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MG Apte[1], LA Gundel[1], RL Dod[1], GM Chang[2], RG Sextro[1]

[1]Environmental Energy Technologies Division
Indoor Environment Department
Lawrence Berkeley National Laboratory
Berkeley, CA, USA

[2]Department of Electrical Engineering and Computer Science University of California, Berkeley
Berkeley, CA, USA

February 2002

This work was supported by funds from the California Tobacco-Related Disease Research Program, Grant Number 6RT-0307. Additional support was by the U.S. Department of Energy under Lawrence Berkeley National Laboratory contract number DE-AC03-76SF00098

LBNL-49585

A PILOT STUDY OF THE BEHAVIOR OF GAS- AND PARTICLE-PHASE ETS TRACERS IN RESIDENCES

MG Apte[1*], LA Gundel[1], RL Dod[1], GM Chang[2], and RG Sextro[1]

[1]Indoor Environment Dept., Lawrence Berkeley National Laboratory, Berkeley, CA, USA

[2]Department of Electrical Engineering and Computer Science, University of California, Berkeley, CA, USA

ABSTRACT

Our previous study of environmental tobacco smoke (ETS) in a three-room environmental chamber showed that smoking history significantly influenced inter-room ETS transport , particularly of gas-phase nicotine. We conducted a three-home pilot study where smoking was limited to one room. Single-smoker residences were monitored during five one-week periods while the smoker participated in a smoking cessation program. Nicotine traced ETS particles were detected reliably in the smoking rooms (SRs) and unreliably in the non-smoking rooms (NSRs). On average, the ventilation- and volume-normalized smoking rate, 0.1 Cigarette-$h^{-1}m^{-3}$, added about 17 and 4 $\mu g\ m^{-3}$of ETS particles into the SR and NSR, while average nicotine concentration increases were 2 and 0.06 $\mu g\ m^{-3}$, respectively. Thus, nicotine tracers may underestimate ETS particle exposure in a NSR (e.g., a child's bedroom) by a factor of 2 to 8. In other words, ETS exposure predicted from nicotine concentrations could be almost an order of magnitude lower than actual exposure.

INDEX TERMS: Environmental tobacco smoke, Exposure assessment, Field study, FPM, Nicotine, Residence, Sorption, UVPM

INTRODUCTION

ETS is a significant contributor to the concentrations of respirable suspended particles (RSP) in indoor environments where smoking occurs (Spengler *et al,* 1981; NRC 1986). ETS exposure assessment in indoor environments has been based primarily on measurements of gas- and particle-phase chemical tracers. Our previous studies of ETS (Apte *et al*., 1999 and 2002) indicated that inter-room mixing in a three-room environmental chamber affected individual ETS constituents differently, depending upon both the chemical identity of the ETS constituent and its phase (gas or particle). Nicotine from ETS in the smoking room was virtually undetected in the corridor or the non-smoking room, even when ETS particles were easily detected, and even when the door openings allowed for free air exchange between the rooms. The rate of sorption onto and into chamber surfaces dominated the mass transfer dynamics.

These results also suggested that ultraviolet absorbing particulate matter (UVPM) and fluorescent particulate matter (FPM) could trace ETS particles more accurately than nicotine when measurements are made in environments with varying or unknown conditioning of interior surfaces (smoking history). ETS in the SR was used to calibrate UVPM so that the

* Contact author email: MGApte@lbl.gov

ratio of UVPM to respirable suspended particles (RSP) averaged 1.0 in the smoking room. That ratio was 0.8 in the corridor and nonsmoking room, indicating a possible 20% reduction of PM tracer specificity through transport and loss mechanisms such as selective volatilization of UV-absorbing material from the particles. In contrast, the ratio of nicotine to RSP was about 0.2 in the smoking room, and dropped to about 0.05 in both the corridor and NSR. This 400% reduction in the nicotine to RSP ratio suggests a potential for significant underestimation of particle-phase ETS exposure when nicotine is measured in non- or lightly-ETS-conditioned environments that connect to rooms where smoking occurs.

Thus, the reliability of ETS tracers in actual home or building environments should be examined in light the following criteria for tracer effectiveness:

1. Tracers must be conservative, remaining in constant ratio to the ETS constituent of interest. Exposures to ETS aerosols are generally regarded as the source of the respriratory health effects; hence tracers must reflect changes in ETS particle concentration as the ETS dilutes, ages, and moves between rooms, even in the presence of other sources of particles. Conservative tracers must also be chemically stable over the time of both collection and analysis.

2. The analytical methods for tracers must have adequate sensitivity and selectivity.

3. The analytical methods must be cost effective. This is especially important for large-scale exposure studies where many samples will be processed. The main issue is balancing precision and accuracy with analysis cost.

