

ENVIRONMENTAL TOBACCO SMOKE PARTICLES*

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ABSTRACT

This article reviews information on environmental tobacco smoke (ETS) particles that is of potential interest to scientists and professionals involved in exposure or risk assessment, epidemiology, or tobacco policy. It presents material on observed levels in common locations, physics and chemistry, dynamic behavior, and methods for modeling and technological control.

INTRODUCTION

Environmental tobacco smoke (ETS) is a mixture of gases and particles. It is generated by the combustion of tobacco products, including cigarettes, cigars, and pipes. Most ETS issues from the smoldering tobacco between puffs. Exhaled mainstream smoke also contributes to ETS.

Once released to the environment, the particles and gases in ETS are subjected to physical and chemical processes that can alter their concentrations, their physical form and their chemical composition. For example, dispersion processes such as turbulent mixing cause ETS concentrations to become more uniformly distributed throughout a room. Deposition onto indoor surfaces reduces airborne particle concentrations and shifts the particle size distribution. Sorptive interactions with indoor materials can alter the relative amounts of volatile and semivolatile compounds that remain airborne.

Although ETS comprises both gases and particles, for the purposes of this chapter, we focus exclusively on the particle phase. ETS particles are small liquid droplets — mostly in the size range 0.02-2 μm diameter. Chemically, they are mainly comprised of organic compounds that have vapor pressures sufficiently low to remain in the condensed phase. These compounds are formed during smoking by volatilization, pyrolysis, and partial oxidation of the components of the tobacco product. It is the scattering of light by particles that causes smoke to be visible.

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Environmental tobacco smoke is a major contributor to indoor air concentrations and human exposures to particles. Consequently, many studies have investigated the nature and significance of ETS particles. In this chapter, we summarize what has been learned, emphasizing the physical and chemical processes that influence indoor ETS particle levels. We point out several unresolved issues that warrant further study.

CONTRIBUTION OF ETS TO INDOOR PARTICLE MASS CONCENTRATIONS AND EXPOSURES

This section summarizes the experimental evidence demonstrating that ETS contributes substantially to indoor particle levels. A simple material-balance model is used to complement the empirical data. Ambient air quality standards are discussed briefly to place the findings in context.

Table 1 summarizes the findings from 23 large field studies that have considered the contribution of ETS to indoor particle levels, especially in residences and in offices. The nature of these studies is to compare particle mass concentrations in environments where smoking does and doesn't occur. Some studies have used microenvironmental monitoring, while others have emphasized personal sampling.

Among the studies reported in Table 1, more than half were supported by the tobacco industry (Turner et al., 1992; Jenkins et al., 1996; Heavner et al., 1996; and all of the studies by Phillips and coworkers). Excluding these studies, each of the others showed that the indoor particle levels were consistently and substantially higher in indoor environments with smoking than without. The average differences between smoking and nonsmoking households ranged from approximately 10 $\mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$ in Arizona homes with light smoking (Quackenboss et al., 1989) and in Finnish homes (Koistinen et al., 2000) to almost 50 $\mu\text{g m}^{-3}$ for $\text{PM}_{3.5}$ in Kingston-Harriman, Tennessee (Spengler et al., 1985).

The results of the studies sponsored by the tobacco industry are largely, but not entirely consistent with the findings of the other studies. So, for example, Jenkins et al. (1996) reported that 16-h average $\text{PM}_{3.5}$ levels in smoking households exceeded

TABLE (1): *Environmental Tobacco Smoke Particle Concentrations Measured During Field Surveys*^a

Investigators	Study Name	Subjects/Locations Surveyed	Methods	Results ^b	
				Concentrations ($\mu\text{g m}^{-3}$)	Sample Characteristics
Özkaynak et al. (1996)	PTEAM ^c	178 random nonsmokers aged 10 - 70; personal monitoring; households; Riverside, CA	PM _{2.5} ; 12-h samples; day/night	27-32 ETS contribution to PM ₁₀ /PM _{2.5} ; day or night	31 homes with smokers; 61 samples day + night
Neas et al. (1994)	Harvard Six City ^d	1,273 households with children aged 7 - 11; Caucasian	PM _{2.5} ; 2-wk samples	annual means: 48.5 SH; 17.3 NSH	580 consistently SH; 470 consistently NSH
Spengler et al. (1985)	Harvard Six City ^d	101 households; personal and household monitoring; Kingston-Harriman, TN	PM _{3.5} ; 24-h samples	means: 74 SH; 28 NSH	28 SH; 73 NSH
Spengler et al. (1987)	Harvard Six City ^d	300 households with children; Watertown, MA; St. Louis, MO; Kingston-Harriman, TN	PM _{2.5} ; 1-wk samples	means: 30 greater in SH than in NSH	NA
Leaderer et al. (1990)	New York State ^e	359 stratified households with valid data; Onondaga and Suffolk Counties, NY	PM _{2.5} ; 1 wk samples	geometric means: 29-61 SH; 14-22 NSH	238 SH; 121 NSH
Jenkins et al. (1996)	Sixteen cities/CIAR ^{f,g}	1564 subjects; personal monitoring; households; workplaces; 16 U.S. cities	PM _{3.5} ; 8-h sample at work; 16-h sample at home	means: 44 SH; 20-21 NSH; 49 SW; 18 NSW	SH: 306; NSH: 2078; SW: 331; NSW: 867
Heavner et al. (1996)	RJR ^h	104 nonsmoking married female subjects over 25; personal monitoring; households; workplaces; New Jersey and Pennsylvania	PM _{3.5} ; ~ 14-h samples at home; ~ 7-h samples at work	means: 89 SH; 28 NSH (without regard to work)	29 SH; 58 NSH
Quackenboss et al. (1989)	-	98 households; Tuscon, AZ	PM _{2.5}	means: 27 SH \leq 1 pack d ⁻¹ ; 61 SH > 1 pack d ⁻¹ ; 15 NSH	45 NSH; 26 SH \leq 1 pack d ⁻¹ ; 17 SH > 1 pack d ⁻¹
Turner et al. (1992)	-	585 office environments	PM _{3.5} ; 10 samples per hour	means: 46 SW; 20 NSW	331 smoking offices; 254 nonsmoking offices
Phillips and Bentley (2001)	CIAR ^g	124 random nonsmoking office workers; personal monitoring; Bremen, Germany	RSP; 24-h samples	smoking locations: 48-53 (winter); 22-30 (summer); nonsmoking locations: 22-28 (winter); 17-20 (summer)	winter: 49 SW & SH; 53 NSW & NSH; summer: 52 SW & SH; 50 NSW & NSH
Phillips et al. (1998h)	CIAR ^g	241 random office workers and housewives; personal monitoring; Kuala Lumpur, Malaysia	RSP; 24-h samples	medians: 52 SH; 48 NSH; 50 SH/SW; 52 SH/NSW; 43 NSH/SW; 43 NSH/NSW	42 SH; 51 NSH; 30 SH/SW; 29 SH/NSW; 45 NSH/SW; 44 NSH/NSW
Phillips et al. (1998c)	CIAR ^g	194 random nonsmoking office workers and housewives; personal monitoring; Hong Kong	RSP; 24-h samples	medians: 45 SH; 46 NSH; 53 SH/SW; 50 SH/NSW; 54 NSH/SW; 43 NSH/NSW	35 SH; 35 NSH; 21 SH/SW; 29 SH/NSW; 31 NSH/SW; 43 NSH/NSW

Continued.

Table 1 Continued.

