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To: Technical Task Group on Vinyl Chloride Research
Technical Panel on Fluorocarbon Research

Subject: EPA Paper Exposure to Halogenated Hydrocarbons in the Indoor Environment

Gentlemen:

Distributed herewith are copies of a draft of the subject paper, which was presented at a recent conference on public health implications of components of plastics manufacture.

Sincerely,

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KDJ/mb

Enclosure

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Exposure to Halogenated Hydrocarbons in the Indoor Environment

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SECOND DRAFT

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INTRODUCTION

The indoor environment has often been ignored as a significant source of exposure to air pollution despite the fact that levels of pollutants in the indoor air can frequently exceed those concentrations which commonly occur in the outdoor environment. The indoor environment includes that present in a number of circumstances including occupational situations, public buildings, hospitals and the home. The discussion to follow stresses, but is not limited to, consideration of exposures in the home situation. The present paper endeavors to review those instances where indoor air pollutants and particularly halogenated hydrocarbons, may reach concentrations of potential public health significance. In the past there has been considerable attention given to the potential health hazards of chlorinated aromatic compounds such as PCB's and pesticides. To date, however, there has been relatively little attention given to the halogenated aliphatic compounds which are the primary focus of the present paper.

Behavior of Pollutants in the Indoor Environment

Assuming that a given pollutant is chemically stable in the indoor air and that it is present as a result of being generated from an indoor source than its concentration as a function of time may be described by the following equation (1): $C = C_o e^{(-K \frac{Q}{V} t)}$ (1)

where:

C = concentration at time, t
C_o = initial concentration at time, 0.
Q = air supply rate in ft^3/min
V = room volume, ft^3
t = time, minutes
k = a constant to adjust for imperfect mixing

This equation assumes that after time zero the source is turned off and that the air supply into the room is not contaminated by the pollutant in question. If a continuous source for a given pollutant is also present then the problem becomes more complex. Even a small continuous source can greatly affect indoor pollution levels which under certain circumstances may build up considerably (2,3). In the discussion to follow only the simplified situation without a constant source of pollution will be considered, recognizing that this may underestimate exposure in those instances when a constant emission source is present.

In practice, under best mixing conditions, the highest achievable value for k, the mixing constant (shown in equation (1)) is 1/2. Under these circumstances equation (1) becomes: $C = C_o e^{\left(-\frac{Q}{2V}t\right)}$ (2)

It is apparent from this equation that an important parameter affecting concentration is the turnover rate of air in the room per minute, Q/V. If there is no exchange of fresh air then the equation reduces to a condition in which the concentration at any time is equal to the concentration at time zero. The more frequent is the exchange for fresh air, the more rapid is the concentration of the pollutant decreased.

Let us examine, for example, the decrease in concentration with time predicted by equation (2) under various rates of fresh air exchange. These calculations are shown in Table 1 and in Figure 1. The strong dependence of outdoor concentration upon air exchange rate is illustrated in this Table and Figure. For example, with but one air exchange per hour, at 30 minutes the concentration is nearly 80% of its initial value whereas at 6

TABLE 1

Dependence of Indoor Concentration Upon Air Exchange Rates in a Room

Time (minutes)	Air Exchange Rate: Once Per Hour (Q/v=1/60)	Twice Per Hour (Q/v=2/60)	Six Times Per Hour (Q/v = 6/60)
0	Co	Co	Co
15	0.88 Co	0.78 Co	0.47 Co
30	0.78 Co	0.61 Co	0.22 Co
45	0.69 Co	0.47 Co	0.11 Co
60	0.61 Co	0.37 Co	0.05 Co
90	0.47 Co	0.22 Co	0.01 Co
120	0.37 Co	0.14 Co	< 0.01 Co
150	0.29 Co	0.08 Co	-
180	0.22 Co	0.05 Co	-

