CHARACTERIZING SIZE-SPECIFIC ETS PARTICLE EMISSIONS

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ABSTRACT
We report a method for estimating the size distribution of particle emissions from indoor sources. The method is applied to concentration data from a series of cigar and cigarette experiments to characterize environmental tobacco smoke particles. The method incorporates a particle dynamics model, which provided good fits to observed concentrations when using, as input, optimal values of mass emission rate and deposition velocity for each particle size range. The optimal particle emissions were fit to log-normal distributions, yielding mass median diameters of approximately 0.2 μm and an average geometric standard deviation of 2.3. The total particle emissions obtained by integrating the empirical size distribution were 0.2 - 0.7 mg/min for cigars and 0.7 - 0.9 mg/min for cigarettes. The measurements of particle size characteristics agree well with prior research, but the integrated mass measurements are consistently lower than those determined from filter-based measurements.

INDEX TERMS
Environmental tobacco smoke, Particle emissions, Particle size distribution, Modeling, Sources

INTRODUCTION

Accurate modeling of environmental tobacco smoke (ETS) particle concentrations can improve our understanding of human exposure to ETS particles, including the regional deposition of ETS particles in the lung. To this end, we seek an improved characterization of the particle size distribution of ETS emissions, as well as an evaluation of model performance and a better understanding of ETS particle dynamics.

Because size is a major factor controlling airborne particle behavior, many previous investigators have characterized the particle size distribution of mainstream, sidestream, or ETS emissions (Chung and Dunn-Rankin, 1996; and Chang et al., 1985), including aged and diluted sidestream smoke (Ingebrethsen and Sears, 1989) and smoke generated by a person in a room-sized chamber (Kleeman et al., 1999). The particles in ETS, having undergone mixing and dilution over varying time scales, have size distributions that can be quite different from the distributions of fresh mainstream or sidestream smoke. While undergoing dispersion, particle mass can evaporate (Hinds, 1978), leading to smaller-sized particles, or coagulate, leading to larger-sized particles. Particle-size distributions may also be transformed by particle deposition on room surfaces and removal by filters.

The approach we follow in the present work is based on a preliminary effort by Sextro et al. (1991), and consists of the analysis of measured time-dependent ETS concentrations using a particle dynamics model developed by Nazaroff and Cass (1989). The model is fitted to the

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experimental data with emissions and deposition profiles as unknown inputs. By adjusting the input parameters such that an optimized fit is obtained, important information can be obtained about ETS particles. The quality of the fits gives an indication of model accuracy.

METHODS

For this study, we used data from experiments by Klepeis et al. (1999) in which a single cigarette or cigar was machine-smoked inside a low-ventilation stainless steel chamber. In these experiments, ETS particle concentrations were measured in 1-minute intervals using an optical particle counter (PMS-LASAIR; 0.1 - 2 μm diameters) and over 10- or 30-minute scans using a differential mobility particle sizer (TSI-DMPS; 0.01 - 1 μm diameters). Total particle mass (TPM) concentrations were also measured using Teflon filters, which were weighed before and after each experiment.

We adapted the aerosol dynamics model of Nazaroff and Cass (1989) to calculate ETS-particle concentrations for each experiment, taking into account the effects of ventilation, particle coagulation, particle deposition, and direct particle emissions from a cigar or cigarette. The model does not incorporate particle evaporation or condensation, although Kousaka et al. (1982) report that humidity only alters the size distribution of tobacco smoke particles under supersaturated conditions, which do not apply in our case. Ventilation was measured from tracer gas decay, and an ETS particle density of 1.1 g/cm^3 (Lipowicz 1988) was used for conversion between number and mass concentration. The unknown model parameters, which we intend to characterize, are size-specific particle deposition (quantified as a deposition velocity) and size-specific mass emission rate.

To determine the unknown parameters for each of the measured particle size ranges (or *bins*), our task is to find the mass emission rate and deposition velocity for each bin that result in the best fit of model predictions to measured concentrations. In mathematical terms, we are solving an *inverse problem* (Rabitz and Alis, 2000). The fits were accomplished using a search method where values for emissions and deposition velocity were varied in a grid pattern until the minimum mean absolute deviation between the modeled and observed time series was encountered (see Figure 1). We term this procedure *parameter optimization*, since we are concerned with finding *optimal* values for model input parameters (see Law and Kelton, 1991, Chapter 12). Initial guesses for each optimization were determined from the observed peak concentrations and decay rates for each size bin (using the LASAIR data), assuming independence between the bins. Initial guesses for particle sizes below 0.1 μm, which were missed by the LASAIR, were obtained from the DMPS data and then optimized against all of the larger LASAIR bins.