Investigators	Study Name	Subjects/Locations Surveyed	Methods	Results ^b	
				Concentrations ($\mu\text{g m}^{-3}$)	Sample Characteristics
Phillips et al. (1998b)	CIAR ^g	238 random nonsmoking office workers and housewives; personal monitoring, Prague, Czech Republic	RSP; 24-h samples	medians: 48 SH; 32 NSH; 60 SH/SW; 40 SH/NSW; 40 NSH/SW; 30 NSH/NSW	54 SH; 39 NSH; 64 SH/SW; 13 SH/NSW; 48 NSH/SW; 20 NSH/NSW
Phillips et al. (1998d)	CIAR ^g	190 random nonsmoking office workers and housewives; personal monitoring; Bremen, Germany	RSP; 24-h samples	medians: 36 SH; 25 NSH; 39 SH/SW; 36 SH/NSW; 29 NSH/SW; 23 NSH/NSW	21 SH; 60 NSH; 18 SH/SW; 6 SH/NSW; 49 NSH/SW; 36 NSH/NSW
Phillips et al. (1998a)	CIAR ^g	222 office workers and housewives; personal monitoring; Paris, France	RSP; 24-h samples	medians: 62 SH; 36 NSH; 80 SH/SW; 64 SH/NSW; 43 NSH/SW; 35 NSH/NSW	51 SH; 44 NSH; 45 SH/SW; 13 SH/NSW; 59 NSH/SW; 10 NSH/NSW
Phillips et al. (1998g)	CIAR ^g	319 nonsmokers; personal monitoring; Sydney, Australia	RSP; 24-h samples	medians: 30 SH; 24 NSH; 34 SW; 16 NSW	30 SH; 48 NSH; 20 SW; 60 NSW
Phillips et al. (1998e)	CIAR ^g	197 random nonsmoking office workers and housewives; personal monitoring; Lisbon, Portugal	RSP; 24-h samples	medians: 38 SH; 38 NSH; 41 SH/SW; 43 SH/NSW; 40 NSH/SW; 34 NSH/NSW	24 SH; 56 NSH; 28 SH/SW; 7 SH/NSW; 61 NSH/SW; 21 NSH/NSW
Phillips et al. (1997a)	CIAR ^g	188 office workers and housewives; personal monitoring; Turin, Italy	RSP; 24-h samples	medians: 71 SH; 54 NSH; 80 SH/SW; 66 SH/NSW; 59 NSH/SW; 55 NSH/NSW	36 SH; 47 NSH; 21 SH/SW; 9 SH/NSW; 51 NSH/SW; 24 NSH/NSW
Phillips et al. (1997b)	CIAR ^g	154 office workers and housewives; personal monitoring; Barcelona, Spain	RSP; 24-h samples	medians: 63 SH; 51 NSH; 85 SH/ any workplace; 40 NSH/any workplace; 94 SW/any home; 52 NSW /any home	43 SH; 42 NSH; 25 SH/SW; 3 SH/NSW; 36 NSH/SW; 5 NSH/NSW
Phillips et al. (1996)	CIAR ^g	190 working and nonworking nonsmokers; personal monitoring; Stockholm, Sweden	RSP; 24-h samples	medians: 39 SH; 18 NSH	9 SH; 31 NSH
Phillips et al. (1999)	CIAR ^g	196 nonsmoking office workers and housewives; personal monitoring; Basel, Switzerland	RSP; 24-h samples	medians: 34 SH; 28 NSH; 39 SH/SW; 24 SH/NSW; 27 NSH/SW; 26 NSH/NSW	26 SH; 60 NSH; 25 SH/SW; 14 SH/NSW; 43 NSH/SW; 28 NSH/NSW
Phillips et al. (1998f)	CIAR ^g	253 random nonsmoking office workers and housewives; personal monitoring; Beijing, China	RSP; 24-h samples	medians: 102 SH; 70 NSH; 114 SH/SW; 93 SH/NSW; 100 NSH/SW; 95 NSH/NSW	56 SH; 46 NSH; 46 SH/SW; 31 SH/NSW; 47 NSH/SW; 27 NSH/NSW
Koistinen et al. (2001)	EXPOLIS ⁱ	201 random adults aged 25-55; household, workplace, and personal monitoring; Helsinki, Finland	PM _{2.5} ; 48-h samples	averages: 21 SH; 8.2 NSH; 30 SW; 9.5 NSW; 31 personal active smoker; 17 personal ETS-exposed NS; 9.9 unexposed NS	57 SH; 135 NSH; 46 SW; 105 NSW; 48 active smokers; 9 ETS-exposed NS; 137 non-ETS exposed NS

^a The listed studies are limited to those that are large ($n > 100$) and/or probability-based with city-wide or larger scope, and where ETS-related and non-ETS-related particle levels were reported.

^b NSH = nonsmoking home; SH = smoking home; NSW = nonsmoking workplace; SW = smoking workplace; NS = nonsmoker; NA = not available

^c PTEAM = USEPA's Particle Total Exposure Assessment Methodology

^d The Harvard Six City study is described with preliminary results in Spengler et al. (1981). The six cities were: Portage, WI; Topeka, KS; Kingston-Harriman, TN; Watertown, MA; St. Louis, MO; Steubenville, OH.

^e The main reference for the New York State study is Sheldon et al. (1989).

^f The sixteen cities surveyed were: Knoxville, TN; Portland, ME; San Antonio, TX; Fresno, CA; Boise, ID; Seattle, WA; Baltimore, MD; Columbus, OH; Daytona Beach, FL; Buffalo, NY; St. Louis, MO; Grand Rapids, MI; Camden/Philadelphia, NJ,PA; Indianapolis, IN; Phoenix, AZ; New Orleans, LA

^g These studies were sponsored by the Center for Indoor Air Research (CIAR), which is affiliated with the tobacco industry. The Phillips et al. studies typically recruited subjects that stayed mostly at home (single monitor subjects; SH or NSH) and/or those that were both at home and at work (dual monitor subjects; SH or NSH with SW or NSW). Sample sizes are total study subjects. The statistics presented represent both time in and out of work for dual monitor subjects. In contrast, for the Phillips et al. (1998g) study the listed 24-h results reflect pumps that were shut off when subjects were not at home or at work (although 72-h and continuously-sampling 24-h samples were also collected).

^h Conducted by R.J. Reynolds Tobacco Company, Research and Development

ⁱ The main reference for EXPOLIS is Jantunen et al. (1998)

those in nonsmoking households by $24 \mu\text{g m}^{-3}$. On the other hand, Phillips et al. (1998c, 1998e) reported in 2 of the 13 cities that they studied (Hong Kong and Lisbon) that the median personal monitoring RSP concentrations were the same for those living in smoking and nonsmoking homes.

In reviewing indoor particles, Wallace (1996) summarized the evidence available at that time by stating that "all major studies found that an important indoor source of fine and coarse particles was cigarette smoking, with estimated increases in homes with smokers ranging from 25 to $45 \mu\text{g m}^{-3}$ $\text{PM}_{2.5}$." With the evidence now available, we would modify this statement to state that almost all of the major studies have found that an important source of fine particles is cigarette smoking. Most of the studies have indicated an increase in average fine particle levels indoors in the range 10 to $45 \mu\text{g m}^{-3}$.

The association between smoking and elevated indoor PM levels does not, by itself, prove that ETS caused the PM levels to be higher. Additional evidence in support of this inference comes from material-balance modeling. To a fair approximation, the increase in time-averaged and volume-averaged indoor $\text{PM}_{2.5}$ concentration ($\mu\text{g m}^{-3}$) caused by ETS can be estimated from the following equation (see Appendix for derivation):

$$\bar{C}_{ETS} = \frac{S \times E}{(\lambda + k) \times V} \quad (1)$$

where S is the average smoking rate (cig h^{-1}), E is the $\text{PM}_{2.5}$ emission factor ($\mu\text{g cig}^{-1}$), λ is the air-exchange rate of the building (h^{-1}), k is the first-order rate constant for removal of $\text{PM}_{2.5}$ by all means other than ventilation (h^{-1}), and V is the building volume (m^3).

Consider the case of a single-family residence with conditions typical of those in the United States: $V = 300 \text{ m}^3$; $\lambda = 0.5 \text{ h}^{-1}$; $k = 0.1 \text{ h}^{-1}$; $S = 0.5 \text{ cig h}^{-1}$; and $E = 1.2 \times 10^4 \mu\text{g cig}^{-1}$.[‡] Equation (1) predicts an estimated increase in fine particle mass concentration caused by ETS of $33 \mu\text{g m}^{-3}$. The consistency of this result with the central tendency of the empirical evidence from field studies provides strong evidence that the indoor smoking is not only *associated* with elevated indoor fine particle levels, but that it *causes* these elevated levels.

