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EFFECT OF SORPTION ON EXPOSURES TO ORGANIC GASES FROM ENVIRONMENTAL TOBACCO SMOKE (ETS)

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ABSTRACT

The effects of sorption processes on dynamic ETS organic gas concentrations and potential exposures were studied in a carpeted and furnished 50-m^3 room ventilated at 0.6 h^{-1}. Ten cigarettes were machine-smoked on six of every seven days over four weeks. Concentrations of ETS-specific tracers and regulated toxic compounds were quantified during daily smoking, post-smoking and background periods. Potential exposures were calculated by period and day. Large sorption effects were observed for the widely used tracers 3-ethenylpyridine and nicotine, and for several toxic compounds including naphthalene and cresol isomers. Short-term adsorption to indoor surfaces reduced concentrations and potential exposures during smoking, while later reemission increased concentrations and exposures hours after smoking ended. Concentrations during nonsmoking periods rose from day to day over the first few weeks, presumably from increased reemission associated with increased sorbed mass concentrations. For sorbing compounds, more than half of daily potential exposures occurred during nonsmoking periods.

INDEX TERMS

Environmental tobacco smoke, Pollutant sorption and desorption, Exposure assessment, VOCs and SVOCs, Laboratory and field experiments

INTRODUCTION

Environmental tobacco smoke (ETS) is a dynamic mixture of particles, inorganic compounds, and organic gases that span a wide volatility range. The ETS gas-phase (filtered ETS) has been identified as carcinogenic in animal-based toxicology studies (Witschi et al., 1997). Many individual ETS organic gases are regulated as toxic air contaminants and hazardous air pollutants (henceforth referred to as "toxic compounds") by the California state and United States federal governments, respectively (CARB, 2001).

Quantifying exposures to toxic organic gases in ETS is challenging since many of these compounds are also emitted from other sources. One approach is to measure exposure to a tracer compound that is specific to ETS. Exposure to ETS toxic components then may be estimated based on the relative abundances of toxic and tracer compounds as measured in ETS under controlled conditions. This approach requires that tracer and toxic compounds exhibit similar dynamic behavior (Daisey, 1999). Nicotine and its primary metabolite cotinine are the most widely used chemical tracers of ETS exposure. Yet it has been known for some time that nicotine dynamics differ from those of other ETS components (Eatough, 1993). In light of this concern, other gas-phase tracers have been suggested, including 3-ethenylpyridine (3-EP), pyridine, and pyrrole (Eatough, 1993; Hodgson et al., 1996).

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Concentrations of nicotine, 3-EP, and some toxic ETS compounds are affected by sorption (Piade et al., 1999; Van Loy et al., 2001). Adsorption to indoor surfaces can reduce gas-phase concentrations during smoking events, while later reemission (desorption) can raise concentrations long after smoking has stopped. If sorption is reversible and reemission occurs on a time scale of hours or days, the sole effect should be to shift potential exposures in time. By contrast, if some fraction of the mass is sorbed irreversibly, or the time scale of reemission is much slower, then sorption should reduce potential short-term exposures associated with a single emission event. We hypothesize that when smoking occurs regularly, e.g. on a daily basis, sorbing organic compounds will accumulate on surfaces. Higher surface concentrations of reversibly sorbed mass should, in turn, lead to increased reemission rates and higher gas-phase concentrations during nonsmoking periods. Thus, we expect that longer-term sorption processes will increase potential exposures beyond those resulting from new emission events.

We investigated the dynamic behavior of ETS organic gases through experiments in which cigarettes were smoked by machine in a facility designed to simulate residential and office environments. Key components that affect sorption, such as ventilation rate and furnishing level (i.e. material type and surface area) were varied. A detailed investigation of sorption effects on exposures for single-day ETS emission events is reported elsewhere (Singer et al., 2002). This paper presents preliminary results from an investigation of sorption effects on exposures when smoking occurs on a repeated daily basis.

