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METHYLENE CHLORIDE – CPSC

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CONSIDERATIONS IN EVALUATING EMISSIONS FROM CONSUMER PRODUCTS

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Abstract—While several indoor air quality studies suggest consumer products (e.g. aerosol sprays, paint removers, etc.) can be significant sources of volatile organic compounds, until recently characterizing emissions from consumer products has received relatively little attention. Many considerations that must be addressed in designing studies of consumer product emissions are similar to those addressed in studies of the emissions from building materials and combustion appliances. These similarities are discussed and, in addition, the considerations unique to studies of consumer product emissions are discussed with reference to an ongoing study of consumer products that contain methylene chloride. These unique considerations include bulk chemical analysis, the form of the product (e.g. liquid, aerosol or paste) and the related consumer usage pattern. The issue of personal exposure of the product user vs the average area concentration resulting from product use must be considered, as well as the challenge of incorporating results into predictive models that adequately treat the effect of different usage patterns. Finally, post-study considerations, such as exploring new issues discovered in the study, studying similar products, and validating predictive models through extension into field studies are summarized.

Key word index: Consumer product, emissions, indoor air quality, methylene chloride, personal exposure, source characterization, ventilation.

INTRODUCTION

While several indoor air quality studies suggest that consumer products can be significant sources of volatile organic compounds (De Bortoli *et al.*, 1985; Lebret *et al.*, 1984), characterization of emissions from consumer products has received relatively little attention. This is in strong contrast to the characterization of emissions from combustion appliances (Johnson *et al.*, 1984; Leaderer, 1982; Traynor *et al.*, 1982) and building materials (Girman *et al.*, 1987; Molhave, 1982), where numerous chamber studies have been conducted. In this paper, considerations necessary for evaluating emissions from consumer products in laboratory studies are presented.

Although a broad definition of consumer products could include many combustion appliances and some building materials, a narrower definition as used in the National Academy of Sciences monograph, *Indoor Pollutants* (National Research Council, 1981) is employed in this paper. As used here, it is limited to expendable products in small containers which are readily available in retail outlets. In this context, consumer products are items such as cleaning/polishing products, insecticides, painting/finishing/refinishing products, personal grooming products, hobbyists' products, and deodorizers/disinfectants.

Many considerations that must be addressed when designing studies of emissions from consumer products are similar to those addressed in other types of indoor air quality studies. These similarities include: defining the potential health hazard; establishing study goals; assessing analytical instrument and chamber capabilities; pretesting and establishing the experimental procedures; and establishing quality assurance/control objectives. There are, however, additional considerations that are unique to studies of consumer product emissions.

These unique considerations in protocol development are discussed with reference to an ongoing study of consumer products that contain methylene chloride (CH_2Cl_2). This study, supported by the U.S. Consumer Product Safety Commission (CPSC), was undertaken by Lawrence Berkeley Laboratory (LBL) to resolve uncertainties about the exposure consumers can receive by using consumer products containing CH_2Cl_2. LBL has conducted a controlled study in an environmental chamber to measure both the airborne concentrations produced by the use of these products and the exposures of individuals using them. Development and testing of models for both average area concentrati n and personal exposure are important goals of this study.

In discussing considerations for studies of consumer

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products, eight major tasks are defined. While the order of presentation of the tasks is reasonable, it is not a rigid prescription. Certain tasks should be accomplished before starting the next task; however many tasks can be accomplished in parallel or, as is often required, in an iterative fashion.

DEFINING THE POTENTIAL HEALTH HAZARD

Many interrelated factors must be considered in defining the potential health hazard of a particular product or class of products: the chemical composition of the product; the toxicity of the compounds contained in the product; an estimate of airborne concentrations and exposures produced by the use of the product; the market penetration (either at the present time or expected in the future due to changing market conditions); and an estimate of the population potentially at risk.

This task should not be confused with actually developing an estimate of a health risk due to use of a product. This would be premature since too many parameters are not sufficiently well known. Rather, the goal at this stage is to judge whether the potential risk is large enough to warrant a detailed study.

This initial task is similar for most indoor air pollutant source characterization studies. In some respects, this task is more easily completed for consumer products than for other indoor air pollutant sources such as combustion appliances, building materials or smoking, since consumer products generally have labels describing their chemical content. However, these labels, while useful as a guide, are not necessarily comprehensive and chemical analyses are usually required.