In comparison with health-based ambient air quality standards, an incremental concentration increase for fine PM of tens of micrograms per cubic meter is substantial. For example, the US Environmental Protection Agency is implementing a National Ambient Air Quality Standard of $15 \mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$, based on annual average conditions (<http://www.epa.gov/ttn/oarpg/naaqsfm/pmfact.html>). The evidence summarized in this section indicates that ETS alone could cause indoor $\text{PM}_{2.5}$ concentrations to exceed this standard in buildings in which smoking is habitually practiced.

The most serious exposures to ETS particles occur in buildings in which smoking occurs. However, particle emissions from cigarette smoking are sufficiently high to have a detectable

[‡]The selection of $S = 0.5 \text{ cig h}^{-1}$ represents a typical condition in a residence with a single smoker, averaged over an entire day. This average rate corresponds to a total daily consumption of 24 cigarettes (= 1.2 packs per day), with half of them smoked indoors at home.

impact on ambient fine-particle mass concentrations in urban air. Using molecular marker methods, Rogge et al. (1994) determined that cigarettes contributed $\sim 0.3 \mu\text{g m}^{-3}$, representing about 1%, to the fine particle mass concentration in outdoor air in Los Angeles.

KEY CHARACTERISTICS OF ETS PARTICLES: EMISSION FACTORS, SIZE DISTRIBUTION, AND CHEMICAL COMPOSITION

A key parameter that influences the impact of environmental tobacco smoke on indoor particle levels is the emission factor, i.e. the mass of ETS particles emitted per appropriate unit of tobacco product consumed. As shown in Equation (1), the increment of $\text{PM}_{2.5}$ attributable to ETS is proportional to the product of an emission factor, E , and a smoking rate parameter, S . This section summarizes available information about the particle emission factor for ETS.

A second important factor addressed in this section is the size of particles present in ETS. In general, particle sizes found in indoor air can vary over a vast range, from $\sim 0.001 \mu\text{m}$ to $> \sim 10 \mu\text{m}$ in diameter. The tendency of particles to deposit on surfaces, the ease with which they can be removed by filters, and the efficiency of respiratory tract deposition following inhalation all depend strongly on particle size.

A third important property of ETS particles is their chemical composition. Composition is potentially of great importance in fully understanding the adverse health effects of ETS exposure. Chemical composition can also influence dynamic behavior, particularly for semivolatile compounds that can be variably partitioned between the particle-, gas- and sorbed phases. Only a few studies of the chemical composition of ETS particles have been reported; their key findings are summarized here.

Table 2 summarizes the available evidence from 18 papers and reports on the particulate-matter mass-emission factors in ETS, including both cigarettes and cigars as sources. The studies have used a variety of settings and methods to infer mass emission factors, and the results span a broad range. For example, studies have been conducted in laboratory chambers using machine-smoked cigarettes and real-time instruments to measure resulting particle concentrations (Sextro et al., 1991; Klepeis et al., 2002). Other laboratory-based studies have used human smokers (e.g., Martin et al., 1997) or filter-based particle sampling methods (e.g., Löfroth et al., 1989).

The results of these studies can be distilled in many ways, one being the following. Consider cigarettes as the source, with emission factors expressed as PM mass emitted per cigarette smoked. Treat each of the seventeen rows in Table 2 with entries in the appropriate column (labeled "Total (mg)") as equivalently valid. (And, where a range is indicated, use the arithmetic mean of the endpoints of the range.) The median of this distribution is 12.7 mg/cigarette. The distribution exhibits positive skew. The lognormal parameters of the distribution are a geometric mean of 12.0 mg/cigarette, with a geometric standard deviation of 2.1. The central 90% of the distribution lies between approximately 5 and 40 mg/cigarette. Ten of the 17 results are in the range

TABLE (2): Reported Environmental Tobacco Smoke Particle Mass Emissions from Cigarettes and Cigars

Study ^a	Source ^b	Sample ^c	Method ^d	Particulate matter emissions, mean (standard dev.)		
				Total (mg)	Rate (mg min ⁻¹)	Normalized (mg g ⁻¹) ^e
<i>Cigarettes</i>						
* Klepeis et al. (2002)	MS+SS	2	M, C, OPC, DM	5.2 (0.3)	0.76 (0.12)	7.2 (0.3)
Klepeis et al. (2002)	MS+SS	2	M, C, F, TPM	8.4 (1.4)	1.3 (0.33)	11.6 (1.9)
Daisey et al. (1998) ^f	SS	6	M, C, F, PM _{2.5}	8.1 (2.0)	—	12.4 (1.3)
Hammond et al. (1987) ^g	MS+SS	4	H, C, F, RSP	12.7 (2.1)	1.7 (0.28)	26 (4)
Hildemann et al. (1991)	MS+SS	4	H, HD, F	20 (4.2)	—	—
Leaderer & Hammond (1991) ^h	MS+SS	10	H, C, F, RSP	17 (2.1)	—	27 (3.4)
Löfroth et al. (1989)	SS	1	M, C, F, TPM	10	—	10 - 11
Martin et al. (1997) ⁱ	MS+SS	50	H, C, F, RSP	13.7 (4.1)	1.2	24.5
Martin et al. (1997) ⁱ	MS+SS	50	H, C, PZ, RSP	11.6 (3.6)	1.1	20.7
Martin et al. (1997) ⁱ	MS+SS	50	H, C, MR, RSP	39.1 (1.7)	3.6	69.8
Ott et al. (1992)	SS	1	M, C, MR	49	7.0	—
* Sextro et al. (1991) ^j	MS+SS	6	M, C, OPC, DM	—	2.4	—
* Sextro et al. (1991) ^j	SS	3	M, C, OPC, DM	—	0.7	—
* Ueno and Peters (1986)	SS	1	M, OPC, DM	1.3 - 2.3	—	—
Ueno and Peters (1986)	SS	1	M, CI	6.0 - 9.6	—	—
* Chang et al. (1985) ^k	MS	1	M, OPC, DM	2.0 - 18.5	—	—
Chang et al. (1985) ^k	MS	1	M, CI	19.1 - 20.7	—	—
Klepeis et al. (1996) ^l	MS+SS	2	H, PZ, RSP	—	1.43 (0.01)	—
Koutrakis et al. (1992) ^m	MS+SS	178	H, PM _{2.5}	12.7	—	—
Özkaynak et al. (1996) ⁿ	MS+SS	178	H, PM _{2.5}	13.8 (3.6)	—	—
<i>Cigars</i>						
* Klepeis et al. (2002)	MS+SS	3	M, C, OPC, DM	5.9 (2.3)	0.46 (0.21)	3.9 (0.9)
Klepeis et al. (2002)	MS+SS	3	M, C, F, TPM	7.5 (2.8)	0.63 (0.26)	5.5 (1.4)
CPRT Laboratories (1990) ^o	—	13	—	—	—	10.3 (2.4)
Klepeis et al. (1999)	MS+SS	1	H, PZ, RSP	88	0.98	8.2
Leaderer and Hammond (1991)	MS+SS	1	H, C, F, RSP	—	—	48 (9)
Nelson et al. (1998)	MS+SS	6	H, C, F, RSP	50 (24)	5.0 (2.4)	6.4 (4.1)
Nelson et al. (1999)	MS+SS	20	H, C, F, RSP	93 (41)	9.3 (4.1)	12 (5)

^a An asterisk (*) indicates that size-specific ETS particle emissions were measured in this study.

^b Mainstream (MS), sidestream (SS), or both kinds of emissions (MS+SS) were studied.

^c The number of different experimental sites or individual types of tobacco sources used.

^d Key: H = human smokers; M = machine-smoked; C = chamber experiments; HD = emissions collected in a hood; F = filter-based sampling; OPC = optical particle counter; DM = differential mobility analysis; MR = Miniram optical scattering monitor; PZ = piezobalance; RSP = respirable suspended particulate matter; PM_{2.5} = particulate matter smaller than 2.5 μm in diameter; TPM = total particulate matter; CI = cascade impactor.