A three-home pilot field study was conducted to: (a) examine the reliability of various ETS particle tracers in real indoor environments; (b) estimate potential bias of nicotine-based exposure assessments due to nicotine sorption and re-emission; (c) estimate potential bias of particle-mass based exposure assessments due to interference from other particle sources; and (d) quantify non-smoker ETS residential exposure reduction due to smoking cessation.

Table 1. Instrumentation and measurement methods for 3 home ETS pilot field study.

	Sampling Method	*Instrumentation*	*Analytical Method*
Nicotine	Passive diffusion, treated filter	Hammond Passive Sampler (see Hammond and Leaderer, 1987)	GC-NPD
RSP (PM 3.5)	Gravimetric air sampling at 1.7 $L \cdot min^{-1}$	10mm Nylon Cyclone, Teflon Coated Glass Fiber Filter	Gravimetry
UVPM,FPM	As PM 3.5	Analysis of PM 3.5 filter	HPLC
Ventilation Rate	Peristaltic pump, Tedlar source and sample collection bags	Continuous injection and sampling of sulfur hexafluoride (SF_6)	GC-ECD

METHODS

Participants were recruited from members of a six-week smoking cessation class conducted by the Department of Health and Human Services, Tobacco Prevention Program, Berkeley, CA, USA. Each participant was the only smoker in the household, and smoking always took place at the same location in the house. Weekly smoking rate was monitored by cigarette butt count. Houses were monitored for ETS and nicotine during weeks 1, 2, 3, 4 and 6, and the

smoking cessation class encouraged abstinence beginning with week 3.

The five one-week integrated measurements included nicotine, particle mass, UVPM, FPM and time averaged whole-house ventilation rate (Table 1). RSP was sampled with a 10 mm nylon cyclone (50% cutpoint = 3.5 μm, flowrate 1.7 L min^{-1}) and collected on pre-weighed, pre-cleaned filters. The homes were visited weekly to recover the samples and deploy new sampling media.

Particle and nicotine samples were collected indoors in the rooms identified as the main smoking room and the non-smoking room. Outdoor PM 3.5 was also measured weekly. These outdoor samples were used to adjust for any infiltrating particles or particle-bound tracers. Particle mass was determined gravimetrically. UVPM and FPM concentrations were analyzed from extracts of the PM 3.5 particle filter samples (Apte *et al*., 2002). Solanesol analyses were conducted, but solanesol was detected in only one location, the SR used by the heaviest smoker. We suspect that low concentrations of solanesol were not stable over the one-week sampling period.

The limit of detection (LOD) of the nicotine, based on field blanks, is about 0.07 μg m^{-3}. Likewise, uncertainty based on duplicate measurements is about ±10% for nicotine greater than 0.15 μg m^{-3} (LOD*2) and ±50% for those less than 0.15 μg m^{-3}. Estimated UVPM and FPM uncertainty ranged from ±30% to ±50%, and from ±40% to ±80%, respectively, depending on the concentration range (Apte *et al*., 2002). Uncertainty in the PM 3.5 measurements was less than ±10%.

Whole-house air exchange rate was measured by decay of sulfur hexafluoride injection (SF_6). An SF_6 injection suitcase was located in a central (non-smoking) room about 1-2 m above the floor. Two sampling suitcases were used, one in the smoking room and one in the non-smoking room. Weekly-average indoor SF_6 concentrations ranged from 18 to 780 μg m^{-3} during the study.

Although the exact smoking time and emission rate profile would be needed to calculate the true week-average ETS concentrations, the steady-state mass balance model can be used to compare, on an equivalent basis, the measured ETS particle and particle tracer concentrations across study weeks and between houses. The steady-state form of the mass balance equation (Traynor et al., 1989) is:

$$C_{ss} = \frac{Pa\,C_o + S/V}{(a+k)} \qquad (1)$$

where
C_{ss} = the steady-state particle or particle tracer concentration (μg m^{-3}),
P = penetration fraction of particles from outdoors (0-1, unitless)
C_o = outdoor concentration of particles or particle tracers (μg m^{-3}),
S = the ETS particle or particle tracer source strength (μg h^{-1}),
V = the house volume (m^3),
a = the whole-house air exchange rate (h^{-1}), and
k = the particle deposition decay rate (h^{-1}).

Weekly cigarette butt count was used as a surrogate for S, assuming that this count was proportional to the weekly average ETS particle emission rate. Values of 0.08 h^{-1} and 0.7 were assumed for k and P, respectively (Traynor et al., 1989). We have used a value for k that

represents particle sizes typical of ETS. Measured house volume and weekly average air exchange rate values were used for V and a, respectively. The results for indoor PM 3.5 and ETS tracers have been adjusted by subtracting the contribution of outdoor air, $PaC_o/(a+k)$.

