inf} values in Table A-2, the slopes are lower than the ratios and once again the standard deviations are nearly twice as high (0.19 compared to 0.11).

The results of regressing the outdoor contribution to personal exposures (by season) on the outdoor monitors are provided in Figures A-1 to A-3, which provide boxplots of the adjusted R^2 values. Note that the definition of the adjusted R^2 parameter allows negative values. The range of these seasonally calculated R^2 values is quite a bit larger than the range of the year-round R^2 and suggests that the increased variability due to the smaller number of observations in each regression outweighs whatever advantage was gained in looking at the smaller time period.

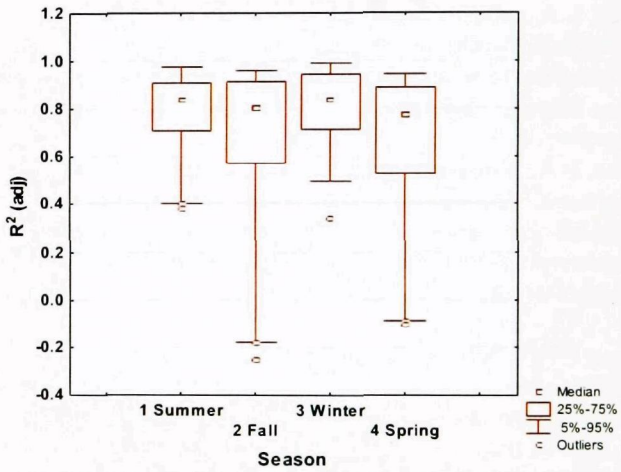

Figure A-1. Adjusted R^2 values from regressing the outdoor contribution to personal exposure on outdoor $PM_{2.5}$ measurements just outside the house.

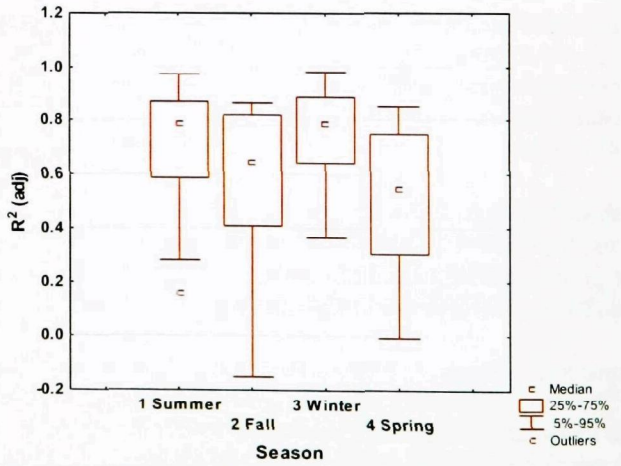

Figure A-2. Adjusted R^2 values from regressing the outdoor contribution to personal exposure on outdoor $PM_{2.5}$ Harvard Impactor (HI) measurements at the central site.

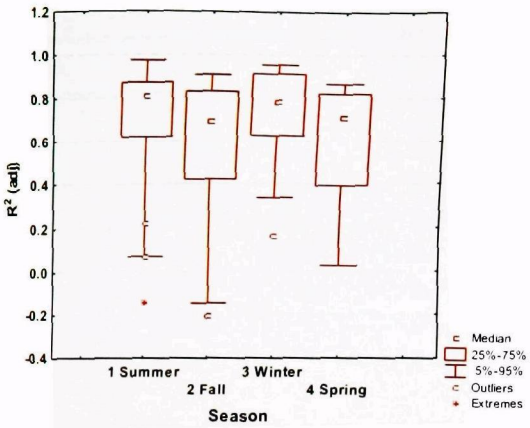

Figure A-3. Adjusted R^2 values from regressing the outdoor contribution to personal exposure on outdoor $PM_{2.5}$ Federal Reference Method (FRM) measurements at the central site.

Table A-1. Values of the Average Sulfur Indoor/Outdoor Ratio (F_{inf}) and the Air Exchange Rates by House and by Season

House	Season	N	S In/Out[a]	SE	Airex[b]	SE
1	Summer	8	0.58	0.03	0.63	0.10
1	Fall	6	0.64	0.03	0.79	0.11
1	Winter	2	0.76	0.06	1.29	0.19
1	Spring	6	0.65	0.03	0.82	0.11
2	Summer	6	0.59	0.03	0.46	0.11
2	Fall	4	0.53	0.04	0.81	0.14
2	Winter	7	0.64	0.03	1.35	0.10
2	Spring	6	0.66	0.03	0.71	0.11
3	Summer	8	0.78	0.03	1.05	0.10
3	Fall	6	0.70	0.03	0.50	0.11
3	Winter	7	0.69	0.03	0.87	0.10
3	Spring	7	0.76	0.03	1.07	0.10
4	Summer	3	0.32	0.05	0.21	0.16
4	Fall	7	0.48	0.03	0.26	0.10
4	Winter	4	0.51	0.04	0.34	0.14
4	Spring	7	0.45	0.03	0.22	0.10
5	Summer	7	0.51	0.03	0.43	0.10
5	Fall	6	0.65	0.03	0.36	0.11
5	Spring	7	0.74	0.03	0.60	0.10
6	Summer	7	0.60	0.03	0.45	0.10
6	Fall	5	0.65	0.04	0.57	0.12
6	Winter	7	0.58	0.03	0.60	0.10
6	Spring	7	0.61	0.03	0.58	0.10
7	Summer	7	0.67	0.03	0.56	0.10
7	Fall	7	0.83	0.03	1.06	0.10
7	Winter	5	0.75	0.04	1.50	0.12
7	Spring	6	0.86	0.03	1.94	0.11
8	Summer	6	0.36	0.03	0.24	0.11
9	Summer	6	0.48	0.03	0.23	0.11
9	Fall	5	0.55	0.04	0.40	0.12
9	Winter	5	0.62	0.04	0.84	0.12
9	Spring	5	0.69	0.04	0.66	0.12
10	Summer	6	0.46	0.03	0.30	0.11
10	Fall	7	0.57	0.03	0.54	0.10
10	Winter	5	0.56	0.04	0.67	0.12
10	Spring	7	0.64	0.03	0.66	0.10
11	Summer	6	0.69	0.03	0.39	0.11
11	Winter	2	0.67	0.06	4.49	0.19
12	Summer	7	0.40	0.03	0.27	0.10
12	Fall	7	0.57	0.03	0.45	0.10
12	Winter	7	0.62	0.03	0.64	0.10
12	Spring	7	0.54	0.03	0.33	0.10
13	Summer	7	0.37	0.03	0.22	0.10
14	Summer	7	0.41	0.03	0.23	0.10