^e Particle mass emitted per unit mass of tobacco product combusted.

^f Results shown for six commercial cigarettes, 62.5% of top-selling California cigarettes, ca. 1990.

^g 40 cigarettes of each type were smoked for each experiment. Emission factors were calculated from information presented in the paper.

^h The cigarette types smoked in this study represent 48% of the market, ca. 1987. Results summarized for US commercial cigarettes only. Danish cigarettes and research cigarette (Kentucky 1R 3F) are omitted from this table. Forty cigarettes of each type were smoked for each experiment.

ⁱ This study presents weighted results for 50 top-selling US cigarette brands, which comprise 65% of the US cigarette market ca. 1991. Each cigarette was tested twice with an 11 min smoking duration and about 0.56 grams of tobacco consumed.

^j As reported in Nazaroff et al. (1993).

^k The range of results shown is due to varying dilution ratios with more dilution leading to a lower equivalent yield for DM measurements.

^l This study estimated the average total particle emission rate for cigarettes from real-time measurements in airport smoking lounges over a period of 2-3 h.

^m Emissions were estimated in the cited reference by fitting nonlinear regression model to average PM_{2.5} concentrations for 178 homes with valid data out of 394 (excluding homes with pipe or cigar smoking and fireplaces) total in New York state.

ⁿ Emissions were estimated in the cited reference by fitting nonlinear regression model to average PM_{2.5} concentrations for 178 homes in Riverside, CA.

^o As cited in NCI (1998), pp 169 and 178.

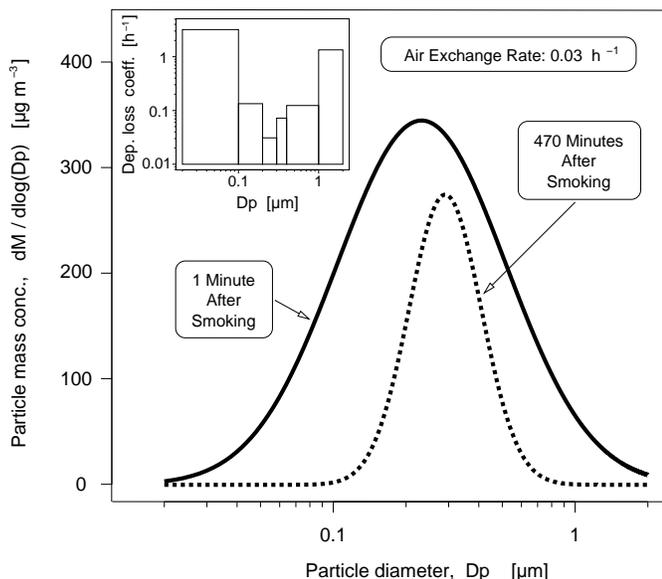


FIGURE (1): The mass size distributions of ETS particles shortly after a cigarette was smoked in a 20-m^3 room and after nearly eight hours had elapsed. The bell-shaped curves are fitted lognormal distributions, based on data measured by Klepeis et al. (2003). The earlier, more broad distribution has $\text{MMD} = 0.23\ \mu\text{m}$, $\text{GSD} = 2.2$, and a total mass concentration of $300\ \mu\text{g m}^{-3}$. The later, more narrow distribution has $\text{MMD} = 0.29\ \mu\text{m}$, $\text{GSD} = 1.4$, and a total mass concentration of $100\ \mu\text{g m}^{-3}$. The size-resolved particle deposition rates, determined for this particular experiment, are shown in the upper left inset. In addition to deposition onto surfaces, particles were removed at a ventilation rate of $\lambda = 0.03\ \text{h}^{-1}$. The particle size distribution becomes narrower in time because larger and smaller particles deposit more quickly than those near the mode of the distribution ($0.2 - 0.4\ \mu\text{m}$). In addition to the effects of deposition, coagulation causes the mode to shift toward larger sizes.

7.8-13.8 mg/cigarette.

While it is convenient to express emission factors for cigarettes on a per cigarette basis, cigars vary too widely in size for this approach to be useful. When expressed per mass of tobacco product burned, cigars exhibit emission factors that are roughly a factor of two lower than cigarettes. The median entry for cigars in Table 2 is 8.2 mg/g (range: 3.9-48); the corresponding value for cigarettes is 20.7 mg/g (range: 7.3-70). The total emissions of particulate matter from cigars can be estimated as the product of an emission factor and the mass of cigar tobacco consumed indoors.

The results of the many studies that have reported on the size of ETS particles are summarized in Table 3. Although the results of the studies vary somewhat, they portray a consistent picture with respect to the main points. ETS particles are emitted in a single size mode, with a large majority of the particle mass distributed among particles with diameters in the range $0.02\text{-}2\ \mu\text{m}$. A lognormal distribution is often used to describe this mode, as depicted in the Figure 1 trace labeled “1 minute after

smoking.” In that case, the mass median diameter is $0.23\ \mu\text{m}$ and geometric standard deviation is 2.2. For the data presented in Table 3, the 23 MMD entries have a median of $0.37\ \mu\text{m}$, with the central 90% ranging between 0.2 and $0.5\ \mu\text{m}$. The median reported GSD is 1.4, with the central 90% ranging between 1.2 and 2.1.

The particles in ETS are largely contained in that portion of the particle size distribution known as the accumulation mode, which has diameters in the range $0.1\text{-}2\ \mu\text{m}$ (Hinds, 1999). In the ambient environment, these particles are characterized by long atmospheric residence times. In the indoor environment, their persistence is commonly governed by how fast they are removed by ventilation. They are of a size that does not deposit rapidly onto indoor surfaces (see §3.5.4). They are the most difficult particle size to remove by filtration (Hanley et al., 1994), and they do not deposit with high efficiency in the respiratory tract when inhaled (Nazaroff et al., 1993). Particles are most efficient at scattering light when they are the same size as the wavelength of visible light ($0.3\text{-}0.7\ \mu\text{m}$) (Hinds, 1999). This, and the high emission rate of particles in ETS, explains why the air in a heavy smoking environment appears hazy.

The mechanism of ETS particle formation is similar to that of oxygen-starved combustion of any solid organic fuel, such as a wood fire. In the smoldering ember of a cigarette, the organic compounds that make up the cured tobacco leaf are partially oxidized and pyrolyzed, sustaining the heat of the ember. The product molecules of these reactions may be significantly in the vapor phase at the temperature of the smoldering ember. The gases rise by natural convection and, as they rise, they cool. Product molecules that have low vapor pressure at ambient temperature undergo a phase change from the gas to the condensed phase. They do this by a combination of homogeneous nucleation (forming new particles) and condensation onto preexisting particles. Nucleation is favored early in the smoke; later, condensation dominates as the rate of cooling slows and as the number of condensation sites increases (i.e., the newly formed smoke particles plus those from the ambient air). Particles in ETS can further undergo coagulation within the smoke plume, whereby two smaller particles collide and adhere to form one larger particle. Coagulation shifts the size distribution toward larger mass-median diameter and narrows the breadth of the distribution.

Because of the competition between evaporation, condensation, nucleation, and coagulation, the particle size distribution of freshly generated ETS can shift with ambient conditions. When the indoor air is relatively still, the smoke plume rises steadily above the smoldering ember. With ample time for coagulation and condensation, the ETS particles will be relatively fewer in number but larger in size. By contrast when the plume is dispersed rapidly by strong ambient air motion, a greater number of particles is generated in ETS, of smaller average size. This effect of dilution on particle size can be seen in the data of Hinds (1978) summarized in Table 3. It has also been observed anecdotally in experiments reported by Wampler et al. (1995). Additional discussion of the basic physics and chemistry of cigarette combustion can be found in Baker and Proctor (1990, 2001).

