6 HUMAN EXPOSURE ASSESSMENT

The likelihood of an adverse response to particles is influenced by the degree of exposure, defined as any contact between the chemical at a specified concentration and the outer (e.g., skin) or inner (e.g., respiratory tract epithelium) surface of the human body (Sexton and Ryan, 1988; Lioy, 1990). Exposure implies the simultaneous occurrence of these two events in time and space (Ott, 1982). Changes in the degree of exposure are influenced by the duration (how long you are exposed), magnitude (concentration) and frequency (how often you are exposed) of exposure. Inhalation is the only exposure pathway considered in the assessment. This chapter will briefly: review the factors influencing exposure and the methods of determining air pollution exposure, review monitoring studies of personal and microenvironmental particle concentrations, present the results of a probabilistic exposure model, and discuss the relationship between current ambient levels of particles and human exposure in Canada.

6.1 CONCEPTS

Ambient particles are typically collected over a 24 h sampling period, although hourly average measurements of particle mass help illustrate the diurnal, weekly and seasonal variability in particle concentrations. Until more hourly data become available, expressing exposure over 24 h facilitates comparison of individual and population exposures with ambient particle levels.

Over a 24 h period, a person spends their time in many locations or microenvironments. US residents spend on average 21½ hours indoors, one hour outdoors, and 1½ hours in a vehicle. The data vary by age, gender and day of the week (US EPA, 1995b). For an individual, microenvironmental exposures lasting minutes to hours can be summed over one day and expressed by the following equation:

$$E_l = \sum_{j=1}^{J} c_{ij} t_{ij} \tag{6.1}$$

where E_i equals the exposure to individual *I*, $t_{i\,j}$ denotes the time the I-th individual spent in the j-th microenvironment, and $c_{i\,j}$ denotes the average pollutant concentration for the I-th individual in the j-th microenvironment (Ott, 1982; Duan, 1991). This equation is based upon the concept that concentrations of pollutants measured in locations where people are, multiplied by the time spent in each place, will approximate personal exposure (Lioy, 1990). Therefore, individual and population exposures will vary, depending on the time spent in various microenvironments (frequency and duration of exposure) and the particle concentrations in those microenvironments (magnitude of exposure).

With people spending, on average, less than 10% of their time outdoors (Özkaynak et al., 1995; U.S. EPA, 1995b), it is important to understand what influences the concentrations of particles in other microenvironments.

6.2 MICROENVIRONMENTAL PARTICLE SOURCES AND CONCENTRATIONS

Microenvironmental levels of particles are a function of: indoor sources, outdoor particle levels, the fraction of ambient air penetrating indoors, filtration, air-exchange rates, decay rates, and resuspension rates (Clayton et al., 1993, Wallace et al., 1993, Thatcher and Layton, 1995; US EPA, 1982, 1996). For a microenvironment with no indoor sources of particles, the concentration can be illustrated by the following equation:

$$\frac{C_{in}}{C_{out}} = \frac{Pa}{a+k} \tag{6.2}$$

where *p* is the penetration factor (a unitless measure of the ability of particles to penetrate the building envelope), *a* is the air exchange rate per hour (ach, or h^{-1}), and *k* is the particle decay rate (h^{-1}, a measurement of particle settling due to diffusion or sedimentation).

6.2.1 Sources

Indoor particles (Table 6.1) are attributed to six major source types: plant, animal, mineral, combustion, home/personal care and radioactive (Owen et al.,

Table 6.1 Sources of Indoor Particles (based on Owen et al., 1992)

Source Type	Description
plant	pollens, spores, molds, miscellaneous byproducts (finely ground grains, coffee, cornstarch)
animal	bacteria, viruses, hair, insect parts and byproducts, epithelial cells (eg. dandruff)
mineral	asbestos, talc, man-made mineral fibres, elemental particles (carbon)
combustion	tobacco smoke, cooking, heating appliances
home/personal care products	sprays, humidifiers
radioactive	radon progeny

1992). Plant materials are mostly of outdoor origin; pollens and spores settle out quickly but can be resuspended by vacuuming, sweeping or dusting. Animal and mineral particles have indoor and outdoor sources.

6.2.2 Penetration, Deposition and Resuspension

Penetration of ambient particles indoors is affected by several factors, including the physical and chemical characteristics of the particles and the mechanisms of home air exchange. Outdoor air enters a building through doors, windows and cracks, and as make-up air for heating and ventilating systems. Air-conditioned and energy efficient homes tend towards very low air exchange rates, while older homes are more “leaky”. Air exchange rates can range from a minimum 0.1 air changes per hour up to 10 changes per hour when doors and windows are open. The Exposure Factors Handbook (US EPA, 1995b) presents arithmetic and geometric mean (SD) estimates of air exchange rates for the United States (0.63 [0.65] and 0.46 [2.25] each respectively).

Several early studies indicated penetration is more effective for fine particles than coarse particles. Penetration factors of approximately 0.6-0.7 have been reported for $PM_{2.5}$, with the coarser fraction (2.5 - 10 µm) probably penetrating less effectively (Colome et al., 1992; Dockery and Spengler, 1981a; Koutrakis et al., 1992; Lioy et al., 1990; and Yocom, 1982). Dockery and Spengler (1981a) reported that the net effect of full air conditioning was to reduce penetration factors from approximately 0.7 to approximately 0.3. The results of the pTEAM study, however (Özkaynak et al., 1993) show that the penetration factor (mean, lower and upper 95% confidence levels), calculated using a nonlinear statistical approach, for both fine and coarse particles is very close to unity (1.00; [0.89; 1.11] and 1.00; [0.85; 1.15], respectively). The authors expect that penetration rates would be lower outside of Riverside because of tighter house construction. The results of Thatcher and Layton (1995) agree with the pTEAM results, with experimentally derived penetrations factors for both fine and coarse particles not significantly different from one (i.e., full penetration of outdoor air indoors). Despite these recent experimental results, current scientific thinking (US EPA, 1996) concludes that the penetration factor for submicron particles is greater than for $PM_{2.5}$ which is greater than for coarse particles.

Once indoors, ambient particles settle out quickly by gravity or electrostatic forces. Average decay rates, due to diffusion or sedimentation, were calculated as part of the pTEAM study (Özkaynak et al., 1993) for $PM_{2.5}$ ($0.39 \pm 0.16\ h^{-1}$), PM_{10} ($0.65 \pm 0.28\ h^{-1}$), and the coarse fraction ($1.01 \pm 0.43\ h^{-1}$). The decay rate of sulphate ($0.16\ h^{-1}$), an indicator of the fine fraction, was less than that of $PM_{2.5}$. Thatcher and Layton (1995) calculated similar deposition velocities, with particles of size range 1 to 5 µm depositing at a rate of $0.46\ h^{-1}$, and particles of size range 5 to 10 µm depositing at a rate of $1.36\ h^{-1}$. Particles of size range 0.1 to 1 µm have negligible settling velocities, while particles >10 µm normally settle out of the air.

Once deposited, resuspension of particles readily occurs in homes and buildings with occupants as a result of indoor activities. Thatcher and Layton (1995) studied resuspension and showed that particles greater than 5 µm are easily resuspended, while smaller particles (< 1 µm) are “not” resuspended. Particles ranging from 1 – 5 µm can be resuspended with vigorous activity.

6.2.3 Concentrations

Three large-scale studies conducted in the United States as well as multiple small-scale studies have measured indoor levels of particles in homes. Each study followed slightly different methodologies, using monitors with different cutpoints and sampling for variable periods, thereby precluding direct comparison between studies. Thus, the results of several of these studies are presented (Table 6.2) for information purposes, and to further the subsequent discussion of individual and population exposure estimates.

Harvard's Air Pollution Health Effects Study, also called the Six Cities Study, investigated the respiratory effects of air pollutants in six cities in the United States: Portage, Wisconsin; Topeka, Kansas; Kingston/Harriman, Tennessee; Watertown, Massachusetts; St. Louis, Missouri; and Steubenville, Ohio (Spengler et al., 1981). During the first phase of this study, which began in 1979, indoor and outdoor air was sampled every six days in eight to eleven homes in each city. Respirable particle measurements were made using a cyclone sampler with a 3.5 µm cutpoint operating at a flow rate of 1.7 L/min. The results from this large-scale study show that mean indoor concentrations were greater than outdoor levels (Table 6.2) (Dockery and Spengler, 1981a; Sexton et al., 1984; Spengler et al., 1985).

Similar results were reported from the New York State Energy Research and Development Authority [ERDA] Study (Sheldon et al., 1989), which examined the impact of four combustion products on indoor particle levels. Week-long $PM_{2.5}$ samples were collected in two indoor locations in 394 homes and outdoors in a subset of these homes using the Harvard impactor. The samplers were equipped with two oiled impactor plates, designed to minimize the collection of particles larger than 2.5 µm. Mean indoor concentrations were approximately double the outdoor levels.

A large-scale field monitoring pTEAM study was undertaken in Riverside, California, in the fall of 1990 (Pellizzari et al., 1992; Özkaynak et al., 1993; Wallace et al., 1993; US EPA, 1996). The main goal of pTEAM was to estimate the frequency distribution of exposures to PM_{10} for all non-smoking residents of Riverside (n = 139 000 ± 16 000) aged 10 and above. A pilot study was undertaken in Azusa, CA to test the sampling equipment (Wallace, 1996). A sample of 178 residents, selected using a probability-based design, participated by wearing a personal exposure monitor (PEM) for two consecutive 12 h periods (nominally 7 p.m. to 7 a.m. and 7 a.m. to 7 p.m.). The PEM consisted of a using a low-flow (4 L/min) impactor characterized by sharp cutpoints at 2.5 and 10.7 µm. Particles were collected on 37 mm Teflon filters mounted behind a greased impaction plate. The same monitor was modified to collect indoor and outdoor samples (stationary indoor monitors [SIMs] and stationary ambient monitors [SAMs], respectively) at participants' homes. Participants completed time-activity diaries and separately administered questionnaires about potential exposure to particle sources. The complete study design and sampling methods for the measurement of particles and subsequent elemental analyses are discussed elsewhere (Pellizzari et al., 1992; Özkaynak et al., 1993; Thomas et al., 1993).

Results from both the pilot study and the pTEAM study showed lower mean 24 h indoor concentrations of both PM_{10} and $PM_{2.5}$ when compared with outdoor levels (Pellizzari et al., 1992; Wallace, 1996). Similar results were reported from some small-scale studies ([Bahadori et al., 1995; Diemel et al., 1981; Janssen et al., 1995; Kulmala et al., 1987; cited in Wallace, 1996] Colome et al., 1992; Lioy et al., 1990; Morandi et al., 1988) while other studies reported higher mean indoor concentrations. However, in many cases, the range of particle concentrations indoors and outdoors are similar.

Table 6.2 Indoor Concentrations of Particles (μg/m^3)

Reference	Location	Size fraction	Time	N	Indoor mean (range)	Outdoor mean (range)
Dockery & Spengler, 1981a	Portage, WI	$PM_{3.5}$	annual 24 h (q 6 day)	68	20	13
	Topeka, KS		mean indoors		23	13
	Watertown, MA				31	
	Kingston/Harriman, TN	$PM_{2.5}$	annual mean outdoor		36	24
	St. Louis, MO				47	20
	Steubenville, OH				39	36
Sexton et al., 1984	Waterbury, VT	$PM_{3.5}$	24 h (8-8), every other day for two weeks	24	25 (6-69)	19 (6-30)
Spengler et al., 1985	Kingston/Harriman, TN	$PM_{2.5}$ out $PM_{3.5}$ in	24 h	101	42	18
Sheldon et al., 1989	Onondaga, NY	$PM_{2.5}$	collected during alt 15 min periods over 7 days, winter	433	36.7 (0.72-172)	16.8 (6.32-28.4)
	Suffolk, NY				46.4 (2.18-284)	21.8 (12-106)
[Wiener, 1988; Spengler et al., 1989; Özkaynak et al., 1990; Wiener et al., 1990][a]	Azusa, CA	PM_{10}	24 h (two 12 h periods)	9	58.7 (3.4)[b]	62.2 (3.5)
		$PM_{2.5}$			36.3 (2.6)	42.6 (3.0)
Pellizzari et al., 1992	Riverside, CA	PM_{10}	24 h (two 12 h periods)	178	79	91
		$PM_{2.5}$			43	50
[Diemel et al., 1981]	Arnhem, the Netherlands	< 3-4 μm, cutpoint not specified	24 h	101	140 (20-570)	64 (53.7-73.3)
[Kulmala et al., 1987]	Helsinki, Finland	fine (< 1 μm)		100	16 (4-67)[c]	20 (5-82)
		coarse (> 1 μm)			13 (3-63)	16 (3-91)
[Janssen et al., 1995]	Amsterdam	PM_{10}	24 h, home		35 (19-65)	42 (32-51)
[Bahadori et al., 1995]	Nashville, TN	$PM_{2.5}$	12 h daytime	10	22	33
			12 h nighttime		15	32
		PM_{10}	12 h daytime		16	23
			12 h nighttime		12	22
Lioy et al., 1990	Phillipsburg, NJ	PM_{10}	24 h	14	54 (25-60)	60 (12-165)
Colome et al., 1992	Orange County, CA	PM_{10}	24 h	8	32.7 (2.2)[a]	51.5 (4.9)

[a] bracketed studies cited in Wallace, 1996; [b] Mean (SE); [c] geometric mean (95% range).

6.2.4 Source Apportionment

Additional data analyses from many of the studies estimated source emission rates and the contributions of various sources to microenvironmental particle concentrations (Tables 6.3, 6.4, 6.5, and 6.6). The data from these tables are discussed in turn below.

Spengler et al., (1981) categorized the Harvard Six Cities data by number of smokers in the home and concluded cigarette smoke was the major source of indoor particles in smoking homes (Table 6.3). Similar results were presented from separate analysis of the Kingston/Harriman data (Letz et al., 1984; Spengler et al., 1985) and the Watertown data (Lebret et al., 1987). Dockery and Spengler (1981a) applied a simple conservation of mass model, to the phase one data set of the Six Cities Study, to determine contributions from three sources: cigarette smoking, heating and cooking. Processes affecting indoor levels were limited to infiltration, air-exchange rates, pollutant removal or decay and indoor sources. The impact of smoking one pack of cigarettes a day was significant, raising 24 h indoor respirable particle levels by approximately 18 µg/m^3, compared to an additional 15 µg/m^3 by other sources. In fully air-conditioned buildings, where infiltration rates are greatly reduced, smoking was estimated to contribute an additional 42 µg/m^3 of respirable particles. Smoking and gas stoves were also identified as possible sources of sulphates.

The second phase of the Harvard Six Cities Study took place between 1983 and 1988 (Neas et al., 1994). It involved 1,237 Caucasian never-smoking children, aged 7 to 11 years, which were drawn from a cohort of 6,273 children from the six cities. Indoor PM$_{2.5}$ levels in 200 to 300 homes selected from each city were measured over two consecutive one-week sampling periods during the summer and winter using the Harvard impactor. Sampling was continuous except for an 8 h period each weekday in which the child was at school. Neas et al. found that annual household average PM$_{2.5}$ concentrations were 31.2 µg/m^3 higher for the 580 children living in consistently smoking households than for the 470 children living in consistently non-smoking households. Based on the indoor measurements and questionnaire responses, they reported that the estimated number of cigarette packs smoked per day was a good indicator of annual average indoor PM$_{2.5}$ concentrations. A concentration of 30 µg/m^3 corresponded to the impact of smoking approximately one pack of cigarettes per day. Analysis of a subset of the phase two data in three communities (Spengler et al., 1987) also revealed mean PM$_{2.5}$ concentrations in homes with smokers about 30 µg/m^3 greater than homes without smokers.

Santanam et al. (1990) reported on a subset of the Harvard Six Cities Study data collected between 1986 and 1987 in Steubenville and Portage. Particle samples were analyzed for 25 elements using XRF spectrometry. Concentrations of selected tracer elements were analyzed by principal component analysis (PCA) to apportion source contributions. Based on these analyses, tobacco smoke was identified as the largest source of indoor PM$_{2.5}$ in smoking households in both cities (Table 6.4). Sulphur-related sources accounted for 8 to 9 µg/m^3 in Steubenville during the summer, but contributions during the winter were not detected. In Portage, sulphur-related sources accounted for 5 to 6 µg/m^3 during the summer and winter. Woodsmoke accounted for approximately 4 µg/m^3 in Steubenville and only 1 µg/m^3 in Portage during the winter. Auto-related sources accounted for 4 µg/m^3 in Steubenville during the summer and 2 to 5 µg/m^3 in Portage during the winter and summer. Unexplained source contributions range from 2 to 12 µg/m^3 in Steubenville and 1 to 6 µg/m^3 in Portage. The sizable unexplained portion was attributed to the fact that since some tracer elements from distinct sources covaried, the PCA method could not distinguish between the associated source types.

Analysis of the ERDA study data from Suffolk and Onondaga Counties, New York, (Sheldon et al., 1989; Leaderer and Hammond, 1991) demonstrated that smoking significantly increased indoor PM$_{2.5}$ concentrations. Koutrakis et al. (1992) furthered the analysis, looking at the contributions of the combustion sources to indoor PM$_{2.5}$ levels. Samples were analyzed by X-ray fluorescence for 16 elements. A simple physical model was applied to determine the contribution of indoor and outdoor sources. Gas stoves were not found to contribute to indoor PM$_{2.5}$ levels during this study. In homes containing only wood-burning or kerosene stoves, or none of the identified sources, approximately 60-70% of the indoor PM$_{2.5}$ levels were attributed to outdoor sources. However, in homes occupied by cigarette smokers, smoking accounted for 54% of PM$_{2.5}$ levels, and outdoor sources and other indoor sources an additional 30 and 16%, respectively. Large home-to-home variations in PM$_{2.5}$ levels were observed.