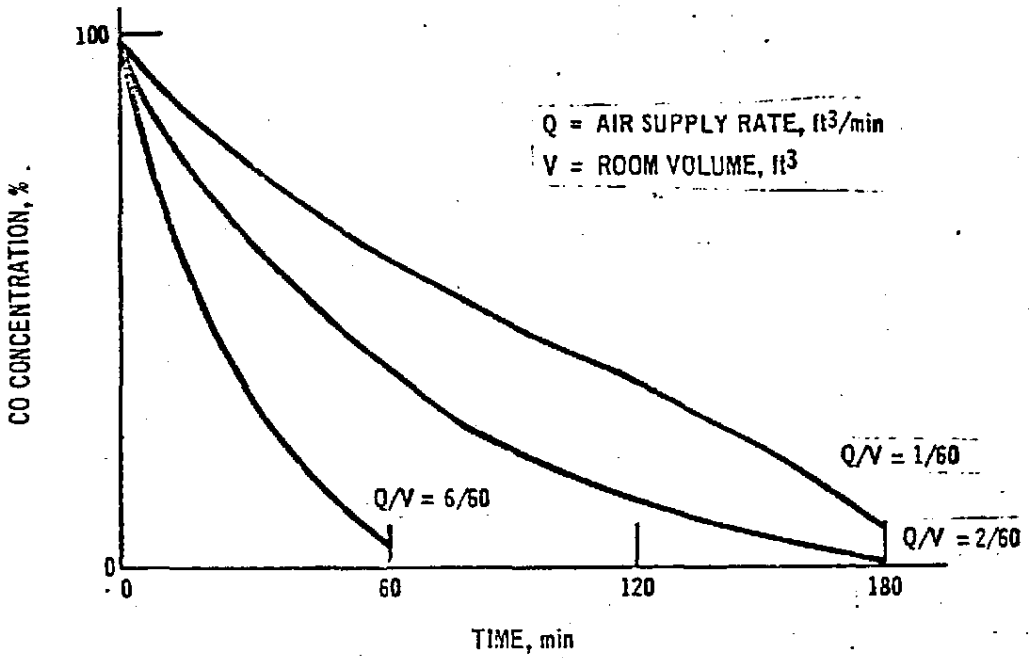

Figure 1. Indoor concentration as a function of air exchange rate and time.

air exchanges per hour the concentration has dropped to about 20% of the initial value. After 2 hours, at one air exchange per hour the concentration is still better than one-third the initial concentration (C_o), but at 6 air exchanges per hour the concentration has fallen below 1% of its initial value.

Given a functional dependence of concentration with time as shown in equation (2) let us examine some real world measurements of indoor pollution levels (4) to see if they obey this type of relationship. Shown in Table 2 are measurements of propellant concentrations following a 60 second release of a hairspray in a 29,000 liter room. The propellants measured are vinyl chloride and Freon-12. The rate of decay of vinyl chloride parellels that of Freon 12 (shown in Figure 2) suggesting that the vinyl chloride was chemically inert over the time frame examined and under the existing experimental conditions. The effect of air exchange upon concentration in this experiment is similar to that predicted by equation (2) assuming an air exchange rate of 6 times each hour, and considering that the initial high measurements in the breathing zone at time 0 do not reflect complete mixing of the propellants in the room.

Shown in Table 3 are the results of a 30-second aerosol insect spray released in a home laundry room of 21,000 liter volume. The spray was released along the baseboards of the room containing a washer and dryer, sink, clothes and other miscellanceous items. The room has two doors, one leading to a hallway in the house proper and the other leading to the outside. During the experiment, the door to the hall was closed and samples were taken by entering the door to the outside of the house. Sample No. 1, collected one minute after the aerosol was released, showed a concentration of 380 ppm vinyl chloride. Sample No. 4, taken 150

TABLE 2

Sixty-Second Release of Hairspray in 29,300 Liter Room

Sample	Time	Column I		Column II	
		Vinyl Chloride	Freon-12	Vinyl Chloride	Freon-12
No. 1	Collected at Breathing Zone During Spray	122.7 ppm	62.15ppm	124.4ppm	58.30ppm
No. 2	10 minutes	25.8	11.0	25.1	11.0
No. 3	30 minutes	5.56	2.53	5.2	2.26
No. 4	60 minutes	0.13	0.06	0.12	0.05

TABLE 3

Thirty-Second Release of Insect Spray in 21,400 Liter Room

Sample	Time	Column I		Column II	
		Vinyl Chloride	Freon-12	Vinyl Chloride	Freon-12
1	Collected one minute after spray	380.1ppm	466.4ppm	383.6ppm	457.6ppm
2	30 minutes later	52.1	54.45	48.7	55.65
3	60 minutes later	24.6	26.40	22.5	25.85
4	150 minutes later	10.3	11.55	9.3	12.10
5	Collected in adjacent hall at 151 minutes	0.83	0.94	0.65	0.83

minutes after aerosol release, had a concentration of 10.3 ppm vinyl chloride. A sample taken in the hallway adjacent to the laundry room at 151 minutes contained 0.83 ppm vinyl chloride, indicating spread of vinyl chloride to other areas of the house despite the closed door. Assuming that the initial measurement was not made at a time when there was equal mixing throughout the room, the rate of decay of concentration with time was similar to that which would be expected at an air turnover of twice per hour according to equation (2).

