ENVIRONMENTAL RESEARCH 21, 432–437 (1980)

Tetrachloroethylene in Exhaled Air of Residents Near Dry-Cleaning Shops

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Received July 12, 1979

Alveolar air was analyzed for tetrachloroethylene (PER) in 136 residents living near 12 dry-cleaning shops. The PER concentrations found were distinctly higher in subjects living closer to the shops. The geometrical mean concentration was 5 mg/m^3 in subjects living in the flats (maximum five floors) above the dry-cleaning shops, 1 mg/m^3 in houses adjacent to the shops, 0.2 mg/m^3 in the next houses, and <0.1 mg/m^3 in those living across the street opposite the shops. Eighteen workers in the shops had a mean concentration of 73 mg/m^3. The concentrations appeared to be influenced by the direction of the prevailing wind. It is concluded that a certain part of the general population is exposed to PER, and that biological monitoring of exhaled air seems feasable to measure it.

INTRODUCTION

Tetrachloroethylene (perchloroethylene, PER) is extensively used as a cleaning solvent for textile in dry-cleaning shops. In The Netherlands the emission of PER into the air by the smaller shops is estimated to be several thousands of kilograms per year each (Bakkers, 1977). Some of these shops are situated in densely populated town districts. Many individuals live next to or above those dry-cleaning shops. So the question arose whether the PER emitted might reach these subjects.

An explorative study was undertaken to estimate the exposure by measuring the PER concentration in exhaled air. This simple procedure probably yields better information on recent exposure than the PER concentration in indoor air. Volunteer studies by Fernandez *et al.* (1976) and Monster *et al.* (1979) showed that after a few hours of exposure to a constant PER concentration, the concentration in exhaled air is about 0.4× the concentration in inhaled air, and that about a quarter of an hour after such exposure the alveolar concentration is half its end-exposure value.

METHODS

Twelve dry-cleaning shops in the districts of Amsterdam built during the late 19th century were selected on topographical criteria, i.e., situated more than 100 m from each other and not on a streetcorner. The population density is high; the houses have three to five flats, are built immediately adjacent to each other, and have a rather poor quality. The dry-cleaning shops are situated amidst other shops at ground-floor level and the majority have a ventilation system at the backside from the cleaning machine up to the roof of the house. At the backside often windows and doors are open as well, to ventilate the working area.

The shops were studied, each on a Thursday or a Friday, during 6 weeks in October and November 1978. A few days before a dry-cleaning shop was visited, a letter explaining the purpose of the study was sent to the residents living above the

0013-9351/80/020432-06$02.00/0

shop (group III), one house or two houses adjacent to it (respectively, groups II and I) and those living across the street opposite the shop (Control group). The subjects engaged in the dry-cleaning operations within the shops were also invited to participate. All subjects were visited between 10.30 and 12.30 AM in the flat or shop where they stayed. Before taking the samples, a short questionnaire was completed.

For sampling exhaled air, a glass tube (length 20 cm, volume 65 cm^3) was used, of which each of both ends (8 mm ϕ) could be closed by means of a perforated screw cap provided with a rubber disk and a Teflon membrane, the latter to prevent absorption of PER. This construction permits small volumes of air (25 μl) to be taken by syringe for analysis without opening the tube. Alveolar air was obtained as follows: The subject inspires deeply, holds his breath for at least 5 sec, and expires through the sampling tube which is open at both ends. Just before expiration is finished, the investigator closes the tube with both caps. In this way a portion of the expiratory reserve volume is retained in the tube. From each participant two samples were taken. Breath holding for 5 sec had been checked to be sufficient for at least 90% of the alveolar equilibration concentration.

The PER concentration in the sampling tube was determined by gas chromatography as described by Monster and Boersma (1975). The lower limit of detection (three times base level noise) was 0.1 mg/m^3. The values of both samples of a subject were averaged. In case the lower value was less than half the higher one, the latter was used, because it is assumed that such a difference can only be caused by leakage from a tube (this occurred eight times on a total of 157 subjects). For the other duplicates, the coefficient of variation was 8% on the average at concentrations above 1 mg/m^3.

