CHARACTERIZING ETS EMISSIONS FROM CIGARS: CHAMBER MEASUREMENTS OF NICOTINE, PARTICLE MASS, AND PARTICLE SIZE

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ABSTRACT
We measured the environmental tobacco smoke (ETS) emissions (diluted sidestream plus mainstream tobacco smoke) from three types of cigars: regular, small plastic-tipped, and large premium. Fifteen experiments (nine using cigars and six using cigarettes) were conducted in a well-mixed 20 m³ chamber with stainless steel walls. The cigars and cigarettes were both smoked by machine at a rate of two 35-ml puffs a minute. For each experiment, we collected filter samples to obtain total particle mass concentration. Integrated nicotine samples were collected on Tenax sorbent tubes, and nicotine analyses were performed using thermal desorption followed by GC-FID. Size-resolved particle measurements were made using a differential mobility particle sizer with a 0.01 - 0.5 μm diameter range. Emission factors for nicotine and total particle mass (for either milligrams emitted per gram of tobacco smoked or milligrams emitted per minute) were up to 6 times larger for cigarettes than for cigars.

INTRODUCTION
The characterization of emissions from sources of indoor air pollution is important in the fields of indoor air quality, exposure assessment, risk assessment, and epidemiology. A critical attribute of sources is the mass of pollutant emissions per unit of fuel consumed or per unit time. The latter is called the emission rate. Smoke exhaled by smokers and emitted from the burning end of a tobacco product, commonly called second-hand smoke or environmental tobacco smoke (ETS), is a major source of indoor pollution.

ETS emission data have been reported for cigarettes by a number of researchers (1). However, the available information on cigar ETS emissions appears limited to three studies. Two investigators have reported cigar ETS emission factors for carbon monoxide (2) and total particle mass (TPM) (2, 3 as cited in 4) and another investigator has reported concentrations for several compounds emitted from cigars that were smoked in an unventilated chamber (5). The need for more emissions data for cigars motivates the current study.

We conducted a series of controlled chamber experiments for the specific purpose of generating TPM and nicotine ETS emission data for three different types of cigars (premium, regular, and small). We measured the size distribution of ETS particles, since particle size is crucial in determining both the regional lung deposition and the dynamic behavior of particles in indoor air. All cigar results are compared to results for cigarettes smoked under identical conditions.

METHODS
Chamber Experiments. We conducted nine cigar (premium, regular, and cigarillo) and six cigarette (regular and lights) smoking experiments in an unventilated 20 m³ chamber (< 0.1 air changes per hour). The chamber's interior surfaces consisted entirely of stainless steel.

In addition, two 4' x 8' sheets of upright gypsum wallboard (a total of 12 m^2 of exposed surface area) were placed vertically in the center of the chamber. The cigars were smoked by machine (ADL/II, Arthur D. Little, Inc., Cambridge, MA) at a rate of two 35-ml puffs per minute. Both sidestream and mainstream tobacco smoke were freely emitted into the chamber where they were mixed thoroughly by 6 small fans. For each experiment, we collected TPM on Teflon-coated glass fiber filters (sampling at ~18 liters/min), after smoking stopped over a sampling time period of between 30 and 60 min. Although nicotine in ETS is mostly in the vapor phase (6), we collected both particle and vapor-phase nicotine on a single Tenax sorbent tube (sampling at ~100 cm^3/min) just after smoking stopped for sampling time periods similar to those for the filters. The nicotine samples were analyzed by thermal desorption onto a GC with a flame ionization detector.

We determined TPM concentration gravimetrically by weighing the accumulated mass on each filter using a Cahn 25 precision electrobalance. Semi-continuous particle size measurements were made using a differential mobility particle sizer (TSI, Inc., St. Paul, MN), which scanned over 34 size bins in the range of approximately 0.01 to 0.5 μm in diameter.

Calculation of Nicotine and TPM Emission Factors. Our goal is to calculate apparent emission factors for nicotine and TPM. Emission factors are defined as the mass of pollutant emitted per unit mass of tobacco smoked [mg/g] or per unit of time [mg/min]. We use a well-mixed reactor model with terms for source emission, pollutant removal by ventilation, and pollutant removal by deposition:

$$(1) \qquad V\frac{dC(t)}{dt} = E - AC(t) - KC(t)$$

where V is the volume of the chamber [m^3], $C(t)$ is the pollutant concentration in the chamber at time t [mg/m^3], E is the pollutant emission rate [mg/min], A is the ventilation rate [m^3/min], and K is the deposition rate [m^3/min], which can be related to the deposition velocity, v_d [cm/s]. While ETS particles adhere to surfaces and are well described by the linear model in Equation 1, nicotine has been described by Van Loy et al. (7) using a nonlinear, reversible sorption model. They observed the nicotine concentration to decrease rapidly immediately following smoking, and subsequently begin a slower decay. Since Equation 1 neglects this initial, rapid nicotine loss, we expect our results for ETS to underestimate total nicotine emissions (see additional discussion below).