In contrast to the experiment with the hairspray, in which Freon-12 concentrations were about one-half those of the vinyl chloride, Freon-12 levels in the insect spray experiment were 10-20% greater than the vinyl chloride concentrations. Given the frequent sequential use of aerosol products containing Freons such as hairsprays and deoderants, peak exposures to Freon mixtures in excess of 1,000 ppm might result following combined use of such aerosol products in the indoor environment. This raises the possibility that peak exposures to a mixture of Freons following use of aerosol products could exceed the peak exposure recommended for such a mixture in the work environment. The current TLV for Freon-12 is, for example, 1,000 ppm, suggesting that excursions in excess of 1,250 ppm should not be permitted for occupationally exposed individuals(5). Naturally, exposures of the general population should be kept well below those permitted for the occupational population since the general population frequently includes potentially high risk groups not usually found in the work forces.

Shown in Table 4 is a breakdown of propellant use in aerosol products as a function of product category. It is apparent from this table that a

Table 4. USE OF PROPELLANTS IN AEROSOL PRODUCTS BY PRODUCT CATEGORY

Product Classification	Propellants Used	Frequency Us
Inhalants Containing Bronchodilator drugs (11 products)	Freon 11 (Trichlorofluoro methane)	5/11
	Freon 12 (Dichlorofluoromethane)	11/11
	Freon 114 (Dichlorotetrafluoro ethane)	9/11
Mouth Products (6 products)	Freon 12	5/6
	Freon 114	5/6
	Freon 1426 (Monochlorodifluoroethane)	1/6
Vaporizers (2 products)	Freon 11	2/2
	Freon 12	2/2
	Trichloroethane	1/2
Hair products (62 products)	Freon 11	53/62
	Freon 12	54/62
	Freon 114	4/62
	Freon 152a (Difluoroethane)	5/62
	Methylene chloride	8/62
	Vinyl chloride	1/62
	Propane	3/62
	Isobutane	32/62
Women's personal hygiene products (22 products)	Freon 11	5/22
	Freon 12	20/22
	Freon 114	6/22
	Isobutane	2/22
Deoderants and anti-perspirants (38 products)	Freon 11	13/38
	Freon 12	37/38
	Freon 114	8/38
	Freon 142b	1/38
	Propane	1/38
	Isobutane	1/38
Foot products (9 products)	Freon 11	8/9
	Freon 12	9/9
	Freon 114	1/9
Miscellaneous products for personal use (18 products)	Freon 11	7/18
	Freon 12	15/18
	Freon 114	3/18
	Isobutane	2/18
	Propane	2/18

number of halogenated hydrocarbons are employed as aerosol propellants. Depending upon the frequency and patterns of use of aerosol products among the population, a substantial number of individuals may be repeatedly exposed to these compounds. By far the most commonly used propellants as illustrated in Table 5 are Freon-11 and Freon-12.

Recently published studies of instantaneous Freon-11 and Freon-12 concentrations in the indoor environment taken at random confirm the presence of measurable quantities of these pollutants in the indoor air (6). Levels of Freon-12 in homes, for example, exceeded 500 ppb and were generally much greater than simultaneous outdoor concentrations which usually measured 1 ppb or less. In one study a measurement of Freon 12 in a beauty shop indicated a concentration of 370 ppb (6). A study of Freon-12 levels in a beauty shop conducted by the Environmental Protection Agency found a concentration of 3,000 ppb or 3 ppm averaged over a 15 minute period. In this experiment no vinyl chloride was identified.

Apart from aerosol propellants, another important source of exposure to halogenated hydrocarbons in the indoor environment may be from the active ingredients contained in aerosol products. For example, aerosol spot removers used for clothing, carpets, upholstery and wallpaper may frequently contain perchloroethylene as the active ingredient. Though the concentration of perchloroethylene in such products often is not identified on the label, it is conceivable that the proportion of active ingredients could be similar to the quantity of propellant in these products. Under these circumstances, peak concentrations of perchloroethylene well in excess of 100 ppm might occur during and immediately following spraying as judged from the data in

Table 5. OVERALL USE OF PROPELLANTS IN 168 AEROSOL PRODUCTS

Propellant	Frequency of Use	Percent
Freon 11	93/168	55.4
Freon 12	153/168	91.1
Freon 114	36/168	21.4
Freon 1426	2/168	1.2
Freon 152 a	5/168	3.0
Isobutane	37/168	22.0
Methylene chloride	8/168	4.8
Propane	6/168	3.6
Trichloroethane	1/168	0.6
Vinyl chloride	1/168	0.6