DISTRIBUTION OF HALOGENATED ENVIRONMENTAL CHEMICALS AMONG PEOPLE OF DIFFERENT AGES, RACES, AND SEXES IN THE UNITED STATES

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Introduction

Many halogenated organic chemicals in the environment are of concern because of their potential persistence, ability to undergo trans-boundary migration, bioaccumulation potential, and possible toxicity. Historically, most of these halogenated chemicals have been the chlorinated chemicals, including polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs), and many organochlorine insecticides. However, more recently, several brominated chemicals, such as the polybrominated diphenyl ethers (PBDEs) and polybrominated biphenyls (PBBs), have garnered much scientific interest. Even more recently, polyfluorinated chemicals, such as perfluorooctane sulfonic acid (PFOS) and perfluoroctane carboxylic acid (PFOA), have been the subject of many investigations.

The general population is exposed to these environmental chemicals through a variety of environmental pathways. In general, for the chlorinated chemicals the primary exposure route is ingestion of contaminated foods, especially dairy products, fish, and wildlife. For the brominated chemicals ingestion of contaminated foods and indoor dust appears to be the major route. The primary pathway for general population exposure to PFOS and PFOA is still being studied, but certainly includes ingestion of contaminated drinking water. Regardless, the chemicals enter the body, are not readily metabolized, and are distributed within the body. The brominated and chlorinated chemicals are in general quite lipophilic and are stored in the fat portions of the body, primarily adipose tissue. Their concentrations tend to equilibrate in the lipid portions of the adipose tissue, other organs, and blood serum. The perfluorinated chemicals travel in the blood, not with the lipid components, but with the protein fraction and are also bound to proteins in the liver. Although both PFOS and PFOA have half lives of several years in the human body, neither chemical bioaccumulates in the food chain.

In industrialized countries, general population exposure to most of the chlorinated compounds peaked in the late 1970s. For the PBDEs, the general population exposure in Sweden peaked in the late 1990s,[1] but this was probably not the case in other European countries and certainly not in the United States.[2] PFOS and PFOA have only more recently become of environmental concern, and whereas 3M Corporation discontinued the manufacturing of PFOS in 2000, PFOA is still being produced. Hence, the exposure history to these 3 different halogenated classes of chemicals varies.

As part of our attempts to examine human exposures to environmental chemicals in the United States' general population, we are analyzing blood and urine samples from participants in the National Health and Nutrition Examination Survey (NHANES) (www.cdc.gov/nationalreport). Individual congener results for the PCBs, PCDDs, and PCDFs have been reported for the NHANES serum samples collected in 2001-2002 for this purpose. However, NHANES results for the individual perfluorinated chemicals and polybrominated chemicals have not been published. In this paper, we compare the results for representative chemicals from the 3 halogenated groups in various stratifications of the U.S. population based on results from NHANES 2001-2002 pooled serum samples.

Materials and Methods

In addition to the individual analyses, we combined 2150 individual serum samples taken from selected participants in 2001-2002 to make serum pools that were based on age range in years (12-19; 20-39; 40-59; 60+); race/ethnicity

(Mexican American, non-Hispanic black, non-Hispanic white, and both sexes; thus, there were 24 demographic groups. Each of the more than 50 pools was made from 0.75 mL of serum from 34 persons for a total of 25.5 g of serum per pool. The serum was divided for each of the measurements as follows: brominated flame retardants/PCBs/organochlorine pesticides- 2 g; PCDDs/PCDFs/coplanar PCBs/polychlorinated naphthalenes- 22 g; perfluorinated chemicals- 0.4 g; and total lipids- 0.5 g. Data on the dioxin toxic equivalents (TEQ) and on the perfluorinated chemicals have been published in part.[3,4]