With the assistance of the study participants we identified rooms in each home in which smoking did or did not occur and designated them as the SR and NSR, respectively. In House 1, a single story, single family residence, the SR was the kitchen. House 2 was a second-story flat of a large 2-floor wooden structure, and the SR was the living room. House 3 was also the second story of a small two-story wooden structure and the SR was the smoker's bedroom.

RESULTS

Figure 1 shows the measured weekly SR and NSR PM3.5 concentrations (adjusted for infiltrating outdoor PM 3.5) for the three study households. The clear downward trends in adjusted indoor PM 3.5 during the course of the smoking intervention reflect the success of the participants in reducing their smoking rate.

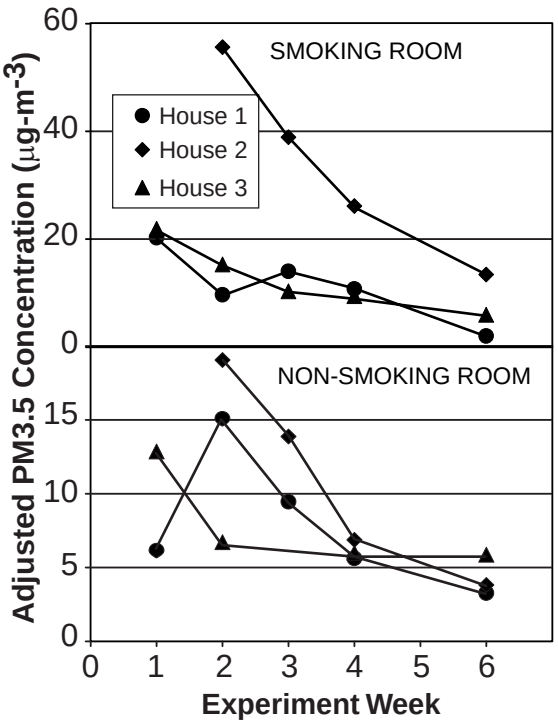

Figure 1. Adjusted weekly PM 3.5 concentrations in three study homes.

Figures 2 and 3 present the nicotine and ambient-adjusted PM 3.5, UVPM, and FPM indoor concentrations for the SR and NSR, respectively. Based upon the limited data from 3 houses, these figures should be considered suggestive of ETS behavior, but not conclusive. The ETS tracer concentrations are plotted against the household ETS source strength calculated from the weekly butt-count and ventilation rate data. Presenting the data this way allows examination of tracer behavior across the three houses. All the tracers performed similarly in predicting weekly smoking rates (and thus ETS emission rates) in the SRs in real homes – correlation between butt-count based source strengths and all tracers have R^2 values from 0.6 to 0.7.

For the NSRs the relationship between the tracer concentrations and the ETS source strength is much less clear. In Figure 3, a week 1 nicotine measurement in the NSR (0.003 μg m^{-3}) was removed from the figure, as it was an extreme outlier and possibly suspect. If this data point were included, nicotine would have no predictive value (R^2 =0.03, slope=0.70). Nicotine and FPM had poor predictive ability when NSR data from all houses were considered together. UVPM, with an R^2 of 0.5, did the best at predicting NSR ETS concentrations, while adjusted PM 3.5 had much lower R^2 values, possibly due to other indoor sources of respirable particles. Overall, none of the ETS tracers provided particularly strong predictive power for the ETS source in the NSR. Even so, UVPM and FPM, when adjusted for infiltration of outdoor PM, did meet the criteria (given in the introduction) for tracer effectiveness in both the smoking and non-smoking rooms.

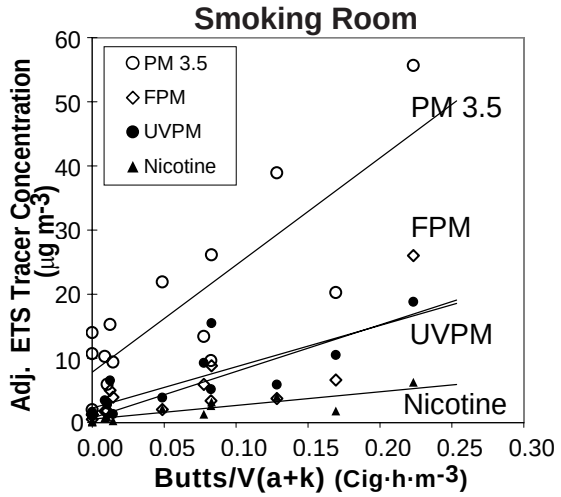

Figure 2. Adjusted ETS tracer concentrations in the SR vs. butt-count based source strength for all three study homes. Slope (R^2) for regression fits are 167 (0.68), 73 (0.63), 64 (0.68), and 21 (0.73) for PM3.5, FPM, UVPM, and nicotine, respectively.