Table 6.3 Indoor Concentrations of Particles as a Function of Smoking (µg/m^3)

			Indoor		
Reference	**Location**	**Size fraction**	**classification**	**mean level**	**Outdoor**
Spengler et al., 1981	six cities	$PM_{3.5}$ Annual (q6d) 24 h mean	no smokers one smoker two or more smokers	24.4 (n = 35) 36.5 (n = 15) 70.4 (n = 5)	21.1 (n = 55)
Neas et al., 1994	six cities	$PM_{2.5}$, 2 cons. weeks, summer & winter	no smokers smokers	17.3 (n = 470) 48.5 (n = 580)	
Spengler et al., 1985	Kingston/ Harriman TN	$PM_{3.5}$	no smokers smokers	28 (n = 73) 74 (n = 28)	18
Lebret et al., 1987	Watertown, MA	$PM_{2.5}$ 24 h	no smokers smokers	21.6 (n = 70/74) 54 (n = 147/152)	
Santanam et al., 1990	Steubenville Portage	$PM_{2.5}$ (4 pm to 8 am) one week in summer and winter	no smokers smokers no smokers smokers	19.5 (w)[a] 24.5 (s)[b] 43.6 (w) 49.9 (s) 14.8 (w) 13.6 (s) 34.6 (w) 24.9 (s)	18.7(w) 29.5(s) 10.3(w) 13.6(s)
[Jenkins et al., 1994, 1995][g]	12 cities	$PM_{3.5}$	non smoking offices restricted smoking unrestricted smoking	18 (13)[c], n = 629 28 (16), n = 297 58 (33), n = 113	
[Lebret et al., 1990]	Ede & Rotterdam, the Netherlands	RSP (cutpoint not specified) week-long, winter	no smokers smokers	30 (geom. mean) 70	45
[Heavner et al., 1995]	New Jersey & Pennsylvania	$PM_{3.5}$ 14 h home samples 7 h workplace samples	home – no smokers home – smokers workplace – no smokers workplace – smokers	27.6 (19.9)[d] n = 58 86.7 (145.4) n = 30 30.3 (17.6) n = 52 67.0 (44.3) n = 28	
[Quackenboss et al., 1989]	Tucson, Arizona	PM_{10} $PM_{2.5}$	no smokers < one pack/day > one pack/day no smokers < one pack/day > one pack/day	30.3 (19.9)[d] n = 43 46.2 (29.1) n = 27 75.0 (57.2) n = 18 15.2 (15.5) n = 45 27.3 (23.6) n = 26 60.8 (50.8) n = 17	
[Quackenboss et al., 1991]	Tucson, Arizona	$PM_{2.5}$, spring/summer/fall $PM_{2.5}$, winter	no smokers smokers no smokers smokers	10[e] 20 13 (n = 26) 36 (n = 24)	
[Kim & Stock, 1986]	Houston area	$PM_{2.5}$, week long day and night 12 h samples	no smokers smokers	10.8 (4.9)[d] n = 6 33.0 (4.7) n = 5	12.0 (5.9) 24.7 (7.4)
[Morandi et al., 1986]	Houston	$PM_{3.5}$ (approximate) week long daytime (12 h) samples	no smokers smokers smokers with AC smokers without AC	19 89 114 52	
[Chan et al., 1995]	Taiwan	PM_5, 24 h, summer winter	one smoker (11.1 butts) one smoker (6.1 butts)	44 (32)[d] 107 (44)	27 (15) 92 (40)

[a] (w) = winter; [b] (s) = summer; [c] mean (median); [d] mean (standard deviation); [e] median levels; [f] arithmetic mean (geometric mean);
[g] bracketed studies cited in Wallace, 1996

Table 6.4 Contribution of Smoking to 24 h Indoor Particle Concentrations (μg/m^3)

Reference	Size fraction	Contribution	Comments
Dockery & Spengler, 1981[a]	$PM_{3.5}$ (24 h average)[a]	0.88 1.23	homes without air conditioning homes with air conditioning
Letz et al., 1984	$PM_{3.5}$	29.4 (n = 57, 24 h)	ETS component
Lebret et al., 1987	$PM_{2.5}$ (24 h average)[a]	0.8	
Santanam et al., 1990	$PM_{2.5}$ (16 h averages)	20-27 10-25	Steubenville Portage
Leaderer and Hammond, 1991	$PM_{2.5}$ (24 h average)[a]	2.1 (1.9 - 2.3)	
Koutrakis et al., 1992	$PM_{2.5}$	26 (54%)	ETS component in smoking homes
Wallace, 1996	PM_{10} $PM_{2.5}$	29-37 27-32	
Özkaynak et al., 1993	PM_{10} (24 h averages) $PM_{2.5}$	2 1.5	
[Lebret et al., 1990] cited in Wallace, 1996	RSP (24 h averages) cutpoint not specified	2 - 5 10	one cigarette one cigar
[Chan et al., 1995] cited in Wallace, 1996	PM_5 (24 h average)	16	ETS-related component

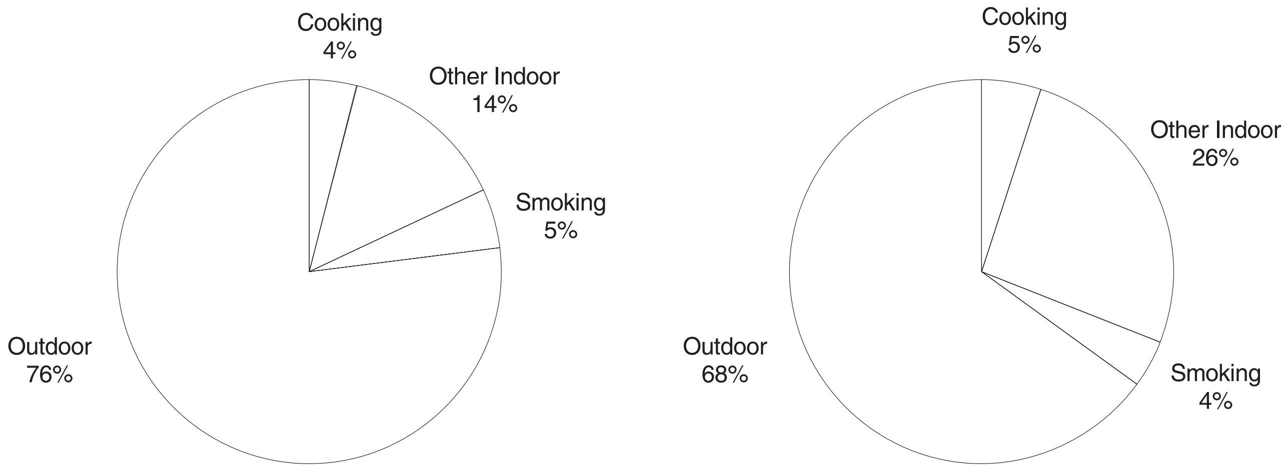
[a] increase in the daily concentration per cigarette

Koutrakis et al. (1992) also looked at the sources of various elements in their analysis. The elements Mn, Se and Ni were associated mostly with outdoor sources. The elements K, Ca and Cu were associated with indoor sources. In particular, smoking, consumer products and dust resuspension were cited as possible sources of chlorine. Smoking, wood-burning and kerosene heaters were associated with K. Possible sources of Ca included dust resuspension, biological particles and particles formed during showering. Finally, vacuum cleaners, kitchen ventilation, fans and kerosene heaters were associated with Cu. Cadmium was exclusively associated with outdoor sources in non-source and wood-burning homes. In homes with kerosene heaters or smokers, outdoor contributions of Cd dropped to 10-40%, suggesting that at least one of these two sources emits Cd. Sulphur was also largely associated with outdoor sources in non-source and wood-burning homes. However, kerosene heaters were also found to be a large source of indoor S, contributing 40-50%, while smoking contributed approximately 10%. Silicon was associated with outdoor sources and kerosene heaters, wood-burning and other indoor sources.

Source contributions were estimated from the pTEAM results using a non-linear method of solving a mass-balance model. Averaged over homes containing no known indoor sources (n = 244), outdoor sources accounted for 76% of indoor $PM_{2.5}$ and 66% of PM_{10} in Riverside (Figure 6.1). Unidentified indoor sources accounted for 14% of $PM_{2.5}$ levels and 26% of PM_{10} levels. Smoking and cooking, the two main identified indoor sources, accounted for only 4-5% each in both size fractions. In homes where these activities took place, each accounted for approximately 20-30% of the indoor particle concentrations (Figure 6.2 and 6.3). Other household activities appeared to make smaller contributions to indoor particle levels.

The pTEAM results showed that tobacco smoke significantly increased indoor PM_{10} and $PM_{2.5}$ concentrations (Özkaynak et al., 1993; Wallace, 1996). Overnight indoor levels for both PM_{10} and $PM_{2.5}$ in smoking homes, as well as personal PM_{10} levels, were about twice as high as corresponding levels in non-smoking homes. Averaged over smoking households (n = 40), outdoor air still remained the largest source of indoor $PM_{2.5}$, accounting for 60% compared to 25-30% by smoking and 7-16% by other unidentified sources.

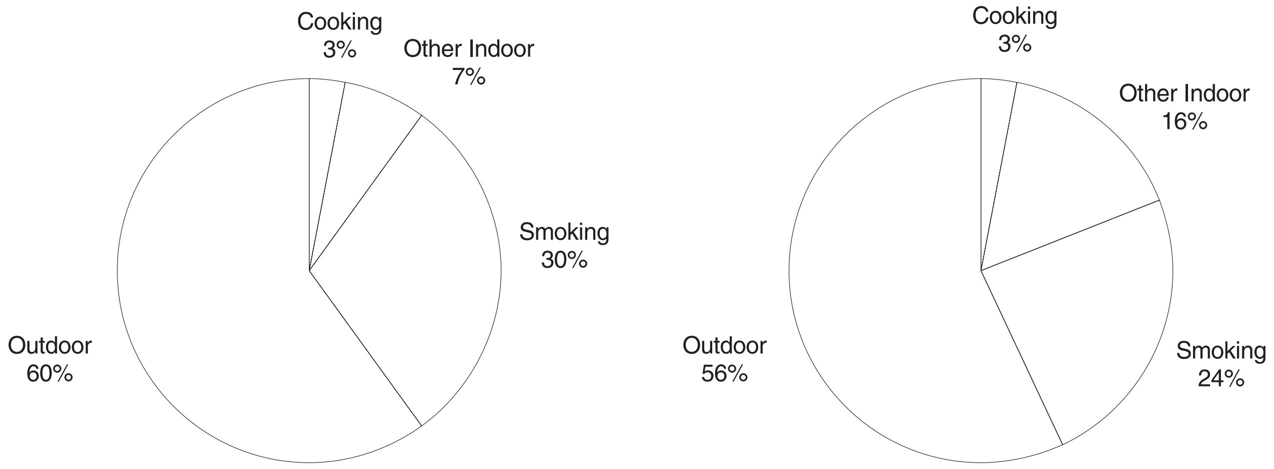
Figure 6.1 Sources of Fine Particles ($PM_{2.5}$) (left) and Thoracic Particles (PM_{10}) (right) in All Homes (Riverside, CA). Source: Özkaynak et al., (1995b)



n = 352 Samples from 178 homes

n = 350 Samples from 178 homes

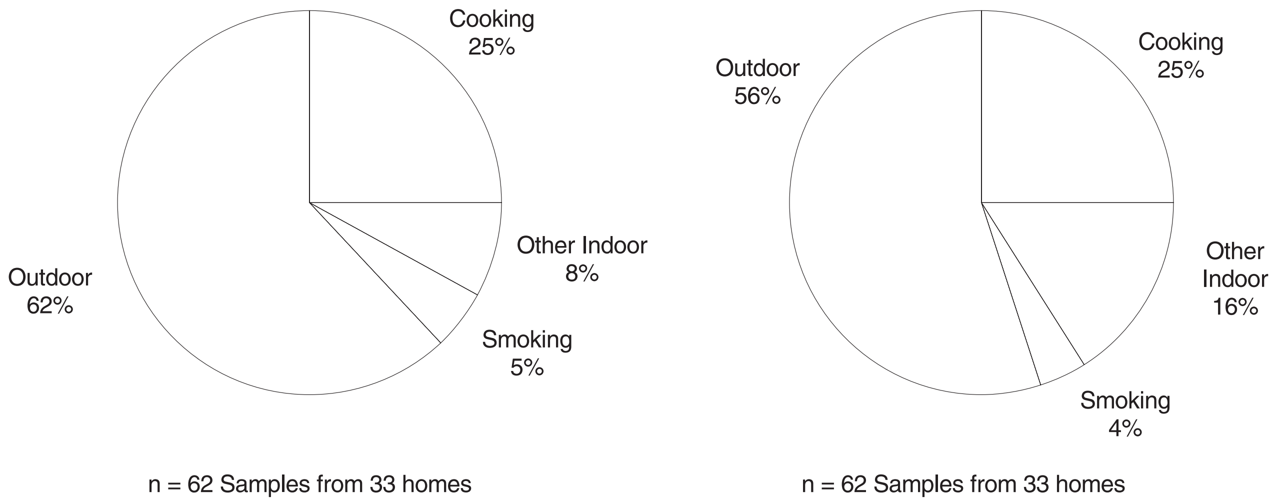
Figure 6.2 Sources of Fine Particles ($PM_{2.5}$) (left) and Thoracic Particles (PM_{10}) (right) in Homes with Smokers (Riverside, CA). Source: Özkaynak et al., (1995b)



n = 61 Samples from 31 homes

n = 61 Samples from 31 homes

Figure 6.3 **Sources of Fine Particles ($PM_{2.5}$) and Thoracic Particles (PM_{10}), left and right panels, Respectively, for Homes with Cooking During Data Collection (Riverside, CA). Source: Özkaynak et al., (1995b)**



Using elemental fingerprinting and applying multivariate analysis methods, Özkaynak et al. (1995b) demonstrated that the contribution to 12 h average indoor PM_{10} and $PM_{2.5}$ of each cigarette smoked is about 2 and 1.5 µg/m^3, respectively. Cooking added 13 µg/m^3 to daytime $PM_{2.5}$ levels and increased daytime PM_{10} levels by about 6 µg/m^3 per minute of cooking. The elements Al, Ti, P, S and Zn were mainly associated with outdoor sources, while the elements Cu, Ca and Cl were mainly associated with unidentified indoor sources. Smoking accounted for 16-21% of the elements Ca, K and Cl. Outdoor air provided 70-100% of the indoor concentrations for most elements.

Several smaller studies also reported indoor levels of particles according to smoking prevalence (Chen et al., 1995; [Heavner et al., 1995; Jenkins et al., 1994. 1995; Kim and Stock, 1986; Lebret et al., 1990; Morandi et al., 1986; Quackenboss et al., 1989, 1991; cited in Wallace, 1996]) (Table 6.3). The data clearly show smoking increases the levels of both PM_{10} and $PM_{2.5}$. Results from instantaneous sampling, performed as part of a study in the Netherlands (Lebret et al., 1990; cited in Wallace, 1996), indicate that the indoor concentration of fine particles varies temporally depending upon the time since (or during) smoking (Table 6.5).

Indoor particle measurements were made in three middle-class, non-smoking homes in Chapel Hill, North Carolina during November and December of 1987 (Kamens et al., 1991). Particle samples were collected using two dichotomous samplers ($PM_{2.5}$ and PM_{10}), and 37 and 47 mm prototype personal sampling units, all co-located. Sampling periods consisted of at least two 8 h daytime periods and three 13 h evening-to-early morning periods. Particle size distributions were measured using an electrical mobility aerosol analyzer (EAA) and two laser optical aerosol counters. Particle morphology was analyzed using scanning electron microscopy (SEM). Measurements from all three homes showed that 37% of the estimated total mass of the collected material was in the fine fraction, 26% in the coarse fraction, and 37% greater than 10 µm. These size distributions varied considerably. For example, EAA measurements showed that during periods of cooking, particles less than 0.10 µm in diameter contributed 30% of the total EAA particle volume. Based on EAA data and SEM images, soot and sulphate particles associated with cooking appeared to be the largest contributors to the fine fraction. Morandi et al. (1986) also reported significantly higher respirable particle levels associated with cooking compared to homes where no cooking occurred. Vacuuming and sweeping were the most significant indoor sources of large particles (>10 µm), while biological and mineral-based particles were most closely associated with the coarse fraction (Kamens et al., 1991).

Table 6.5 Influence of Smoking on Instantaneous RSP Concentrations

Time Since Smoking	N	RSP[a] (geometric mean, µg/m^3)
No smoking	98	41
More than one hour ago	18	52
Between ½ and 1 hour ago	7	76
Less than ½ hour ago	27	141
During the measurements	54	191

[a] cutpoint not specified
source: Lebret et al., 1990; cited in Wallace, 1996

The impact of kerosene heaters on indoor air quality was studied by Mumford et al. (1991). Measurements were made in eight mobile homes in Apex, North Carolina. Selection criteria included heater type, regular heater usage, trailer size of 150-225 m^3, use of electric stoves or heating systems so that emissions from wood and gas stoves could be ignored, and no cigarette smokers. The sulphur content of the kerosene fuel was 0.024%. Measurements were made 6.5 hours per day, 3 days per week, and were restricted to evenings only. The indoor samples were collected on Teflon-coated glass fibre filters using a 10 µm inlet separator. Outdoor samples were collected using a high-volume sampler. The results of the study showed that, on average, the kerosene heaters did not affect indoor PM_{10} levels. However, in 2 of the 8 homes, significant effects were observed.