A good example of defining a potential health hazard is provided by CH_2Cl_2, a chemical widely used by consumers. More than half of a billion pounds of CH_2Cl_2 are produced annually in the U.S., much of it for use in paint removers and aerosol finishes (Anonymous, 1985). The chemical composition of these products is readily obtained. For example, semi-paste paint removers contain approximately 85% CH_2Cl_2 and aerosol finishes with CH_2Cl_2 generally contain 20–40% of this chemical. Market data also exist for these products, although these data are not discussed here.

There is concern about consumer exposure to CH_2Cl_2 from these products, since CH_2Cl_2 retained in inhalation is metabolized to carbon monoxide, which can lead to anoxic stress from elevated levels of carboxyhemoglobin (Ratney *et al.*, 1974; Stewart *et al.*, 1972). In addition, a recent animal-inhalation laboratory study has associated increased incidence of carcinoma with exposure to CH_2Cl_2 (Peer Review Panel, National Toxicology Program Board of Scientific Councilors, 1985). These facts strongly suggest that there is a potential health risk that is worth investigation.

ACQUIRING APPLICATION TECHNIQUE AND USAGE PATTERN INFORMATION

In completing this task, the goal is to acquire sufficient information about product application and use to assure that the product is used realistically in test situations. The term "application technique" is used to denote the detailed method employed by the consumer when using the product, e.g. for a paint remover, "Use a single brush stroke, applying in one direction only. Wait at least 10 min before scraping." Since consumer products are found in a number of different forms including aerosols, liquids, semi-pastes or gels, pastes or waxes, and solids, a broad range of application techniques must be anticipated.

In contrast to this, usage pattern information is broader in scope. It would, ideally, include such data as amount used in various applications, frequency of use, typical room volumes and air exchange rates where the products are used, the frequency with which consumers increase ventilation by opening windows and/or doors, and the size and types of objects to which the products are applied.

There are several ways to acquire information about application techniques and usage patterns. Manufacturing trade associations for the product type may share their market research data about techniques for application and how consumers actually use the product. User surveys are another, probably better, method of acquiring information. These range from structured surveys with questionnaires to informally questioning colleagues and acquaintances about their personal use of specific products. However, one of the best and most readily available sources of information is probably manufacturers' information sheets and the instructions printed on product containers since these are the source of information most often used by consumers.

For the methylene chloride study two major product types with very different forms were investigated: paint removers, which are primarily semi-pastes, and aerosol finishes. Information about application techniques and product usage was acquired through informal surveys (including personal experience on the part of the researchers and program managers) and manufacturers' information sheets and instructions on product containers.

ESTABLISHING STUDY GOALS

This task allows the greatest latitude and, accordingly, the greatest creativity for researchers and program managers, since a wide range of choices is available to shape the direction and goals of the study. Initially, it must be decided if the study should be biased toward a 'worst case' use or toward a more typical product use or both. A worst case scenario may be appropriate when acute health effects are of interest and are likely to occur within the population with some

frequency. A typical use scenari may be more appropriate f r chronic health effects or to estimate the typical exposure of larger populations.

Very often, source characterization is a primary goal and this characterization can be done with respect to time (in which case the form of the product and the method of application are important); with respect to temperature (where the form of the product may be important); with respect to the ventilation rate (or perhaps with respect to local air flows near the product and user); and/or with respect to other environmental parameters.

Another decision must be made regarding measuring area concentrations vs personal concentrations or both. The person using the product can receive an exposure far different than would be predicted from an area sample taken in the same room. The concentration gradients in rooms where consumer products are being used may be large and, therefore, may have a strong effect on personal exposures. This may lessen the importance of area concentrations in the absence of a model correlating personal exposure to area concentrations and increase the importance of personal sampling.

Of course, as discussed previously, the application technique can affect emissions from a consumer product. Therefore, it must be decided if the effect of this source of variability is to be measured.

Finally, the goals must be prioritized. Because of limited resources, the scope of a study must usually be restricted in some way. The actual decisions as to which goals will be pursued need not be made at this time, but they should be prioritized.