METHODS

Experiments were conducted in a 50-m^3 room that was constructed and furnished with typical indoor materials including painted gypsum wallboard; nylon carpet; upholstered chairs; wood and veneer desks, bookcases and tables; and heavy cotton draperies. The floor (carpet) area was 20 m^2 and the projected area of the chairs, draperies and wood furniture was 43 m^2. With 84 m^2 of wallboard, the total surface-to-volume ratio was 2.6 m^{-1}. ETS was simulated with the sidestream emissions from machine smoking of a popular brand of filter cigarettes. Room ventilation air passed through a bed of activated carbon to reduce organic gas concentrations in the inlet air. Air samples were collected on sorbent tubes containing Tenax-TA and Carbosieve S-III 60/80 mesh. Organic compounds were analyzed by thermal desorption gas chromatography/mass spectrometry. Target compounds included regulated toxics, ETS tracers, and other prominent organic gases in ETS. Detailed descriptions of all methods are provided elsewhere (Singer et al., 2002).

The effects of short- and long-term sorption processes on dynamic ETS organic gas concentrations and potential exposures were studied through two types of experiments. In the first set, organic gas concentrations were measured over multi-hour periods for 3-4 days following a single 3-h smoking period. The primary goal was to quantify the effect of sorption processes on exposures to new emissions of ETS. Exposure-relevant emission factors that account for sorptive losses and re-emission over a 24-h period were determined for three furnishing levels (wallboard with aluminum flooring, wallboard with carpet, wallboard with carpet and full furnishings) and three ventilation rates (nominally 0.3, 0.6 and 2.0 h^{-1}). Full results have been reported (Singer et al., 2002). An ongoing second group of experiments focuses on the effect of longer-term sorption and reemission processes that pertain when smoking occurs regularly and repeatedly. This paper presents results from an experiment in which 10 cigarettes were smoked in the fully furnished room during a 3-h period each day, six days per week, for 4 weeks, with a chamber ventilation rate of 0.6 h^{-1}. Organic gas concentrations were measured over three daily periods: a 4 h “smoking” period extending for

1 h after smoking ended, a 10 h "post-smoking" period, and a 10 h daily "background" period. These periods were chosen to simulate a residential scenario in which a working adult smokes for several hours each evening. Samples were collected during the background period each day, and during each period on days 1, 3, 6, 10, 13, 17, 20, 24 and 27.

RESULTS AND DISCUSSION

The short-term effects of sorption can be seen in Figure 1, which shows potential daily exposures to seven representative ETS gases on Day 1 of the repeated daily smoking experiment. These are potential exposures that would occur for a nonsmoker occupying the room throughout each period. Potential exposures were calculated as the product of the measured gas-phase concentrations and the duration of the period. For 1,3-butadiene and benzene, time-dependent concentrations were controlled entirely by emissions and ventilation; about 73% of the potential exposure occurred during the 4-h smoking period with the remaining 27% during the next 10 h period. By contrast, potential exposures during the smoking period comprised only about half of the daily total for the regulated toxics p-cresol (48%) and naphthalene (55%), and for the ETS tracer 3-EP (48%). Potential exposures during the post-smoking period accounted for 30-38% of daily totals for these compounds. The remaining 14-18% was attributed to the background period. Short-term sorption and reemission processes thus shifted about one quarter of potential exposures of these three compounds from smoking to nonsmoking periods. The ETS tracer pyridine and the toxic compound styrene (not shown) exhibited intermediate behavior with potential exposures distributed by period as follows: smoking (65%), post-smoking (35%), and background (5%). For pyrrole (also not shown), potential exposures were split evenly between smoking and post-smoking periods (45% each). The small fraction of nicotine exposure during nonsmoking periods indicates a slower, or less-reversible sorption.