TABLE (3): Reported Size-Specific Tobacco Particle Emissions for Cigarettes and Cigars

Study	Source ^a	Sample ^b		Method ^c	Dilution	Size distrib., mean (std. dev.) ^d		
		N	T			CMD [μm]	MMD [μm]	GSD
<i>Cigarettes</i>								
Anderson et al. (1989)	MS	—	7	M, C, DM	80K	0.10 (0.0084)	0.38 (0.02)	2.0 (0.05)
Benner et al. (1989)	SS	1	1	M, C, OPC, DM	—	0.11	0.26	—
Klepeis et al. (2002)	MS+SS	3	2	M, C, OPC, DM	20-50K	—	0.20 (0.01)	2.1 (0.0)
Chang et al. (1985)	MS	13	1	M, C, DM	10 ^e	0.23	0.26	1.23
"	MS	31	1	M, C, DM	10 ^e	0.23	0.25	1.19
"	MS	—	1	M, C, DM	6 ^e	0.26	0.30	1.27
"	MS	—	1	M, C, DM	18 ^e	0.22	0.26	1.26
Chung and Dunn-Rankin (1996)	SS	—	2	M, C, OPC	—	0.27	0.5	1.6
"	MS	—	2	M, C, OPC	—	0.15	0.7	2.0
Hinds (1978)	—	2	1	M, C, AC-NS	10	—	0.52	1.37
"	—	3	1	M, C, CI	10	—	0.52	1.38
"	—	2	1	M, C, CI	50	—	0.44	1.49
"	—	1	1	M, C, CI	100	—	0.39	1.43
"	—	1	1	M, C, AC	100	—	0.38	1.33
"	—	1	1	M, C, AC	320	—	0.38	1.37
"	—	1	1	M, C, AC	500	—	0.38	1.35
"	—	1	1	M, C, AC	700	—	0.37	1.31
Ingebrethsen and Sears (1989)	SS	—	—	M, C, OPC, DM	—	0.1	0.20	—
Keith and Derrick (1960)	MS	10	1	M, C, CON	295	0.23	—	—
Kleeman et al. (1999)	MS+SS	1	1	H, C, MOUDI	—	—	0.3-0.4	—
Ishizu et al. (1978)	SS	—	1	M, C, OPC	1000	0.1	—	1.5
Okada and Matsunuma (1974)	MS	—	5	M, C, OPC	1500	0.17	—	1.5
"	SS	—	5	M, H, OPC	1500	0.10 – 0.12	—	1.4 – 1.6
Sextro et al. (1991) ^f	MS	6	—	M, C, OPC, DM	—	—	0.22	—
"	MS+SS	3	—	M, C, OPC, DM	—	—	0.48	—
Ueno and Peters (1986)	SS	—	1	M, OPC, DM, CI	6-18 ^e	0.10	0.16	1.4 – 1.7
<i>Cigars</i>								
Klepeis et al. (2002)	MS+SS	5	3	M, C, OPC, DM	20-50K	—	0.20 (0.02)	2.4 (0.44)

^a Mainstream emissions (MS), sidestream emissions (SS), or both (MS+SS)

^b Total number of cigars or cigarette experiments (across all source types) (N); number of types of cigars or cigarettes (T)

^c M = machine smoked; H = human smoked; C = chamber experiment; OPC = optical particle counter; DM = differential mobility analyzer; CON = confuge; CI = cascade impactor; AC = aerosol centrifuge; AC-NS = aerosol centrifuge in non-spectrometric mode; MOUDI = micro-orifice uniform-deposit impactor.

^d Particle size distribution characteristics are as follows: CMD is the count median diameter, MMD is the mass median diameter, and GSD is the geometric standard deviation.

^e Primary dilution ratio.

^f As reported by Nazaroff et al. (1993)

See Table 1 notes for other abbreviations.

The chemical composition of ETS particles has been the subject of several investigations, as summarized in Table 4. Hildebrand et al. (1991) reported that $\sim 60\%$ of tobacco smoke particles are organic compounds. Mainstream smoke studies have identified the following biologically active organic substances in the particle phase of cigarette smoke (Baker and Proctor, 2001), summarized by chemical class (and specific examples of the class): polycyclic aromatic hydrocarbons (benzo(a)pyrene), aza-arenes (quinoline), heterocyclic compounds (nicotine, pyridine), aromatic amines (2-naphthalene, 4 aminobiphenyl), n-heterocyclic amines (amino pyridoindoles and imidazoles), n-nitrosamines (n-nitrosornicotine), and miscellaneous organic compounds (phenol, catechol, acetamide, and maleic hydrazide). Given that exhaled mainstream smoke contributes to ETS and given the broad similarity in combustion processes that produce mainstream and sidestream smoke, these same compounds would be expected to be present in ETS particles. However, the particle/gas phase partitioning will vary for semivolatile organics between concentrated MS (which favors the condensed phase) vs. dispersed ETS (which favors the gas phase). Also, the combustion conditions differ in detail: MS is generated at higher temperature with a higher oxygen-fuel ratio as compared to the smoldering conditions under which SS smoke is produced. Baker and Proctor (1990) have noted that “the same chemicals present in mainstream smoke are also present in sidestream smoke, although their relative yield per cigarette is highly dependent on the compound considered.”

An important specific constituent of ETS is nicotine, which Benner et al. (1989) found to make up about 8% ($470 \mu\text{mol g}^{-1}$) of SS smoke particles. As smoke disperses, nicotine volatilizes such that in ETS nicotine is primarily in the vapor phase. Nicotine is widely used as a chemical tracer of environmental tobacco smoke (Daisey, 1999). The complex dynamic behavior of nicotine poses a challenge for this application, as it partitions variably between particles and the gas phase, and it sorbs strongly to indoor surfaces (Van Loy et al., 2001; Singer et al., 2002). Consequently, its concentrations do not track perfectly the levels of other ETS constituents (Nelson et al., 1992). Nevertheless, for the purpose of environmental health studies, it may adequately track the concentration of other ETS components on a time-averaged basis (Van Loy et al., 1998). Solanesol has been proposed as an alternative ETS particle-phase marker (Tang et al., 1990). Because of its low vapor pressure, it does not exhibit the same complex behavior as nicotine. However, it is present in ETS at much lower levels than nicotine and so its use introduces some analytical challenges.

In addition to organic compounds, ETS particles are strong indoor sources of a few trace elements. For example, Koutrakis et al. (1992) determined that, in 77 smoking homes, ETS contributed 70-75% of the airborne particle load of Cl, K, and Cd. The emissions of cadmium in ETS are sufficiently distinctive and reproducible that it has been proposed as a tracer of ETS particles (Wu et al., 1995).

Once released into indoor air, the concentration and fate of ETS particles is influenced by their dynamic behavior. This section provides a brief summary of the dynamic processes in indoor air that affect ETS particles: ventilation, mixing, deposition, evaporation/condensation, coagulation, and reactions with ozone.

Broadly, ventilation refers to the exchange of indoor air with outdoor air. Ventilation rates of buildings are commonly expressed in terms of an air-exchange rate (λ), which represents the volume flow rate of ventilation air divided by the interior volume of the building. Typically, ventilation rates of buildings are in the range $0.2\text{-}2 \text{ h}^{-1}$ (Murray and Burmaster, 1995; Persily, 1989). Because ETS particle concentrations are low in outdoor air, increasing the ventilation rate decreases the indoor ETS particle concentration in smoking environments, as reflected in Equation (1).

Environmental tobacco smoke issues from localized sources: the smoldering end of a tobacco product and the exhaled smoke from a smoker. Exposure of nonsmokers to ETS particles, and the removal of ETS particles by ventilation or other means, depends on the transport and mixing of ETS particles within the indoor space. These processes influence the effectiveness of segregation techniques in preventing exposure of nonsmokers to ETS.

Indoor air is rarely still. Generally, it is moving at net local speeds that range from a fraction to several tens of a cm s^{-1} . This motion can be induced by several means, including fans in mechanical ventilation systems, wind-induced flow through openings in the building shell, and natural convection resulting from heat exchange at surfaces. These motions often exhibit turbulent fluctuations superimposed on a systematic and structured mean flow. The mean flow can cause organized transport of pollutants; the turbulent fluctuations tend to disperse or spread pollutants so that the concentrations become more uniform throughout the interior space. The particles in ETS are sufficiently small to move readily along with the air currents.