Table A-1. Continued

House	Season	N	S In/Out[a]	SE	Airex[b]	SE
14	Fall	6	0.46	0.03	0.33	0.11
14	Winter	7	0.45	0.03	0.51	0.10
14	Spring	7	0.47	0.03	0.35	0.10
15	Summer	7	0.61	0.03	0.42	0.10
15	Fall	7	0.73	0.03	0.49	0.10
15	Winter	3	0.76	0.05	0.72	0.16
15	Spring	7	0.77	0.03	0.56	0.10
16	Summer	7	0.59	0.03	0.41	0.10
16	Fall	7	0.71	0.03	0.65	0.10
16	Winter	7	0.68	0.03	1.28	0.10
16	Spring	7	0.61	0.03	0.95	0.10
17	Summer	7	0.42	0.03	0.11	0.10
17	Fall	5	0.53	0.04	0.14	0.12
17	Winter	1	0.43	0.08	0.51	0.27
17	Spring	6	0.59	0.03	0.27	0.11
18	Summer	6	0.52	0.03	0.30	0.11
19	Summer	6	0.62	0.03	0.61	0.11
19	Fall	7	0.79	0.03	1.33	0.10
19	Winter	7	0.84	0.03	1.54	0.10
19	Spring	7	0.73	0.03	0.93	0.10
20	Summer	7	0.56	0.03	0.24	0.10
20	Fall	6	0.43	0.03	0.23	0.11
20	Winter	7	0.47	0.03	0.52	0.10
20	Spring	7	0.51	0.03	0.35	0.10
21	Summer	7	0.43	0.03	0.28	0.10
21	Fall	6	0.68	0.03	0.53	0.11
21	Winter	6	0.67	0.03	0.63	0.11
21	Spring	7	0.67	0.03	0.58	0.10
22	Summer	6	0.44	0.03	0.27	0.11
22	Fall	7	0.65	0.03	0.48	0.10
23	Summer	7	0.43	0.03	0.57	0.10
23	Fall	6	0.55	0.03	1.02	0.11
24	Summer	7	0.41	0.03	0.33	0.10
24	Fall	7	0.55	0.03	0.35	0.10
24	Winter	7	0.60	0.03	0.51	0.10
24	Spring	6	0.64	0.03	0.69	0.11
25	Summer	2	0.46	0.06	0.27	0.19
25	Fall	6	0.81	0.03	0.67	0.11
26	Summer	6	0.61	0.03	0.50	0.11
26	Fall	6	0.78	0.03	0.79	0.11
26	Winter	5	0.70	0.04	1.15	0.12
26	Spring	5	0.73	0.04	0.79	0.12
27	Summer	6	0.54	0.03	0.61	0.11
27	Fall	7	0.77	0.03	1.25	0.10

Table A-1. Continued

House	Season	N	S In/Out[a]	SE	Airex[b]	SE
27	Winter	7	0.66	0.03	2.61	0.10
27	Spring	7	0.68	0.03	0.67	0.10
28	Summer	5	0.41	0.04	0.28	0.12
28	Fall	7	0.65	0.03	0.47	0.10
28	Winter	6	0.61	0.03	0.86	0.11
28	Spring	6	0.62	0.03	0.37	0.11
29	Summer	13	0.40	0.02	0.31	0.08
29	Fall	6	0.59	0.03	0.42	0.11
29	Winter	5	0.75	0.04	0.61	0.12
29	Spring	7	0.70	0.03	0.53	0.10
31	Summer	7	0.97	0.03	2.77	0.10
31	Fall	7	0.80	0.03	1.11	0.10
32	Summer	7	0.25	0.03	0.23	0.10
32	Fall	7	0.40	0.03	0.27	0.10
32	Winter	7	0.48	0.03	0.33	0.10
32	Spring	7	0.31	0.03	0.25	0.10
33	Summer	5	0.23	0.04	0.15	0.12
33	Fall	5	0.35	0.04	0.25	0.12
33	Winter	4	0.49	0.04	1.12	0.14
33	Spring	6	0.40	0.03	0.64	0.11
34	Summer	7	0.54	0.03	0.36	0.10
34	Fall	3	0.74	0.05	0.45	0.16
36	Summer	5	0.25	0.04	0.25	0.12
37	Fall	5	0.58	0.04	0.27	0.12
37	Winter	7	0.66	0.03	0.69	0.10
37	Spring	7	0.55	0.03	0.30	0.10
38	Fall	3	0.52	0.05	0.18	0.16
38	Winter	5	0.64	0.04	0.54	0.12
38	Spring	7	0.50	0.03	0.26	0.10
	Mean	720	0.59		0.64	
	SD		0.14		0.56	