Since E, A, and K in Equation 1 are taken to be constant, the mass emitted, q, can be written as follows (for either TPM or nicotine):

$$(2) \qquad q = E[t_2 - t_1] = V[C(t_2) - C(t_1)] + [A+K]\int_{t_1}^{t_2} C\, dt$$

where $t = t_1$ is the time that smoking begins and $t = t_2$ is the time that smoking ends. In our case, we have $t_1 = 0$ and $C(0) = 0$, so that $C(t_2)$ is the peak pollutant concentration, C_{max}. In addition, the integral on the right-hand side of Equation 2 is equal to the average pollutant concentration, $\overline{C}$, between $t = t_1 = 0$ and $t = t_2$ multiplied by $t_2 - t_1 = t_2$. With an elapsed smoking time $t = t_2$:

$$(3) \qquad q = VC_{max} + [A+K]\overline{C}\, t_2$$

Dividing q by the mass of tobacco smoked gives the mass-normalized emission factor [mg/g]. Dividing q by the source duration gives the emission rate [mg/min]. If the total removal rate $(A + K)$ is fairly low, the concentrations determined from our integrated samples (taken immediately after smoking) are approximately equal to C_{max}, and the second term on the right-hand side of Equation 3 is likely to be small. We have included this term in our calculations by using half of the peak concentration, $\frac{1}{2} C_{max}$, to represent the mean pollutant concentration during smoking, $\overline{C}$, since the increase in C during the smoking episode is nearly linear.

RESULTS
The smoking times and calculation of the mass of tobacco smoked are summarized in Table 1. We determined the mass of tobacco smoked on a length basis (i.e., we multiplied the length smoked by the mass of tobacco per unit length of tobacco in the fresh product) to eliminate errors produced by changes in density of the unsmoked tobacco column. Estimates of the measurement uncertainty for concentrations of TPM and nicotine are 3% and 5%, respectively. The total removal rate $A+K$, as determined by exponential decay constants (first order), ranged from 0.04 to 0.08 m^3/min (mean of 0.06 m^3/min) for TPM and 0.08 to 0.16 m^3/min (mean of 0.12 m^3/min) for nicotine.

Table 1. Smoking Time and Mass Smoked for the Cigars and Cigarettes Used in This Study

Tobacco Product (N) [a]	Smoking Time, Ave (SD) [min]	Original Mass, [b] Ave (SD) [g]	Original Length, [b] Ave (SD) [mm]	Mass per Length, Ave (SD) [g/mm]	Length Smoked, Ave (SD) [mm]	Mass Smoked, [c] Ave (SD) [g]
Regular Cigar (5)	12.6 (1.8)	6.2 (0.26)	133 (0.44)	0.047 (0.002)	34 (5.0)	1.6 (0.21)
Premium Cigar (1)	13.2 (0)	16.3 (0)	181 (0)	0.09 (0)	14 (0)	1.3 (0)
Cigarillo (3)	13.2 (2.7)	2.7 (0.39)	105 (0)	0.026 (0.004)	44 (12)	1.12 (0.21)
Cigarette (5)	6.4 (0.8)	0.74 (0)	80.9 (0)	0.009 (0)	79.3 (0.13)	0.72 (0.001)

[a] Regular cigar = Swisher Sweets regular, premium cigar = Macanudo, cigarillo = Tiparillo (aromatic or mild blend), cigarette = Marlboro regular or Camel light. [b] These columns contain statistics for the mass and length of tobacco only – not including filters or plastic tips. [c] The mass smoked is calculated by multiplying the length smoked by the mass of tobacco per unit length of fresh tobacco. Abbreviations: N, sample size (number of trials); Ave, average; SD, standard deviation.