A few reported experiments have investigated the transport and dispersion of cigarette smoke plumes, as summarized by Klepeis (1999). Data from one such study (Baughman et al., 1994) are depicted in Figure 2. In this case, a tracer gas was released from a point in a low-ventilation 31-m^3 room in a manner intended to represent the smoke from a smoldering cigarette. The tracer gas concentrations were then measured as a function of time at an array of points within the room. The experiment was repeated with different amounts of input heat energy to induce natural convection flow.

The panels in Figure 2 show snapshots in time of the local concentrations of tracer gas measured at breathing height at different positions throughout the room. The upper set of panels illustrates a case (“quiescent”) in which the mixing was intentionally made weak. The room was unoccupied, there was no mechanically induced flow, and natural convection was minimized by covering the windows and by selecting a time for the experiment when the room surfaces were at nearly the same temperature as the room air. Under these conditions, which are atypical

TABLE (4): Reported Chemical Composition of ETS Particles for Cigarettes^a

Study	Source	Method ^b	Chemical Composition	
			Organics	Inorganics
Benner et al. (1989)	SS	C, M, F, D, GC-FID, NPD, GC-MS, IC	Main classes: n-alkanes, branched alkanes, bases, sterols, fatty acids, sterenes $\mu\text{mol/g}$ (std. dev.): nicotine 467 (144); myosmine 35 (21); nicotyrine 14 (11); cotinine 20 (11); cholesterol 1.41 (0.33); stigmasterol 2.9 (1.6); campersterol 1.53 (0.58); β -sitosterol 2.2 (1.8); 24-methylcholesta-3,5-diene 2.1 (2.0); 24-ethylcholesta-3,5,22-triene 1.60 (0.81); solanesol 22.2 (3.3)	Species detected: Cl^- , NO_2^- , NO_3^- , SO_4^{2-} , NH_4^+ Species detected above 50 $\mu\text{mol/g}$: Cl^- , NO_3^- , NH_4^+ Elements detected: K, Ca, Ti, Ba, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Pb, As, Se, Br Elements detected above 50 $\mu\text{mol/g}$: Ca, K
Hildemann et al. (1991)	MS+SS	H, HD, F, XRF, GC-MS	organic carbon: 59.5% by mass	Species present above 0.01% by mass: S 0.14%; Cl 0.23%; K 0.41%; elemental carbon 0.49%; Cl^- 0.28%; NO_3^- 0.071%; SO_4^{2-} 0.059%; NH_4^+ 0.04%
Kleeman et al. (1999)	MS+SS	H, C, MOUDI, F, FID, IC	ETS particles are predominantly organic compounds in every particle size range	elemental carbon and the following trace elements and other species were detected: Na, K, V, Mn, Br, Sb, La, Ce, Cl^- , NO_3^- , SO_4^{2-} , NH_4^+ ; the size distribution of these species, as for the total particle size distribution, had a single mode between 0.3 and 0.4 μm
Özkaynak et al. (1996) ^c	MS+SS	H, Fd, XRF		Major elements associated with smoking were K, Cl, and Ca
Koutrakis et al. (1992) ^d	MS+SS	H, Fd, XRF		In 77 homes with smoking, smoking contributed the following mass percentages: S (11%); Cl (72%); K (70%); V (16%); Zn (14%); Br (44%); Cd (75%) Estimated emission rates (μg per cigarette): S (65); Cl (69); K (160); V (0.37); Zn (1.2); Br (3.0); Cd (0.32)
Rogge et al. (1994)	MS+SS	H, HD, F, GC-MS	The following classes of species were detected (with compounds having emission rates greater than 100 μg per cigarette listed in parentheses): n-alkanes (hentriacontane, triacontane); iso and anteisoalkanes; isoprenoid alkanes; n-alkanoic acids (hexadecanoic acid); n-alkenoic acids; dicarboxylic acids; other aliphatic and cyclic acids; n-alkanols; phenols (1,4-benzenediol); phytosterols (stigmasterol, β -sitosterol); N-containing compounds (nicotine, 3-hydroxypyridine, myosmine); polycyclic aromatic hydrocarbons	

^a The listed studies all involved cigarettes (i.e., no cigars).

^b C = chamber; M = machine-smoked; F = filter-based sampling; D = Denuder; FID = flame ionization detector, NPD = nitrogen-phosphorus detector; GC-MS = gas chromatograph – mass spectrophotometer; H = human smoker; HD = emissions captured in a hood; FID = flame-ionization detector; MOUDI = micro-orifice uniform-deposit impactors; Fd = field sampling; XRF = X-ray fluorescence; IC = ion chromatography.

^c This effort was part of the USEPA's Particle Total Exposure Assessment Methodology (PTEAM) study of 178 homes in Riverside, CA

^d This effort was part of a New York State study carried out in Onondaga and Suffolk counties (see Sheldon et al. 1989).