[a] Average ratio of indoor/outdoor sulfur concentrations
[b] Harmonic average of air exchange rate (h^{-1})

Table A-2. Comparison of Seasonal Average Sulfur Indoor/Outdoor Ratios (Sin/Sout) with Slopes of Regressions of Sin on Sout

House	Season	N	Sin/Sout (F_{inf})	SE	Intercept	SE	p (Int)	Slope	SE	p (Slope)	R^2 (adj.)	Ratio[a]
1	S	8	0.58	0.01	99	111	0.41	0.52	0.06	0.0001	0.92	0.90
1	F	6	0.64	0.03	96	101	0.40	0.57	0.06	0.0008	0.94	0.88
1	W	7	0.66	0.06	-64	210	0.77	0.72	0.15	0.0045	0.79	1.09
1	V	6	0.65	0.03	106	135	0.48	0.57	0.09	0.0027	0.89	0.87
2	S	6	0.59	0.04	552	222	0.07	0.30	0.10	0.0447	0.60	0.51
2	F	7	0.51	0.02	18	153	0.91	0.51	0.05	0.0001	0.95	0.99
2	W	7	0.64	0.02	39	113	0.75	0.60	0.09	0.0010	0.88	0.94
2	V	6	0.66	0.01	22	77	0.79	0.64	0.06	0.0005	0.95	0.97
3	S	8	0.78	0.03	537	193	0.03	0.48	0.10	0.0027	0.77	0.62
3	F	7	0.71	0.02	60	173	0.74	0.68	0.07	0.0002	0.94	0.96
3	W	7	0.69	0.04	190	88	0.08	0.54	0.06	0.0002	0.94	0.78
3	V	7	0.76	0.02	365	119	0.03	0.62	0.04	0.0001	0.97	0.81
4	S	3	0.32	0.06	880	108	0.08	0.00	0.03	0.9711	-1.00	0.00
4	F	7	0.48	0.02	104	157	0.54	0.42	0.07	0.0014	0.87	0.88
4	W	6	0.50	0.02	45	116	0.72	0.47	0.07	0.0025	0.90	0.94
4	V	7	0.45	0.06	166	87	0.12	0.31	0.04	0.0004	0.92	0.68
5	S	7	0.51	0.01	198	105	0.12	0.44	0.04	0.0001	0.96	0.85
5	F	6	0.65	0.04	-250	222	0.32	0.83	0.15	0.0055	0.85	1.29
5	W	7	0.71	0.05	-68	185	0.73	0.77	0.13	0.0018	0.85	1.09
5	V	7	0.74	0.08	-148	403	0.73	0.81	0.17	0.0050	0.78	1.10
6	S	7	0.60	0.03	164	270	0.57	0.54	0.10	0.0030	0.82	0.89
6	F	7	0.63	0.02	-100	187	0.62	0.67	0.06	0.0001	0.95	1.07
6	W	7	0.58	0.01	-63	47	0.24	0.63	0.03	0.0001	0.99	1.08
6	V	7	0.61	0.01	52	66	0.47	0.57	0.04	0.0001	0.97	0.93
7	S	7	0.67	0.02	37	238	0.88	0.66	0.07	0.0002	0.95	0.98
7	F	7	0.83	0.02	-288	54	0.00	0.97	0.02	0.0001	1.00	1.17
7	W	7	0.76	0.02	-3	53	0.96	0.76	0.05	0.0001	0.97	1.00
7	V	7	0.84	0.03	-257	119	0.08	1.07	0.10	0.0001	0.95	1.28
8	S	6	0.36	0.02	291	247	0.31	0.27	0.06	0.0141	0.77	0.75
9	S	6	0.48	0.02	-14	227	0.95	0.49	0.07	0.0017	0.92	1.01
9	F	6	0.57	0.05	284	223	0.27	0.34	0.17	0.1111	0.39	0.59
9	W	7	0.61	0.01	23	26	0.40	0.58	0.03	0.0001	0.99	0.95
9	V	5	0.69	0.10	263	477	0.62	0.53	0.16	0.0484	0.70	0.77
10	S	6	0.46	0.01	76	164	0.67	0.43	0.06	0.0014	0.92	0.94
10	F	7	0.57	0.03	45	94	0.66	0.54	0.06	0.0003	0.93	0.94
10	W	7	0.60	0.04	192	119	0.17	0.37	0.13	0.0389	0.53	0.61
10	V	7	0.64	0.06	1002	266	0.01	0.22	0.09	0.0518	0.48	0.35
11	S	6	0.69	0.03	382	368	0.36	0.55	0.12	0.0098	0.80	0.80
11	W	2	0.67	0.01								
12	S	7	0.40	0.02	253	184	0.23	0.31	0.06	0.0033	0.82	0.78
12	F	7	0.57	0.02	69	90	0.47	0.51	0.05	0.0002	0.94	0.89
12	W	7	0.62	0.03	23	57	0.70	0.60	0.04	0.0001	0.97	0.96
12	V	7	0.54	0.06	132	100	0.24	0.41	0.04	0.0002	0.94	0.76
13	S	7	0.37	0.03	60	78	0.48	0.32	0.05	0.0010	0.88	0.87
14	S	7	0.41	0.04	155	113	0.23	0.28	0.07	0.0111	0.71	0.69