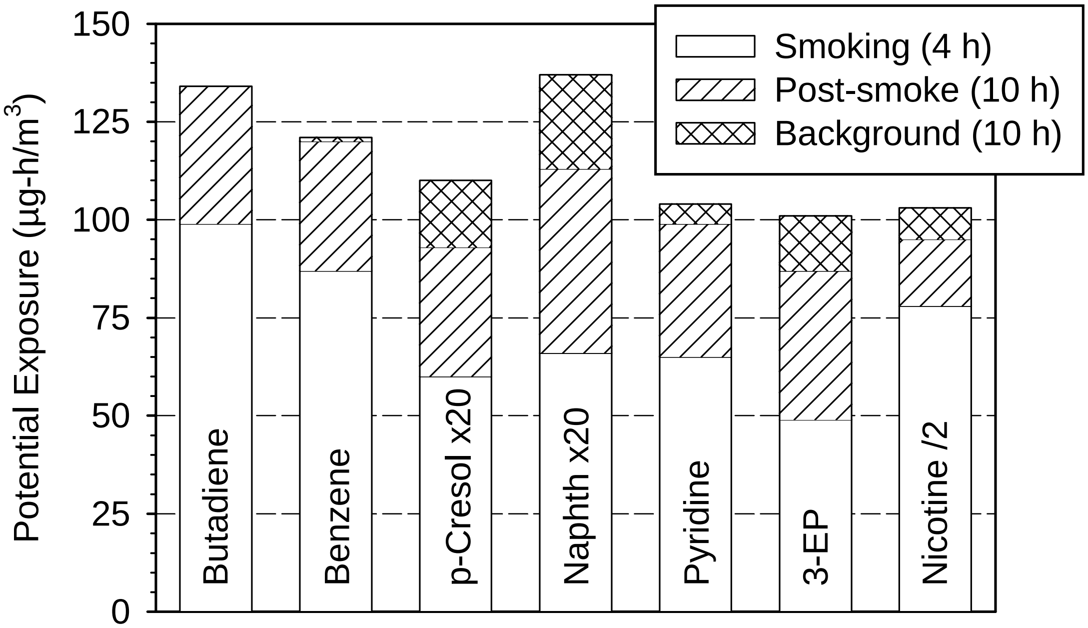

Figure 1. Potential ETS exposure, by period of day, in a furnished 50-m^3 room on **Day 1** of the repeated daily smoking experiment. 10 cigarettes were smoked by machine during the first 3 h of the designated smoking period. Naphth = Naphthalene; 3-EP = 3-Ethenylpyridine.