After the optimization procedure was completed for each experiment, we fit a log-normal distribution to the mass emission rate to obtain estimates for the mass median diameter (MMD) and geometric standard deviation (GSD) of the emissions size distribution. These fitted parameters are identical for the distributions of total mass emissions (mg), emission rate (mg/min), and mass-normalized emissions (mg/g-smoked).

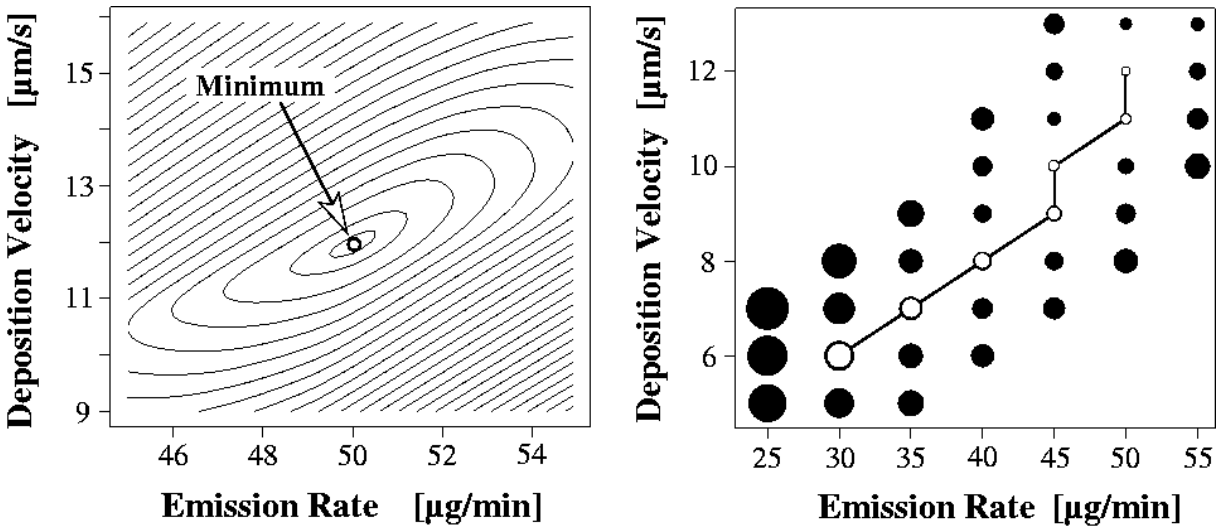

Figure 1. The left plot shows contours of the optimization response surface for particles with diameters of 0.1 - 0.2 µm (for the *Cigarillo #2* experiment) over a range of emission rates and deposition velocities. The right plot depicts the *optimization pathway*, illustrating how the local grid search method was used to find the minimum point on the surface and the optimal values of 12 µm/s and 50 µg/min. The circle size indicates the error of the model in fitting the measurements.

RESULTS AND DISCUSSION

Table 1 contains a summary of each experiment and the filter-based results for TPM emissions. Table 2 contains our best estimates of the size distribution of particle mass emissions and integrated emissions for eight experiments. The total particle mass emitted by the cigarillos and premium cigar and their emission rates were markedly lower than for the other types of cigars and for cigarettes. This finding may be an artifact of leakage around the end-fittings during smoking (the cigarillos had plastic tips and the premium cigar was rather bulky). Total particle emissions determined by integrating the particle mass size distributions (Table 2) were generally lower than those determined using filters (Table 1) with total particle mass emissions ranging from 54 - 84% of the filter-based emissions. The larger values for filters may be a consequence of the collection onto the filters, by sorption or condensation, of semivolatile organic compounds that are present in ETS. The integrated emissions were consistently higher for cigarettes (7 - 8 mg/g-smoked and 0.7 – 0.9 mg/min) than for cigars (3 - 5 mg/g-smoked and 0.2 – 0.7 mg/min). The mass-normalized emissions (mg/g-smoked), which may be more appropriate for direct comparisons, showed consistent results among different types of cigars.