Table A-2. Continued

House	Season	N	Sin/Sout (F_{int})	SE	Intercept	SE	p (Int)	Slope	SE	p (Slope)	R^2 (adj.)	Ratio[a]
14	F	6	0.46	0.04	54	111	0.65	0.42	0.07	0.0040	0.87	0.91
14	W	7	0.45	0.04	-71	139	0.63	0.53	0.12	0.0062	0.77	1.17
14	V	7	0.47	0.02	316	164	0.11	0.33	0.07	0.0050	0.78	0.69
15	S	7	0.61	0.04	85	116	0.50	0.55	0.07	0.0007	0.90	0.89
15	F	7	0.73	0.02	-15	60	0.82	0.75	0.04	0.0001	0.98	1.02
15	W	6	0.66	0.05	-212	145	0.22	0.91	0.15	0.0033	0.88	1.38
15	V	7	0.77	0.03	-116	147	0.34	0.88	0.09	0.0002	0.93	1.15
16	S	7	0.59	0.02	246	126	0.11	0.50	0.04	0.0001	0.96	0.83
16	F	7	0.71	0.03	-49	106	0.67	0.45	0.07	0.0001	0.96	0.63
16	W	7	0.68	0.01	34	15	0.08	0.64	0.01	0.0001	1.00	0.94
16	V	7	0.61	0.02	-217	77	0.04	0.81	0.06	0.0001	0.96	1.31
17	S	7	0.42	0.03	4	340	0.99	0.42	0.12	0.0190	0.64	1.00
17	F	6	0.52	0.05	285	198	0.22	0.35	0.07	0.0090	0.81	0.67
17	W	7	0.55	0.06	-31	222	0.89	0.59	0.19	0.0250	0.60	1.07
17	V	6	0.59	0.03	102	87	0.31	0.51	0.05	0.0006	0.95	0.88
18	S	6	0.52	0.03	226	173	0.26	0.42	0.06	0.0025	0.90	0.80
19	S	6	0.62	0.03	-118	510	0.83	0.65	0.14	0.0086	0.82	1.06
19	F	7	0.79	0.01	-19	31	0.57	0.80	0.02	0.0001	1.00	1.02
19	W	7	0.84	0.05	135	115	0.29	0.69	0.10	0.0009	0.89	0.82
19	V	7	0.73	0.02	-102	504	0.85	0.77	0.18	0.0084	0.74	1.05
20	S	7	0.56	0.04	755	197	0.01	0.32	0.05	0.0016	0.86	0.58
20	F	6	0.43	0.02	-25	108	0.82	0.45	0.06	0.0017	0.92	1.04
20	W	7	0.47	0.02	-33	59	0.60	0.50	0.05	0.0001	0.95	1.07
20	V	7	0.51	0.01	-152	192	0.46	0.57	0.07	0.0006	0.91	1.12
21	S	7	0.43	0.02	-151	167	0.41	0.48	0.05	0.0001	0.95	1.11
21	F	7	0.70	0.03	-73	312	0.82	0.73	0.11	0.0012	0.88	1.04
21	W	6	0.67	0.03	173	66	0.06	0.50	0.05	0.0006	0.95	0.75
21	V	7	0.67	0.02	-55	91	0.58	0.72	0.08	0.0003	0.93	1.08
22	S	6	0.44	0.03	586	177	0.03	0.22	0.06	0.0247	0.69	0.49
22	F	7	0.65	0.03	89	128	0.52	0.60	0.06	0.0001	0.95	0.92
23	S	7	0.43	0.02	88	168	0.62	0.40	0.04	0.0003	0.93	0.93
23	F	6	0.55	0.02	15	63	0.82	0.54	0.04	0.0002	0.97	0.97
24	S	7	0.41	0.02	248	174	0.21	0.32	0.06	0.0035	0.81	0.77
24	F	7	0.55	0.03	125	127	0.37	0.48	0.05	0.0002	0.94	0.86
24	W	7	0.60	0.04	67	66	0.35	0.51	0.05	0.0002	0.94	0.86
24	V	6	0.64	0.04	-129	218	0.59	0.76	0.19	0.0150	0.76	1.20
25	S	2	0.46	0.01								
25	F	6	0.81	0.02	-95	173	0.62	0.86	0.08	0.0003	0.96	1.06
26	S	6	0.61	0.01	-66	212	0.77	0.63	0.05	0.0002	0.97	1.03
26	F	6	0.78	0.02	-63	349	0.87	0.80	0.11	0.0016	0.92	1.03
26	W	7	0.69	0.02	100	87	0.30	0.57	0.10	0.0020	0.85	0.83
26	V	5	0.73	0.04	-680	1169	0.60	0.99	0.43	0.1038	0.52	1.34
27	S	7	0.54	0.01	-81	73	0.32	0.57	0.02	0.0001	0.99	1.05
27	F	7	0.77	0.03	-2	126	0.99	0.77	0.08	0.0002	0.94	1.00
27	W	7	0.66	0.01	-3	50	0.96	0.66	0.04	0.0001	0.98	1.00