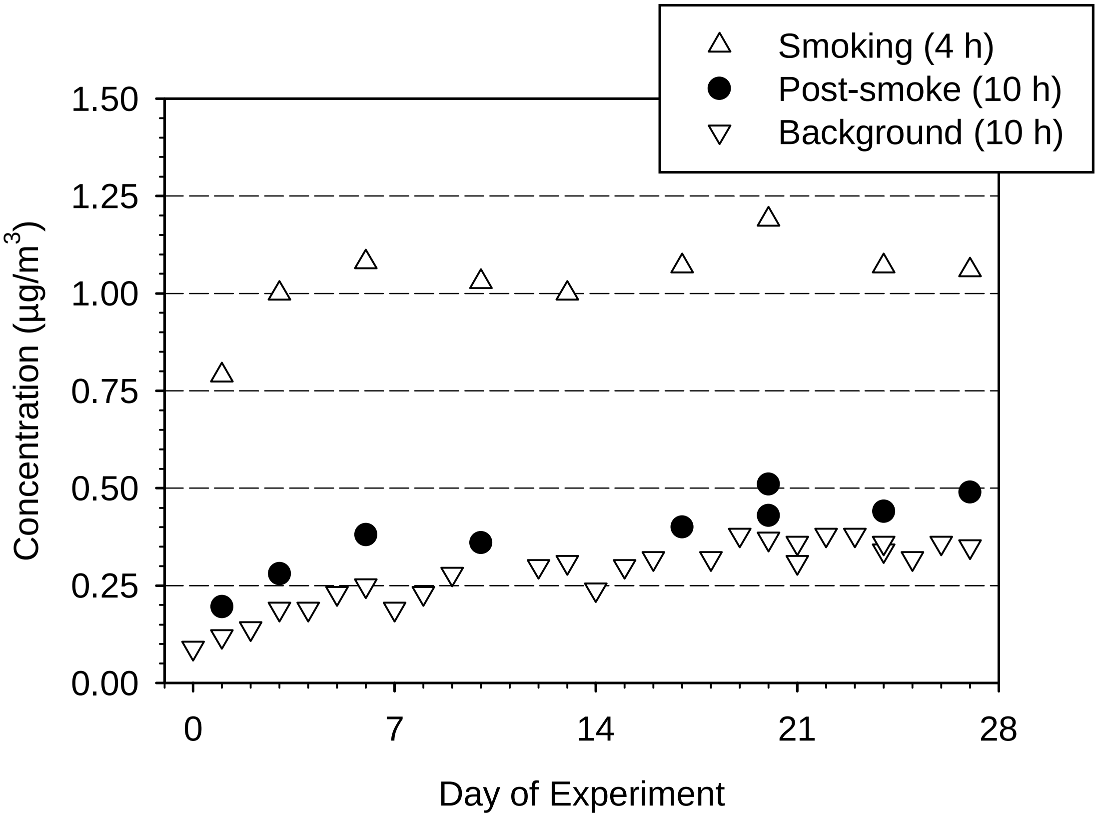

Figure 2. Time-averaged concentrations of p-cresol measured during smoking, post-smoking and background periods in a 50-m^3 furnished chamber. 10 cigarettes were smoked each day, six days per week over a 4-week period. No cigarettes were smoked on days 7, 14, and 21.

Mass loss due to longer-term sorption necessarily leads to the accumulation of sorbed mass on indoor surfaces. To the extent that the sorption is reversible there should be a consequent increase in reemission; this should in turn lead to higher gas-phase concentrations of the sorbing compounds over time. Figure 2 demonstrates this phenomenon for p-cresol over the 4-week repeated daily smoking experiment. Background period concentrations generally rose on each smoking day of the first two weeks, but dropped on days without new ETS emissions. Background concentrations leveled off during the third week then fluctuated from day to day, but did not increase overall during the fourth week. The rise in concentrations during daily smoking and post-smoking periods mirrored the rise during background periods.

These higher concentrations translate into higher potential ETS exposures for sorbing compounds when smoking is habitual. Figure 3 shows potential exposures on Day 27, resolved by daily period, for the same compounds shown in Figure 1. A comparison of the total daily exposures by compound indicates that for 1,3-butadiene and benzene, potential exposures were essentially unaffected by long-term sorption and reemission processes. (The increases of 8% for 1,3-butadiene and 20% for benzene may result from emission variability.) By contrast, potential daily exposure increased by about 110% for naphthalene and p-cresol, by 80% for 3-EP and by 180% for nicotine. Results for phenol and other cresol isomers were similar to those shown for p-cresol, whereas larger increases of 150% were observed for methylnaphthalene isomers. This result suggests that even larger effects are likely for certain polycyclic aromatic hydrocarbons (PAH), primarily three-ring compounds, that are larger than the methylnaphthalenes but still volatile enough to exist primarily in the gas-phase of ETS. Reemission of long-sorbed mass increased potential exposures to other ETS tracers by only 30% for pyridine and by 35% for pyrrole (not shown in figure).