Results and Discussion

Representative distributions of the analytical results from the pools are given in the Figures. When multiple pools were analyzed for a particular demographic group, the data are represented by the minimum value, maximum value, and if applicable the mean value. Figure 1A, 1B, and 1C represent data for the dioxin TEQ, perfluorooctane carboxylic acid (PFOA), and 2,2',4,4'-tetrabromodiphenyl ether (PBDE 47), respectively. These distributions represent the combined effects of exposure and pharmacokinetics (absorption, distribution, metabolism, and elimination). In examining the dioxin TEQ data across age ranges, we see that serum concentrations increase with age; this is probably a result of relatively higher exposures when the now older people were younger and exposures over a longer time period coupled with the slow elimination of these chemicals from the body. In examining the two sexes, it is interesting that at the young age group males consistently have slightly higher levels than females but as the population ages females have the higher levels; one could interpret this as the exposure effect predominates for the younger population but the pharmacokinetic properties predominate in the older populations; these data are consistent therefore with those showing a longer half-life for 2,3,7,8-tetrachlorodibenzo-p-dioxin in females compared to males.[5] In examining the data across race/ethnicity, consistent with other reports on recent NHANES data,[3,6] the Mexican-Americans have lower serum dioxin TEQ than non-Hispanic blacks and non-Hispanic whites. This dioxin TEQ plot (Figure 1A) is very similar in trends to those seen as well for PCB congeners and for many of the organochlorine insecticides (data not shown); however, the data (not shown) for p,p'-DDE differs probably because of more recent exposures to p,p'-DDT in Mexico.

Similar distribution data are presented for PFOA in Figure 1B. Although PFOA reportedly has a long half life in the body, its distribution within the body is much different than the polychlorinated chemicals discussed above. Whereas the latter chemicals are lipophilic and hence sequester in the lipid portions of the body, PFOA binds to proteins, such as in the blood and liver. For PFOA, the most striking results are the serum concentrations do not increase with age, concentrations in males are consistently higher than in females, and non-Hispanic whites have higher concentrations than the other two groups.

Distribution data are presented for BDE-47 in Figure 1C. These chemicals are like the dioxins in that they are lipophilic, but unlike the dioxins, the environmental concentrations in the U.S. have not been decreasing over the last several decades. Therefore, because the older people were not exposed when environmental levels were higher, we do not see increases in concentrations with age.

In conclusion, the serum distribution data for representative chemicals with different halogen atoms in these serum pools differ. We should caution, however, that although the samples constituting the pools originated from NHANES 2001-2002, which was designed to be representative of the civilian, noninstitutionalized U.S. population, we cannot be assured that estimates based on pooled data are unbiased; however, they do represent a good geographical and demographic coverage of the U.S. population.