The impact of woodstoves on indoor particle levels was investigated by Sexton et al. (1984). As part of the Harvard wood-burning study, indoor levels of respirable particles were measured in a total of 19 wood-burning and 5 non-wood-burning homes in Waterbury, Vermont during the winter of 1981/1982. All occupants were nonsmokers. Indoor and outdoor particle levels were measured using Bendix cyclone preseparators attached to Harvard/EPRI portable sampling pumps. Sampling took place over 24 hours every other day for two-week periods. Indoor particle levels were significantly higher than outdoor levels, with an average indoor/outdoor ratio of 1.6. However, no statistical difference was found between 24 h average particle levels in wood-burning and non-wood-burning homes. A linear regression model that incorporates data on air-exchange rates, house volume, fuel use and outdoor levels accounted for only about 20% of the variance of indoor particle levels, suggesting large home-to-home differences in indoor particle levels.

Traynor et al. (1987) estimated the emissions of TSP from four different wood-burning stoves: 3 airtight and 1 non-airtight. TSP was collected on 47 mm Teflon filters. Source strengths were calculated by assuming that the indoor air was well-mixed, and that steady-state conditions applied. The airtight stoves were found to emit little TSP (2.5-8.7 mg/h) relative to the non-airtight stove (26-230 mg/h). Background emission rates were estimated to be 1.1-1.6 mg/h. Different operating styles were suggested as an explanation for the large variation in TSP emission rates estimated for non-airtight stoves.

The ambient data from Highsmith et al. (1988a; cited in Wallace, 1996), which increased by about 50% at night, suggest an influence of wood-burning on ambient levels. In this study, indoor levels of fine particles were greater in homes with wood-burning stoves compared to homes without (Table 6.6) in contrast with the data from Sexton et. al. Subsequently, Highsmith et al., (1988b; cited in Wallace, 1996) studied the effect of different types of portable humidifiers supplied with tap or distilled water on indoor particle concentrations in one home in Boise, Idaho. The study showed that the use of tap water in the humidifier had a dramatic effect on indoor particle levels compared to the use of distilled water (Table 6.6).

6.2.5 Conclusions

Indoor concentrations of PM_{10} and $PM_{2.5}$ are significantly influenced by ambient concentrations and air exchange rates. The data in Table 6.3 show that indoor levels can be higher than outdoor levels or lower than outdoor levels. Wallace (1996) notes that in areas where the outdoor concentrations are fairly high, indoor concentrations are less, whereas indoor concentrations greatly exceed outdoor concentrations in areas where the outdoor levels are relatively low. Clearly, a source of indoor particles is ambient air that has penetrated indoors.

Recent studies indicate both PM_{10} and $PM_{2.5}$ can readily penetrate into buildings (penetrations factors close to unity). Wallace (1996) calculated the fraction of outdoor fine and coarse particles found in homes at equilibrium for a range of air exchange rates (Figure 6.4) based upon the pTEAM estimates for penetration rates and deposition. The fraction of ambient PM_{10} indoors will lie somewhere between

Table 6.6 Influence of Indoors Sources on Indoor Concentrations

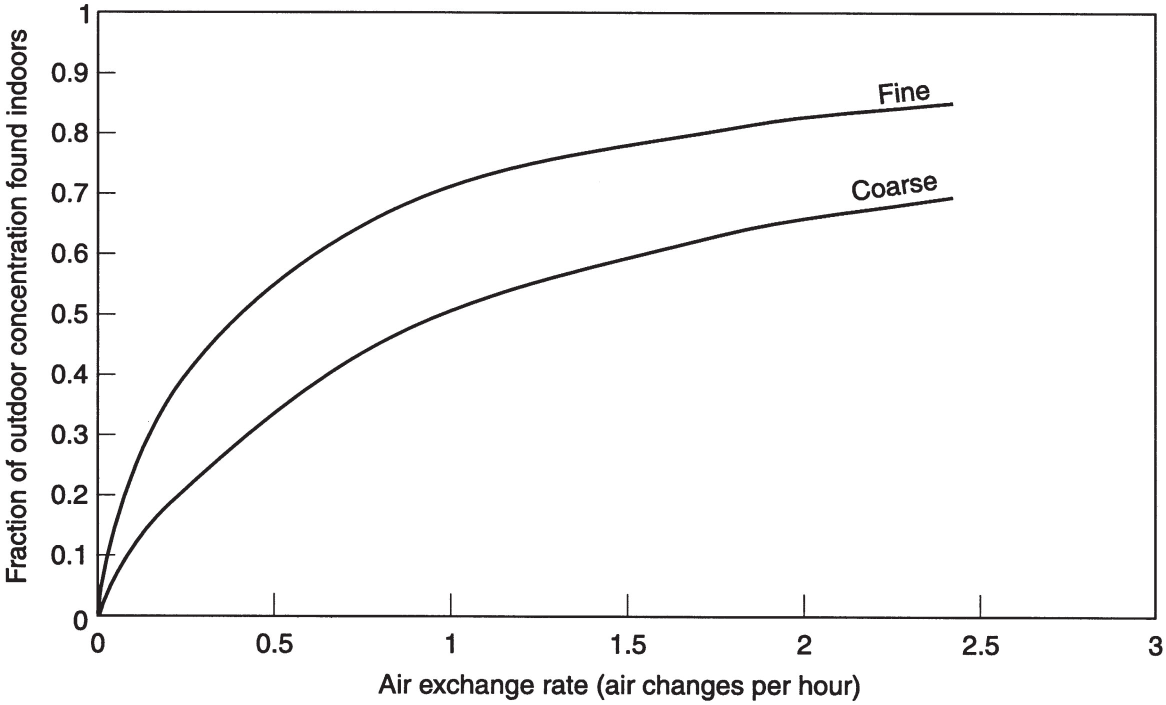
Reference	Sample	Particle size	Indoor levels
[Highsmith et al., 1988[a], 1991]*	10 homes with wood-burning stoves 10 homes without	$PM_{2.5}$, eight consecutive 12 h periods coarse particles, cutpoint not specified	26.3 18.2 10.2 9.7
[Morandi et al., 1986]	13 homes, with cooking without cooking	$PM_{3.5}$ (approximately)	27 20
[Highsmith et al., 1988b]	one home, background level; with ultrasonic humidifier & tap water; ultrasonic humidifier & distilled water	$PM_{2.5}$, 6-8 h samples	11 593 27

* [studies] cited in Wallace, 1996

the fine and coarse particle curves. In Canada, where building construction emphasizes energy efficiency and, therefore, low air-exchange rates, the fractions of fine and coarse particles of ambient origin that will be found indoors under equilibrium will tend toward 50% or less, particularly in the winter.

Once inside, the larger particles tend to settle out more quickly than smaller particles, however, the larger particles are easily resuspended as a result of indoor activities. Of the indoor sources of particles, in smoking homes, cigarette smoking has been identified as the most significant in the three large-scale studies conducted in the United States, and in many of the small-scale studies. Cooking was also consistently identified as a source of indoor particle levels, both PM_{10} and $PM_{2.5}$. In contrast, kerosene heaters and wood-burning stoves (perhaps surprisingly) were not found to consistently raise indoor particle levels. Unknown indoor sources remain, contributing approximately 25% to both PM_{10} and $PM_{2.5}$ levels.

Figure 6.4 Fraction of Indoor Particulate Matter (PM) from Outdoor Airborne PM, Under Equilibrium Conditions, as a Function of Air-exchange Rate, for Two Different Size Fractions. Source: US EPA, (1996)



Deposition rate = 0.39/h for fine particles (≤2.5 µm), 1.01/h for coarse (>2.5 µm and <10 µm)

6.3 DIRECT MEASUREMENT OF PERSONAL EXPOSURE

There are three types of personal exposure: (1) Personal exposure to ambient particles, both ambient particles outdoors and ambient particles that have infiltrated indoors, (2) Personal exposure to non-ambient particles, (i.e., generated indoors), and (3) Personal exposure to total PM (ambient plus non-ambient). Exposure to total PM can be accurately measured if individual study participants carry personal monitors throughout their normal daily activities. With people spending 90% of their time indoors, total personal exposure (3 above), should theoretically be similar to indoor levels.

6.3.1 Personal Exposure Studies

As part of the Six Cities Study, Spengler et al. (1985) measured personal respirable particles (<3.5 µm), in addition to indoor and outdoor levels, in Kingston/Harriman, Tennessee. Ninety-seven volunteers participated during the 46-day sampling period in the winter of 1981. Twenty-four-hour indoor concentrations averaged 42 ± 2.6 (SE) µg/m^3 (n = 266); personal exposures averaged 44 ± 2.8 (SE) µg/m^3 (n = 249). Ambient concentrations measured at a central site were approximately 25 µg/m^3 (n = 71) less than mean personal and indoor concentrations.

Personal (n = 189), indoor (n = 101) and outdoor (n = 54) PM_{10} levels were monitored for two weeks in the winter of 1988 in Phillipsburg, New Jersey (Lioy et al., 1990). The eight participating homes of 14 non-smoking, non-smoke-exposed individuals were within 2 km of a grey iron pipe manufacturing company. Twenty-four-hour average outdoor PM_{10} concentrations ranged from 12 to 165 µg/m^3 at three rooftop sites (Table 6.7). Mean 24 h indoor concentrations ranged from 25 to 60 µg/m^3. Personal exposure levels averaged over 24 h were higher than outdoor concentrations and quite variable, ranging from 40 to 130 µg/m^3.

The pTEAM pilot study reported mean (SE) 24 h personal exposures approximately twice as high as indoor and outdoor for both PM_{10} (112 vs 58.7 vs 62.6 µg/m^3) and $PM_{2.5}$ (70 vs 36.3 vs 42.6 µg/m^3) (Wallace, 1996). Twenty-four-hour personal PM_{10} concentrations measured in the main study were also higher than all other monitored concentrations, ranging from 34 to 287 µg/m^3 (Table 6.7). However, the positive bias measured in the personal and microenvironmental samplers should be considered when evaluating the study results; measured PM_{10} concentrations may be overestimated (Clayton et al., 1993; Thomas et al., 1993).

Population-weighted daytime PM_{10} personal exposures (median 129.7 µg/m^3) were about 50% higher than both indoor (81.7 µg/m^3) and outdoor (84.1 µg/m^3) levels (Table 6.8). Daytime personal PM_{10} exposure was about twice as high as the nighttime personal exposure (median 66.2 µg/m^3). The latter was slightly less than nighttime SAM concentrations (median 74 µg/m^3) and slightly higher than nighttime SIM concentrations (median 52 µg/m^3) (Pellizzari et al., 1992).

Table 6.7 Distributions of 24 hour PM_{10} Concentrations

Monitor	n	24 hour PM_{10} concentration (µg/m^3)										
		Arithmetic mean	Standard error	Geometric mean	Standard error	Min.	Percentiles					Max.
							10	25	50	75	90	
data from Lioy et al., 1990												
SAM	54	60	–	48	–	12	–	–	–	–	–	165
SIM	101	54	–	42	–	25	–	–	–	–	–	60
PEM	189	86	–	66	–	40	–	–	–	–	–	130
modified from Pellizzari et al., 1992												
SAM	153	91.2	4.1	81.4	3.7	15	47.2	60.9	78.3	112.5	159.7	280.4
SIM	157	79.0	4.1	68.5	3.8	20	32.9	45.1	65.3	106.4	143.6	324.8
PEM	161	112.5	6.0	100.7	5.6	34	52.1	70.1	102.5	148.2	183.5	286.9

Personal PM_{10} exposures were different, although the ambient concentrations were similar, for 37 non-smoking adults compared with 45 children, presumably non-smokers (Janssen et al., 1995; cited in Wallace, 1996). Subjects were monitored for one day per week over 7-8 weeks. The mean wintertime 24 h personal exposures (range) were 61 (36-113) µg/m^3 for the adults compared to 104 (59-194) µg/m^3 for the children. Mean ambient concentrations were 42 (32-51) µg/m^3 and 38 (25-56) µg/m^3 respectively. Twenty-four hour indoor concentrations in the adult homes averaged 35 (19-65) µg/m^3.

Other studies also support the observation that personal exposure to fine particles averages higher than indoor and outdoor measurements (Dockery and Spengler, 1981b; Sexton et al., 1984; Morandi et al., 1988). Spengler et al. (1980) also reported that 12 h personal respirable samples averaged 2.5 times greater than outdoor levels.

In all these studies, personal exposure was measured to be higher than either the indoor or the outdoor PM measurements. The exceptions are studies in disabled or elderly persons (Bahadori et al., 1995; Tamura and Ando, 1994; cited in Wallace, 1996). In a study of patients with COPD, Bahadori et al., 1995; (cited in Wallace, 1996) monitored indoor and outdoor PM_{10} and $PM_{2.5}$ levels for 12 consecutive 12 h periods. The patients wore PEMs during the daytime, alternating between a PM_{10} and $PM_{2.5}$ nozzle. Preliminary analysis of the data, which is population specific, indicated personal exposures were similar to outdoor concentrations for both size fractions. Individual PEM values were 30 to 50% of the SAM values measured outside the subject's house (Tamura and Ando, 1994; cited in U.S. EPA, 1996).

6.3.2 "Personal Cloud"

In non-smoking households, daytime personal exposure levels are always higher than indoor and outdoor particle concentrations (Spengler et al., 1985; Özkaynak et al., 1993; Neas et al., 1994; Thatcher and Layton, 1994). Wallace et al. (1991) suggested that this finding – higher personal exposures – may be explained by a "personal cloud effect" in which a person's movements or activities contribute to increased personal exposures.

Table 6.8 shows that overnight personal PM_{10} levels are similar to both indoor and outdoor levels, while indoor levels of PM_{10} and $PM_{2.5}$ are similar to outdoor levels during the day and drop off at night. Time-activity data may help explain the nighttime similarities in personal and indoor levels; while sleeping for about two-thirds of the overnight monitoring period, participants were not engaged in particle generating or re-entraining activities (Clayton et al., 1993). Examination of daytime and nighttime differences in personal, indoor and outdoor particle concentrations in the pTEAM study (Table 6.9) revealed a significantly lower PM_{10} exposure for working people than for participants who stayed home (Clayton et al., 1993; Özkaynak et al., 1993; Wallace et al., 1993). While at home, personal activities such as cooking, cleaning the house and smoking were related to higher personal exposures. Re-entrainment of house dust as a result of activities such as sitting on chairs and sofas or walking across carpets also increased personal exposure (Thatcher and Layton, 1995; Wallace et al., 1993).

An alternative explanation for the personal cloud could be attributable to the PEM over sampling particles larger than 10 µm. PM_{10} concentrations measured with the personal and microenvironmental monitors were consistently higher than the SAM measurements. Additional factors that may influence the differences in personal exposure and indoor and outdoor particles concentrations include: the presence of strong indoor sources, air-exchange rates, occupational exposures, the spatial variability of ambient particles and commuting habits.

As noted above, smoking is a major source of increased particle concentrations. Its contribution to the "personal cloud" is evident primarily at night (Table 6.9). Repace and Lowery (1980) estimated that the amount of particulate matter directly inhaled by the average smoker is 2-3 orders of magnitude greater than the concentration of environmental tobacco smoke (ETS) measured with PEMs.

The results of Bahadori et al. (1995; cited in Wallace, 1996) indicate that the "personal cloud" is made up mostly of coarse particles ($PM_{2.5}$ -PM_{10}). The results of Thatcher and Layton (1995), which indicated coarse particles are easily resuspended compared with fine particles, tend to support those results. The pTEAM results showed a 50-100% increase in 14 of 15 elements found in the indoor aerosol in the personal cloud. This would imply that the "personal cloud" consists largely of particles resuspended by personal activity.

Table 6.8 Weighted Distributions of Personal, Indoor and Outdoor Particle Concentrations (modified from Pellizzari et al., 1992)

			Particle concentrations ($\mu g/m^3$)												
				Percentiles											
		n	Min.	10	25	50	75	90	Max.	Mean	Standard error	Geom. Mean	Standard error	Standard deviation	Geom. standard deviation
$PM_{2.5}$ (day)	SAM	167	7.4	14.9	23.4	35.5	60.1	102.2	187.8	48.9	3.5	37.7	2.5	38.1	2.8
	SIM	173	2.8	11.5	19.3	33.5	61.5	101.0	238.3	48.2	4.1	35.0	3.3	41.3	2.6
$PM_{2.5}$ (night)	SAM	161	3.4	14.5	23.0	35.0	64.9	120.7	164.2	50.5	3.7	37.2	3.1	41.0	3.5
	SIM	166	2.9	10.0	14.8	25.9	48.9	82.7	133.3	36.2	2.2	26.7	1.9	29.9	3.1
PM_{10} (day)	SAM	165	16.2	42.8	56.9	84.1	110.8	157.2	406.6	94.9	5.5	82.7	4.1	59.6	3.1
	SIM	169	16.6	30.9	49.5	81.7	127.2	180.7	512.8	94.7	5.7	78.2	5.0	63.1	3.0
	PEM	171	35.1	59.9	86.1	129.7	189.1	263.1	454.8	149.8	9.2	128.7	8.5	88.5	3.3
PM_{10} (night)	SAM	162	13.6	39.3	53.6	74.1	103.7	167.8	222.9	86.3	4.4	74.5	4.0	51.3	3.6
	SIM	163	14.1	25.2	33.5	51.6	84.8	116.9	180.3	62.7	3.2	53.1	3.1	40.3	3.8
	PEM	168	19.1	36.6	48.1	66.2	98.8	135.0	278.3	76.8	3.5	67.9	3.1	42.4	3.0

Table 6.9 Effects of Particle-generating Activities on Mean PM_{10} Personal Exposures (Pellizzari et al., 1992)

	PM_{10} personal exposures (μg/m^3)									
	Daytime					Nighttime				
Activity	n	Mean	Standard error	Geom. mean	Standard error	n	Mean	Standard error	Geom. mean	Standard error
Work[a]	59	126.8	11.6	107.6	10.7					
No work	111	162.1*	9.9	141.6*	8.5					
Housework[b]	110	161.5*	11.0	142.2*	9.6	64	73.0	5.6	65.9	4.9
No housework	61	125.0	11.4	104.4	10.6	104	79.5	3.2	69.4	2.8
Spraying[c]	70	158.3	12.0	138.5	10.6	55	81.5	5.9	69.7	5.5
No spraying	101	143.1	10.5	121.6	8.9	113	74.6	4.1	67.1	3.5
Smoke[d]	61	155.2	15.3	131.4	15.9	29	104.2*	8.0	96.6*	6.7
No smoke	110	146.8	7.7	127.3	6.3	139	71.4	3.3	63.3	2.7
Exhaust[e]	31	96.5	8.7	85.5	7.0	7	—	—	—	—
No exhaust	121	166.3*	9.0	144.8*	8.1	140	77.5	3.8	68.2	3.1

* indicates that the means or geometric means for persons with and without the activity are significantly different; the asterisk is shown beside the higher member of the pair.