For each day that measurements were performed, data on direction and velocity of the wind and on the fraction of nonblue sky were obtained.

RESULTS

Participation

A number of 139 residents was studied as well as 18 workers within the shops. Five residents and the workers of one shop refused to participate. Among the residents were 65 males and 74 females with a fairly equal age distribution; the youngest was 17 years. Of the participants 98% had been at home that morning at least from 8.30 AM on.

Of the 12 flats immediately above the (12) shops, in 5 flats one or more subjects could be studied, i.e., 42%. Of the other flats in the same house this percentage was 63%. In the houses immediately adjacent to the shops the participation rate with regard to the flats was 55%; in the second next houses only a few subjects were visited, due to the limited number of sampling tubes available. In most cases the number of control subjects was three. The data were excluded of three subjects who mentioned to have had contact with PER during the preceding days from other sources than the dry-cleaning shop.

PER in Alveolar Air

The concentrations of the different groups are shown in Fig. 1 as cumulative frequency distributions. The concentrations were distinctly higher in the groups

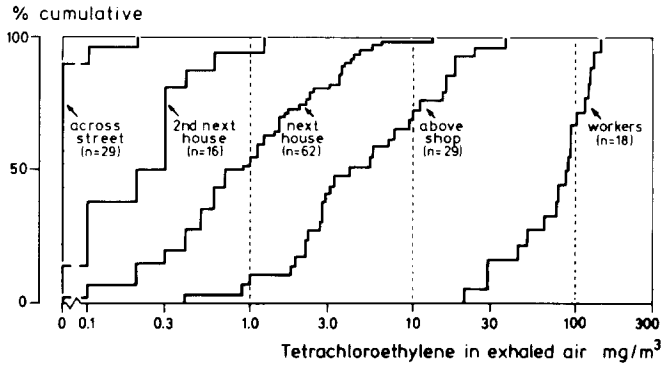

FIG. 1. Distribution of tetrachloroethylene in exhaled air of residents living near dry-cleaning shops and workers within those shops. The number of subjects in each group is indicated between parentheses.

living more close to the source. The geometrical mean values were: control group < 0.1 mg/m^3; Group I, 0.22 mg/m^3; Group II, 1.0 mg/m^3; Group III, 4.9 mg/m^3; and workers 73 mg/m^3.

Around the separate shops a gradient could be shown as well. For seven shops the concentrations of all subjects of Group III were $\geq$ Group II $\geq$ Group I $\geq$ control group, as far as these groups were present. Around the other five shops a similar gradient, however less distinct, was present. Among those living above a shop, no relation was found between the exhaled concentration and the floor where one lived, although in subjects living on the second floor the highest three concentrations among all residents were found.

Symptoms

Smell and/or annoyance during the recent week and ascribed to the vapors was mentioned by 15 subjects, 7 around one and 8 around six other shops. All these subjects had more than 1 mg/m^3 PER in exhaled air. Smell was noticed by 12 subjects, i.e., 15% of those with PER between 1 and 10 mg/m^3 and 50% (5 out of 10) of those with PER above 10 mg/m^3. Annoyance was mentioned by 9 subjects; in most cases it consisted in irritation of nose, throat, or eyes. No relation was found between symptoms and concentration, probably because the willingness to mention complaints also depends on the relation between the subject and the manager of the shop. Of the residents 81%, including all who mentioned symptoms, said that the situation during the recent week with respect to the dry-cleaning shop was similar as during the preceding months. This suggests that the results obtained may be reasonably representative for that period.

