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Total TEQ reference range (PCDDs, PCDFs, cPCBs, mono-PCBs) for the US population 2001–2002

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ABSTRACT

We report reference ranges for the total toxic equivalency (TEQ) and TEQ sub-fractions of polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofurans (PCDFs), coplanar biphenyls (cPCBs), and mono-ortho-substituted biphenyls (mPCBs) in a statistically designed sampling of the US population in 2001–2002. The TEQ and TEQ sub-fractions have been stratified by age, sex, and race/ethnicity. The TEQ levels are lower using the 2005 toxic equivalency factors (TEFs) compared to using the 1998 TEF values, principally due to the much lower 2005 TEF values assigned to the mPCBs. Mexican Americans (MA) have significantly lower TEQ levels than both non-Hispanic whites (NHW) and non-Hispanic blacks (NHB). Using the 1998 or 2005 TEF values, males and females have nearly the same distribution of TEQ sub-fractions. We found a significant increase in TEQ levels with age for males, females, and NHW. About 80–90% of the total TEQ can be estimated by using seven congeners, namely 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1,2,3,6,7,8-HxCDD, 2,3,4,7,8-PeCDF, PCB-126, PCB-118, and PCB-156. We also measured geometric mean TEQ levels in pooled samples from the US population. The geometric mean TEQ levels also increase with age. In the youngest age group (12–19 years), the TEQ levels were higher in males than in females while females had higher TEQ levels than males in all older age groups. In the pools, as age increases the percent contribution of the PCDD TEQ levels increases while the percent contribution of the PCDF TEQ levels decreases for all race/ethnicity and sex strata.

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1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) are chemicals that are produced as contaminants in several chemical processes such as in the synthesis of 2,4,5-trichlorophenol. Trace levels of these compounds are also produced during the incineration of waste materials, combustion of leaded gasoline, and the bleaching of wood pulp. Thus, these compounds can enter the environment by a variety of means. Once there, they are remarkably stable, and many of these compounds, especially those that are substituted with chlorine at the 2,3,7,8 positions and that are also the most toxic, bioaccumulate in the human food chain (Fisher, 1999). Polychlorinated biphenyls (PCBs) are chemicals that were used as electrical insulating and heat-exchange fluids. Together with the PCDDs and PCDFs, certain PCB congeners, the coplanar (cPCBs) and mono-ortho-substituted (mPCBs) PCBs, are often referred to as "dioxin-like" because they act pharmacologically through a similar mechanism. These compounds can generally be found at the parts-per-trillion (ppt) levels in the lipid stores of humans, especially those living in an industrialized society. The general population is exposed to these chemicals as mixtures primarily through the ingestion of high-fat foods, such as dairy products, eggs, and animal fats, and some fish and wildlife.

The PCDDs, PCDFs and PCBs are lipophilic compounds and tend to accumulate in the lipid stores of the body; therefore, adipose tissue has historically been the matrix of choice for measuring these compounds in humans. Because adipose tissue is mostly lipid (generally more than 75% of the tissue), the PCDDs, PCDFs and PCBs are found at the highest levels in this tissue. In recent years, however, the need for a less invasive procedure for obtaining a sample has prompted the development of methods to measure PCDDs, PCDFs, and PCBs in serum, plasma, and whole blood (Patterson et al., 1987; Nygren et al. 1988; Papke et al. 1989). A validation study was conducted to compare the concentrations of 2,3,7,8-TCDD in adipose tissue with paired serum samples from the same individuals (Patterson et al., 1988) which found a 1:1 partitioning of 2,3,7,8-TCDD between these two tissues when a correction was made for their lipid content. The use of blood for these types of measurements has resulted in high participation rates in several large-scale epidemiologic studies of potentially exposed populations (CDC, 1987, 1988a, 1988b, 1988c; Fingerhut et al. 1989; Haring-Sweeney et al. 1990; Patterson et al., 1990a, 1990b, 1990c). The small amount of lipid (generally about 0.6% of the

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sample) in blood, however, requires very sensitive procedures to measure the PCDDs, PCDFs and PCBs at the parts-per-quadrillion level. These low levels in blood require a very extensive laboratory quality assurance program with as high as 60% quality control (QC) samples to ensure the validity of the measurements (Patterson et al., 1989, 1990a, 1990b; Turner et al. 1989).