The fits between observed and modeled time series data were generally quite good with minima for the mean absolute deviation in each bin ranging from 0.9 to 3 µg/m^3. The model seemed to accurately account for the effect of particle coagulation, which influenced concentrations in the smaller size bins for times as long as 4 hours after the source was extinguished. However, we observed small systematic deviations in the fits for the first hour or two of a few of the time series in the > 0.4 µm particle diameter range, where the observed particle loss appeared to be faster than later in the time series. This effect may be due to evaporative losses.

Figure 2 shows the log-normal fits to particle emissions for selected experiments. The fitted parameter values are given in Table 2. MMDs were close to 0.2 µm (mean = 0.20 µm; sd =

0.02 μm) for all source types. The variation in GSDs was larger with a range of 1.9 to 3.1 (mean = 2.3; sd = 0.37). ETS particle mass emissions appear to be due mostly to particles that have diameters between 0.02 and 2 μm — with no clear difference in the estimated mass size distributions for cigars and cigarettes.

Table 1. Summary of chamber experiments and filter-based ETS-particle emissions

Experiment	Smoking Duration [min]	Tobacco Mass Smoked [g]	Filter-Based Emissions[a]		
			TPM [mg]	Rate [mg/min]	Mass-Normalized [mg/g-smoked]
Regular Cigar #1	12.5	1.71	10.8	0.86	6.3
Regular Cigar #2	14.8	1.46	8.1	0.55	5.6
Regular Cigar #3	10.3	1.92	9.5	0.92	4.9
Regular Cigar #4	11.3	1.41	11.8	1.04	8.4
Regular Cigar #5	14.0	1.50	7.3	0.52	4.8
Premium Cigar	13.4	1.26	5.6	0.42	4.0
Cigarillo #1	10.1	1.02	4.0	0.67	6.6
Cigarillo #2	14.8	0.96	4.0	0.27	4.1
Cigarillo #3	14.8	1.36	6.3	0.42	4.6
Cigarette #1	5.5	0.73	9.8	1.79	13.4
Cigarette #2	6.1	0.72	7.0	1.15	9.7
Cigarette #3	7.4	0.72	9.3	1.3	13.0
Cigarette #4	7.1	0.72	7.3	1.03	10.1

[a] Emission rate and mass-normalized emissions were calculated by dividing the total mass emissions by the smoking time and mass of tobacco smoked, respectively.

Table 2. The estimated size distributions of ETS-particle emissions

Experiment	MMD[a] [μm]	GSD[b]	Integrated Emissions[c]		
			TPM [mg]	Rate [mg/min]	Mass-Normalized [mg/g-smoked]
Regular Cigar #1	0.18	2.5	8.8	0.71	5.2
Regular Cigar #2	0.21	2.2	6.7	0.46	4.6
Regular Cigar #3	0.20	2.4	6.3	0.61	3.3
Premium Cigar	0.18	1.9	4.7	0.35	3.7
Cigarillo #2	0.23	3.1	2.8	0.19	2.9
Cigarette #2	0.21	2.1	5.5	0.90	7.7
Cigarette #3	0.20	2.1	5.0	0.68	7.0
Cigarette #4	0.19	2.1	5.1	0.71	7.1

[a]MMD is the fitted mass median diameter, also called the mass geometric mean diameter.
[b]GSD is the fitted geometric standard deviation.
[c]The "integrated'' total mass, rate, and mass-normalized emissions were obtained by integrating the estimated size-distribution of total mass emissions and multiplying by unity, the smoking time, or the mass of tobacco smoked, respectively. See Table 1 for the values of the smoking time and mass of tobacco smoked.

