## TEMPORAL TRENDS IN INTAKE OF DIOXINS FROM FOODS IN THE U.S. AND WESTERN EUROPE: ISSUES WITH INTAKE ESTIMATES AND PARALLEL TRENDS IN HUMAN BODY BURDEN

Lesa Aylward<sup>1</sup>, Sean Hays<sup>2</sup>, and Brent Finley<sup>3</sup>

<sup>1</sup>Exponent, Inc., 1800 Diagonal Road, Suite 355, Alexandria, VA 22314, U.S.A., laylward@exponent.com
<sup>2</sup>Exponent, Inc., 4940 Pearl East Circle, Suite 300, Boulder, CO 80301, U.S.A., shays@exponent.com
<sup>3</sup>Exponent, Inc., 631 First Street, Suite 200, Santa Rosa, CA 95404, U.S.A., bfinley@exponent.com

### Introduction

Food is likely to be the source of more than 90 to 95 percent of exposures to dioxins in the United States (U.S.) and Western Europe. Several Western European countries have attempted to estimate intake of dioxins and dioxin-like compounds over time. More recently, the U.S. Environmental Protection Agency (USEPA) has estimated intake on a toxic equivalency (TEQ) basis. This paper describes some of the issues associated with evaluating and comparing intake estimates from different countries and from within a country over time. In addition, this paper presents compiled data on human TEQ body burden for selected polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and polychlorinated biphenyl (PCB) compounds. Because of the long half-life for elimination of these compounds in the human body, changes in human tissue levels are likely to lag changes in levels of sources to the environment and food.

### **Methods and Materials**

We compiled published values for mean or median human adipose or serum lipid levels of PCDD/ Fs and PCB-126 from general population samples in North America and Western Europe. We also compiled estimates of intake levels over time from several government agencies and review articles. We assessed issues that introduce uncertainty and limit the comparability of data and estimates from different researchers. We also assessed the implications of the parallel trends in food intake estimates and human body burdens over the past three decades for future exposures.

### **Results and Discussion**

#### Issues in comparing intake estimates for dioxins

Although dioxins have been a focus of environmental concern for nearly 30 years, with food the known source of the majority of human intake, no comprehensive, systematic, or representative food sampling or monitoring programs have been conducted in the U.S. Therefore, estimates of current levels of dioxins in foods and dietary intakes in the United States are a hodge-podge based on a variety of data sources. Several European governments have had ongoing food monitoring programs for 10 to 20 years, but data from these programs must also be interpreted carefully. Intake estimates are a function not only of the concentration of dioxins in foods are typically reported on a TEQ basis, and often, compound-specific concentrations are not reported. Because the TEF values for specific compounds have been adjusted several times since the TEQ system was first used, different dioxin

### ORGANOHALOGEN COMPOUNDS Vol. 55 (2002)

levels reported from one study to another may be due to differing TEF values in effect at the time the studies were conducted. Thus, reported changes in dioxin intake levels over time may be due to a combination of factors other than changes in the level of dioxins in the food. A major issue that arises in comparing food levels or intake rates from one study to another is the variability in detection limits and the handling of non-detects in the analytical results. Due to the low levels of dioxins present in foods and the difficulty of achieving sensitive detection limits, non-detectable levels for many compounds are common in food sampling.

### Estimates of intake

Estimated intake rates over time for PCDD/Fs from the U.S., the United Kingdom (U.K.), and the Netherlands are presented in Figure 1. Estimated intake rates for PCB TEQ are presented in Figure 2. These estimates present a remarkably consistent picture of declining intake. While the intake levels from the U.K. appear to be somewhat higher than levels reported for other countries during the same time period, this is primarily due to the use of the full limit of detection (LOD), rather than ½ the LOD, for non-detects in the Food Standards Agency of the United Kingdom (FSA, formerly the Ministry of Agriculture, Fisheries, and Food) reporting protocol. Although estimates from earlier time frames are very limited, the data from the U.K. and the Netherlands indicate even larger drops preceding the late 1980s. Details of these estimates are as follows:

• The USEPA has presented estimates of mean dietary intake levels in each of the last two dioxin reassessment drafts (1994 and 2000). The 2000 estimate of 0.6 pg TEQ/kg-day (representing intake levels in about 1996) is 66 percent lower than the 1994 estimate of 1.7 pg/kg-day for PCDD/Fs (for intake in the mid- to late 1980s)<sup>1</sup>. Intake of dioxin-like PCB compounds of 0.34 pg TEQ/kg-day for 1996. A significant proportion of the reduction in the intake estimate is due to lower estimates of food intake across every food category.