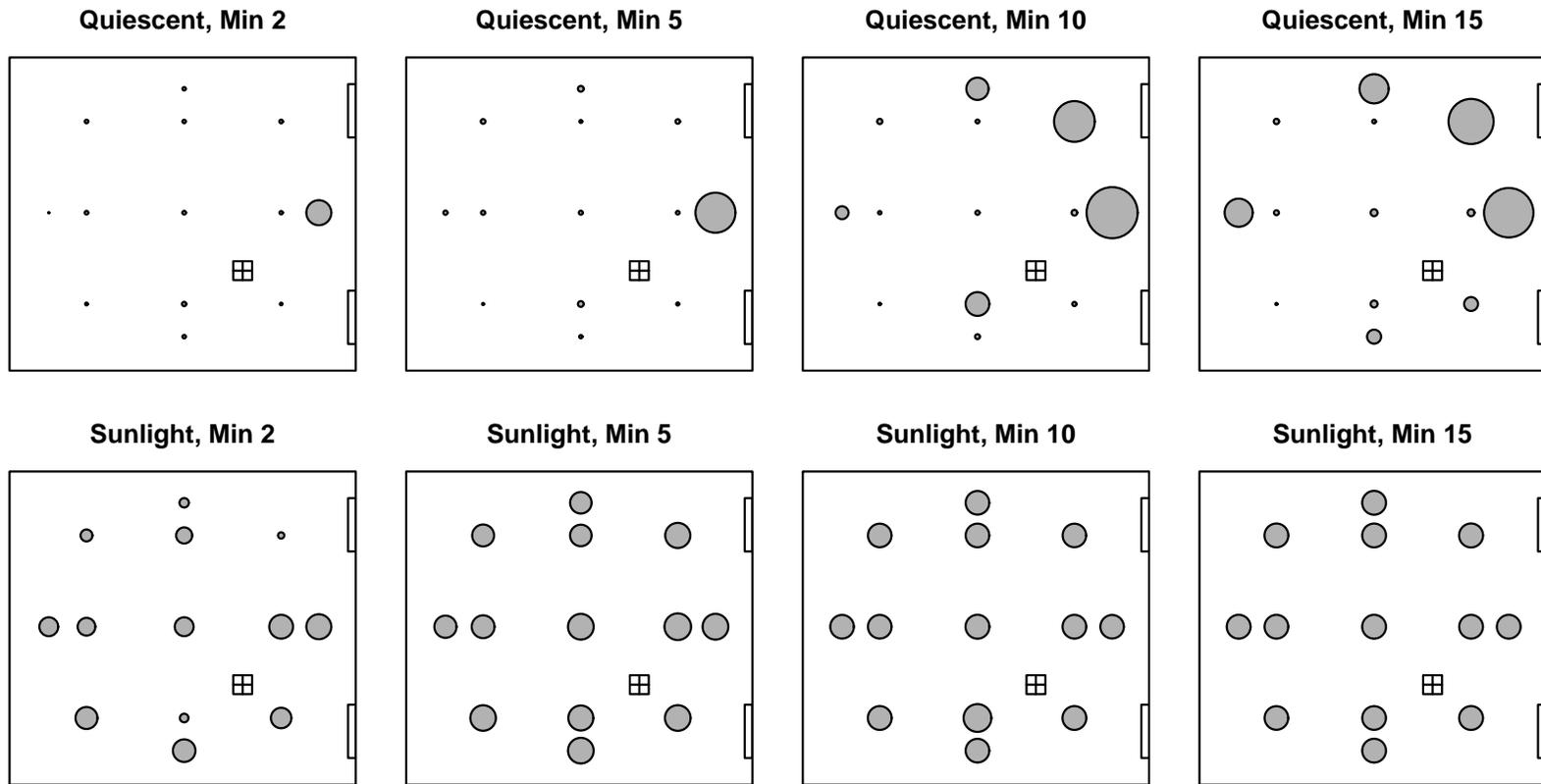


FIGURE (2): Panels showing snapshots of the extent of SF_6 tracer gas mixing at four times and for two sets of conditions, as measured in a 31-m^3 room. The conditions are (1) “quiescent,” with windows covered and a minimum of air movement; and (2) “sunlight,” in which sunlight entered through two 1.25 m^2 windows, providing about 600 W of incident solar energy. The area of the circles in each panel is proportional to the concentration of SF_6 gas measured at each of 13 monitoring locations. The SF_6 was emitted over a brief period from a point in the lower right portion of the room, to simulate the emissions from a smoldering cigarette. Nine breathing-height sampling points ($1.2 - 1.6\text{ m}$ high) were positioned in a rectangular pattern in the middle of the room, and four additional sampling points were established 1 cm from the center of each wall. Under quiescent conditions, the tracer remained incompletely mixed after 15 min, whereas the buoyancy induced mixing in the sunlight case caused the tracer to become fully mixed within this period. [Note: Raw data from Baughman et al. (1994) were interpolated with cubic splines to create the regular time series shown in this figure.]

— especially for an occupied room — the tracer concentration remained strongly unmixed for the 15 minutes depicted. Interestingly, the peak concentrations do not occur at the points closest to the source. Instead, they occur at sampling points close to the walls. This illustrates that airflow patterns in rooms can be complex. In this case, weak natural convection induced by small temperature differences between the walls and the adjacent air appears to strongly influence the overall airflow and pollutant transport pattern.

The lower set of panels illustrates a case with moderately strong, buoyancy-induced mixing. Here, the windows were uncovered, and a spot of direct sunlight was incident on the floor, with an estimated heat release of 600 W. The natural convection airflow that resulted caused the tracer gas to become well mixed at the breathing height across the room on a time scale of ~ 5 min.

Another important aspect of transport and mixing concerns the movement of air from one room or zone in a building to another. This is an important factor in evaluating the efficacy of controlling ETS exposures to nonsmokers through designating different areas within a building as “smoking” or “nonsmoking.” Although potentially of considerable importance in understanding ETS exposure, the subject has been addressed in only a few recent research studies (Liu et al., 2001; Miller and Nazaroff, 2001).

The term k in Equation (1) refers to the rate of loss of ETS particles by all means other than ventilation. The two main contributors to this loss are active filtration by air cleaning devices, which is discussed in below, and the deposition of particles onto room surfaces.

ETS particle deposition reduces the airborne particle concentrations. The rate of deposition varies with particle size, as illustrated in Figure 3 (reproducing data from Xu et al., 1994). The deposition rate is higher for the smallest and largest ETS particles than for those of intermediate size (0.1–0.4 μm diameter). This feature is a consequence of the different mechanisms that control deposition: turbulent plus Brownian diffusion for the smallest particles and gravitational settling for the largest (Lai and Nazaroff, 2000). A consequence of the differential rates of deposition is the sharpening of the ETS particle size distribution with time, as seen in comparing the two particle size distribution traces in Figure 1. Figure 3 also shows that the rate of particle deposition depends on the intensity of indoor air motion, with the most intense motion (resulting in this case from the highest fan speed) inducing the most rapid deposition. A third important point about the deposition process is that it is typically a slower removal mechanism than ventilation.

Other dynamic processes can alter the size and composition of environmental tobacco smoke particles. However, these have been less thoroughly studied than the other dynamic processes reviewed here. There is some evidence, for example, that mass can be lost from ETS particles owing to evaporation of certain volatile components as the ETS ages (Ingebrethsen and Sears, 1989). Nazaroff and Cass (1989) showed that coagulation causes a shift in the particle size distribution of ETS particles, inducing a net loss of the smallest particles and a net gain

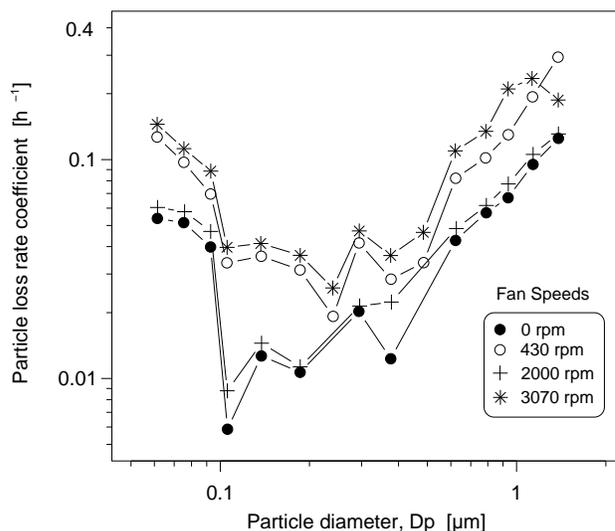


FIGURE (3): Particle deposition rates for ETS measured by Xu et al. (1994) in a low-ventilation chamber for four different fan speeds.

in particle mass for diameters in the vicinity of 0.1 μm . This process is slow except when the ETS particle concentrations are very high. Finally, we note an anecdotal report that suggests the possibility of new particle mass formed in ETS by the ozone-induced oxidation of vapor-phase organics into species such as carbonyls that have much lower vapor pressure (Weschler and Shields, 1999). An earlier incidental report also provides evidence of secondary particle formation following desorption of ETS vapors from room surfaces (Johansson et al., 1993).

OPPORTUNITIES FOR TECHNOLOGICAL CONTROL

The many approaches to control ETS exposure can be broadly grouped into administrative (or social) and technological. Examples of the former class include restrictions on smoking in public buildings and programs aimed at reducing the incidence of smoking in the population. Examples of the latter class include changing ventilation rates or airflow patterns in a building and the addition of particle filtration devices to indoor environments. Some controls straddle this broad classification scheme. For example, the segregation of smokers from nonsmokers augmented by enhanced exhaust ventilation from the smoking area contains both administrative and technological elements. Within each of the broad categories are many specific options. Our attention in this section is restricted to technological controls and their effectiveness in reducing ETS exposure.

Inhalation exposure to ETS particles can be considered as a sequence of three events that occur in series: the emission of particles from the tobacco product, the transport of particles from the point of release to the exposed individual, and the inhalation of air that contains the ETS particles. Interventions to control exposure can be applied at any of these three points. The third option, direct respiratory protection, is generally not practiced for controlling ETS exposure and so won't be further consid-

ered here. However, we note that this approach is routinely and effectively used for protecting workers from exposure to hazardous materials and infectious agents (Schaefer, 1997).

Emission control to reduce ETS exposure can be achieved through the use of "smokeless ashtrays" (Wampler et al., 1995). The basic principle behind these devices is to reduce the release of ETS components into room air. Since most ETS derives from smoldering cigarettes between puffs, one strategy for a smokeless ashtray is to capture the smoke plume and filter it. The commercially available devices tested by Wampler et al. were generally ineffective, either owing to failures to capture the smoke plume or because of the use of poor quality filter media. Wampler et al. did demonstrate high capture efficiency in a proof-of-principle device that used high efficiency particle filters for particle removal along with sorbents (activated alumina and activated carbon) to capture gaseous pollutants. Another strategy employed in one commercial device (SmokeRight ashtray) quenches, but doesn't extinguish the smoldering cigarette ember between puffs. The lower temperature of the ember markedly reduces the rate of visible smoke generation.

