HUMAN LEVELS OF SELECTED NON-PERSISTENT CHLORINATED COMPOUNDS

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1. Introduction

Many people, either as a group or independently, have recently called for the banning of chlorinated organic chemicals. This proposed ban is based primarily on alleged adverse health outcomes brought to the ecologic system and mankind by the presence of and exposure to some of these chlorinated organic chemicals, particularly those that bioaccumulate, e.g., DDT, polychlorinated biphenyls (PCBs), and 2,3,7,8-substituted polychlorinated dibenzo-p-dioxins. Other people have the opinion that the continued use of each chlorinated organic chemical should be evaluated separately; e,g., based upon a scientific analysis that uses major principles for those evaluations (1). From the standpoint of human health risk, one major principle could center around human exposure to these compounds for, of course, without such exposure there would be no resulting health effects in humans. There have been numerous reports on levels of chlorinated organic pesticides and PCBs in selected human populations. In the United States, these compounds have been measured in the general population in two surveys, the second National Health and Nutrition Examination Survey (NHANES II) (2) and the National Human Adipose Tissue Survey (NHATS) (3). Temporal data from NHATS indicate that PCB levels in human adipose tissue greatly decreased from the early 1970's to the mid 1980's (4). However, much less is known regarding human exposure to many other chlorinated organic chemicals, particularly those that tend not to bioaccumulate. Several of these were measured in human specimens in our priority toxicant reference range study (5). In this study, we measured blood concentrations of 32 volatile organic compounds (VOCs) and urinary concentrations of 12 pesticides or their metabolites. The specimens were from 1,000 adults who participated in the Third National Health and Nutrition Examination Survey (NHANES III) from 1988-1994. Reference range data from this general population study have been used for many purposes, including comparing with data from allegedly overtly exposed populations. Tables I and II list the halogenated compounds we measured as VOCs and pesticides, respectively.

Table I. Halogenated VOCs in Blood Measured in Reference Range Study

Chlorobenzene	1,1,2,2-Tetrachloroethane
1,2,-Dichlorobenzene	Tetrachloroethene
1,3-Dichlorobenzene	Hexachloroethane
1,4-Dichlorobenzene	Methylene chloride
1,1-Dichloroethane	Chloroform
1,2-Dichloroethane	Carbon tetrachloride
1,1-Dichloroethene	1,2-Dichloropropane
cis-1,2-Dichloroethene	Bromoform
trans-1,2-Dichloroethene	Dibromomethane
1,1,1-Trichloroethane	Bromodichloromethane
1,1,2-Trichloroethane	Dibromochloromethane
Trichloroethene	

Table II. Chlorinated Pesticides or Their Metabolites (and Parent Pesticide) Measured in Urine

3,5,6-Trichloro-2-pyridinol (chlorpyrifos)
2,4-Dichlorophenoxy acetic acid
Pentachlorophenol
2,4,5-Trichlorophenol
2,4,6-Trichlorophenol
2,5-Dichlorophenol (p-Dichlorobenzene)
2,4-Dichlorophenol

2. Results

As shown in Table I, 23 of the 32 VOCs measured in our reference range study of nonoccupationally exposed people are halogenated organic chemicals. We found 11 of the 32 VOCs in more than 75% of the samples examined (6); however, only three of these are halogenated organic chemicals: 1,1,1-trichloroethane; 1,4-dichlorobenzene; and tetrachloroethene. Table III gives the blood concentrations of these three VOCs in the reference range population (from reference 6).

Table III. Blood Concentrations of 3 Halogenated VOCs Found in Greater Than 75% of the Reference Population

Halogenated VOC	Detection Limit, ppt	Mean ppt	Median ppt	5th/95th Percentile, ppt
1,1,1-Trichloroethane	86	340	130	ND/800
1,4,-Dichlorobenzene	73	1900	330	ND/9200
Tetrachloroethene	30	198	63	ND/620

Five other VOCs were found in 10-75% of the samples examined; all of these are halogenated organic chemicals and are listed in Table IV (from reference 6).

Table IV. VOCs Found in 10-75% of Blood Specimens

VOC	Detection Limit, ppt	Percent Detected
Bromodichloromethane	9	14
Chloroform	21	54
Dibromochloromethane	13	12
Chlorobenzene	7	21
Trichloroethene	10	13

Fifteen more VOCs, all of them halogenated VOCs, were detectable in less than 10% of the specimens. These are 1,1,2,2-tetrachloroethane; 1,1,2-trichloroethane; 1,1-dichloroethane; 1,1-dichloroethene; 1,2-dichlorobenzene; 1,2-dichloroethane; 1,2-dichloropropane; 1,3-dichlorobenzene; bromoform; carbon tetrachloride; cis-1,2-dichloroethene; dibromomethane; hexachloroethane; methylene chloride; and trans-1,2-dichloroethene.

The halogenated urinary analytes were all detected in order of decreasing frequency (percent positive): 2,5-dichlorophenol (98%), 3,5,6-trichloropyridinol (82%), pentachlorophenol (64%), 2,4-dichlorophenol (64%), 2,4,5-trichlorophenol (20%), 2,4-dichlorophenoxyacetic acid (12%) and 2,4,6-trichlorophenol (9.5%). 2,5-Dichlorophenol, a metabolite of 1,4-dichlorobenzene, was also found at the highest levels (7). Furthermore, 2,5-dichlorophenol urinary concentrations and 1,4-dichlorobenzene blood concentrations were highly correlated with a Pearson correlation coefficient of 0.82 ($p < 0.0001$) (8).

3. Discussion

For the VOCs, it is interesting to compare these human internal dose measurements with indoor and outdoor results from EPA's Total Exposure Assessment Methodology (TEAM) Study (9). For example, the three halogenated VOCs found in greater than 75% of the reference population were the three halogenated VOCs found at the highest levels in the TEAM Study. This implies that inhalation, especially of indoor air, is the primary route of exposure. Likewise, other halogenated VOCs, trichloroethene and chloroform, that were found in 10-75% of the reference population are the other two halogenated VOCs reported in the TEAM Study. Two other VOCs, bromodichloromethane and dibromochloromethane, that we found in 10-75% of the blood specimens, are trihalomethanes, as is chloroform.

The pesticide data are interesting in that chlorpyrifos in indoor air and its metabolite (3,5,6-trichloro-2-pyridinol) in urine were the most commonly found pesticide/metabolite in EPA's Nonoccupational Pesticide Exposure Study (NOPES) (10) and in our study, respectively. This implies that breathing indoor air may be a primary route of exposure to this pesticide. Many of the other halogenated pesticides, including 1,4-dichlorobenzene, were not measured in NOPES. Our data indicate that 1,4-dichlorobenzene is ubitquitous in the United States and is likely to be a worldwide contaminant (8).

As previously mentioned, these reference range data will have many uses. One use should be in the exposure assessment portion of risk assessment. Also the temporal trend data of many of these VOCs will be interesting, especially in light of the production ban of 1,1,1-trichloroethane, carbon

tetrachloride, and other ozone-layer depleting solvents in accordance with the "Montreal Protocol" treaty .

4. References

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