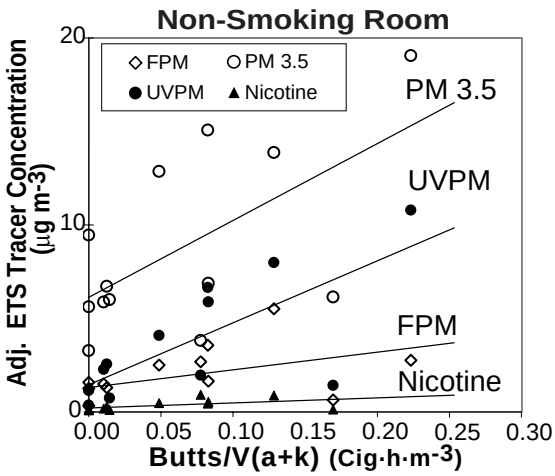

Figure 3. Adjusted ETS tracer concentrations in the NSR vs. butt-count based source strength for all three study homes. Slope (R^2) for regression fits are 41 (0.37), 33 (0.54), 9 (0.20), and 2.7 (0.26) for PM3.5, UVPM, FPM, and nicotine, respectively.

Interestingly, after adjustment for infiltrated particles, PM 3.5 rather than UVPM or FPM, provided the most consistent and highest correlation with ETS source strength. This is not likely to occur in homes with significant indoor sources of RSP such as combustion-generated particles from fireplaces and candles.

Nicotine traced ETS well in the smoking areas of all three houses, but was inconsistent in the non-smoking rooms where surfaces were probably not saturated with nicotine. The variability in nicotine concentrations in the NSRs could be attributed to the absence of information about the rates of sorption/desorption processes and inter-room air transport. In contrast, the PM tracers did follow ETS reliably in areas with unknown smoking histories and inter-room transport rates.

DISCUSSION AND CONCLUSIONS

In this study, for every 0.1 Cig-h^{-1}-m^{-3} of smoking about 17 μg m^3 of elevated ETS RSP could be expected in the SR, and 4 μg m^{-3} in the NSR. Similarly, predicted increases in nicotine concentrations at the same normalized smoking rate would be 21 μg m^{-3} and 0.3 μg m^{-3}, respectively. Thus, in these houses the concentration of ETS particulate matter in NSRs was about 25% of that in the smoking room, while on average, only about 13% of the nicotine was transported. Based on a detailed examination of the pilot study data, we conclude that an estimate of ETS particle exposure based on nicotine measurements in the NSR might be biased low by a factor ranging from two, to eight if the low nicotine value is included.

The pilot field study did not uncover significant bias from other ambient particle sources during spring weather in residential neighborhoods. Evaluation of the selectivity of UVPM for ETS compared to other particle sources requires supplementary measurements of ETS in the presence of wood smoke and diesel exhaust (Gundel *et al,* 2000). Our initial assumption was that indoor respirable particle sources such as cooking or vacuuming would be negligible compared to ETS. However, we found that indoor sources of respirable particles without a UV absorbing or fluorescing component may have been present in concentrations similar to

the ETS particles. Certainly, as the ETS concentrations declined with the residents' smoking cessation efforts, other indoor sources would become more dominant sources of particles. Additionally, the actual smoking rates in two houses were lower than expected.

Based on this pilot study, indoor PM 3.5, UVPM, and FPM and nicotine correlated well with ETS emission rates (calculated from cigarette butt counts and ventilation measurements as discussed above) in household smoking rooms. UVPM tracked reliably even in non-smoking rooms after all three smokers reduced their smoking rate by at least half during the six-week program. Additional method development is necessary to reduce the uncertainty in PM tracer quantitation. Furthermore, a larger study sample size would be necessary to make any clear conclusions and to validate our preliminary finding that exposure estimation based on nicotine measurements can be complicated and potentially biased.

ACKNOWLEDGEMENTS

We thank the participants in the study and Marcia Brown-Machen and Tina Benitez of the Tobacco Prevention Program, Berkeley (CA) Department of Health and Human Services, and Dr. Katharine Hammond and Charles Perino, UC, Berkeley. We also thank Drs. Brett Singer and Mark Mendell for their reviews. This work was supported by funds from the California Tobacco-Related Disease Research Program, Grant Number 6RT-0307. Additional support was by the U.S. Department of Energy under Lawrence Berkeley National Laboratory contract number DE-AC03-76SF00098

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