Table A-2. Continued

House	Season	N	Sin/Sout (F_{inf})	SE	Intercept	SE	p (Int)	Slope	SE	p (Slope)	R^2 (adj.)	Ratio[a]
27	V	7	0.68	0.04	257	273	0.39	0.56	0.11	0.0040	0.80	0.82
28	S	5	0.41	0.02	126	241	0.64	0.37	0.07	0.0210	0.88	0.89
28	F	7	0.65	0.01	-20	25	0.46	0.67	0.01	0.0001	1.00	1.03
28	W	7	0.62	0.02	173	68	0.05	0.42	0.07	0.0024	0.84	0.67
28	V	6	0.62	0.05	980	319	0.04	0.14	0.15	0.4100	-0.03	0.22
29	S	13	0.40	0.02	89	210	0.68	0.36	0.06	0.0001	0.73	0.90
29	F	7	0.61	0.05	182	311	0.58	0.47	0.22	0.0840	0.38	0.77
29	W	7	0.71	0.06	259	142	0.13	0.39	0.15	0.0508	0.48	0.55
29	V	7	0.70	0.03	135	167	0.46	0.60	0.10	0.0019	0 85	0.86
31	S	7	0.97	0.02	59	106	0.60	0.92	0.07	0.0001	0.97	0.95
31	F	7	0.80	0.03	-319	160	0.10	0.95	0.06	0.0001	0.97	1.19
32	S	7	0.25	0.02	2	71	0.97	0.25	0.05	0.0032	0.82	0.98
32	F	7	0.40	0.02	35	91	0.72	0.38	0.05	0.0005	0.91	0.94
32	W	7	0.48	0.03	-6	112	0.96	0.48	0.07	0.0011	0.88	1.01
32	V	7	0.31	0.02	335	129	0.05	0.16	0.05	0.0306	0.57	0 52
33	S	5	0.23	0.04	95	31	0.06	0.12	0.02	0.0078	0.91	0.55
33	F	6	0.40	0.07	454	373	0.29	0.05	0.27	0.8700	-0.24	0.12
33	W	5	0.49	0.05	-89	146	0.58	0.60	0.17	0.0350	0.76	1.23
33	V	7	0.42	0.04	165	83	0.10	0.29	0.03	0.0003	0.93	0.69
34	S	7	0.54	0.03	-41	174	0.83	0.56	0.06	0.0002	0.94	1.03
34	F	5	0.72	0.03	-21	331	0.95	0.74	0.11	0.0060	0.92	1.02
36	S	6	0.26	0.02	10	93	0.92	0.26	0.03	0.0017	0.92	0.99
37	F	6	0.59	0.04	-152	326	0.66	0.69	0.21	0.0290	0.67	1.17
37	W	7	0.66	0.03	-94	129	0.50	0.73	0.08	0.0003	0.93	1.10
37	V	7	0.55	0.03	565	113	0.00	0.30	0.04	0.0011	0.88	0.54
38	F	3	0.52	0.03	-23	87	0.84	0 54	0.07	0.0869	0.96	1.05
38	W	7	0.64	0.03	136	112	0.28	0 46	0 13	0.0172	0.65	0.73
38	V	7	0.50	0.01	96	59	0.17	0.41	0 05	0.0006	0.90	0.81
Mean		775	0.59	0.03	89	167	0.47	0.54	0.08	0.03	0.83	0.91
SD			0.14	0.02	241	137	0.30	0.20	0.06	0.13	0.25	0.26

[a] Ratio of slope to Sin/Sout.

Table A-3. Comparison of Seasonal Average Sulfur Personal/Outdoor Ratios (Spers/Sout) with Slopes of Regressions of Spers on Sout