References

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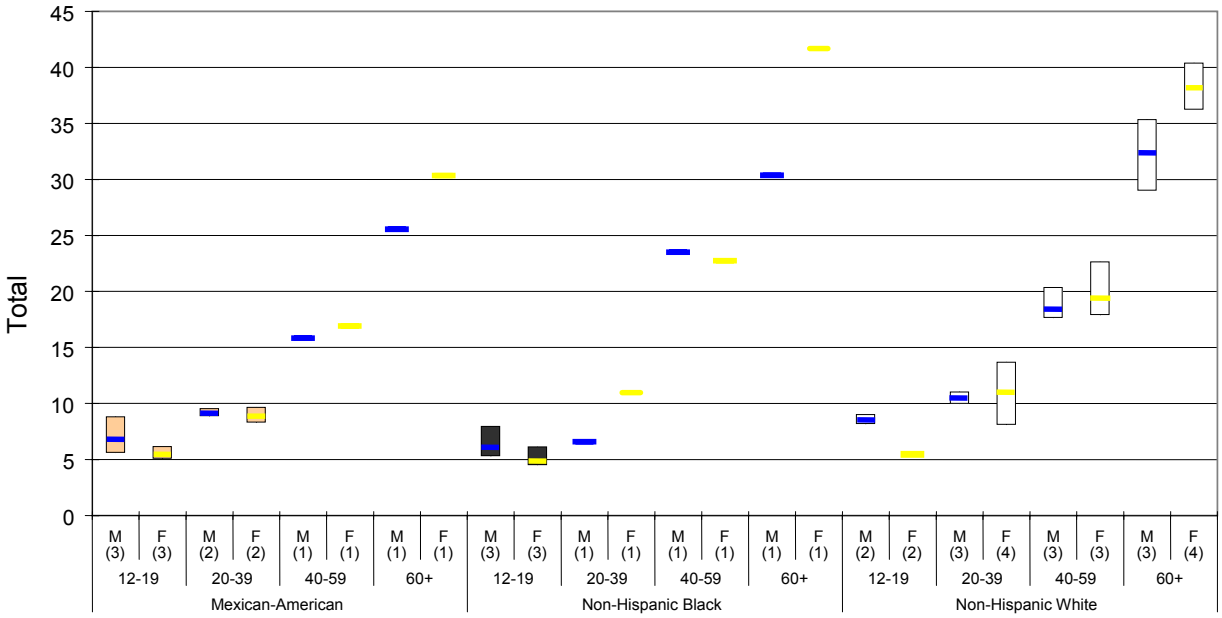
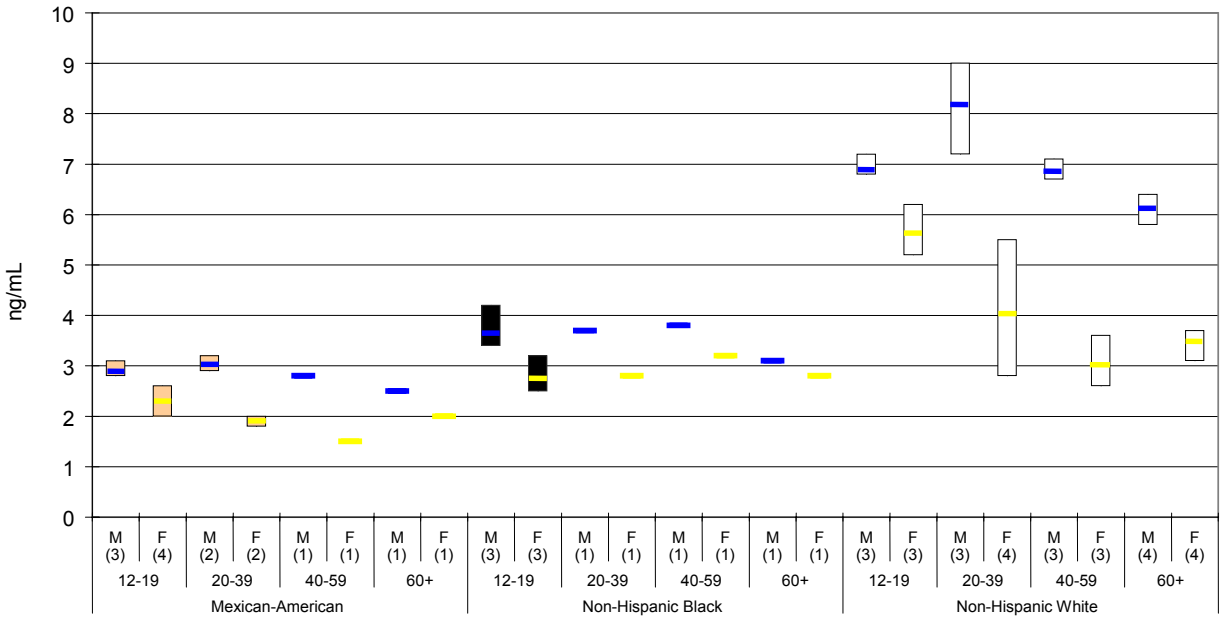
Figure 1A. Mean and Range of Dioxin TEQ (PCDs, PCDFs, cPCBs) by Age Group, Race and Sex in NHANES 2001/2002 Serum Pools (number of pools per category in parentheses)



1B. Mean and Range of Perfluorooctane carboxylic acid (PFOA) by Age Group, Race and Sex in NHANES 2001/2002 Serum Pools (number of pools per category in parentheses)



1C. Mean and Range of 2,2',4,4'-tetrabromodiphenyl ether (PBDE 47) by Age Group, Race and Sex in NHANES 2001/2002 Serum Pools