[a] *work*: identifies participants who have a paid job outside of the home and does not include self-employed in the home, full-time student, full-time homemaker, out of work just now but usually employed, retired or disabled, and other.

[b] *housework*: consisted of vacuuming, dusting, carpet cleaning, indoor cooking and using a clothes dryer.

[c] *spraying*: included either pump- or propellant-type spraying of paints, cleaners, disinfectants, air fresheners, hair care products, perfumes/colognes, deodorants, cooking products, lubricants, insecticides, repellents and plant care products.

[d] *smoke*: is one or more cigarettes, cigars or pipefuls.

[e] *exhaust*: identifies participants or other household members potentially exposed to exhaust from vehicles running in attached garages.

Analysis of the pTEAM data suggested that the “personal cloud” is not due to variations in sampling characteristics of the monitors, nor to organic particles from people and their clothes. The excess personal exposure is likely due to generation or re-entrainment of particles during personal activities (Clayton et al., 1993; Wallace et al., 1993). However, the exact nature of the “personal cloud” has not yet been determined.

6.4 SURROGATES OF PERSONAL EXPOSURE

6.4.1 Exposure Estimates Using FAM

Particle concentrations measured at centrally located fixed ambient monitors (FAMs) in epidemiological studies are indicators of the extent of the contribution of ambient particles to personal exposure. This assumes that, ignoring microscale fluctuations in particle concentrations, population exposure to ambient PM can be represented by the 24 h average ambient concentrations measured at the central site (FAM). On this basis, 24 h average population exposures across Canada would range from 15-42 μg/m^3, the mean PM_{10} concentrations averaged from 1984-1995 at urban sites in the ambient monitoring network (Table 6.10). Similarly, mean 24 h $PM_{2.5}$ exposure would range from 8.5-20.2 μg/m^3, based upon FAM measurements at urban central sites over the same timeframe. Median and high end (90th percentile) 24 h FAM measurements representing personal ambient exposures are also presented.

Table 6.10 Estimated 24 h Population Exposure Based Upon Central Site Fixed Ambient Monitoring Data (range, µg/m^3)

	mean	median	high end exposure (90%)
PM_{10}	15-42	8-36	19-84
$PM_{2.5}$	8.5-20.2	4.8-17	10.9-39

data from dichot samplers, 1984-1995

However, as described in Chapter five, there are wide variations in daily measurements of PM at FAMs due in part to the location of the monitor, meteorology, intra-city differences, and diurnal patterns. In addition, it has been previously established that concentrations measured at FAMs do not correlate well with readings obtained simultaneously with personal monitors (Binder et al., 1976; Cortese and Spengler, 1976; Wallace, 1979; Ott and Flachsbart, 1982; U.S. EPA, 1982a). Exposure to non-ambient particles is not included in the personal exposure as represented by the FAM.

Table 6.11 summarizes the recent studies that measured personal exposure to total PM, including some additional exposure studies explained in detail elsewhere (US EPA, 1996). Almost all of the studies were conducted on urban and suburban residents. Personal exposures were usually greater than either the indoor (SIM) or outdoor (SAM) particle concentrations. Nine studies confirm the poor correlation between personal exposures and ambient particle concentrations, as evidenced by the R^2 values shown in the table. Of the remaining studies, the R^2 values range from 0.04 to 0.70, many of them significantlly different from zero. Some of the individual studies are discussed in the following paragraphs.

Correlational analysis between 24 h SAM, SIM and PEM pTEAM measurements show outdoor levels weakly correlated ($r_s = 0.41$) with personal PM_{10} exposures (Clayton et al., 1993; Pellizzari et al., 1992). Daytime correlations between personal and outdoor concentrations measured at the residence (SAM) (0.35) and at the fixed site (FAM) (0.37) suggest that outdoor monitors do not adequately represent personal exposure to total PM_{10}, particularly during the day. Correlations of personal and residential concentrations with fixed-site concentrations showed higher nighttime correlations between fixed-site and SAM data (Table 6.12). Correlations of indoor data (SIMs) with fixed-site data (FAMs) were lower; correlations with personal data were lower still (Clayton et al., 1993). The authors concluded that measurements of ambient air would be unable to predict personal exposure to total PM_{10} for most persons accurately.

In Kingston and Harriman (Spengler et al., 1985), correlation coefficients between personal exposure and ambient concentrations were low and not statistically significant ($r = 0.07$, $p = 0.30$); the correlation between indoor and personal concentrations was much stronger ($r = 0.7$, $p = 0.0001$). Cross-sectional (daily) regressions calculated from the Lioy et al., 1990 data (Wallace, 1996) also reported low personal-outdoor correlation coefficients; the median R^2 (range) was 0.06 (0.00 to 0.39). Unique results from Tokyo show personal PM_{10} exposures averaging well below outdoor concentrations, with a correlation coefficient of 0.68, likely because of minimal generation and resuspension of particles indoors (Tamura and Ando, 1994; cited in Wallace, 1996).

Most of the studies report poor cross-sectional personal-outdoor correlations. If there is a lot of variability in particle concentrations from sources poorly correlated with centrally located FAMs, than the percent of variance that can be explained by the FAM data is small. Sampling error, a non-random sample, very strong indoor sources and personal activities all contribute to a poor correlation between FAMs and PEMs. FAMs measure ambient particles and PEMs measure personal exposure to ambient and non-ambient PM.

Table 6.11 Comparison of Personal Exposure Concentrations with Simultaneous Ambient Concentrations

Location	Year	PM (µm)	n	time	mean PEM (µg/m^3)	mean SAM (µg/m^3)	R^2 PEM vs SAM	p	Reference
Ansonia	1973	5	20	24 h	115	59	NS	NS	Binder et al., 1976
Watertown	1975	3.5	18	24 h	35	17	0	NS	Dockery & Spengler, 1981
Steubenville	1976	3.5	19	12 h	57	64	0.19	NR	Dockery & Spengler, 1981
Topeka	1979	3.5	46	12 h	30	13	0.04	NS	Spengler et al., 1980
Toronto	1981	25	13	8 h					WHO/UNEP, 1982
non-asthmatic	winter				122	68	0.15	NS	
non-asthmatic	summer				124	78	0.1	NS	
asthmatic	winter				91	54	0	NS	
asthmatic	summer				124	80	0.07	NS	
Kingston/Harriman	1981	3.5	97	12 h	44	18	0	NS	Spengler et al., 1985
Zagreb	1982	5	12	1 wk					WHO/UNEP, 1982
	summer				114	55	0	NS	
	winter				187	193	0.5	NR	
Waterbury	1982	3.5	48	24 h	36	17	0	NS	Sexton & Spengler, 1984
Bombay	1982	3.5	15	24 h				NR	WHO/UNEP, 1984
	winter				127	117	0.26	NR	
	summer				67	65	0.2	NS	
	monsoon				58	51	0.02		
Beijing	1985	3.5	20						WHO/UNEP, 1985
	winter			24 h	177	421	0.07	0.09	
	summer			1 wk	66	192	0.03	NS	
Houston	1988	3.5	30	12 h	27	16	0.34	<0.05	Morandi et al., 1988
Philipsburg	1988	10	14[a]	24 h	86	60	0.04	0.008	Lioy et al., 1990
			14[b]		76	60	0.25	0.001	
Azusa	1989	2.5	9	24 h	79	43	0.01	NS	Perritt et al., 1993
		10	9		115	62	0.01	NS	
Riverside	1990	10	141	24 h	113	84	0.23	NR	Clayton et al., 1993
Tokyo	1992	10	7	48 h	37	56	0.68	0.000	Tamura et al., 1996
Nashville, TN	1995	2.5	10	12 h	22	23	0.42	0.000	Bahadori et al.,
		10		day	33	32	0.48	0.000	

Note: Please refer to US EPA (1996) for complete reference information.

*n = number of individuals carrying personal monitors; Year = year study was performed.

NS = not statistically significant from 0; NR = p value not reported, but mentioned as significant

[a] = 14 subjects carried PEMs for 14 days for 191 valid measurements;

[b] = three outliers are removed and regression is for 188 measurements

Table 6.12 Correlations of PM_{10} and $PM_{2.5}$ Residential Data and Personal PM_{10} Concentrations with Fixed-Site Concentrations

Personal and residential data	Particle fraction	Daytime fixed-site	Nighttime fixed-site
Residential Outdoors (SAM)	2.5	0.83	0.96
SAM	10	0.66	0.93
Residential Indoors (SIM)	2.5	0.70	0.71
SIM	10	0.51	0.59
Personal (PEM)	10	0.37	0.54

compiled using data from Clayton et al., 1993

When individual (longitudinal) regressions of personal exposure with the nearest outdoor site are calculated, the correlations improve (R^2=0.46, range: 0.02-0.82) when compared with the cross-sectional correlations (Lioy et al., 1990). Similar improvements were noted when the longitudinal regressions of personal exposures and outdoor concentrations were calculated for the pTEAM pilot study data (Wallace, 1996) and in Tokyo (Tamura and Ando, 1994; cited in Wallace, 1996). Median individual longitudinal correlations for individual adults or children were also higher (R^2=0.25, range 0.17-0.85 and R^2=0.40, range 0.00-0.96, respectively) compared with cross-sectional personal-outdoor correlation coefficients for adults (R^2= 0.02) and children (R^2= 0.11) (Janssen et al., 1995, cited in Wallace, 1996). The improvement in correlation coefficients suggests that, for individuals who are not exposed to microenvironmental sources of particles, and whose day-to-day activities are fairly repetitive, ambient levels of particles directly reflect their exposure to particles originating in the ambient air.

The correlation between outdoor measurements at FAM or SAM and personal exposure measurements also increases when the mean of the PEM values from the personal exposure studies is calculated. Mage and Buckley (1995) conclude that the uncertainty in predicting mean community exposure to total PM is smaller than the uncertainty associated with predicting individual personal exposure to PM. They propose the following: when a large portion of the total particulate matter is composed of fine particles, and when coarse particles are relatively constant in a community, the correlation between PEMs and FAMs is higher, because the fine particles readily penetrate indoors. Thus, ambient fine particles measured at the centrally located FAM can serve as an indicator of community (population) exposure. However, personal exposure of smokers and individuals exposed to ETS is not represented by the FAM (Mage and Buckley, 1995; US EPA, 1996).

In summary, a 24 h time-weighted average exposure estimate using ambient data from a central site (FAM), may be able to represent a part of the distribution of personal exposure to particles (fine fraction more so than coarse fraction), particularly for non-smoking individuals who are not exposed to indoor particle sources. However, the 24 h time-weighted average exposure estimate using solely ambient data does omit potential “peak” exposure to non-ambient particles in microenvironments such as indoors at home, at work, in-transit, and other indoor locations (discussed in sections 6.2 and 6.3). Therefore, estimates of personal exposure based upon ambient data do not tell us anything about personal exposure to non-ambient particles; total personal exposure is underestimated.

6.4.2 Time-Weighted Average Exposure Estimates Using Ambient and Indoor Data

Introducing microenvironmental exposures into a time-weighted average exposure estimate will theoretically improve estimates of personal exposure to total PM. Results from the pTEAM study show 24 h PEM and SIM PM_{10} data are highly correlated (0.70). Nighttime correlations between personal and outdoor (SAM) and indoor (SIM) data (0.62 and 0.80, respectively) are uniformly high (Pellizzari et al., 1992; Clayton et al., 1993). Moderate correlations of personal exposures to indoor concentrations were reported for both size fractions by Bahadori et al.

(1995; cited in Wallace, 1996). The high correlation between personal exposure and indoor concentrations combined with the time spent indoors indicates that indoor microenvironments are the most important contributors to overall exposure.

However, like the indirect personal exposure estimates using solely FAMs, indirect exposure estimates (E_t) using indoor and ambient particle measurements will likely underestimate personal exposure. Spengler et al., (1985) estimated that only 54% of the variance in personal exposure could be predicted using the equation:

$$E_t = C_i T_i + C_o T_o \tag{6.3}$$

where concentration and time are modelled for the indoor (I) and outdoor (o) environments. The additional variability could be because concentrations measured during personal exposure studies in unoccupied microenvironments do not include the increase in indoor particle concentrations generated by vacuuming, dusting, indoor cooking, carpet cleaning and using the clothes dryer. Similarly, ambient monitors do not include the increased particle levels generated during outdoor activities such as gardening, burning leaves/rubbish, outdoor cooking or recreational activities, nor vehicular emissions and resuspension of roadway dusts.

6.5 PREDICTED DISTRIBUTION OF PERSONAL EXPOSURES TO PM_{10} IN CANADA

Using the findings from population-based field investigations, personal and population exposure models have been developed (Johnson et al., 1994; Özkaynak et al., 1996) that combine ambient measurements of pollutants with information on age-specific time-activity and estimates of microenvironmental pollutant concentrations. A Monte-Carlo based framework developed by Harvard researchers was applied to Canadian ambient PM_{10} measurements, demographics, and smoking rates by region to generate personal exposure distributions (Özkaynak et al., 1995). The results from that modelling-based analysis are described in the subsequent sections. Predictions are limited to exposures occurring via inhalation.

6.5.1 Methodology

Model Structure

The PM_{10} population exposure model estimates personal exposures on a daily (24 h average) period by summing the time-activity weighted microenvironmental exposures using equation 6-1:

$$E_i = \sum_{j=1}^{m} E_{ij} = \sum_{j=1}^{m} f_{ij} \cdot C_{ij}$$

where E_{ij} = exposure to individual I in microenvironment j (µg/m^3), f_{ij} = the fraction of time spent by person I in microenvironment j during the 24 h prediction period, C_{ij} = the average PM_{10} concentration (µg/m^3) in microenvironment j when individual I is present and m is the number of microenvironments considered in the model.

These are the six different microenvironments included in the exposure model: 1) outdoors, 2) indoors at home during daytime (7:00 a.m.-7:00 p.m.), 3) indoors at home during nighttime (7:00 p.m.-7:00 a.m.), 4) in-transit, 5) indoors not home or bar/restaurant, 6) bar/restaurant. Exposures to PM_{10} from smoking and cooking sources were also considered by combining the time during which individuals are exposed to either source, with predicted PM_{10} concentrations from ETS or cooking, in environments where cooking or smoking occurs. The general structure of the physical model for indoor PM_{10}, developed initially for the pTEAM study, is:

$$C_{\text{in}} = \frac{P \cdot a}{a+k} C_{\text{out}} + \frac{N_{\text{cig}} S_{\text{smk}} + T_{\text{cook}} S_{\text{cook}}}{(a+k)Vt} + \frac{S_{\text{other}}}{(a+k)V} \tag{6.4}$$

where C_{in} = PM_{10} concentration indoors, C_{out} = PM_{10} concentration outdoors, P = penetration fraction (unitless), a= air exchange rate (hr^{-1}), k= PM_{10} decay or deposition rate (hr^{-1}), Ncig = number of cigarettes, V = house volume (m^3), T_{cook} = cooking time (hr), t = monitoring period (12 hrs), S_{smk} = estimated source strength for cigarette smoking (mg/cig), S_{cook} = estimated source strength for cooking (mg/min), and S_{other} = estimated source strength for other indoor sources (mg/hr).

Simulation Procedure

The model produces estimates of the distribution of 24 h average exposures by using input distributions that represent variability of microenvironmental concentrations, time-activity patterns and contact rates among the modelled individuals. Characterizations of uncertainty for the PM_{10} model were limited to parameter uncertainty; scenario and model uncertainty were assumed to be negligible. A two-dimensional Monte-Carlo simulation approach was used to propagate the estimated uncertainty about model inputs (parameter variability and uncertainty) through to the distributions of the prediction endpoints. The details of the simulation methodology and the uncertainty analysis can be found elsewhere (MacIntosh et al., 1995; Özkaynak et al., 1995a).

6.5.2 Data Bases and Input Distributions

The types of distributions used to represent inter-individual, spatial and temporal variability, and parameter uncertainty were based on physical data as much as possible and supplemented by the professional judgement of the contractors when required. Normal or lognormal distributions were assigned to distributions representing inter-individual differences in microenvironmental concentrations and exposure durations. For distributions representing knowledge uncertainty, uniform distributions were used to characterize inputs when the quantity and quality of data were low. When moderate amounts of data were available, triangular distributions were typically used; normal or lognormal distributions were used when substantial amounts of quality data were available.