Subject	Season	N	Spers/Sout (F_{pex})	SE	Slope	SE	p	Intercept	SE	p	R^2 (adj)	Ratio[a]
1	S	6	0 50	0 02	0 52	0 09	0 0049	-53	184	0 79	0.86	1.06
1	F	6	0 58	0 03	0.57	0 05	0.0003	17	80	0 84	0.96	0.98
1	W	7	0 59	0 05	0 66	0 14	0.0051	-76	197	0.71	0.78	1.11
1	V	6	0 65	0.05	0.54	0 11	0.0078	141	169	0 45	0 82	0 83
2	S	6	0 47	0 04	0.29	0 12	0 0683	345	254	0.25	0 51	0 62
2	F	6	0 48	0.03	0 46	0 09	0 0073	55	269	0.85	0.83	0.96
2	W	6	0 47	0 04	0.56	0 16	0.0240	-103	216	0.66	0 70	1 18
2	V	6	0 59	0 02	0 74	0 09	0.0012	-173	110	0.19	0.93	1.26
3	S	5	0.72	0 04	0 45	0.44	0 3799	439	736	0 59	0.01	0.63
3	F	7	0 64	0.04	0 68	0 12	0.0026	-95	304	0 77	0.83	1 06
3	W	7	0 60	0.02	0.50	0.03	0.0001	122	53	0.07	0.97	0 84
3	V	2	0.59	0 01								
4	S	2	0 41	0.01								
4	F	7	0.48	0.04	0 33	0 09	0 0150	282	218	0.25	0.67	0.69
4	W	7	0 50	0 05	0 35	0 13	0.0440	182	206	0 42	0.51	0 70
4	V	7	0 47	0 06	0.55	0 12	0.0051	-131	282	0.66	0.78	1.17
5	S	6	0 59	0 04	0 46	0.10	0 0085	307	279	0.33	0.82	0 78
5	F	7	0.65	0.05	0.66	0 23	0.0336	0	315	1 00	0 55	1 00
5	W	7	0 66	0 03	0 62	0.07	0 0004	61	105	0 59	0.92	0.93
5	V	7	0 82	0 04	0 79	0 06	0.0001	12	152	0 94	0.96	0.97
6	S	6	0 44	0.04	0 42	0.13	0 0327	21	369	0.96	0 65	0 96
6	F	7	0 55	0 03	0 68	0 08	0.0004	-341	254	0.24	0 92	1.24
6	W	7	0.50	0 02	0 55	0.04	0 0001	-76	63	0.28	0.97	1 12
6	V	7	0 54	0 02	0 45	0 05	0.0003	109	79	0 23	0.93	0.85
7	S	6	0 54	0 02	0 50	0 11	0 0111	131	429	0.77	0 79	0 93
7	F	7	0 66	0 05	0 99	0 06	0 0001	-680	150	0 01	0 98	1 51
7	W	7	0 56	0 04	0 78	0 12	0 0013	-180	119	0 19	0 87	1 38
7	V	7	0 70	0.04	1 00	0 11	0 0003	-334	137	0 06	0 93	1 44
8	S	6	0 36	0 01	0 33	0 04	0 0007	77	138	0.61	0 94	0 93
9	S	6	0 47	0 02	0 43	0.06	0 0016	120	198	0.58	0.92	0 92
9	F	7	0 58	0 07	0 56	0 30	0 1230	31	400	0.94	0.29	0 96
9	W	7	0 57	0.07	0 46	0 20	0 0660	100	192	0.62	0.43	0 80
9	V	5	0 70	0 05	0 72	0 14	0 0128	-54	393	0 90	0.87	1 03
10	S	5	0 44	0 02	0 48	0 05	0 0031	-91	161	0 61	0 95	1 08
10	F	7	0 46	0 03	0.36	0.05	0 0008	104	76	0 23	0.90	0 79
10	W	7	0 51	0 03	0 48	0 08	0 0023	17	76	0 83	0 84	0.95
10	V	2	0 62	0 25								
11	S	6	0 76	0 04	0 49	0 12	0 0148	746	370	0 11	0 76	0 65
11	W	3	0 62	0 05	0 77	0 13	0 1075	-118	127	0.52	0.94	1 23
12	S	7	0 40	0 02	0.28	0 04	0 0015	337	135	0.05	0 87	0 69
12	F	7	0 51	0 02	0 45	0 06	0 0009	72	105	0.52	0 89	0 87
12	W	7	0 56	0.06	0 60	0 09	0 0012	-56	129	0 68	0 88	1 07
12	V	6	0 44	0 03	0 42	0 05	0 0008	10	121	0 94	0 94	0 95
13	S	7	0 43	0 04	0 39	0 10	0 0094	37	159	0 82	0.72	0.92

Table A-3. *Continued*

Subject	Season	N	Spers/Sout (F_{pex})	SE	Slope	SE	p	Intercept	SE	p	R^2 (adj)	Ratio[a]
14	S	7	0.40	0.02	0.35	0.05	0.0011	62	83	0.49	0.88	0.87
14	F	7	0.43	0.02	0.51	0.05	0.0001	-99	68	0.21	0.96	1.21
14	W	6	0.43	0.04	0.65	0.14	0.0097	-247	178	0.24	0.80	1.51
14	V	7	0.48	0.03	0.23	0.05	0.0084	526	132	0.01	0.74	0.49
15	S	6	0.59	0.03	0.69	0.08	0.0011	-129	137	0.40	0.93	1 16
15	F	7	0.61	0.02	0.67	0.03	0.0001	-56	45	0.27	0.99	1.09
15	W	7	0.61	0.05	0.86	0.14	0.0019	-219	146	0.19	0.85	1.41
15	V	7	0.64	0.03	0.81	0.05	0.0001	-224	82	0.04	0.97	1.26
16	S	6	0.57	0.01	0.53	0.06	0.0008	107	177	0.58	0.94	0.93
16	F	7	0.66	0.03	0.66	0.06	0.0001	-16	100	0.88	0.95	1.01
16	W	7	0.57	0.03	0.46	0.04	0.0001	107	47	0.07	0.96	0.80
16	V	7	0.56	0.03	0.90	0.05	0.0001	-389	59	0.00	0.98	1.62
17	S	6	0.44	0.03	0.51	0.09	0.0053	-176	271	0.55	0.85	1.17
17	F	6	0.43	0.06	0.27	0.08	0.0293	250	216	0.31	0.67	0.61
17	W	7	0.46	0.06	0.47	0.19	0.0574	6	228	0.98	0.46	1.01
17	V	6	0.57	0.03	0.55	0.08	0.0023	34	134	0.81	0.90	0.95
18	S	6	0.51	0.02	0.47	0.07	0.0028	76	201	0.73	0.89	0.93
19	S	6	0.59	0.03	0.61	0.11	0.0055	-49	417	0.91	0.85	1.03
19	F	7	0.72	0.01	0.68	0.02	0.0001	45	41	0.32	0.99	0.95
19	W	7	0.72	0.03	0.66	0.07	0.0002	49	82	0.57	0.94	0.92
19	V	2	0.66	0.06								
20	S	7	0.55	0.03	0.42	0.07	0.0018	412	265	0.18	0.85	0.77
20	F	6	0.45	0.03	0.39	0.09	0.0103	69	156	0.68	0.80	0.88
20	W	7	0.39	0.02	0.50	0.04	0.0001	-102	44	0.07	0.97	1.26
20	V	2	0.52	0.03								
21	S	7	0.55	0.03	0.54	0.07	0.0007	33	265	0.91	0.90	0.98
21	F	7	0.74	0.01	0.71	0.03	0.0001	81	97	0.44	0.99	0.95
21	W	7	0.75	0.05	0.52	0.08	0.0015	197	98	0.10	0.87	0.70
21	V	7	0.73	0.05	1.21	0.15	0.0004	-505	167	0.03	0.92	1.66
22	S	6	0.48	0.04	0.22	0.11	0.1198	660	323	0.11	0.37	0.47
22	F	7	0.60	0.03	0.68	0.05	0.0001	-92	111	0.45	0.97	1.12
23	S	7	0.47	0.03	0.49	0.13	0.0120	-66	473	0.89	0.70	1.04
23	F	7	0.50	0.02	0.46	0.04	0.0001	39	62	0.56	0.95	0.92
24	S	7	0.53	0.02	0.64	0.04	0.0001	-275	114	0.06	0.98	1.20
24	F	6	0.53	0.04	0.48	0.04	0.0002	71	99	0.52	0.97	0.90
24	W	6	0.63	0.10	0.49	0.16	0.0377	125	215	0.59	0.63	0.79
24	V	4	0.64	0.03	0.67	0.16	0.0547	-32	193	0.88	0.84	1.05
25	S	7	0.52	0.02	0.49	0.07	0.0012	68	278	0.82	0.88	0.95
25	F	7	0.70	0.02	0.86	0.06	0.0001	-317	141	0.07	0.97	1.23
26	S	6	0.52	0.02	0.47	0.05	0.0006	187	199	0.40	0.95	0.89
26	F	6	0.70	0.01	0.65	0.05	0.0002	136	157	0.43	0.97	0.93
26	W	7	0.56	0.02	0.42	0.09	0.0051	120	79	0.19	0.78	0.75
26	V	2	0.74	0.02								
27	S	7	0.46	0.01	0.43	0.03	0.0001	90	105	0.43	0.97	0.93