Each of the PCDD, PCDF, and PCB congeners has been assigned a potency value relative to 2,3,7,8-TCDD (toxic equivalency factor, TEF). The TEF values are multiplied by the respective congener concentration to give the congener WHO-toxic equivalency (TEQ), and these are summed to give a total TEQ (Van den Berg et al., 1998). Thus, the dioxin-like toxicity contribution of each chemical class can be compared. Recently in 2005 the TEFs were reevaluated (Van den Berg et al., 2006), and a new set of TEF values were published. Since most of the literature publications to date have used the 1998 TEF values, the results in this paper are presented using both the 1998 and the 2005 TEF values. Several recent papers have presented reference range estimates for the US population. Needham et al. (2005) presented US reference range data based on the National Health and Nutrition Examination Survey 1999–2000 (NHANES 1999–2000) using 1998 TEF values for samples collected in 1999–2000. Ferriby et al. (2007) have used data published by the Centers for Disease Control and Prevention (CDC 2003) for NHANES samples collected in 2001–2002 to estimate reference ranges for the US population. As described in Section 2, we have taken a different approach and used the same data set (CDC 2003) to present reference ranges for the US population using both 1998 and 2005 TEF values. In addition, we also present data using pooled serum from NHANES 2001–2002 samples that allow a higher detection frequency for many congeners due to the relatively small serum sample size for individual samples. Attempts at determining a reference range have also been made using a combination of separate dioxin studies in several US States with samples collected between 1996 and 2001 (Patterson et al., 2004). The study of Patterson et al. measured the PCDDs, PCDFs, and cPCBs but did not measure the mPCBs. Because the levels of PCDDs, PCDFs, cPCBs, and mPCBs in the environment and in people have been declining in the past 2–3 decades (Furst et al. 1994; Papke 1997; Aylward and Hayes, 2002; Choi et al., 2002; Jackson et al., 2002; Lorber 2002; Sjodin et al., 2004; Hagmar et al., 2006), estimates from a number of years past may not be a valid measure of today's background dioxin levels.

Table 1
TEQs in pg g^{-1} lipid for all individuals 20+ years of age in the US population

TEQ	Percentiles	TEQ (1998)	95% CI	TEQ (2005)	95% CI	*n*
Total	90th	54.6	47.3–61.8	41.0	35.8–47.1	1194
	95th	68.9	62.9–80.8	56.1	47.6–65.4	1194
PCDD	90th	25.7	20.6–30.3	25.8	20.8–30.4	1194
	95th	34.8	28.7–43.3	34.8	28.7–43.4	1194
PCDF	90th	9.8	8.5–11.4	7.1	6.1–8.3	1194
	95th	12.3	11.0–14.4	8.9	7.7–10.2	1194
Coplanar PCB	90th	7.2	6.4–8.4	8.0	7.2–9.2	1194
	95th	11.0	9.7–12.0	11.9	10.8–12.9	1194
Mono-Ortho PCB	90th	14.1	12.0–15.6	2.0	1.7–2.3	1194
	95th	18.0	15.1–20.4	2.6	2.3–3.0	1194

Table 2
TEQs in pg g^{-1} lipid for Females 20+ years of age in the US population

TEQ	Percentiles	TEQ (1998)	95% CI	TEQ (2005)	95% CI	*n*
Total	90th	57.5	49.5–65.4	43.9	36.6–51.2	656
	95th	76.5	64.6–90.1	58.5	49.3–74.6	656
PCDD	90th	26.7	21.1–30.3	27.1	21.2–30.4	656
	95th	35.6	28.5–48.8	35.8	28.6–49.0	656
PCDF	90th	10.2	8.9–11.2	7.2	6.3–8.0	656
	95th	12.7	11.1–14.3	9.2	8.0–10.0	656
Coplanar PCB	90th	8.6	7.5–10.2	9.3	8.3–11.0	656
	95th	11.8	10.2–13.0	12.5	11.6–14.2	656
Mono-Ortho PCB	90th	14.9	12.8–16.1	2.2	2.0–2.4	656
	95th	18.4	15.7–21.2	2.9	2.4–3.4	656