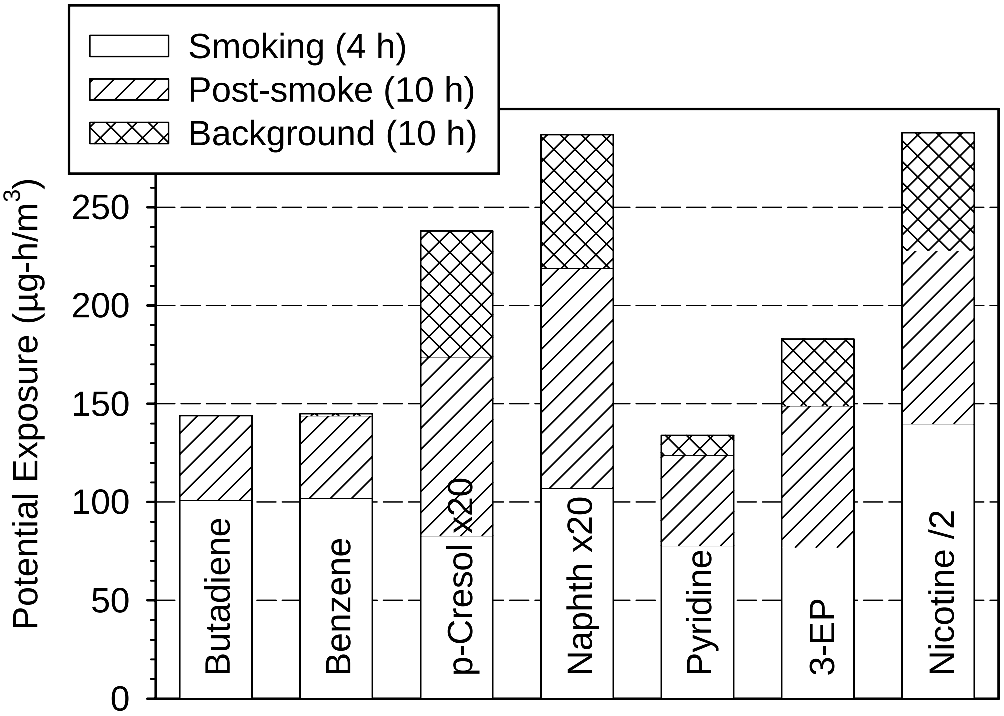

Figure 3. Potential ETS exposure, by period of day, in a furnished 50-m^3 room on **Day 27** of the repeated daily smoking experiment. 10 cigarettes were smoked by machine during the first 3 h of the designated smoking period. Naphth = Naphthalene; 3-EP = 3-Ethenylpyridine.

Figure 3 also shows that increased reemission rates shifted a larger fraction of potential daily exposures from smoking to nonsmoking periods. Potential exposures for 1,3-butadiene and benzene again occurred mostly (70%) during smoking periods, with the remaining 30% during the nominal post-smoking period. For the sorbing toxic compounds p-cresol and naphthalene, the smoking period accounted for only 35% of potential daily exposures; 40% occurred during the post-smoking period and 25% was attributed to the background period. Similar results were obtained for 3-EP and for pyrrole (not shown). The fraction of potential daily exposure during the smoking period was again higher for nicotine compared to the other sorbing compounds. But by the end of the fourth week, only about half of the potential daily nicotine exposure could be attributed to the smoking period, with 30% and 20% occurring during the post-smoking and background periods, respectively. Despite the overall increase in daily exposure potential for pyridine and pyrrole, the distribution of potential exposures throughout the day did not change appreciably for these compounds over the 4-week experiment. These results suggest that among the compounds studied, pyridine may be the best tracer of the dynamics of non-sorbing ETS compounds.

CONCLUSION AND IMPLICATIONS

The experimental data presented in this paper substantiate the hypothesized effects described in the introduction, and provide an initial estimate of the magnitude of these effects in typical residences. Overall exposures will be lower in most circumstances, since the volume of our experimental room is only about 10-20% of a typical home volume and our smoking rate was higher than would be expected for most smokers. But the observed linearity of sorption effects across varying ETS concentration levels (Singer et al., 2002) suggests that the effects reported here should apply to the lower concentrations that prevail when ETS is diluted throughout a residence. Surface-to-volume ratios also vary, but the quantity and types of surfaces in our test room are typical enough that our results should be generally indicative of sorption effects in real residential environments. Longer-term sorption effects at lower ETS concentrations are being examined using data from an experiment in which 5 cigarettes were smoked daily for 4 weeks with the room ventilated at 2.0^{-1}.

These experimental results provide the most complete picture to date of the range of effects that sorption processes may have on indoor exposures to organic gases in general and to ETS compounds in particular. They indicate that exposures to ETS organic gases under daily smoking conditions can differ markedly from what might be predicted using emission factors or toxic-to-tracer ratios measured in specialized test chambers. A logical next step is to validate these results in a real residence where human smoking or another regular source of organic gas emissions is present. Assessing the implications of these findings to ETS exposures in real-life scenarios will require incorporating information about two additional variables: human activity patterns and inter-zonal transport. Continued efforts along these lines promise to improve our understanding of exposure to constituents of ETS, and also to other sources of volatile and semi-volatile organic compounds.

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