Ambient Air

Available ambient PM_{10} measurements for 1990-1993 were obtained from Environment Canada. Table 6.13 lists the station numbers and sites used to develop ambient PM_{10} distributions for each of the five regions (British Columbia, the Prairies, Ontario, Québec, and the Maritimes) used for the analysis. Four sites (Sutton, Kejimkujik, Egbert & Walpole) were used to represent rural PM_{10} concentrations. Analysis of the data showed a lognormal fit for most of the data with some seasonal variations. Site specific geometric means (GM) and geometric standard deviations (GSD) were estimated and combined with demographic information to produce seasonal and regional GM and GSD estimates for the ambient PM_{10} distributions.

Table 6.13 List of Canadian PM_{10} Monitoring Sites Used in the Personal Exposure Model

Region	Station #	Site
Maritimes/ Atlantic	10101 40201 40203 40204 40205 30311 30101 30501	St. John's Saint John Saint John Saint John Saint John Sydney Halifax Keji
Québec	50104 50109 50307 54101	Montréal1 Montréal2 Québec City Sutton
Ontario	60104 60204 60211 60417 60424 61901 64401	Ottawa Windsor Windsor2 Toronto Toronto2 Walpole Island Egbert
Prairies	70119 80109 80110 80209 80211 90130 90204 90227	Winnipeg Regina Regina2 Saskatoon Saskatoon2 Edmonton Calgary Calgary2
British Columbia	100106 100111 100118 100109 100303	Vancouver Vancouver2 Vancouver1 Vancouver3 Victoria

Demographics

Demographic data (age, gender, and location, rural or non-rural dwellers) were obtained from the 1991 Census (Statistics Canada, 1993). The relative size of the population near each monitor was sorted by region and by urban versus rural classification. The population was divided into four age categories (10-19 years, 20-24 years, 25-64 years and 65-99 years) to match the available time-activity information.

Time-Activity Patterns

Time-weighted microenvironmental exposures were calculated using time-activity diaries from a random sample of about 3,000 US residents recently obtained during the National Human Activity Pattern Survey (cited in Özkaynak et al., 1995a). An analysis of the daily time-activity diaries to determine whether significant differences exist in time spent indoors and outdoors by geographic location and season revealed subjects living in the north US and near Canada spending slightly more time outdoors during the summer and less time outdoors in the winter than the rest of the US population. Because the minor seasonal differences in outdoor times were small, estimates of time spent each day in each of the six study microenvironments were calculated from the entire sample of time-activity diaries.

Smoking

Data obtained from Health Canada reports (cited in Özkaynak et al., 1995a) on the prevalence of cigarette smoking by age and gender for all of Canada, cigarette consumption rates by region (regardless of age) and age-specific US cigarette smoking rates published by the Department of Health and Human Services, (cited in Özkaynak et al., 1995a) were used to develop age-dependent smoking prevalence rates by region. Table 6.14 summarizes the percentage of smokers and Table 6.15 breaks down the smoking population, according to the number of cigarettes, by region, age and gender. In modelling ETS exposures, smokers were assumed to be exposed to ETS in the residential and in-transit microenvironments. Exposure to ETS for smokers in the remaining microenvironments and non-smokers in all microenvironments were simulated using data obtained from sources detailed in Özkaynak et al., 1995a.

Indoor PM_{10} Concentrations

Residential indoor concentrations are modelled by a semi-empirical physical model (discussed above) which assumes contributions to indoor PM_{10} from: outdoors, environmental tobacco smoke (ETS), cooking, and other unaccounted indoor sources. The data bases and input source strength estimates required to construct and set parameters for the indoor concentration models were obtained from the pTEAM study results (Özkaynak et al., 1995b, 1996).

PM_{10} concentrations inside automobile cabins were assumed to be similar to concentrations on the highway, which were predicted from a relationship developed between a highway monitor (Duncan Décarie site in Montréal) and the more centrally located site in the city. The highway monitor recorded on average, 20% higher PM_{10} concentrations than the average of the two monitors. ETS concentrations in automobile cabins were simulated based upon consideration of source strength and air exchange rates. Indoor PM_{10} concentrations in public and commercial buildings, and in restaurants and bars were based on survey data from the US and England.

Table 6.14 Smoking Population by Gender, Age & Region (%)

Age*	Gender	British Columbia	Prairies	Ontario	Quebec	Atlantic
14-19	M F	24 27	26 29	21 23	34 38	28 31
20-24	M F	37 31	39 33	31 26	51 43	41 35
25-64	M F	31 29	33 31	26 25	43 41	35 33
>65	M F	15 13	16 14	13 11	21 18	17 15

* For age under 13, smoking rate is assumed to be zero.
Source: Health Canada, 1994 (cited in Özkaynak et al., 1995a)

Table 6.15 Smoking Population by Number of Cigarettes (%)

Region	Gender	Number of Cigarettes Smoked per Day		
		1-10	11-25	>25
British Columbia	M F	14.4 34.4	64.6 62.2	21
Atlantic	M F	16.8 33.6	70.4 59.2	12.8 7.3
Québec	M F	18.3 30.4	36.6 59.9	18.1 9.7
Ontario	M F	26.5 31.2	62.9 64.4	10.6 4.5
Prairies	M F	23.4 27.3	69.6 70	7

Building Factors

Average house size was estimated using the Canadian census (living habits) data on the average number of rooms per household. Estimated Canadian home volumes were compared to the distribution of 1,836 US residence volumes reported by Pandian (cited in Özkaynak et al., 1995a). Since the distributions were found comparable, the more complete house volume data from the US database was used in modelling exposures. The distribution of house volumes was represented by a lognormal fit to the data.

Air exchange data were obtained from the Brookhaven Laboratory (BNL) data base. An analysis of the potential for air exchange rates to vary systematically by region (north, south, and other) revealed similar air exchange rates in the spring, fall and winter months for the North region. Regional differences were apparent for some seasons (summer, winter).

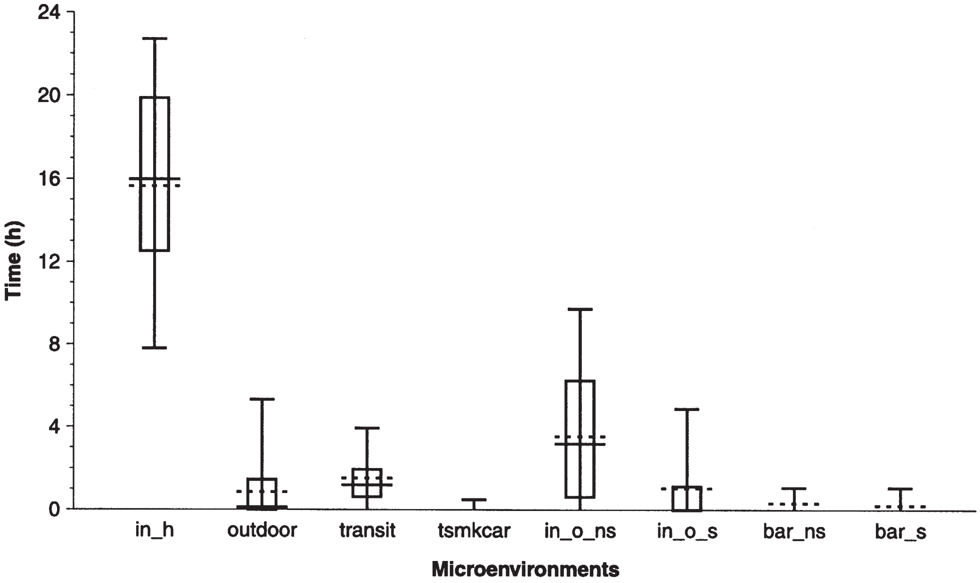
Residential air exchange data in Canada, fit by lognormal distribution, were assumed to be represented by those in the north US for the seasons fall, winter and spring (Özkaynak et al., 1995a). Summer was modelled as being two times greater than in the rest of the year based on summer air exchange rate data for the other region.

6.5.3 Results

The probabilistic PM_{10} exposure simulation model was iteratively run 10,000 times to produce estimated distributions of 24 h average personal, indoor, outdoor and in-transit PM_{10} concentrations. Box plots are used to denote the median, 5th, 25th, 75th and 95th percentiles of the average time spent in various microenvironments. The solid horizontal line is the median, and where appropriate, the dashed line the mean. Figure 6.5 displays the time-activity data (in hours) used in the PM_{10} exposure predictions. Average time spent indoors at home (mean = 15.7 h, nighttime = 10.3 h, daytime = 5.4 h) is the longest time individuals spend within a given microenvironment.

Table 6.16 shows the descriptive statistics for all of the predicted microenvironmental PM_{10} concentrations. Indoor smoking environments dominate PM_{10} concentrations. However, the highest PM_{10} concentrations are estimated in cars with smokers (mean PM_{10} = 71 µg/m^3). Figure 6.6 displays the predicted distributions for 24 h indoor and outdoor microenvironmental concentrations in graphical form. Average residential indoor daytime concentrations are predicted to be about 40 µg/m^3, whereas nighttime levels are lower (25 µg/m^3). In comparison, average outdoor PM_{10} calculated based on ambient measurements (1990-1993), is about 24 µg/m^3.

Figure 6.5 Time-activity Data Used in PM_{10} Exposure Model



in_h	time spent indoors at home
outdoor	time spent outdoors
transit	time in transportation
tsmkcar	time in car spent is presence of a smoker
in_o_ns	time spent indoors other than at home, without smokers
in_o_s	time spent indoors other than at home, with smokers
bar_ns	time spent in smoking free bar/restaurant
bar_s	time spent I bar/restaurant with cigarette smoke

Table 6.16 Summary Statistics of Estimated Microenvironmental PM_{10} Concentrations (µg/m^3, 24-h)

	Mean	Std. dev.	Min	5th	25th	50th	75th	95th	Max
Cout	24.4	17.8	0.8	6.0	12.4	19.9	31.1	58.7	148.6
Cin_day	40.4	55.2	0	7.3	14.8	24.8	43.8	124.7	1206.5
Cin_nite	24.8	38.4	0	4.4	8.8	14.3	25.1	76.3	844.8
Cin_o_ns	43.7	39.5	0	0	13.1	34.9	57.2	118.2	483.2
Cin_o_s	52.3	79.8	0	0	0	0	94.1	213.1	744.8
Cbar_ns	7.8	23.2	0	0	0	0	0	54.5	368.6
Cbar_s	29.0	74.0	0	0	0	0	0	204.5	607.2
Ctransit	25.9	21.9	0	0	11.5	21.4	34.9	66.6	175.6
Csmkcar	71.3	157.4	0	0	0	0	99.8	394.0	1333.4

Cout	outdoor concentration
Cin_day	total indoor daytime concentration from all sources
Cin_nite	total indoor nighttime concentration from all sources
Cin_o_ns	concentrations from other indoor no smoking sources
Cin_o_s	concentrations from other indoor smoking sources
Cbar_ns	concentrations in bar/restaurant with no smoking
Cbar_s	concentrations in bar/restaurant with smoking
Ctransit	concentrations in transportation
Csmkcar	concentrations due to smoking in car

Figure 6.6 Predicted PM_{10} Concentrations at Home, Indoors and Outdoors

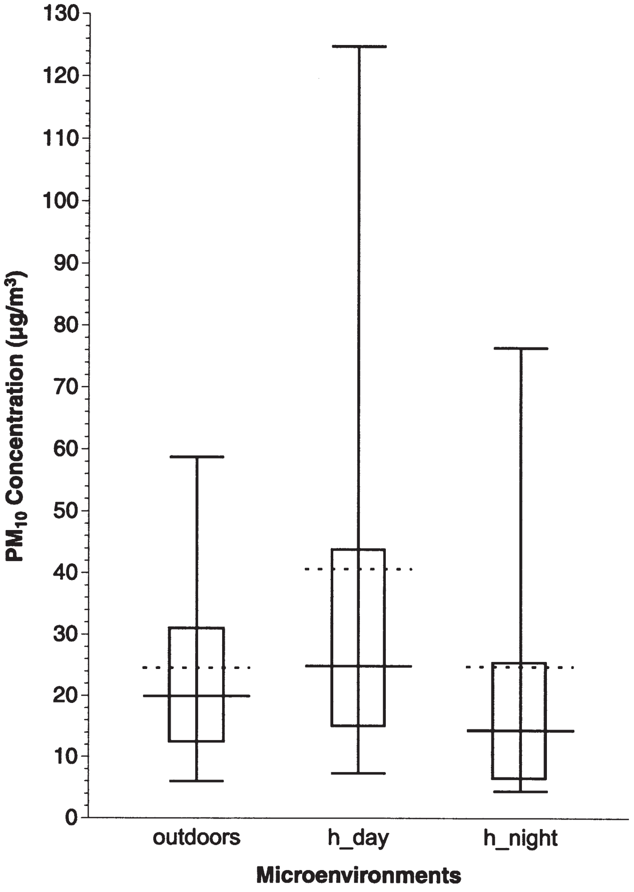
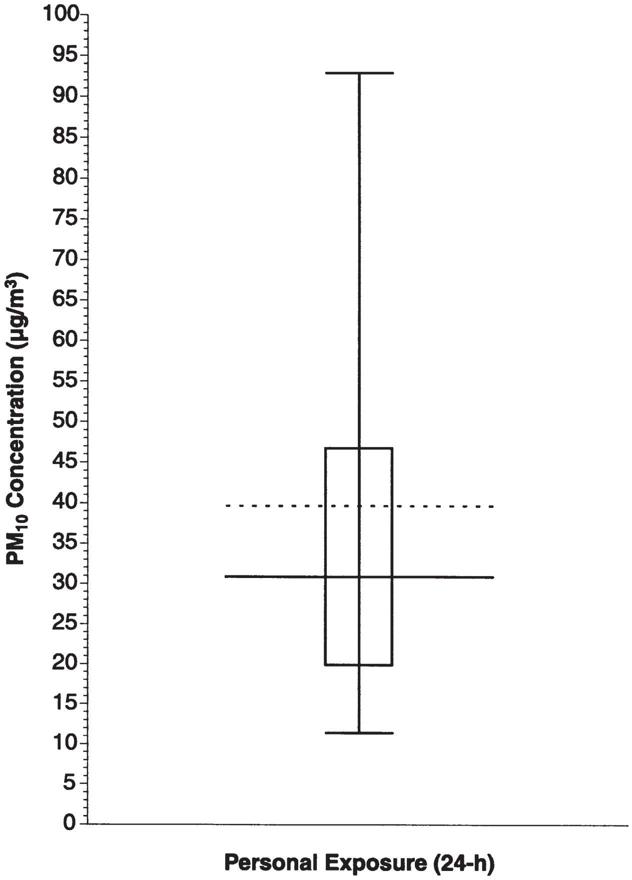


Figure 6.7 National Average Predicted Personal PM_{10} Exposure



Predicted 24 h average personal PM_{10} exposures (across all regions and seasons) is 39 µg/m^3 (Figure 6.7). Median personal PM_{10} exposure is predicted to be 31 µg/m^3 (solid line). The estimated PM_{10} exposure distributions are weighted by the frequency of residences in urban and rural locations, and region-specific population size. Little differences were seen between the predicted exposures in each region. Urban exposure distributions were slightly higher than rural exposures due to differences in the urban versus rural ambient PM_{10} levels. Exposures tended to be slightly higher in the winter than in the spring or summer. A more detailed breakdown of personal exposures by age and gender predicted teenagers and seniors to have lower exposures than the rest of the adult population because of their time-activity profiles and lower smoking rates.

The contributions to personal exposures were calculated; indoors at home and non-residential indoor exposures were predicted to contribute the greatest to personal exposures. Further breakdown of the indoors at home exposures revealed other indoor sources, for example, ETS and unaccounted other sources contribute significantly to residential indoor PM_{10} exposures. Penetration of ambient PM_{10} indoors contributed to indoor levels to a small degree. In-transit or auto-cabin exposures were predicted to be greater than both ambient and in bar/restaurant exposures.

The model predicted vast differences in the personal PM_{10} exposures of individuals exposed to smoke vs non-smoke exposed individuals. The difference is about 20 µg/m^3. Exposure to cooking does not influence the overall distribution of personal PM_{10} exposures.

Uncertainty analysis revealed that uncertainties in the PM_{10} removal or deposition rate, and uncertainties in the estimate of the other indoor residential PM_{10} source strengths strongly influence the uncertainties in the Canadian population distribution of personal PM_{10} exposures. pTEAM data was used to estimate source strengths and infiltration of the high ambient PM10 levels may have biased the statistical model estimates. The uncertainty analysis also revealed that personal PM_{10} exposure variability is strongly correlated (p=0.6) with indoor day or night PM_{10} concentrations.

Additional uncertainties in the estimated population distribution of PM_{10} exposures stem from the use of NHAPS time-activity data rather than Canadian data, which is fairly limited, and the use of non-residential indoor particle concentrations collected in other locations (non-Canadian). Also, the “person cloud” portion of the personal exposure was not incorporated into these estimates. If the pTEAM data, from which the source strengths were estimated, correctly estimated the proportion of “personal cloud” above ambient and indoor particle levels, the predicted microenvironmental PM_{10} exposures would be approximately 10 $\mu g/m^3$ greater.

6.6 DISCUSSION AND CONCLUSIONS

To the extent that microenvironmental concentrations (ambient and non-ambient PM) are different from outdoor concentrations, population exposures (total PM) are different from those estimated by ambient monitoring (ambient PM). Concentrations of particles from different sources are averaged to produce a 24 h personal exposure estimate. The fractions of fine and coarse particles of ambient origin that will be found indoors under equilibrium will tend toward 50% or less, particularly in the winter. The 24 h time-weighted average personal exposure estimate using only ambient data ignores potential exposure to particles in other microenvironments, such as indoors at home, at work, in transit and at other indoor locations. The high correlations between personal exposure and indoor concentrations combined with the time spent indoors indicate that indoor microenvironments are the most important contributors to overall average exposure.