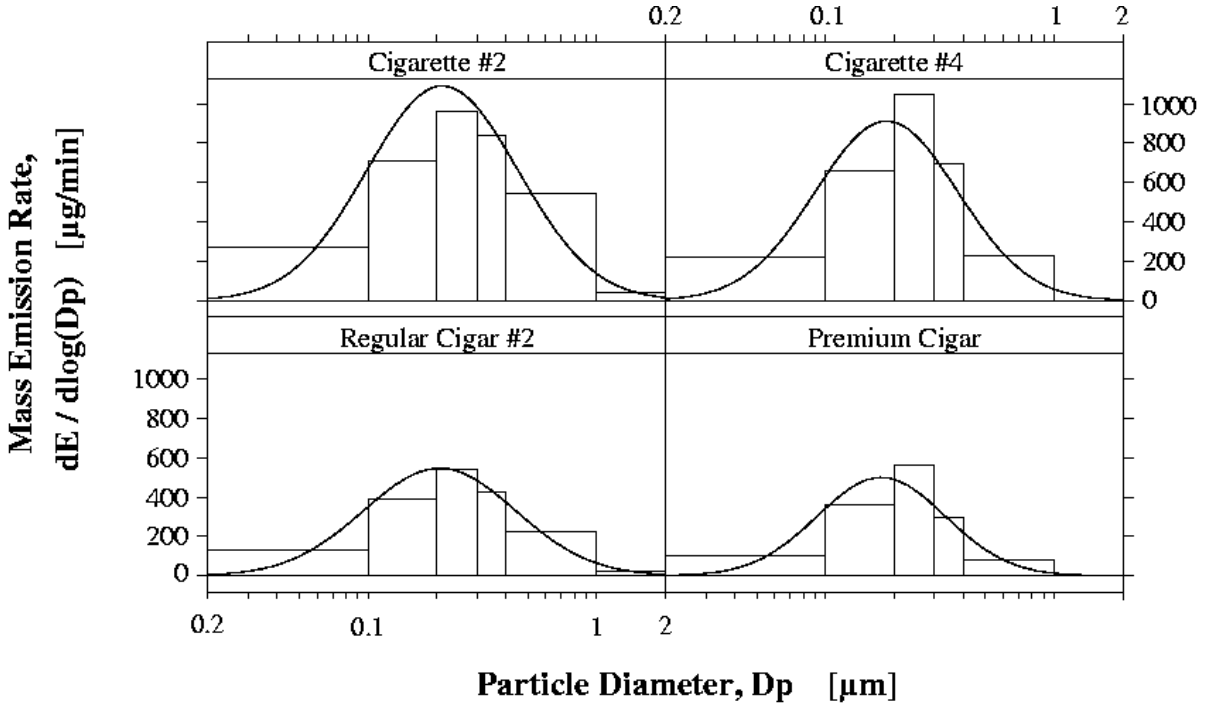

Figure 2. Plots of the estimated size distribution of particle mass emission rate (in µg/min) and the corresponding log-normal fits for four experiments. The text at the top of each panel states the type of source used in each experiment. See Table 2 for the fitted mass median diameter (MMD), geometric standard deviation (GSD), and integrated mass emissions.

Since our approach neglects evaporation, our results may have been influenced by the evaporation of volatile particle constituents, which is likely to have occurred as the emitted ETS was rapidly mixed and diluted with chamber air. The true emissions may be larger in magnitude and occur at larger particle sizes than we determined. However, using the optimal values of emissions and deposition rate for input, our model provides a good fit to ETS-particle concentrations for most size ranges and for most times, and we therefore judge it to be an appropriate tool for predicting concentrations and exposures. In addition, our results for the size distribution of ETS-particle emissions are in generally good agreement with the findings of other investigators. Ingebrethsen and Sears (1989) find an MMD of 0.2 µm for aged and diluted sidestream smoke and Ueno and Peters (1986) find an MMD of 0.16 µm with a GSD of 1.4-1.7. Chang et al. (1985) reported an average MMD of 0.26 µm for diluted mainstream smoke with a GSD of 1.2. In alignment with our findings, both Ueno and Peters and Chang et al. report total mass emissions from real-time instruments (using either electrostatic mobility or light scattering) that are as little as 50% of determinations based on direct mass measurements (a cascade impactor in their case). As far as we know, this discrepancy has not been resolved and will require further investigation.

CONCLUSION AND IMPLICATIONS

ETS particle concentrations depend on the conditions of mixing and dilution, including smoking style, which can vary from one smoking situation to another. Therefore, additional emissions characterization and model evaluation studies, similar to ours, should be carried out using designed experiments that capture real-world variation in room conditions and smoking behavior. In the interest of improving human exposure and risk assessments, the size-resolved emissions of other indoor particle combustion sources that emit submicron particles, such as heaters, stoves, incense, or candles, should also be studied.

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