Total Particle Mass (TPM) and Nicotine Emissions. The ETS TPM emissions per unit mass of tobacco smoked [mg/g] for premium cigars, regular cigars, and cigarillos are, on average, approximately 50% of those for cigarettes (see Table 2). The nicotine emissions (mostly vapor) per unit mass of tobacco smoked for regular cigars and cigarillos are, on average, about one third of those for the cigarettes, and about one fifth of cigarettes for the premium cigar. The TPM emission rates [mg/min] for cigars are, on average, about one third to two thirds of those for cigarettes and the nicotine emission rates are, on average, about one sixth to one third of those for cigarettes. Since cigars typically contain more tobacco mass than cigarettes (up to 16 g for cigars and under 1 g for cigarettes; see Table 1), the TPM and

nicotine emitted by a fully smoked cigar is expected to be greater than that emitted from a single cigarette. ETS particles are all in the respirable size range (< 2.5μm), so TPM can be considered equivalent to respirable suspended particle (RSP) measurements.

Particle Size Distributions. The fitted geometric mean of the ETS particle-size distribution (the median particle size) measured just after smoking appears to be slightly smaller (by 0.025 μm) for regular cigars than for cigarettes (see Table 3). Cigarettes and cigarillos appear to have comparable geometric means. These results are based on only three experiments and should be considered preliminary. All three distributions were approximately lognormal and had similar fitted geometric standard deviations (1.6 to 1.7). Both cigar and cigarette generated particles were observed to coagulate with time. The observed GM of their respective size distributions was seen to increase by about 0.05 μm during the two hours after smoking ceased.

Table 2. Total Particle Mass (TPM) and Nicotine Emission Factors

Tobacco Product (N_{tpm}, N_{nic}) [a]	TPM Emission Factors, [b] Ave (SD)		Nicotine Emission Factors, [b] Ave (SD)	
	[mg/g]	[mg/min]	[mg/g]	[mg/min]
Regular Cigar (5,4)	5.7 (1.5)	0.75 (0.23)	0.41 (0.25)	0.052 (0.032)
Premium Cigar (1,1)	4.3 (0)	0.40 (0)	0.26 (0)	0.024 (0)
Cigarillo (2,1)	4.1 (0.45)	0.33 (0.11)	0.42 (0)	0.027 (0)
Cigarette (3,2)	10.3 (1.7)	1.1 (0.11)	1.3 (0.25)	0.14 (0.0052)

[a] Regular cigar = swisher sweets, premium cigar = Macanudo, cigarillo = Tiparillo (aromatic or mild blend), cigarette = Marlboro regular or Camel light. [b] Emission factors have the units of milligrams emitted per gram of tobacco smoked and milligrams emitted per minute. Abbreviations: N_{tpm}, sample size for total particle mass (TPM) determinations (number of trials); N_{nic} sample size for nicotine determinations (number of trials); Ave, average; SD, standard deviation.

Table 3. Particle-Size Distribution Immediately After Smoking Stopped

Tobacco Product [a]	Particle Size [μm]	
	Fitted GM [b]	Fitted GSD [b]
Regular Cigar	0.075	1.7
Cigarillo	0.094	1.6
Cigarette	0.10	1.6

[a] Regular cigar = Swisher Sweets regular, cigarillo = Tiparillo (aromatic or mild blend), cigarette = Marlboro regular or Camel light. Distribution statistics represent measurements taken from a single experiment. [b] The statistics GM and GSD were obtained from the lognormal fit to binned particle count data. Abbreviations: GM, geometric mean; GSD, geometric standard deviation.

DISCUSSION
The emission factors reported in this paper can be used to model the ETS exposure that humans receive. Through knowledge of (1) source activity (i.e., when and how many

cigarettes or cigars are smoked), (2) source emission factors, such as those presented in this paper, (3) receptor proximity (i.e., when and where the exposed person is present during smoking), and (4) building characteristics, reasonable estimates of human exposure can be made. Since the smoking of cigars has recently become more popular (4), it is important to understand how ETS emissions from cigars and cigarettes differ. Rather than look at total emissions, in which case a single cigar would typically have greater emissions than a single cigarette, we use normalized quantities such as the amount of emissions per unit mass of tobacco smoked to compare the source strengths of different tobacco products. These quantities can be used to calculate the resulting emissions after an arbitrary portion of a cigar (or cigarette) has been smoked.

In the present study, we have smoked cigars and cigarettes in an identical manner and under identical room conditions. This methodology increases our confidence in making comparisons between the two different sources.