Indoor particles come from the following sources: penetration of outdoor air into the building, indoor sources and generation or resuspension of particles during personal activities such as cooking, dusting and vacuuming (US EPA, 1982a, 1996; Clayton et al., 1993; Wallace et al., 1993; Thatcher and Layton, 1994). The latter source, the personal cloud or “Pigpen effect,” explains why personal exposure is greater than indirect estimates using ambient data, or indirect estimates combining indoor and outdoor microenvironments and time-activity information. The increase in particle concentration as a result of a person occupying the microenvironment is overlooked. Time-weighted average exposure estimates often do not capture the increase in particles due to resuspension, while in transit, and as a result of smoking or exposure to ETS.

The additional particle sources vary independently of the centrally located fixed ambient monitor (FAM). Limited data on particle concentrations with short-term averaging times reveal that peak concentrations occur indoors, outdoors, in traffic and while smoking. These sources are not represented by current ambient monitoring networks. Therefore, the relationship between ambient data and indoor and personal exposure is not linearly related. If there is a lot of variability in particle concentrations from sources poorly correlated with FAMs, then the percentage of variance in personal exposure to total PM that can be explained by the FAM data is small.

Epidemiological studies provide concentration-response information rather than exposure- or dose-response data by not linking the source of particulate matter with the exposure process and adverse outcome. Effects measured at a given concentration do not constitute a full risk assessment, because the number of people exposed, and the concentration they are exposed to is not known. The result is to underestimate the associated risks and their statistical significance. However, insufficient data exist to determine whether short-term exposure to high levels of particles results in the same degree of health impact as do longer-term, lower-concentration exposures.

Epidemiology studies are based on PM measurements at a central site (FAM). PM concentrations at FAM sites can represent the exposure of the people in the community to PM of ambient origin. However, comparing parts of the distribution of exposure predicted using urban NAPS sites with the predicted exposures from the probabilistic exposure model clearly shows ambient data underestimate average population exposures to total PM (Table 6.17). Although estimates of exposure to $PM_{2.5}$ have not been predicted through exposure modelling, using the mean ratio of ambient $PM_{2.5}/PM_{10}$ presented in Table 5.20 (0.50), population exposure estimates to fine particles were estimated.

Table 6.17 Predicted Mean, Median, High End Exposure Estimates to Ambient PM (µg/m^3)

	mean	SD	median	P95	max
Urban Ambient PM_{10} Data	27.6	16.3	24.0	58.0	175.0
Predicted PM_{10} Exposures	39.4	33.5	30.9	93.0	682.0
Urban Ambient $PM_{2.5}$ Data	13.9	9.5	11.0	32.2	89.0
Calculated $PM_{2.5}$ Exposures	19.7	16.75	15.5	46.5	341

Given the current information, concluding that the ambient data alone can represent the lower range in the distribution of total particle exposures is reasonable.

6.7 FUTURE REQUIREMENTS

Sources of PM_{10} and $PM_{2.5}$ vary among different provinces as well as between various indoor and outdoor environments. Consequently, the contributions of ambient particles to indoor particle concentrations and subsequently, to total personal exposures will vary by different geographic and climate zones.

It is recommended that special studies be carried out to determine the exact nature of the indoor sources of indoor air particles.

It is recommended that special studies be carried out to determine the penetration rate of ambient particles indoors, specifically in colder climates, where the air exchange rates are a function of ambient temperature.

7 THE AESTHETIC EFFECTS OF PARTICULATE MATTER (VISIBILITY)

Increasing concentrations of fine particles and gases in the atmosphere often result in reduced visual range (or degradation of visibility), by creating a haze that obscures the clarity and structure of what is observed. Visibility is one of the most readily perceived indicators by the public of poor air quality, and is often cited as a concern in urban and wilderness areas. Historically, visual range has been an important weather parameter relevant to the safe operation of aircraft. Visual ranges less than 24.1 km (PM$_{2.5}$ ≈ 18 µg/m^3) present a hazard to aircraft using airports; visual ranges of less than 1 km may cause airports to close.

Prevention of visibility deterioration in cities, national parks and wilderness areas is also relevant due to potential loss of tourism and quality of life. Although the aesthetic effects of visibility degradation are subjective in nature, several perception studies have attempted to quantify the significance of reduced visual range. These include determining what is considered “acceptable” visual range (Pryor et al., 1994b), assessing the economic value of incremental changes in visual range (Chestnut and Latimer, 1994), and estimating the amount of time visitors are willing to drive the see certain vistas (Malm et al., 1984).

This chapter will focus on the relationship of TSP, PM$_{10,}$ and PM$_{2.5}$ to visual range. More detailed reviews on effects of particles on visual range are available: Malm, 1979; Pueschel, 1983; Hoff, 1983; Hoff and Barrie, 1985; Sloane, 1986; White, 1986; Trijonis et al., 1990; Laulainen, 1993.

7.1 THE RELATIONSHIP BETWEEN PARTICULATE MATTER AND VISUAL RANGE

There is a direct relationship between the amount of particulate matter in the atmosphere and the ability of the human eye to see through the atmosphere. Perception of distant objects involves contrast, where the contrast of the distant object against the background (usually the sky) decreases as the distance between the object and the observer increases. Visual range is the measure of transparency of the atmosphere, defined as the distance at which the contrast of the target is equal to the threshold contrast value for the human eye.

Physically, visual range is a function of the light scattering and absorption properties of gas molecules and particles. The light extinction coefficient (b_{ext}) is a measure of light attenuation as a result of scattering and absorption by gas molecules and aerosol particles along the direction of propagation of the light beam. The Beer-Lambert law mathematically expresses this relationship:

$$\begin{aligned} d_{II} &= -b_{ext}dx \\ &= -(b_{scat} + b_{abs})dx \\ &= -(b_{RG} + b_{sp} + b_{ag} + b_{ap})dx \end{aligned} \tag{7.1}$$

$$b_{scat} = b_{RG} + b_{sp} \tag{7.2}$$

$$b_{abs} = b_{ag} + b_{ap} \tag{7.3}$$

where:

b_{ext} = light extinction coefficient (distance^{-1}),

b_{scat} = light scattering coefficient (distance^{-1}),

b_{abs} = light absorption coefficient (distance^{-1}),

I = intensity of the light beam,

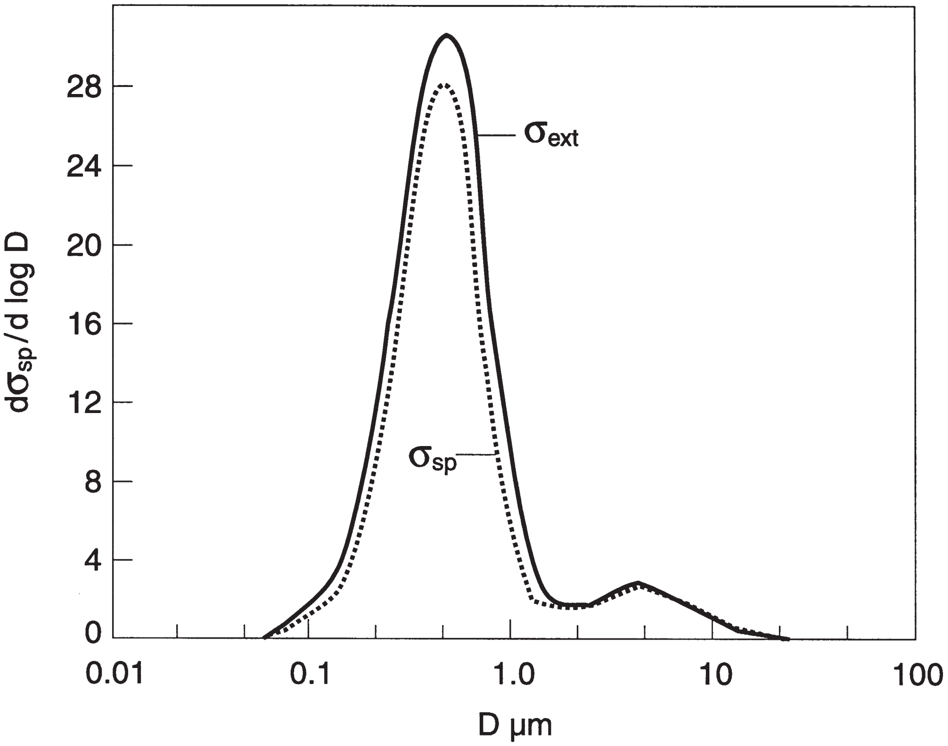
dI = the change of intensity as the light beam traverses a distance

$b_{RG} + b_{sp}$ = the air molecules (Rayleigh) and particle light scattering coefficients in units of distance^{-1}, and

$b_{ag} + b_{ap}$ = the gas molecule and particle light absorption coefficients in units of distance^{-1}.

Equation 7.1 shows that the light extinction coefficient, b_{ext}, is the sum of the light scattering and light absorption coefficients (b_{scat} and b_{abs}, respectively). Waggoner et al. (1981) showed that particle size is the most important factor in determining the relationship between particle mass and b_{scat}. Figure 7.1 shows the particle extinction cross sections for light

Figure 7.1 Particle Extinction Cross Sections for Extinction (σ_{ext}) and Scattering (σ_{sp}) as a Function of Particle Size Diameter (from Waggoner et al., 1981)



extinction and scattering as a function of particle size diameter. This figure also shows that light scattering is the dominant component of light extinction (i.e., light absorption is usually relatively small). Waggoner and Weiss (1980) showed that single particle optical cross section per unit particle volume reaches a maximum for particles with diameters in the range of 0.3-0.7 µm. Thus, fine particles (<2.5 µm) dominate light scattering and hence light extinction. In other words, fine particles ($PM_{2.5}$) and submicrometre particles ($PM_{<1}$) in particular are most effective in reducing visual range.

Visual range (*VR*) is determined as follows:

$$VR = \ln(C(0)/Cd)/b_{ext} \qquad (7.4)$$

where:

b_{ext} = the light extinction coefficient in units of distance^{-1},

$C(0)$ = the object's inherent contrast (contrast at zero distance), and

Cd = the threshold for detection of the object's contrast.

If the object is black then $C(0)$ = -1 and the threshold for detection of the object's contrast has been estimated to be 2% ($|Cd|$=0.02). Equation 7.4 can be simplified for a perfect (i.e., non reflecting) black object to the Koschmieder equation:

$$VR = 3.91/b_{ext} \qquad (7.5)$$

where visual range is in distance units and the factor 3.91 is based upon a 2% threshold value for distinction of an object from background (Charlson et al., 1968). Equation 7.5, the Koschmieder equation, describes the relationship between a measured atmospheric parameter, b_{ext}, and human perception of visual range. The Koschmieder equation incorporates several assumptions: the atmosphere is homogeneous such that scattering and absorption of radiation are the same everywhere, sky brightness is the same at the object, the background and the observer (cloudless sky), and the viewing distance is horizontal, i.e., the earth's curvature is ignored (Seinfeld, 1986). A number of authors have used this relationship to compare measured light-scattering coefficients with visual range studies (Dzubay et al., 1982; Pitchford and Malm, 1994; Pryor, 1996).

7.2 PARTICLE CHARACTERISTICS AND THE LIGHT EXTINCTION COEFFICIENT

Particle mass, hygroscopicity and particle composition all affect the degree to which ambient particulate matter may affect visual range. Light scattering is the sum of Rayleigh scattering, b_{RG} , (due to gaseous molecules) and particle scattering, b_{sp}. In a virtually particle free atmosphere Rayleigh scattering predominates. Rayleigh scattering at sea level is $1.0 \times 10^{-5} m^{-1}$ at 550 nm and decreases from $0.88 \times 10^{-5} m^{-1}$ to $0.61 \times 10^{-5} m^{-1}$ for altitudes of 1000 to 4000 m (Waggoner et al., 1981). As particle concentration increases, particle scattering will increase until $b_{sp} \geq b_{RG}$.

Light scattering dominates light absorption except where there are light absorbing particles or gases present. Graphitic or elemental carbon (commonly known as soot) is very efficient at absorbing light. Particle absorption (b_{ap}) is $\leq$10% of particle scattering *(b_{sp})* in rural areas but can be nearly equal to particle scattering in urban areas where graphite carbon is present (Waggoner and Weiss, 1980). NO_2 is the only light absorbing gaseous species present that is optically significant. This gas absorbs light wavelengths at the blue end of the visible spectrum, resulting in a brownish colour. The presence of NO_2 is important in studies on plume optics but not in the case of a well-mixed layer (Waggoner, 1980).

Measurements of visual range taken with a nephelometer are expressed in terms of the scattering coefficient (b_{scat}), while measurements done using transmissometers are expressed in terms of b_{ext}. In order to calculate visual range (equations 7.4 & 7.5) the value of b_{ext} must be measured or calculated from the ratio of b_{scat}/b_{ext}, which is assumed to be constant. As the presence of graphite carbon influences the particle absorption coefficient, b_{abs}, and thus b_{ext} in urban areas, the ratio of b_{scat}/b_{ext} is estimated separately for urban and rural areas. Waggoner et al. (1981) calculated a ratio of b_{scat}/b_{ext} for two locations near Buffalo National River in northeastern Arkansas from b_{scat} measured with a nephelometer and b_{ext} calculated from equation 7.5 with telephotometer measurements of sky and target radiance. The correlation of b_{scat} and b_{ext} from these instruments ranged from 0.84 to 0.99. There was a higher correlation for cloud-free days. A ratio of b_{scat}/b_{ext} of was calculated for all days (0.77 to 1.20), for cloud-free days (0.8 to 1.04), and was dependent on time of the day. On average for this rural site, b_{scat} was about 0.9 of b_{ext}. For sites located in the metropolitan area of Denver, the ratio of mean b_{scat}/b_{ext} was 0.7 (Middleton et al., 1984), confirming the larger role of absorption in light extinction in urban areas. Similarly, in Houston, Texas, the ratio of b_{scat}/b_{ext} was 0.77 (Dzubay et al., 1982), and in the Fraser Valley, the mean ratio of b_{scat}/b_{ext} was 0.6 (Chilliwack site; Pryor et al., 1995a). From these values, a reasonable estimate of b_{ext} can be determined from:

$$b_{scat}/b_{ext} = 0.7 \text{ in urban areas, and} \qquad (7.6)$$

$$b_{scat}/b_{ext} = 0.9 \text{ in rural areas.} \qquad (7.7)$$

7.2.1 Relationship Between Particle Mass and Light Extinction and Scattering Coefficients

A number of studies have found that there is a linear relationship between particle mass and b_{ext} or b_{scat}. Table 7.1 shows a range in values determined for the ratio of b_{ext} or b_{scat} to particle mass. In early studies, Charlson et al. (1968) suggested that TSP/b_{scat} ranges from 0.2 to 0.7 g/m^2 (b_{scat}/TSP = 1.4-5 m^2/g). For particles of radius < 50 µm, the ratio of b_{scat}/TSP was calculated to be 1.6-2 m^2/g (Horvath and Charlson, 1969). Recent studies have focused on fine particles, of size range 0.1-2.5 µm, due to their efficiency at light extinction. Consequently, a stronger relationship was found when fine particulate mass is used instead of TSP (Table 7.1). Waggoner et al. (1981) compared Charlson et al. (1968) relationship of TSP and b_{scat} at a number of US urban sites with that of fine particulate mass <2.5 µm. A linear relation was observed between TSP and b_{scat}, with a ratio of b_{scat}/TSP $\approx$ 2, but there was more variation in the ratio than for b_{scat} /fine particulate. Waggoner et al. (1981) found that fine particulate mass and b_{scat} has a correlation >0.95. The relationship of b_{scat}/fine mass has been shown to be 3.1 ± 0.2 m^2/g for a number of urban and rural sites in the United States (Waggoner and Weiss, 1980).

The ratios of b_{ext}/mass and b_{scat}/mass generally decrease when mass includes coarse particles (PM_{10} or TSP). A stronger correlation has been shown to exist between fine particulate mass and light scattering, therefore, linear relationships derived for coarse particles and light scattering can be expected to have more error associated with them. For some US sites (Lewis et al., 1986; White and Macias, 1990), ratios of b_{ext}/fine particle mass in the range of 3.3-4.3 m^2/g have been observed. These

Table 7.1 Comparison of Relationship of Coefficient of Scattering or Extinction with Particle Mass

Site	Ratio	Value (m^2/g)	Reference
Mesa Verde, CO (pristine)	b_{scat} / $PM_{2.5}$	2.94	Waggoner and Weiss (1980), Waggoner et al. (1981)
Seattle, WA (residential)		3.13	
Seattle, WA (industrial)		3.23	
Puget Island, WA (rural)		3.03	
Portland, OR (industrial)		3.23	
Denver, CO	b_{ext} / $PM_{2.5}$	4.3	Lewis et al. (1986)
	b_{ext} / PM_{15}	2.1	
Spirit Mtn., NV (rural US)	b_{ext} / $PM_{2.5}$	3.3	White and Macias (1990)
	b_{ext} / PM_{15}	1.56	
Urban US sites	b_{scat} / TSP	1.4-5	Charlson et al. (1968)
from regression line	b_{scat} / TSP	2.6	
Airport urban data and TSP	b_{ext} / TSP	2	Noll et al. (1968)
Urban	$b_{p,scat}$ / TSP	3.4	Kretzschmar, (1975)
Calculated for particles $r < 50$ µm	b_{scat} / mass	1.6-2.0	Horvath and Charlson (1969)
Santiago, Chile	b_{ext} / PM_{10}	5	Trier and Horvath (1993)
Berlin, Germany	b_{ext} / PM_{10}	6.0	Heits and Israel (1982)
Vancouver, B.C.	b_{scat} / $PM_{2.5}$	3.0	Guise-Bagley and Hoff (1993)
Houston, TX	b_{scat} / $PM_{2.5}$	3.52	Dzubay et al. (1982)

ratios are only slightly higher than the ratio of b_{scat}/ fine particle mass of 3.1 m^2/g reported by Waggoner and Weiss (1980). A value of 3.1m^2/g was used for b_{scat}/fine mass in this report as it represents an average of a large number of urban and rural sites and is similar to measurements taken at Vancouver (3.0 m^2/g). Currently, site specific data for b_{scat}/fine particulate mass is not available for Canadian visibility monitoring sites (Waterton, Egbert, St. Andrews), thus for the purposes of this assessment:

$$b_{scat}/PM_{2.5} = 3.1\ m^2/g \quad (7.8)$$

Pitchford (1982) showed that coarse particles could also disrupt visibility. Coarse particles were shown to be one-half to one-third as effective as fine particles in disrupting visibility. However, in some areas, where coarse particle concentrations are unusually high, they can contribute from 30 to 80% of total light extinction (Pitchford, 1982). Following Malm (1993), if:

b_{scat} / $PM_{2.5}$ = 3.1 m^2/g and b_{scat} / ($PM_{10} - PM_{2.5}$) = 0.6 then

$PM_{2.5} = 0.6\ PM_{10} - 0.6\ PM_{2.5} = b_{scat}$.