In the methylene choride study, for example, the focus of the study was to be typical use, not worst case. It was further decided that source characterization with respect to time and ventilation was most important and that the effects of temperature and humidity would be studied later, if at all. While characterizing the emission of CH_2Cl_2 would receive the most attention, the emission of other major solvents such as toluene would also be monitored. The average area concentration and personal exposure were judged, at this point, to be equally important and the study would attempt to measure both. The ventilation rates to be used were both low, 0.5 air changes per hour, and high, 3.0 air changes per hour. These ventilation rates may correspond to the situation when a consumer takes no action to increase ventilation while using the product indoors and the situation when windows are opened. (A consumer could also, in some situations, take the object to be painted or stripped outside. However it ceases to be an indoor air quality problem under these conditions and, more importantly, is not an option in cases when the object is a permanent fixture, e.g. a floor, kitchen cabinets or wall panels.) While the panels to be paint stripped in this study were to be primed and painted several months before the experiments with two coats of enamel paint, which tends to be difficult to remove, they were to be modest in size (compared to wall panels, kitchen cabinets or a floor) and they were to be relatively smooth (compared to lathe-turned legs or a carved piece of furniture) and therefore easily stripped. Overall, the choices made tended to correspond more to typical, not worst case, use of the product.

ASSESSING INSTRUMENTAL AND FACILITY CAPABILITIES

Based upon the prioritized list of goals, the researchers must assess the resources available to conduct the study, in terms of both instruments and a chamber or other specialized facility. This is a straightforward task.

For the methylene chloride study, a continuous i.r. analyzer was to be used to monitor both the personal and area concentrations on an alternate basis by means of sample line switching. Sampling with charcoal tubes was originally considered for personal sampling, but was rejected for this phase of the study because of insufficient precision and time resolution. A gas chromatograph (GC) was also available. A chamber designed for studies of organic emissions was available. Its ventilation system was adequate in terms of air flow and size, but at the time, it lacked temperature and humidity control. However, since strict requirements were not established for these parameters, control could be accomplished simply by controlling the temperature and humidity of the laboratory housing the chamber.

Data acquisition systems and a host computer were available for logging of analog and digital input signals from the i.r. analyzer, the GC and the environmental instrumentation, but some software had to be written.

WRITING THE PROTOCOL

In writing the protocol, all of the information collected regarding product types, application techniques, usage patterns, study goals and the analytical instrument and facility specifications are considered and brought together to construct a unified plan. The detailed experimental procedure is written, incorporating the specific instruments and the chamber capabilities. Calibration procedures are established in detail. Consideration should also be given to data acceptance standards. However, in research, as opposed to monitoring, it is inappropriate to set rigid standards for all types of data. Standards can be set for the limits of acceptable data for enviromental parameters, for instrumental drift and for the precision of calibration data. However, setting standards for other data can be difficult due to lack of knowledge about the behavior of the source and the effect of activity on the part of the person using the product.

Audit procedures should be set and written as part of the protocol. The audit can take two forms: internal where checks of instruments, procedures and data are

conducted and recorded by the researchers themselves according to a formal plan: and external where an outside agency or individuals check the instruments, procedures and data.

A detailed checklist of specific actions to be executed before, during and after an experiment should be constructed from the key elements of the protocol. Writing the checklist at this time serves to insure that the protocol is practical and reasonable. However, the checklist should not be viewed as a static endpoint but, rather, as the first draft of a document that will evolve as the researchers gain experience.

In the current example, the protocol for the methylene chloride study was written by the LBL staff and sent to the CPSC for review. The protocol specified the instruments and the calibration procedures to be used. It established internal audit procedures and named a quality control officer. It described the sampling system, the ventilation rates, the environmental parameters, the substrate to be finished or stripped and incorporated manufacturers' instructions for product use. Incorporating the instructions for the use of paint removers, in particular, required careful consideration. Paint remover was to be applied sequentially to small sections (0.37 m^2) of a 1.5-m^2 panel. Paint remover would set for a minimum of 10 min prior to scraping. Tools, remover containers and paint scrapings would all be weighted so that the weight data could be used as a check of the emission rates developed through the use of a mass-balance, ventilation model. CPSC chose to obtain outside review of the protocol. Comments and suggestions from CPSC and the outside reviewers were incorporated into the protocol by LBL.

PRETESTING

The pretest period is one of the most interesting periods of a study because the learning curve is so steep. The products are selected for screening and subjected to bulk chemical analysis, generally by GC and/or GC/mass spectrometry. Based upon the results of the bulk analysis and market considerations, the specific products to be studied are selected. The performance of the analytical instruments is evaluated using the calibration procedures. During this period, more than one calibration system may be used as an overall check of one system against the other. Sampling systems are fabricated, if necessary, and/or tested. Data acquisition software is debugged, and the acquisition system is tested.