Calculations of apparent ETS nicotine emission factors that don't account for rapid sorption of nicotine, such as ours and those of Daisey et al. (1), underestimate nicotine emissions. For example, Daisey et al. (8) report total sidestream nicotine emissions of 5.35 mg/cigarette or about 7–9 mg/g (assuming 0.6–0.7 g of tobacco smoked per cigarette). However, while our apparent emission factors do not describe absolute nicotine emissions, they can still be useful for comparisons and predictions of exposure under conditions similar to those in our study.

Table 4. Comparison of Particle Mass and Nicotine Emissions to Those from Other Studies

Study	Source Type [b]	Particle Mass Emission Factor, [a] Ave (SD) [mg/g]	Nicotine Emission Factor, [a] Ave (SD) [mg/g]
This Study	3 Cigars	5.2 (1.4)	0.39 (0.20)
CPRT (3)	13 Cigars	10.3 (2.4)	0.13 (0.08)
Klepeis et al. (2)	2 Cigars	8.1 (0.14)	-
Nelson et al. (5) [c]	6 Cigars	6.4 (4.1)	-
This Study [d]	2 Cigarettes	10.3 (1.7)	1.3 (0.25)
Daisey et al. (1)	6 Cigarettes	12.4 (1.3)	1.4 (0.26)

[a] Emission factors in this table have the units of milligrams emitted per gram of tobacco smoked. [b] The number of different types of cigars or cigarettes used in the study. [c] Mass emission factor obtained from presentation slides (5). Nicotine emission factor unavailable. [d] Only 1 type of cigarette was used for nicotine statistics
Abbreviations: Ave, average; SD, standard deviation.

The normalized, apparent cigarette emission factors [mg/g] that we determined for both TPM and nicotine are comparable to those determined using a similar method by Daisey et al. (1) (see Table 4). The emission factors we determined for cigars are clearly much lower than those determined for cigarettes. The work by CPRT (3) suggests even lower nicotine emission factors for cigars than for cigarettes, but particle mass emission factors that are comparable to what has been obtained for cigarettes. In addition, Klepeis et al. (2) report particle mass emission factors for two premium cigars that are approximately 80% of those for cigarettes.

From the above comparisons between different studies, it appears as though cigar brand and/or smoking style (two principal variants within and/or between these studies) may be important factors in determining emissions of particles. While normalized nicotine emissions seem to always be lower for cigars than for cigarettes, particle mass emissions can range from being approximately equal to emissions for cigarettes to being half those of cigarettes.

REFERENCES
1. Daisey JM, Mahanama KRR, and Hodgson AT (1998) Toxic volatile organic compounds in simulated environmental tobacco smoke: emission factors for exposure assessment *J. Expos. Anal. Environ. Epidemiol.* 8(3): 313-334.

2. Klepeis NE, Ott WR, and Repace JL (in press) The effect of cigar smoking on indoor levels of carbon monoxide and particles *J. Expos. Anal. Environ. Epidemiol.*

3. CPRT Laboratories, Inc. (1990) Development of Methods for the Characterization of Toxic Constituents in Cigars, Pipe Tobacco, and Smokeless Tobacco. Tobacco Branch, Health Canada. SSC Contract #H4078-1-C230/01-SS. Cited on pages 169 and 178 in reference number 4.

4. NCI (1998) Cigars: Health Effects and Trends, Monograph 9, Bethesda: National Institutes of Health, National Cancer Institute.

5. Nelson PR, Kelly SP, and Conrad FW (1997) Generation of environmental tobacco smoke by cigars. Presented at the 51st Tobacco Chemists' Research Conference. Winston-Salem, NC, September 14-17, p. 15. Cited on page 102 in reference number 4.

6. Gundel LA and Lane DA (1998) Sorbent-Coated Diffusion Denuders for Direct Measurement of Gas/Particle Partitioning by Semi-Volatile Organic Compounds. Final Report LBNL-41278. Berkeley: Lawrence Berkeley National Laboratory, Environmental Energy Technologies Division.

7. VanLoy MD, Lee VC, Gundel LA, Daisey JM, et al. (1997) Dynamic behavior of semivolatile organic compounds in indoor air 1. Nicotine in a stainless steel chamber *Environ. Sci. Technol.* 31(9): 2554-2561.

8. Daisey JM, Mahanama KRR, and Hodgson AT (1994) Toxic Volatile Organic Compounds in Environmental Tobacco Smoke: Emission Factors for Modeling Exposures of California Populations. Final Report. Contract Number A133-186. LBL Report No. 36379. Sacramento: California Air Resources Board.