Thus, for the contributions to reduced visual range to be equal, PM_{10} must equal 6 × $PM_{2.5}$. More simply put:

$$b_{scat} = 3.1\ PM_{2.5} + 0.6\ (PM_{10} - PM_{2.5}) \quad (7.9)$$

Site specific data is preferable for deriving relationships between light extinction or scattering and particle mass given the variability among sites in particle characteristics and particle mass.

7.2.2 Effects of Particle Composition on Visual Range

Sulphate, nitrate, soil, organic carbon and elemental carbon dominate fine particulate matter composition (Pryor, 1994; Malm, 1992). A number of studies have examined the relative importance of chemical species to reduced visual range (Hidy and Brock, 1971; Cass, 1976; White and Roberts, 1977; Leader et al., 1979; Trijonis, 1980, 1982; Groblicki et al., 1981; Dzubay et al., 1982; Malm, 1992; Sisler and Malm, 1994). These studies found that sulphates are the largest contributor to visual range reduction, and that nitrates are also significant contributors but less efficient per unit mass than sulphates in light scattering.

Appel et al. (1985) showed however, that in some areas, nitrate contributions to light scattering were greater than those of sulphates (sulphate 30.4% and nitrate 45.1%; San Jose, Riverside and downtown Los Angeles). Pryor et al. (1996a) found that for the Fraser Valley, fine mass was dominated by a secondary organic component, but sulphate and ammonium nitrate dominated light scattering. Particle-phase organic compounds are relatively inefficient contributors to light scattering. Elemental or black carbon contributes to light absorption and, to a small extent, light scattering. Measurements from the IMPROVE network in the United States show that ammonium sulphate and organic carbon are the largest contributors to fine particulate mass and reduced visibility (Malm, 1992).

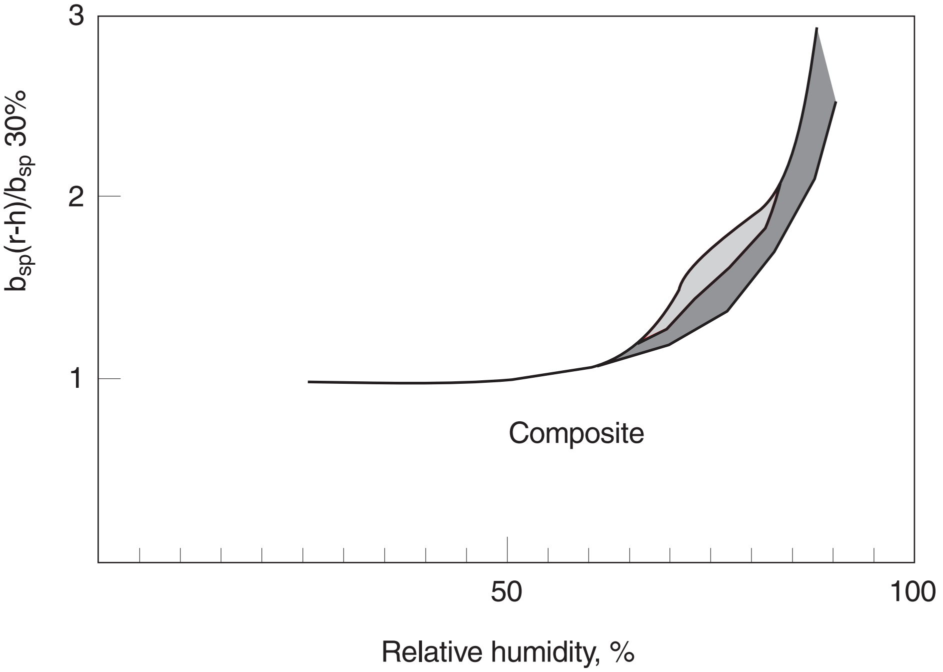
7.2.3 Effects of Relative Humidity on Light Extinction and Scattering Coefficients

Relative humidity influences particle light extinction. When relative humidity exceeds 70%, light scattering efficiencies begin to increase (Charlson et al., 1984). As humidity increases to 95% or more, water soluble components of the fine aerosol can absorb water and swell to more than seven times their dry radius. Covert et al. (1980) showed that scattering increases by a factor of two to three times at many US sites for a change in relative humidity of 30 to 80%, as shown in Figure 7.2. Pitchford and McMurry (1994) studies further confirm this behaviour. The effect of humidity on light scattering properties is also very dependent on chemical and microphysical variables. Components of fine particles, such as sulphates (ammonium sulphates), nitrates (ammonium nitrate), organics, elemental carbon and soil dust, will vary in their ability to absorb water. The hygroscopic fraction of the aerosol will affect visual range largely in regions of Canada with higher relative humidity.

7.3 PARTICULATE MATTER LEVELS AND ESTIMATED VISUAL RANGES

An estimate of visual range and particle levels can be determined using the Koschmieder equation (equation 7.5) by assuming a ratio of light scattering coefficient to fine particle mass of 3.1 m^2/g and ratios of b_{ext} to b_{scat} given in equations 7.6 and 7.7 for urban and rural sites. Brook et al. (1996) calculated that $PM_{2.5}$ accounted for 49% of PM_{10} and PM_{10} accounted for 44% of the TSP, on average,

Figure 7.2 Measured Enhancement of Scattering (*B_{sp}*) as a Function of Relative Humidity.



Note: Hatched area contains the complete range of ambient measurements illustrating the difference between sites in the US West and Midwest (Covert et al., 1980).

across Canadian sites. (Note: the majority of these sites are urban, therefore the following relationships may better characterize urban than rural locations.) Based on the linear regression of $PM_{2.5}$ and PM_{10} with TSP for all Canadian sites, the equivalent PM_{10} and $PM_{2.5}$ levels can be calculated as follows:

$$\text{equivalent } PM_{10} = 0.44 \times TSP \quad (7.10)$$

$$\text{equivalent } PM_{2.5} = 0.21 \times TSP \quad (7.11)$$

$$\text{equivalent } PM_{2.5} = 0.73 + 0.49 \times PM_{10} \quad (7.12)$$

where particle concentrations are in units of µg/m^3. Visual range and b_{scat} were calculated from the fine particle masses (units g/m^3) and are presented in Table 7.2. b_{scat}, and hence b_{ext}, are estimates, and the visual range calculation assumes that the particles are non-absorbing, a 2% threshold value for distinction of an object from the background and relative humidity <70%. Actual visual ranges may be significantly less in regions with relative humidity in excess of 70%.

7.3.1 Visual Range Estimates for Current PM Air Quality Criteria

Table 7.2 lists the current Canadian TSP ambient air quality objectives, equivalent PM_{10} and $PM_{2.5}$ levels (Dann, 1994), 24 h and annual US PM_{10} and $PM_{2.5}$ standards and the 24 h British Columbia objectives and Ontario (interim) PM_{10} standard. The PM air quality criteria in Table 7.2 were converted to PM_{10} and $PM_{2.5}$ equivalents using the relationships presented above. Visual range and b_{scat} was calculated using the fine particle mass equivalents. As previously stated, the size fraction of particles and components primarily associated with light scattering causing reduced visibility is <2.5 µm (dominated by particles with diameter 0.3-0.7 µm).

7.3.2 Visual Range Estimates for Estimated Natural Visibility

Table 7.3 shows estimated ranges of natural visibility (and b_{scat}) at a number of non-urban sites in the United States and Canada (Trijonis, 1982; Malm, 1992; Hoff et al., 1997; Fox et al., 1997) and calculated fine particulate levels. b_{scat} is determined from nephelometer readings and is used to calculate visual range based upon equations 7.7 and 7.5. The fine particulate levels are estimated from equation 7.8 for a number of urban and rural sites in the United States (Waggoner and Weiss, 1980).

Natural visual ranges in Canada, estimated from the 10th percentile in the b_{scat} distribution, are 86 to 350 km and from these, estimated natural $PM_{2.5}$ levels

Table 7.2 Visibility Equivalents of Current Canadian, US and California Particulate Matter Objectives and Guidelines

Particulate standard or objective (µg/m^3)	Equivalent PM_{10} (µg/m^3)	Equivalent $PM_{2.5}$ (µg/m^3)	Visual Range (km)		b_{scat} (×10^6/m)
			Rural	Urban	
TSP 400 24 h – Cdn TSP Tolerable level	180	84	14	11	260
TSP 120 24 h – Cdn Acceptable 24 h Level	53	25	45	35	78
TSP 70 annual – Cdn Acceptable Annual Level	31	15	77	60	46
PM_{10} 150 24 h – US NAAQS	150	74	15	12	230
PM_{10} 50 24 h – BC + Ontario (interim) objective and US annual NAAQS	50	25	45	35	78
$PM_{2.5}$ 65 24 h – New US NAAQS		65	14	18	201
$PM_{2.5}$ 15 annual – New US NAAQS		15	76	59	47
Lower range mean Cdn urban and rural $PM_{2.5}$ levels (Dann, 1994)		8	140	110	25
Upper range mean Cdn rural $PM_{2.5}$ levels (Dann, 1994)		11	100	80	34
Upper range mean Cdn urban $PM_{2.5}$ levels (Dann, 1994)		22	50	40	68

are 3-13 µg/m^3 (Table 7.3). Natural visual range exists in regions not directly impacted by anthropogenic emissions. Western Canada and eastern Canada have estimated natural levels of $PM_{2.5} \leq 6$ µg/m^3, while southeastern Canada has a higher natural $PM_{2.5}$ level of 10-13 µg/m^3. A 10% change in visual range is considered the minimum observable incremental change (see section 7.4). Given that there is a linear relationship between visual range and $PM_{2.5}$, a 10% increase in $PM_{2.5}$ levels above background levels (Table 7.3) represents the change required to detect effects (i.e., decreased visibility). Thus, in western and eastern Canada, impacts on visual range, above background levels, would be detectable when $PM_{2.5}$ levels exceed 6 to 7 µg/m^3 and for southeastern Canada, 14 µg/m^3, using upper limits of estimated $PM_{2.5}$ levels.

7.3.3 Visual Range Estimates for Canadian Sites

As detailed in Chapter 5, the relative distribution of PM_{10} to TSP ratios and $PM_{2.5}$ to TSP ratios is a function of geographic location as well as proximity of the monitoring station to local sources. At Canadian sites, the mean ratio of PM_{10} to TSP ranges from 0.36 to 0.79. PM_{10} to TSP ratios are significantly lower at Prairie sites than at eastern Canadian and Vancouver sites (Prairie sites, PM_{10}/TSP = 0.36-0.53, compared with Vancouver and Halifax, PM_{10}/TSP = 0.67-0.77). Mean $PM_{2.5}$ to TSP ratios range from 0.20 to 0.50. The majority of NAPS sites show 50 to 60% of PM_{10} as $PM_{2.5}$ size fraction (Table 7.4). Prairie urban sites such as Edmonton, Winnipeg, and Calgary had less than 50% of PM_{10} as $PM_{2.5}$ (Dann, 1994). The values for PM_{10}/TSP and $PM_{2.5}$/TSP ratios presented here differ slightly from those in chapter 5, Table 5.20 as somewhat different databases were used in each case. At urban sites across Canada, estimated visual range extends from 34 to 73 km, and from 93 to 136 km at rural sites.

As $PM_{2.5}$ is primarily responsible for the scattering of light, visual ranges at current TSP air quality objective levels were estimated for specific urban sites using a $PM_{2.5}$ level estimated from the $PM_{2.5}$/TSP ratio for each site (Table 7.5). Equivalent PM_{10} levels were also calculated for the NAPs sites based on ratios of PM_{10}/TSP for each of the sites. The Prairie sites were selected as they had the lowest values of PM_{10}/TSP and $PM_{2.5}$/TSP, while Vancouver and Halifax had the highest values. This table shows that visual range in the Prairies (which had the lowest values of PM_{10}/TSP and $PM_{2.5}$/TSP) are much greater than in Vancouver or Halifax (which had the highest values of PM_{10}/TSP and $PM_{2.5}$/TSP), for a specified TSP level. For example, for PM_{10} levels ranging from 22-32 µg/m^3 (equivalent to TSP levels of 60 µg/m^3), and assuming $b_{scat}/b_{ext} = 0.7$ for urban sites (equation 7.6), visual ranges of 64 to 92 km versus 33 to 36 km occur at Prairie versus Vancouver and Halifax sites respectively.

Table 7.3 Estimated Natural Visual Ranges in North America

Site location (time period)		Visual range, km (estimated from b_{scat} (x 10^6/m))	Estimated $PM_{2.5}$ (µg/m^3)	Reference
Natural, eastern US		100-130	8.8-12.0	Malm (1992)
Natural, western US		170-180	6.4-6.7	Malm (1992)
Visibility observations, western Great Basin & Desert southwest US		100-130	8.8-12.0	Trijonis (1982)
Western US (Glacier National Park)	Seasonal best 10% of b_{scat} 93-94	150-160(22-23)	7.0-7.5	Fox et al. (1997)
Western Canada (Waterton, Alberta)	Seasonal best 10% of b_{scat} 93-95	210-350(10-17)	3.2-5.5	Hoff et al. (1997) Fox et al. (1997)
Southeastern Canada (Egbert, Ontario)	Seasonal best 10% of b_{scat} 93-95	86-120(30-41)	9.7-13.0	Hoff et al. (1997)
Eastern Canada (St. Andrews, N.B.)	Seasonal best 10% of b_{scat} 95	185-210(17-19)	5.5-6.1	Hoff et al. (1997)

Note: VR calculated from b_{scat}/b_{ext} =0.9, $VR = 3.91/b_{ext}$, and $PM_{2.5}$ calculated from $b_{scat}/PM_{2.5} = 3.1 m^2/g$.

Table 7.4 Estimated Visual Ranges Associated with Mean 24 h $PM_{2.5}$ and PM_{10} Levels (1984-1993)

Station no.	City	Mean $PM_{2.5}$ (µg/m^3)	Mean PM_{10} (µg/m^3)	Estimated Visual Range (km)
40203	Saint John	10.7	18.3	73
30101	Halifax	14.3	26.0	53
30501	Kejimkujik	7.5	11.8	136
50104	Montréal	16.1	28.1	48
50109	Montréal	21.4	45.7	34
50307	Québec City	12.1	24.3	61
54101	Sutton	7.9	11.6	132
60104	Ottawa	12.7	22.8	60
60204	Windsor	18.6	32.7	41
60211	Windsor	17.2	30.4	45
604117	Toronto	16.7	27.9	47
60424	Toronto	17.6	29.3	44
61901	Walpole Is.	18.1	30.3	43
64401	Egbert	10.9	17.9	93
70119	Winnipeg	10.4	29.0	63
90130	Edmonton	10.5	27.3	64
90204	Calgary	14.4	32.1	50
90227	Calgary	10.1	24.8	68
100106	Vancouver	17.3	27.7	46
100111	Vancouver	15.8	27.2	49
100118	Vancouver	14.1	23.0	56
100303	Victoria	11.6	17.9	69

1. Visibility calculated from equivalent $PM_{2.5}$ levels,$b_{scat}/PM_{2.5} = 3.1 m^2/g$, $b_{scat}/b_{ext} = 0.7$, and $VR = 3.91/b_{ext}$.
2. $b_{scat}/b_{ext} = 0.9$ was used for the three rural sites of Kejimkujik, Sutton and Egbert.
3. These estimates include contributions to reduced visual range from both the fine and coarse fraction according to equation 7.9.

Table 7.5 Visual Range Estimates for PM_{10} and $PM_{2.5}$ Estimated Levels (µg/m^3) Based on TSP NAAQO

TSP (µg/m^3)	Prairie Sites			Vancouver and Halifax		
	Equivalent PM_{10} (µg/m^3)	Equivalent $PM_{2.5}$ (µg/m^3)	Visibility (km) estimate from $PM_{2.5}$	Equivalent PM_{10} (µg/m^3)	Equivalent $PM_{2.5}$ (µg/m^3)	Visibility (km) estimated from $PM_{2.5}$
120	43-64	19-28	32-46	80-92	49-53	17-18
70	25-37	11-16	55-79	47-54	29-31	29-31
60	22-32	10-14	64-92	40-46	25-26	33-36

1. Canadian TSP air quality objectives: annual mean 70 µg/m^3 (geometric mean 60 µg/m^3), 24 h 120 µg/m^3.
2. $PM_{2.5}$ calculated from $PM_{2.5}$/TSP of 0.16-0.23 for Prairies and 0.41-0.44 for Vancouver and Halifax.
3. Visibility calculated from equivalent $PM_{2.5}$ levels,$b_{scat}/PM_{2.5} = 3.1 m^2/g$, $b_{scat}/b_{ext} = 0.7$, and $VR = 3.91/b_{ext}$.