Table A-3. Continued

Subject	Season	N	Spers/Sout (F_{pex})	SE	Slope	SE	p	Intercept	SE	p	R^2 (adj)	Ratio[a]
27	F	7	0.62	0.01	0.60	0.04	0.0001	20	65	0.77	0.97	0.97
27	W	7	0.58	0.02	0.54	0.06	0.0004	49	75	0.54	0.92	0.92
27	V	7	0.54	0.03	0.50	0.10	0.0034	98	235	0.69	0.81	0.92
28	S	5	0.39	0.02	0.36	0.06	0.0089	109	211	0.64	0.90	0.92
28	F	7	0.50	0.02	0.54	0.06	0.0002	-61	108	0.60	0.94	1.10
28	W	7	0.50	0.02	0.33	0.08	0.0085	144	72	0.10	0.73	0.66
28	V	6	0.48	0.04	0.10	0.13	0.4929	785	282	0.05	-0.09	0.20
29	S	7	0.43	0.03	0.49	0.12	0.0090	-156	428	0.73	0.73	1.12
29	F	6	0.61	0.06	0.97	0.37	0.0571	-503	541	0.40	0.55	1.58
29	W	7	0.59	0.05	0.28	0.11	0.0522	252	100	0.05	0.47	0.47
29	V	7	0.62	0.06	0.72	0.20	0.0152	-150	325	0.66	0.67	1.17
31	S	7	0.91	0.02	0.94	0.06	0.0001	-49	87	0.60	0.98	1.04
31	F	7	0.66	0.03	0.76	0.07	0.0001	-207	183	0.31	0.95	1.15
32	S	7	0.47	0.03	0.56	0.06	0.0003	-110	98	0.31	0.93	1.20
32	F	7	0.53	0.04	0.47	0.09	0.0033	74	170	0.68	0.82	0.88
32	W	7	0.60	0.05	0.38	0.09	0.0099	267	145	0.13	0.72	0.63
32	V	7	0.38	0.04	0.15	0.19	0.4704	516	466	0.32	-0.07	0.39
33	S	4	0.30	0.08	0.16	0.06	0.1091	117	86	0.31	0.69	0.53
33	F	6	0.40	0.07	0.15	0.30	0.6425	318	416	0.49	-0.18	0.38
33	W	5	0.48	0.04	0.51	0.12	0.0233	-35	105	0.76	0.81	1.08
33	V	7	0.32	0.03	0.26	0.01	0.0001	81	37	0.08	0.98	0.80
34	S	7	0.52	0.02	0.54	0.04	0.0001	-38	130	0.78	0.96	1.03
34	F	6	0.74	0.04	0.72	0.14	0.0064	85	417	0.85	0.84	0.97
35	S	7	0.41	0.04	0.28	0.07	0.0122	313	219	0.21	0.69	0.67
36	S	6	0.45	0.03	0.51	0.09	0.0057	-133	256	0.63	0.85	1.13
37	F	7	0.55	0.09	0.26	0.33	0.4687	381	484	0.47	-0.07	0.46
37	W	7	0.54	0.02	0.60	0.10	0.0016	-83	151	0.61	0.86	1.12
37	V	7	0.53	0.03	0.32	0.07	0.0058	473	174	0.04	0.77	0.60
38	F	7	0.54	0.04	0.57	0.06	0.0002	-39	83	0.66	0.94	1.06
38	W	7	0.57	0.04	0.47	0.19	0.0566	79	160	0.64	0.46	0.82
38	V	7	0.52	0.04	0.36	0.18	0.0934	157	193	0.45	0.35	0.70
Mean		750	0.55	0.04	0.53	0.10	0.03	41	190	0.48	0.79	0.96
SD			0.11	0.03	0.19	0.07	0.10	231	125	0.29	0.24	0.22

[a] Ratio of slope to Spers/Sout.