If at all possible, trial experiments should be conducted and concentrations measured. During these trial experiments, the product application technique is evaluated and standardized. This is especially important if more than one person will be applying the product during the experiments and measurement of the effect in variability of application technique is not a study goal. The overall experimental procedures are evaluated, as well as the adequacy of the sampling system and the analytical instruments employed. These trial experiments also provide data for testing data reduction and analysis schemes. Often it is discovered that the protocol and checklist should be modified based upon the knowledge and experience gained in the pretest period.

In the example study, the bulk chemical analysis of paint removers confirmed a high percentage of CH_2Cl_2 and lesser amounts of toluene and aliphatic alcohols. The bulk analysis of selected aerosol paints and other aerosol finishes showed lesser percentages of CH_2Cl_2 but larger amounts of toluene and, in some cases, minor amounts of other solvents.

A sampling system was fabricated that switched alternately between a personal sample taken near the breathing zone of the person using the product and an average area sample consisting of the sum of 15 sampling locations in the chamber. These 15 locations are located throughout the chamber at three different heights.

The i.r. analyzer was calibrated for CH_2Cl_2 using both the manufacturer's closed-loop injection system and a mass-flow controlled dynamic gas dilution system with certified gas standard mixtures. Agreement between techniques was excellent, but the dynamic technique was chosen for use in the experiments because of ease and speed of use.

Application techniques for paint removers were standardized. Preliminary experiments demonstrated that CH_2Cl_2 emissions could be well characterized and that even short-term variations could probably be characterized. However, personal exposure concentrations appeared to be highly variable and could not be tracked adequately when sampling was alternated between personal and average area locations. Therefore, it was decided to obtain a second i.r. analyzer so that both personal and average area concentrations could be measured continuously. The protocol was modified to reflect this change.

The preliminary experiments also graphically demonstrated that the products must be used realistically to obtain valid emission rates. If paint remover was applied to a panel and allowed to set undisturbed, the remover didn't appear to evaporate appreciably and emissions were relatively low. However, if the paint remover was agitated by scraping, the remover would evaporate to near dryness within 5–10 min and emissions were large. Similarly, vertical stratification of concentrations was also more evident if paint remover was undisturbed.

CONDUCTING THE STUDY

Once the necessary preparations have been made, completing this task is relatively straightforward and, again, is similar to most indoor air pollutant source characterization studies. If at all possible, the first experiments should be replicate experiments to quantify the reproducibility of the experimental procedure. For these experiments, the same person should use the

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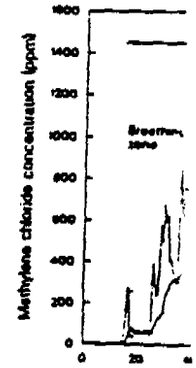

Fig. 1. Temporal pr
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product and every attempt should be made to duplicate events even if a later goal is to determine the effect of variations in application technique. If the data are sufficient, statistical tests can be employed to determine whether the reproductability is adequate to detect changes due to experimental variables.

The remaining experiments are conducted after the replicate experiments are completed and the data are reduced. The average concentrations obtained in the experiments are incorporated into a mass-balance ventilation model such as has been used in combustion appliance studies (Traynor *et al.*, 1982). Because of the temporal variation of emissions from some consumer products, it may be necessary to use a version of this model than can address these variations (Traynor *et al.*, 1985). The model may require some modifications to adequately treat a unique data set.

The data from personal sampling can be used to calculate personal exposures and these can be compared to exposures estimated from average area concentrations. Statistical analyses of the data can be conducted to assess results with respect to selected parameters, e.g. by product type, by product brand, by ventilation rate or by exposure (personal exposure vs exposure derived from the average area concentration).

The chamber experiments for the methylene chloride study have been completed (Girman and Hodgson, 1985). The emission rates for the two product types varied, of course, but the differences in the rates were consistent with the amount of CH_2Cl_2 used (as determined by bulk analysis) and the duration of product application and use. Temporal profiles of the average chamber concentrations and the breathing-zone concentrations of CH_2Cl_2 produced by the use of a paint remover at a high ventilation rate in the 20-m^3 chamber are shown in Fig. 1. The concentration fluctuations produced by the sequential use of the paint remover are clearly evident. Temporal profiles of CH_2Cl_2 concentrations were calculated from source strengths, the ventilation rates and the chamber volume using single-equation, mass-balance models. For paint removers, two types of source strengths are used: the first, the time-averaged source strength, assumed that the product was used uniformly over time for the duration of the entire work period; the second, the time-dependent source strength, accounted for the sequential nature of product application. As illustrated in Fig. 2 which contains the modeled profiles from the same experiment illustrated in Fig. 1, these theoretical concentrations were in good agreement with measured concentrations. Exposure models based upon the concentration models were also developed and then evaluated by comparing theoretical and measured exposures for the experiments. When measured personal exposures were compared to chamber concentrations of CH_2Cl_2 integrated over the work periods, agreement was good at the low ventilation rate but averaged 21% higher at the high ventilation rate. For the experiment illustrated in Figs 1 and 2, the exposures were 1120 ppm·h (personal exposure), 921 ppm·h (exposure based upon average chamber concentrations) and 1180 ppm·h (theoretical exposure). For the same paint remover used in an experiment at the low ventilation rate, the exposures were 2400 ppm·h, 2350 ppm·h and 2530 ppm·h, respectively. The exposure models appeared to have sufficient accuracy and precision for use in assessment of health risk from the use of consumer products containing CH_2Cl_2.