Differences between Shops

No direct comparison of the 12 dry-cleaning shops could be made, because there were appreciable differences in the distribution of subjects with respect to the house or flat where one lived. So a normalization procedure was performed to bring the subjects of Group I (mean concentration 0.22 mg/m^3) and of Group II (1.0 mg/m^3) to the level of Group III (4.9 mg/m^3), by multiplying the concentrations of the members of Group I by 22 (4.9 mg/m^3 : 0.22 mg/m^3) and those of Group II by 5

(4.9 mg/m^3: 1.0 mg/m^3). The normalized concentrations are shown in Fig. 2. Then for each shop the geometrical mean of these normalized concentrations was calculated. The results varied from 0.5 mg/m^3 for the "cleanest," to 12.6 mg/m^3 for the "dirtiest" dry-cleaning shop. No relation was found between these mean values and the concentration in exhaled air of the workers within the corresponding shops.

Influence of the Wind

During the study period the atmospheric conditions were neutral. The direction of the prevailing wind perpendicular to the front facade of each dry-cleaning shop was determined. When the wind goes from, i.e., over the front facade to the back facade, this vector is called negative (−), in the opposite direction positive (+). In the first case an airstream going upward along the backside of the house is induced, and one may expect that PER escaping at ground-floor level from the shop will be transported upward, enters the flats above, and increases the exposure of the inhabitants. In case of a positive vector the escaping PER will be transported from the house and be diluted. Along the front facade such transports will be of little importance: The front door of the shop is opened only for short times, the traffic may disturb the vertical airflow, and the windows of the subjects living above are almost always closed because of the traffic.

In Fig. 2 the direction of the vector is indicated by a + or − sign. Conforming the

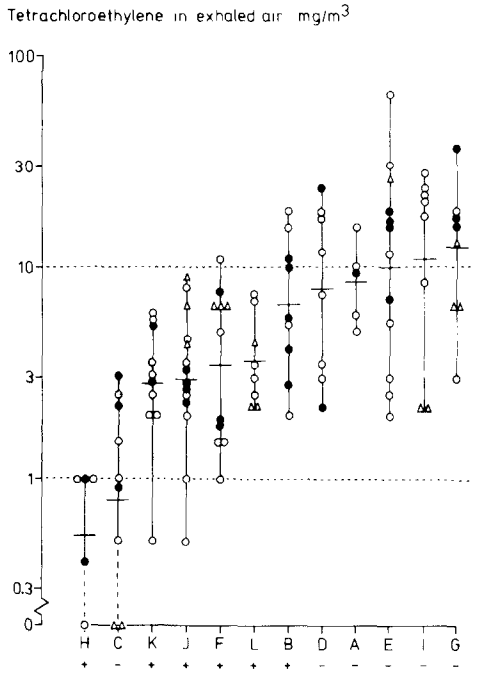

FIG. 2. Tetrachloroethylene in exhaled air of residents near separate dry-cleaning shops (A–L) arranged according to geometrical average (−). The concentrations of Group I (△) and Group II (○) were "weighted" as Group III (●); —— = range. The direction of the wind with respect to the facade is indicated by + or −.

hypothesis a negative vector occurs more often at the shops with the highest concentrations (Wilcoxon two-sample test, $P < 0.05$). This suggests that the differences found between the dry-cleaning shops are partly caused by the direction of the prevailing wind.

DISCUSSION

In this explorative study each subject was measured only once. For individual participants other concentrations are to be expected if the measurement would have been repeated. Still the results of the population studied permit some conclusions. The emission of PER by the 12 dry-cleaning shops resulted in an absorption of this substance by the neighbors, decreasing with the distance to the shop. Moreover, this study shows that biological monitoring of exhaled air can be used to estimate exposure not only in industry, but also in the general population.

In this study ill effects due to PER were not investigated except smell and annoyance. Of the subjects mentioning odors from the dry-cleaning shop, four had a concentration of PER in exhaled air less than 3 mg/m^3, corresponding to about 7 mg/m^3 in inhaled air after a few hours of exposure. Leonardos (1969) mentions 34 mg/m^3 as the odor threshold for PER, in his study the lowest concentration at which the smell of a vapour was recognized by all four trained panel members. The sensation of smell of our subjects may be due to a lower odor threshold, to higher concentrations during the preceding days, or to other substances in the vapors of the dry-cleaning shop.