Table 3
TEQs in pg g^{-1} lipid for males 20+ years of age in the US population

TEQ	Percentiles	TEQ (1998)	95% CI	TEQ (2005)	95% CI	*n*
Total	90th	50.1	41.1–61.5	38.7	30.8–46.4	538
	95th	64.9	55.1–75.7	49.8	41.7–59.1	538
PCDD	90th	24.1	18.2–30.6	24.2	18.3–30.7	538
	95th	32.4	25.7–42.5	32.6	25.8–42.5	538
PCDF	90th	9.4	8.3–11.7	6.9	5.8–8.6	538
	95th	12.2	9.7–14.9	8.8	7.0–10.4	538
Coplanar PCB	90th	5.6	4.9–6.4	6.3	5.5–7.2	538
	95th	8.4	5.9–10.6	9.2	7.0–12.5	538
Mono-Ortho PCB	90th	12.4	9.9–16.1	1.6	1.3–2.0	538
	95th	17.5	13.0–21.2	2.3	1.7–3.0	538

Table 4
TEQs in pg g^{-1} lipid for all 20+ year old Mexican Americans in the US

TEQ	Percentiles	TEQ (1998)	95% CI	TEQ (2005)	95% CI	*n*
Total	90th	31.9	25.3–40.2	24.4	19.6–32.1	259
	95th	40.5	31.9–52.6	33.6	24.9–41.6	259
PCDD	90th	14.6	11.8–17.7	14.7	12.0–17.8	259
	95th	20.5	14.9–28.9	20.5	15.0–29.2	259
PCDF	90th	5.8	4.6–6.9	4.2	3.6–5.0	259
	95th	7.1	6.0–9.2	5.0	4.3–6.7	259
Coplanar PCB	90th	5.1	4.0–6.2	5.5	4.6–6.8	259
	95th	7.3	5.3–10.6	7.6	5.8–11.1	259
Mono-Ortho PCB	90th	8.4	7.1–9.6	1.2	1.0–1.5	259
	95th	9.8	8.6–11.8	1.5	1.3–2.0	259

Table 5
TEQs in pg g^{-1} lipid for all 20+ year old non-hispanic blacks in the US

TEQ	Percentiles	TEQ (1998)	95% CI	TEQ (2005)	95% CI	*n*
Total	90th	61.4	50.9–85.7	47.6	38.1–64.5	212
	95th	90.0	61.0–113	71.1	49.3–82.8	212
PCDD	90th	29.1	22.7–43.7	29.4	22.9–43.9	212
	95th	44.5	29.5–54.9	44.7	29.7–55.7	212
PCDF	90th	10.8	8.7–12.9	7.9	6.2–9.5	212
	95th	14.3	11.2–16.5	10.4	8.0–12.1	212
Coplanar PCB	90th	9.2	6.1–11.4	9.8	6.6–12.1	212
	95th	12.9	10.1–16.7	13.6	10.9–17.7	212
Mono-Ortho PCB	90th	17.1	12.5–22.0	2.5	1.8–3.2	212
	95th	22.3	16.1–31.3	3.5	2.1–4.6	212

Table 6
TEQs in pg g^{-1} lipid for all 20+ year old non-hispanic whites in the US

TEQ	Percentiles	TEQ (1998)	95% CI	TEQ (2005)	95% CI	*n*
Total	90th	57.3	49.5–64.8	43.6	36.9–51.2	640
	95th	72.1	64.9–82.7	56.7	50.6–66.4	640
PCDD	90th	27.4	21.1–32.1	27.5	21.1–32.2	640
	95th	37.0	29.3–44.0	37.4	29.4–44.3	640
PCDF	90th	10.4	9.2–11.9	7.3	6.5–8.7	640
	95th	12.7	11.1–14.8	9.2	8.0–10.6	640
Coplanar PCB	90th	7.5	6.6–8.6	8.4	7.5–9.4	640
	95th	11.5	9.3–13.0	12.4	10.4–14.2	640
Mono-Ortho PCB	90th	14.7	12.3–16.3	2.1	1.7–2.4	640
	95th	18.7	15.4–21.2	2.7	2.3–3.3	640