POST-STUDY CONSIDERATIONS

If funding is available, the research may be extended to explore issues ignored because of limited resources; to explore new issues discovered in the study; to study

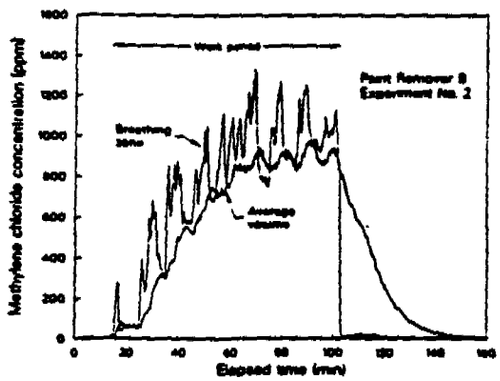

Fig. 1. Temporal profiles of chamber and breathing-zone concentrations of CH_2Cl_2 during an experiment with a paint remover in a 20-m^3 chamber at a ventilation rate of 3.23 h^{-1}. Duration of work period is shown above curves. At end of work period, breathing-zone sampling line was switched to laboratory air external to chamber.

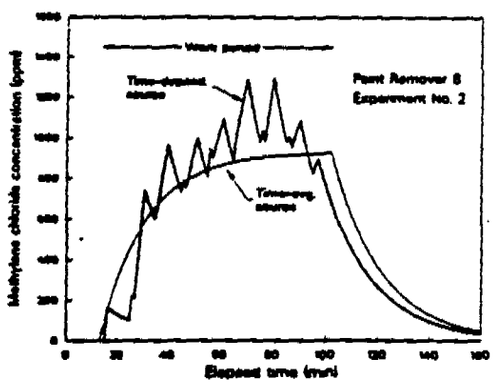

Fig. 2. Temporal profiles of theoretical chamber concentrations of CH_2Cl_2 for an experiment with a paint remover in a 20-m^3 chamber at a ventilation rate of 3.23 h^{-1}. Concentrations were calculated using both time-dependent and time-averaged source strengths. Duration of work period is shown above curves.

similar products; or perhaps most importantly, to validate the model developed by conducting a field study.

For the methylene chloride study, the remaining issue is the validation of the exposure models developed in a field study which will examine the effects of variations in ventilation patterns and rates, in volumes and in product use patterns.

PERSONAL PROTECTION

Personal protection for the researchers deserves special mention. Unlike studies of combustion appliances and building materials where it is not necessary for researchers to have prolonged exposures to emissions, using consumer products can often result in extended periods of exposure. Researchers should be protected even if some accuracy is sacrificed.

In the methylene chloride study, a pressure-demand breathing apparatus supplied by a cylinder of air [illegible] worn by the person using the product during the experiments. Because the mask [illegible] the face of the wearer, it may have affected the personal sample. Nonetheless, personal protection took precedence. Disposable overalls were worn during the experiment by the person using the product to prevent dermal contact. Viton gloves were also worn during the use of paint removers.

[illegible]

Consideration [illegible] consumer products have been discussed with reference to eight tasks that should be accomplished when conducting such a study: (1) defining the potential health hazard; (2) acquiring application technique and usage pattern information; (3) establishing study goals; (4) assessing instrumental and facility capabilities; (5) writing protocol; (6) pretesting; (7) conducting the study; and (8) post-study considerations. These considerations are discussed using examples provided by an ongoing study of methylene chloride emissions from paint removers and aerosol finishes. Emphasis is given to those considerations that are unique to studies of consumer products. These considerations include the need to use the products realistically to obtain valid data, the difference between personal exposure and exposure based upon the average concentration and the factors that affect this difference, and the need for personal protection of the researchers.

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