Control at the second stage in the exposure process is generally based on one or more of these methods: increased or more effective ventilation, filtration, and segregation. Improved ventilation has long been used as the basic strategy for controlling ETS exposure (Leaderer et al., 1984). The highest per-person ventilation rates required by ASHRAE standard 62-1989 are specified for smoking lounges ($30 \text{ L s}^{-1} \text{ person}^{-1}$) (ASHRAE, 1989). Portable air cleaners are also used for ETS control. Most units operate by using a fan to draw room air through a filter before discharging it back into the room. The effectiveness of such a device depends on the "clean-air delivery rate" (CADR), which is the product of the airflow rate through the device and the single-pass filtration efficiency. The effectiveness is also a function of the rate of removal by mechanisms other than those of the control device. For a device to be highly effective, it must increase the total removal rate substantially over the baseline value (Nazaroff, 2000). Commercially available devices vary markedly in their CADR values and, correspondingly, in their effectiveness in reducing ETS exposure (Offermann et al., 1985). It is possible and practical to obtain a moderate degree of improvement by the use of such devices.

The segregation of smokers from nonsmokers is widely practiced as a method for controlling ETS exposure. The efficacy of this approach has been investigated in a few studies (Lambert et al., 1993; Jenkins et al., 2001; Liu et al., 2001; Miller and Nazaroff, 2001). The limited evidence indicates that it is possible to achieve a high (but not complete) level of exposure reduction using a well-conceived segregation strategy. However, simply separating smokers from nonsmokers without regard for the potential air transport pathways that may link the spaces they occupy may only yield moderate reduction in exposure for nonsmokers.

It is self-evident that regulations, which effectively prohibit smoking in public buildings, can be very effective in reducing ETS exposure in those sites. In public buildings where smoking is permitted, the use of segregation combined with well-

designed ventilation and filtration systems can effectively reduce exposures. We know less about how to effectively control exposure in residential environments. Clearly household rules that restrict smoking to outdoors can reduce exposure to occupants. When smoking occurs indoors, it should be possible to substantially reduce exposures by the judicious application of one or more of these methods: emission controls, enhanced ventilation, air filtration, and segregation. However, our current base of knowledge is insufficient to prescribe guidance for the full range of diverse conditions that occur in residences or to quantify how effective such techniques will be in practice.

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APPENDIX: MATERIAL BALANCE FOR INDOOR ETS PARTICLE MASS CONCENTRATIONS

The purpose of this appendix is to provide a formal derivation of Equation (1). In doing so, we will demonstrate the assumptions that are necessary for strict validity of the equation. Although these assumptions are not strictly satisfied in most cases, in many situations the equation provides good estimates.

Let variables be defined as follows:

$C_{ETS,AVG}$ = volume-averaged mass concentration of particles associated with environmental tobacco smoke ($\mu\text{g m}^{-3}$)

$C_{Q,AVG}$ = flow-rate-averaged mass concentration of ETS particles in air flowing out of building ($\mu\text{g m}^{-3}$)

V = volume of the indoor space of interest (m^3)

Q = volume flow rate of air out of the building ($\text{m}^3 \text{h}^{-1}$)

λ = air-exchange rate = Q/V (h^{-1})

k = loss-rate coefficient for ETS particle removal by all mechanisms other than ventilation (h^{-1})

$P(t)$ = time-dependent emission rate of ETS particles from smoking events ($\mu\text{g h}^{-1}$)

t = time (h)

We make the following assumptions at the outset. (1) Phase change processes involving ETS particles are of negligible importance. This implies that ETS particle mass does not change because of evaporation or condensation of semivolatile constituents. (2) The ETS particle mass concentration in outdoor air is negligible. Therefore, it is not necessary to include ventilation supply as a source of ETS particles. (3) Removal of ETS particle mass from indoor air by means other than ventilation (such as deposition) proceeds at a rate that is proportional to the volume-average indoor ETS mass concentration.

With these assumptions and variable definitions, we can express the conservation of ETS particle mass by means of the following equation:

$$\frac{d(C_{ETS,AVG} \times V)}{dt} = P(t) - QC_{Q,AVG} - k \times (C_{ETS,AVG} \times V) \quad (2)$$

The term on the left-hand side expresses the rate of change of mass of suspended ETS particles indoors. On the right-hand side, the terms account, respectively, for emission, removal by means of ventilation, and removal by all other mechanisms.

Let us now consider a period of time, T , such as a day or a week, which is very much longer than the replacement time of indoor air by ventilation ($T \gg V/Q$). During this time, cigarettes are smoked intermittently within the building. The total number of cigarettes smoked, divided by the duration of the period, is the average smoking rate, S (cig h^{-1}). The total mass of ETS particles emitted can be expressed in this way:

$$S \times T \times E = \int_0^T P(t) dt \quad (3)$$

where $S \times T$ represents the total number of cigarettes smoked during time T , and E is the particle emission factor ($\mu\text{g cig}^{-1}$) for ETS.

Returning to Equation (2), it is convenient to multiply both sides by dt and then integrate over the period 0 to T , yielding:

$$[C_{ETS,AVG} \times V]_T - [C_{ETS,AVG} \times V]_0 = \int_0^T P(t) dt - \int_0^T (Q \times C_{Q,AVG}) dt - \int_0^T (k \times C_{ETS,AVG} \times V) dt \quad (4)$$

The pair of terms on the left represents the net change in suspended ETS particle mass between the beginning ($t = 0$) and the end ($t = T$) of the period. On the right-hand side, the first term can be replaced with $S \times T \times E$, using Equation (3). The second and third terms represent the total removal of ETS particles from indoor air by ventilation and by all other means, respectively, during interval T .

Each of the terms on the right-hand side of Equation (4) increases as T increases. However, for a building in which smoking is cyclic, the terms on the left-hand side will vary between some lower and upper bounds, which are independent of the magnitude of T . Therefore, provided T is long enough, the dominant terms on the right-hand side can be made very much larger than the terms on the left. For example, if $T = 7$ d for a single-family residence in which 10 cigarettes are smoked daily, the total particle mass emitted will be of the order of 7 g. The indoor particle ETS concentration may vary between ~ 0 and $\sim 300 \mu\text{g m}^{-3}$ averaged over a volume of 300m^3 . Therefore, the terms on the left-hand side will vary between 0 and ~ 10 mg, negligibly small compared with the terms on the right, at least

two of which will be of order 7 g. By this argument, for sufficiently long integration periods, we may neglect the terms on the left as small compared with the largest terms on the right, such that

$$\int_0^T (Q \times C_{Q,AVG})dt + \int_0^T (k \times C_{ETS,AVG} \times V)dt = S \times T \times E \quad (5)$$

Two final assumptions or approximations need to be made to derive the result we seek. First, we assume that the ventilation flow-rate weighted average ETS particle concentration is the same as the indoor volume average concentration ($C_{Q,AVG} = C_{ETS,AVG} = C_{ETS}$). This assumption is satisfied if the indoor volume is well mixed. However, perfect mixing is not strictly required. We only require that the two volumetric averages are the same on a time-averaged basis. Second, we assume that the removal rate parameters Q and kV are either constant in time or, at least, not correlated in time with the ETS particle concentration. This second assumption permits us to take Q and kV out of their respective integrals.

Finally, we note that the time-averaged concentration \bar{C}_{ETS} can be expressed as

$$\bar{C}_{ETS} = \frac{1}{T} \int_0^T C_{ETS} \times dt \quad (6)$$

Therefore, applying the assumptions from the previous paragraph along with this definition of time average, the left-hand side of Equation (5) can be rewritten as

$$\int_0^T (Q \times C_{Q,AVG})dt + \int_0^T (k \times C_{ETS,AVG} \times V)dt = (\lambda + k) \times V \times T \times \bar{C}_{ETS} \quad (7)$$

Substituting into Equation (5) and rearranging leads to Equation (1).