Observations in volunteers showed that moderate PER concentrations (700–1500 mg/m^3) can cause irritation of the mucous membranes of eyes and upper respiratory tract, headache, dizziness, and slight unconsciousness. In workers also tremor and impairment of liver function was found, however, the influence of alcohol consumption could not be ruled out. Practically no data are available of the effects at lower concentrations, but it is assumed that after long-term exposure to 300 mg/m^3, equivalent to about 120 mg/m^3 in exhaled air, only very few healthy subjects will experience some of these symptoms and only to a little extent. However in the general population there may be more vulnerable subjects, probably those with already impaired functioning of the liver or nervous system; for them a still lower exposure level is required.

Long-term administration of PER by gavage resulted in an increased incidence of liver cancer in mouse sensitive to this type of cancer (Weisburger, 1977). However this was not confirmed in an inhalation study with rats (Rampy, 1977). Modified Ames tests (Greim *et al.*, 1975; Bartsch *et al.*, 1979) did not show PER or its metabolites to be mutagenic. Preliminary results of a study after cancer mortality in dry-cleaning workers (Blair *et al.*, 1978) raises the possibility that PER may be carcinogenic in man. The results of our study show that near dry-cleaning shops, subjects are exposed to PER, so we conclude that at least some of them may have certain risk of acquiring the potential adverse effects of this substance.

REFERENCES

Bakkers, T. (1977). "Veilig Werken met Perchlooretheen (Working Safely with Perchloroethylene)." Institute for Cleaning Techniques T.N.O., Delft.

Bartsch, H., Malareille, C., Barbin, A., and Planche, G. (1979). Mutagenic and alkylating metabolites

of haloethylenes, chlorobutadienes and dichlorobutenes produced by rodent or human liver tissues. *Arch. Toxicol.* **41**, 249–277.

Blair, A., Decoufle, P., and Granman, D. (1978). "Mortality among Laundry and Dry Cleaning Workers." Paper presented at the 11th Annual Meeting of the Society for Epidemiologic Research, June 14–16, 1978.

Fernandez, J., Guberan, E., and Caperos, J. (1976). Experimental human exposures to tetrachloroethylene vapor and elimination in breath after inhalation. *Amer. Ind. Hyg. Ass. J.* **37**, 143–150.

Greim, H., Bonse, G., and Radwan, Z., *et al.* (1975). Mutagenicity *in vitro* and potential carcinogenicity of chlorinated ethylenes as a function of metabolic oxirane formation. *Biochem. Pharmacol.* **24**, 2013–2017.

Leonardos, G., Kendall, D., and Barnard, N. (1969). Odor threshold determinations of 53 odorant chemicals. *J. Air Pollut. Control Asso.* **19**, 91–95.

Monster, A. C., and Boersma, G. (1975). Simultaneous determination of trichloroethylene and metabolites in blood and exhaled air by gaschromatography. *Int. Arch. Occup. Environ. Health* **35**, 155–163.

Monster, A. C., Boersma, G., and Steenweg, H. (1979). Kinetics of tetrachloroethylene in volunteers: influence of exposure concentration and workload. *Int. Arch. Occup. Environ. Health* **42**, 303–309.

Rampy, L. W., Quast, J. F., Leong, B. K. J., and Gehring, P. J. (1977). "Results of Long-Term Inhalation Toxicity Studies on Rats of 1,1,1-Trichloroethane and Perchloroethylene Formulations." Poster presentation, Intern. Congr. of Toxicology, Toronto, Canada, April 1977.

Weisburger, E. (1977). Carcinogenicity studies on halogenated hydrocarbons. *Environ. Health Perspect.* **21**, 7–16.