7.4 PUBLIC PERCEPTION OF DETERIORATION IN VISIBILITY

In addition to the regional differences in visual range for a given particulate loading, there are regional differences in the public's response to changes in visual air quality. Differences in public perception of unacceptable visibility may be related to the aesthetic appeal of environments and vistas viewed, as the public may be more willing to pay for protection of a wilderness area or unique urban skyline. Perception of colour, contrast of markers with background (usually sky) and perception of contrast detail in the atmosphere will affect decisions of acceptable or unacceptable visibility. Since acceptable visibility is subjective, it is important to collect feedback from a population sample that would represent the entire spectrum of "the public". Public perception studies typically use photographs of a limited number of scenes taken during similar times of the day to determine what is an acceptable visual range or what change in visual range is acceptable. Public perception studies have been conducted in wilderness areas and national parks in the United States (Latimer et al., 1981) and the Denver region (Ely et al., 1991; Middleton and Burns, 1991). Public perception studies take different approaches (Malm et al., 1981; Middleton et al., 1985; Ely et al., 1991). Some place a monetary value upon an incremental improvement in visibility (Chestnut and Latimer, 1994). Other studies have examined the relationship between acceptable visual range thresholds and population response of what is acceptable or unacceptable visibility (Pryor et al., 1996a).

The subjective nature of perceived visual range and the need to determine the tolerance of society to changes in visual range (which can vary significantly in different regions) prompted Pitchford and Malm (1994) to propose a scale based on perceived changes in visibility. It's referred to as the deciview scale, *dv*, where:

$$dv = 10 \times \ln(b_{ext}/0.01\ \mathrm{km}^{-1}) = 10 \times \ln(391/VR) \qquad (7.13)$$

A change in 1.0 *dv* corresponds to a 10% change in b_{ext}, which is a small but noticeable scenic change under many circumstances:

$$dv = 10 \ln(b_{ext}/0.01) \text{ and } d(dv)/db_{ext} = 10\,(0.01/b_{ext})(1/0.01) = 10/b_{ext}$$

therefore $d(dv) = 10\ db/b_{ext}$ or if $d(dv) = \pm 1$ then $db/b_{ext} = \pm 0.1$ or 10% change in b_{ext}.

Figure 7.3 illustrates the relationship of haziness and extinction coefficient or standard visual range (Pitchford and Malm, 1994).

Figure 7.3 Visual Range and Extinction Coefficient as a Function of Haziness Expressed in Deciview (from Pitchford and Malm, 1994)

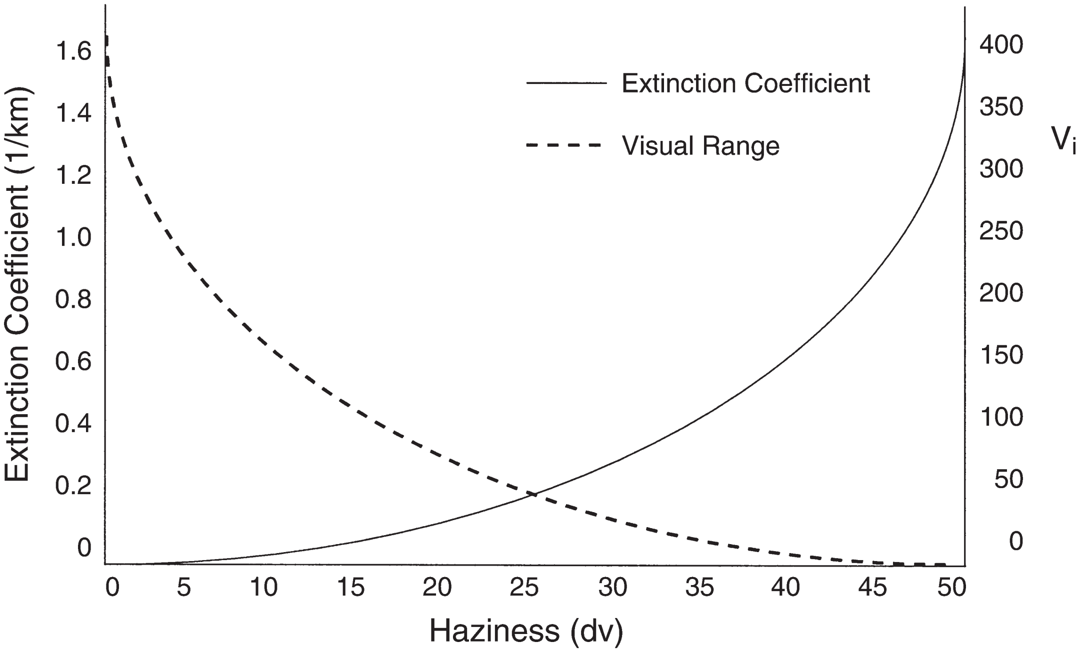
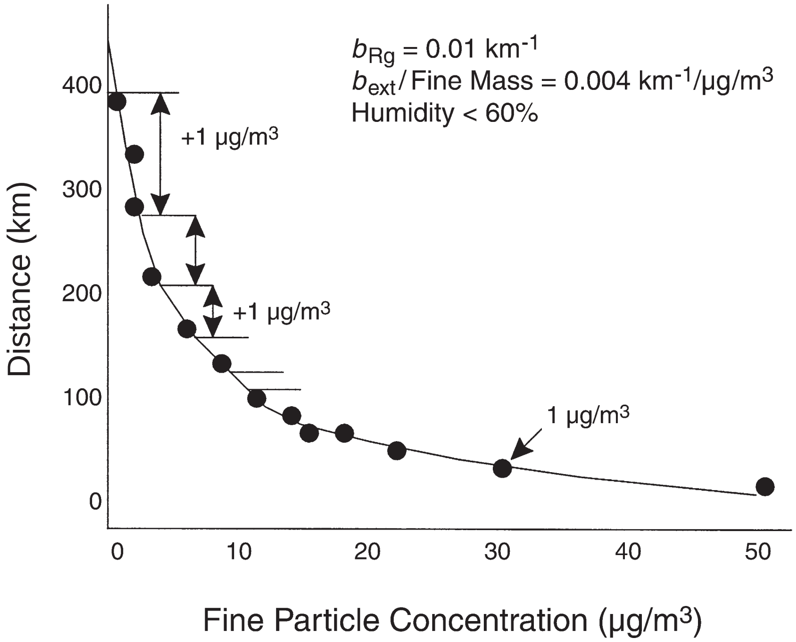


Figure 7.4 Effects of Fine Particle Increments on Calculated Visual Range (from Littlejohn et al., 1981)



A 10% change from existing fine particle levels results in a noticeable change in visibility, illustrated in Figure 7.4. Again, the figure demonstrates that when fine particle levels are low, an addition of 1 µg/m^3 will result in a larger change in visual range than at higher particle levels (Littlejohn et al., 1981).

Figure 7.4 shows that at a level of 10 µg/m^3, a 1 µg/m^3 change would be noticeable. At a level of 30 µg/m^3, a 3 µg/m^3 change would be required to cause a noticeable change in visibility. Consequently, in regions of Canada such as the Prairies, where fine particle levels are lower, a noticeable change in visibility will occur at a lower particle level than for Vancouver or Halifax. Figure 7.4 also shows that the addition of 1 µg/m^3 to a clean atmosphere reduces visual range by 30%. The addition of the same amount when the background visual range is 35 km (20 miles) produces a 3% reduction.

From a comparison of $PM_{2.5}$ and PM_{10} levels at Canadian rural and urban sites, a noticeable change in visual range could occur for a 1-2 µg/m^3 increase in PM_{10} at rural sites and a 2-5 µg/m^3 increase at urban sites. The deciview scale does not consider what the particle level is, rather, it considers the incremental change in visibility, or percent change in particulate matter, from existing levels.

In a recent study in the Lower Fraser Valley of British Columbia, judgements of acceptable or unacceptable visibility were made from photographs (Pryor et al., 1994b). Other studies have shown that there is a high correlation between judgements made from photographs and those made in field (Malm et al., 1981; Middleton et al., 1985). In a public perception study in Denver (Ely et al., 1991), acceptable visibility as measured by visual range was approximately 50 km. Acceptable visibility or acceptable changes in visibility in different regions of Canada still need to be determined.

The effects of changes in particle levels on visibility across Canada have also been evaluated from visual range estimates from meteorological synoptic observing stations located at airport sites (Stuart and Hoff, 1994). Trained observers using a series of visibility markers, for which the observer knows the distance, estimate airport visibility. Using a technique based on the frequency distributions of visual range, median visual range, from observations less than 24.1 km and estimated visual range when greater than 24.1 km, were computed for 140 stations. From this analysis, Ontario sites have lower visibility in the summer, reflecting higher $PM_{2.5}$ levels in the summer season. Consequently, measured summer visibility would be expected to be lower than those calculated from mean $PM_{2.5}$ levels (1984 to 1993). In Vancouver, $PM_{2.5}$ levels reach a maximum in winter, and consequently actual summer visual ranges would be greater than calculated from annual averages (1984 to 1993).

Contours of visual ranges were determined from the 140 stations across Canada. Median summer and winter visual ranges for 1951 to 1991, with relative humidity less than 80%, are shown in Figures 7.5 and 7.6. For many regions of Canada, summertime visual ranges are greater than 80 km. However, southeastern Canada is notable for significantly lower summertime visual ranges. Wintertime visibility is dramatically lower in the western parts of Canada compared to the summertime. Visibility across the country becomes uniform in winter.

These regional differences in visual range have also been observed from visibility measurements taken in Canada at non-urban sites. The visual range measured at Egbert (86-120 km) was lower than the visibility measured at Waterton (210-350 km) and St. Andrews (185-210 km) (Table 7.3). Visual ranges calculated for minimum and maximum mean $PM_{2.5}$ levels (average 1984 to 1993, see Table 7.4) at NAPS stations in Canada ranged from 40 to 140 km. The visual ranges are similar to those observed in the US during 1951 to 1991 (Malm, 1992; Malm et al., 1994a).

Figure 7.5 Contour Plot of Summer Visual Range (from Stuart and Hoff, 1994)

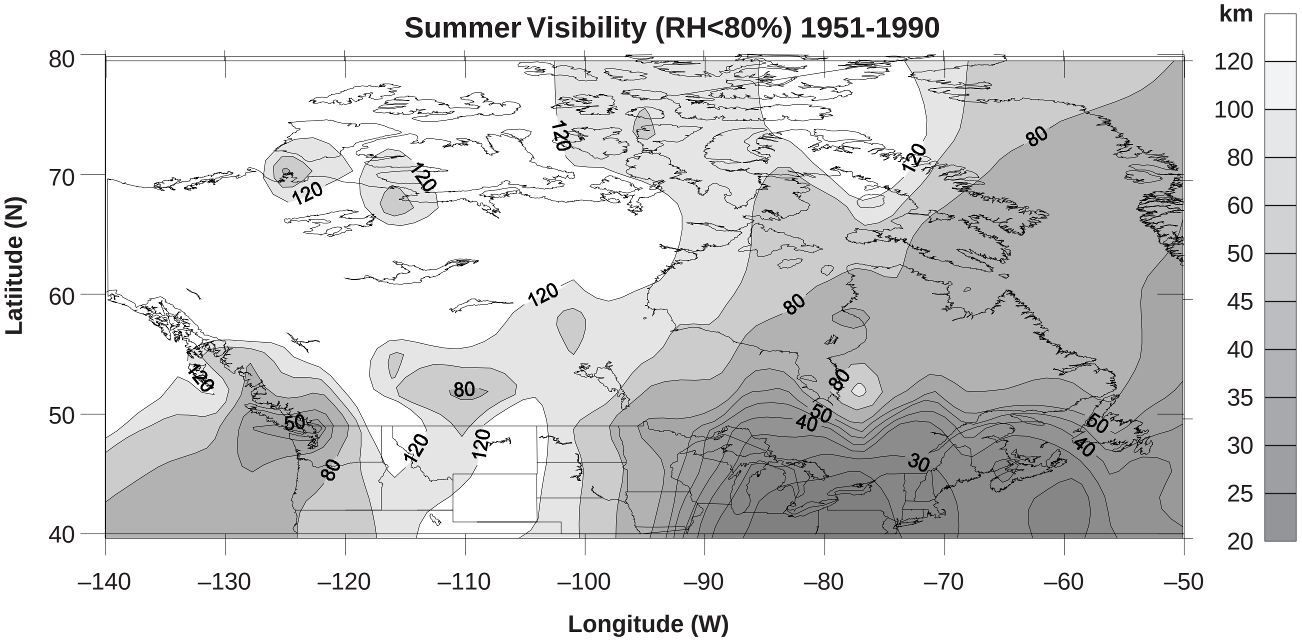
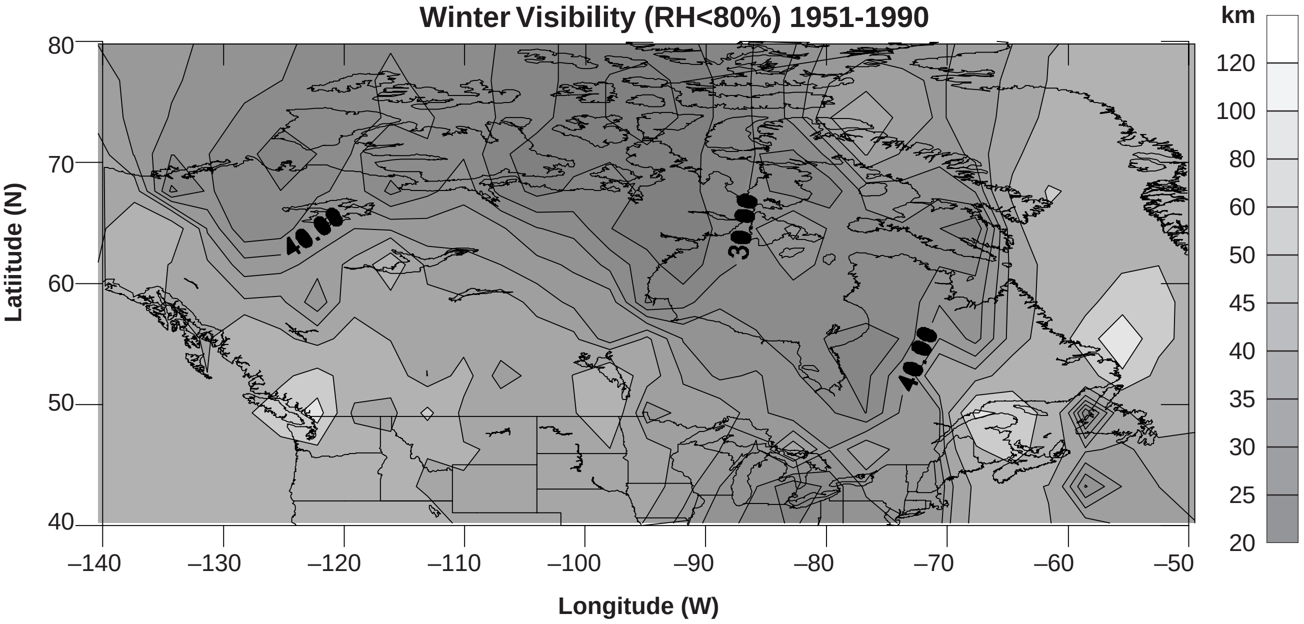


Figure 7.6 Contour Plot of Winter Visual Range (from Stuart and Hoff, 1994)



7.5 SUMMARY

Submicron particles, primarily those in the 0.3 to 0.7 µm size range, are the most effective in reducing visibility. There is empirical evidence showing that there is a linear relationship between the light scattering coefficient (b_{scat}) and fine particle mass, with b_{scat}/fine mass = 3.1 m^2/g. Thus, with the benefit of the Koschmeider equation and known relationships for b_{scat}/b_{ext}, visual range may be determined as function of PM_{10} mass concentrations if the ratio of $PM_{10}/PM_{2.5}$ is known, or directly from coarse and fine particle masses. However, it should be recognized that the fine fraction is primarily responsible for degraded visibility.

Natural visual range estimates in Canada, which are based on b_{scat} measurements at rural sites are in the range of 86 to 350 km depending on local relative humidity conditions. Southeastern Canada has the lowest estimated natural visual range of 86-120 km, which is similar to the visual ranges observed in the United States (100-180 km, Table 7.3). Western and eastern Canada have visual ranges of 210-350 km and 185-210 km respectively. These visual range estimates correspond with natural $PM_{2.5}$ levels of approximately 3.2 to 5.5 µg/m^3 in western Canada, 5.5–6.1 µg/m^3 in eastern Canada, and 10-13 µg/m^3 in southeastern Canada. These levels are consistent with other estimates of natural background levels of $PM_{2.5}$ for North America ranging from 1-5 µg/m^3 (see Chapter 5).

Visual range can be calculated using the Koschmieder equation and assuming that b_{scat}/b_{ext} = 0.9 for rural areas = 0.7 for urban areas, and $b_{scat}/PM_{2.5}$ = 3.1 m^2/g. Using a background $PM_{2.5}$ level of 5 µg/m^3 gives a natural visual range of 230 km for an unpolluted, rural atmosphere. There are regional differences. Regional natural background $PM_{2.5}$ levels, estimated from visual range measurements, range from 5.5 to 13 µg/m^3 (Table 7.3). A 10% increase in these values (i.e. ranging from 6 to 14 µg/m^3) would normally be visually detectable. Any increase in particle levels above these levels would reduce visual range. Conversely, a 10% decrease in fine particle concentrations (improvement) would also be visually detectable.