Table A-4. Questionnaire Variables and Definitions

#	Variable Name	Definition	Units
1	Persdate	Julian date*100+Subject ID	
2	DateID	Date (YYYYMMDD)*100 + SUBJECT id	
3	EPA ID	epa id	
4	Subject	SUBJECT ID	
5	Cohort	Cohort (Cardiovascular or hypertensive)	
6	House	House ID	
7	Season	Season	
8	Date	Julian date	
9	Month	Month	
10	Day	Day	
11	Year	Year	
12	ambHI25	Central-site Harvard Impactor $PM_{2.5}$	$\mu g/m^3$
13	FRM25	Central-site Federal Reference Method $PM_{2.5}$	$\mu g/m^3$
14	Flaghi25	Validity code: 2 = valid; used when central-site $HI_{2.5}$ used in regressions	
15	FlagFRM	Validity code. 2 = valid; used when FRM is a variable in regressions	
16	Flagairex	Validity code: 2 = valid; used when airex is a variable in regressions	
17	persS	personal sulfur	ng/m^3
18	Sin	Indoor sulfur	ng/m^3
19	Sout	Outdoor sulfur	ng/m^3
20	Sinout	Indoor/outdoor sulfur ratio	
21	FlagSinout	Validity code: 2 = valid; used for all indoor/outdoor regressions	
22	SpersSout	personal/outdoor sulfur ratio	
23	Flagspersout	Validity code: 2 = valid; used for all personal/outdoor regressions	
24	PM25in	Indoor $PM_{2.5}$	$\mu g/m^3$
25	PM25out	Outdoor $PM_{2.5}$	$\mu g/m^3$
26	PM25pers	Personal $PM_{2.5}$	$\mu g/m^3$
27	Outcontin	Outdoor contribution to indoor $PM_{2.5}$ concentrations	$\mu g/m^3$
28	Outcontpers	Outdoor contribution to personal $PM_{2.5}$ concentrations	$\mu g/m^3$
29	Incontrib	Indoor-generated contribution to indoor $PM_{2.5}$ concentrations	$\mu g/m^3$
30	Perscontrib	Non-outdoor contribution to personal $PM_{2.5}$ concentrations	$\mu g/m^3$
31	Perscloud	Remainder after accounting for time-weighted indoor-outdoor exposures	$\mu g/m^3$
32	airex	air exchange rate	h^{-1}
33	Cleaning	Time spent cleaning	minutes
34	Grooming	Time spent grooming	minutes
35	Other_indoor	Time in other indoor locations	minutes
36	Other_loc	Time in other locations	minutes
37	Travel	Time spent in travel	minutes
38	Unknown	Time in Unknown location	minutes
39	cooking	Time spent cooking	minutes
40	outdoor	Time spent outdoors	minutes
41	smoke	Time spent near smokers	minutes
42	TempC	Outdoor Temperature (°C)	°C

Table A-4. Continued

#	Variable Name	Definition	Units
43	Tempdelta	Absolute Outdoor-Indoor Temp Difference using thermostat setting	°F
44	Numpeopl	Number of persons living in house	
45	numsmok	Number of smokers in house	
46	cigsmokd	Number of cigarettes smoked in house	
47	Mealsckd	Number of meals cooked	
48	Burning	Was food burned today?	
49	Exhstfan	Was exhaust fan used today?	
50	Candles	Were candles used today?	
51	CANDLDUR	Duration of candle use	minutes
52	Incense	Was incense used today?	
53	INCENDUR	Duration of incense use	minutes
54	Windopen	Were windows open today?	
55	windowall	Sum of products of open windows X width opened X duration open	inch-hours
56	spacehtr	Was a space heater used today?	
57	Cleaning_1	Did cleaning occur today?	
58	Pets	Any dog or cat pets?	
59	broil	broiled food today	
60	fry	fried food today	
61	grill	grilled food today	
62	sautee	sauteed food today	
63	sweep	swept floors today	
64	vacuum	vacuumed today	
65	dust	dusted today	
66	TYPE	Type of building (1 = detached, 2 = duplex, 4 = apartment, 6 = trailer)	
67	AGE	age of building (years)	years
68	BUSY_RD	high-traffic road nearby	
69	DIRT_RD	dir road nearby	
70	DUSTY_RD	Dust from nearby construction etc	
71	GAR_USE	Park cars in attached garage?	
72	A_C	air conditioning unit(s) in home	
73	FAN	whole-house or attic fan	
74	S_WIN	Storm windows (0 = none, 0.5 = partial, 1 = all)	
75	C_FUEL	cooking fuel (1 = gas, 2 = electricity)	
76	C_FAN	range hood?	
77	PILOT	Number of pilot lights (0-3)	
78	DRYER	clothes dryer?	
79	DRY_VENT	clothes dryer vented outside?	
80	VAC	vacuum bag type (0 = none, 1 = standard, 2 = HEPA)	
81	AREA	area of house (square footage)	$feet^2$
82	Rooms	Number of rooms	
83	FLRCOVav	Percent of floor covered by carpet	%
84	MILDEWavg	mildew noticed by technician	
85	DSTFACavg	Estimate of dusty/dirty condition by technician (0 = clean, 6 dusty)	