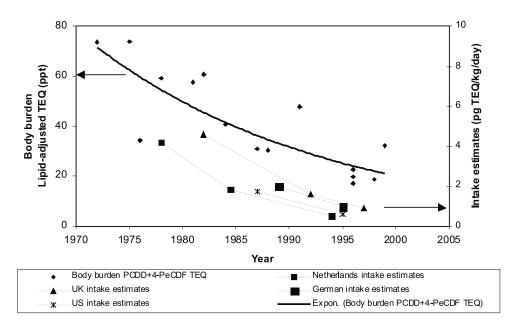
• The U.K. Food Sampling Agency (FSA) conducted food sampling in the U.K. in 1982, 1992, and 1997<sup>2</sup>. The FSA estimates that upper-bound (assuming that non-detects are present at the limit of detection) mean intakes of PCDD/Fs and PCBs by adults dropped from 7.2 pg TEQ/kg-day in 1982 to 2.5 pg TEQ/kg-day in 1992 and to approximately 1.8 pg TEQ/kg-day in 1997 (TEQ estimates made using World Health Organization [WHO] 1998 TEF values<sup>3</sup>). PCDD/F mean intakes (without PCBs) were 4.6, 1.6, and 0.9 pg TEQ/kg-day in 1982, 1992, and 1997, respectively.

• Liem et al. (2000)<sup>4</sup> recounted the analytical results of composites of 24-hour duplicate diets from 1978, 1984/5, and 1994 in the Netherlands. The estimate of total daily TEQ intake decreased from 11 pg TEQ/kg-day in 1978 to 4.2 pg TEQ/kg-day in 1984/5, to 1.5 pg TEQ/kg-day in 1994. These estimates included PCB compounds and were made using the international TEQ system (I-TEQ). PCDD/F intakes (without PCBs) were estimated to be 4.2, 1.8, and 0.5 pg TEQ/kg-day in 1978, 1984/ 5, and 1994, respectively.

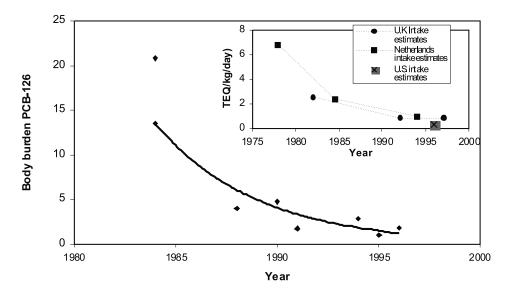
• Furst and Wilmers (1997)<sup>5</sup> analyzed data from a German government food monitoring program and documented a drop in estimated mean intake of dioxins and furans from approximately 2 pg TEQ/kg-day in the late 1980s to about 1 pg TEQ/kg-day in 1994/5 (I-TEQ).

### Temporal trends in body burden

Figures 1 and 2 also present values for general-population TEQ levels of PCDDs and 4-PeCDF (Figure 1) and PCB-126 (Figure 2) from the literature. The trend of decreasing body burden is substantial and parallels the decrease in intake estimates. Because of the long half-life of elimination of these compounds, changes in body-burden levels will probably lag the changes in intake levels. This indicates that further declines in body burden are likely in response to decreases in food levels already seen.



**Figure 1.** Trends in mean or median human lipid-adjusted TEQ body burden (ppt) for PCDDs and 4-PeCDF from several studies<sup>1,6-14</sup> and PCDD/F intake estimates (pg TEQ/kg/day) from the U.S. and Western Europe, as discussed in the text.



**Figure 2.** Mean or median human PCB-126 levels in general population studies <sup>1,15-19</sup> and PCB TEQ intake estimates, as discussed in the text.

ORGANOHALOGEN COMPOUNDS Vol. 55 (2002)

### References

- 1. United States Environmental Protection Agency (USEPA) (2000) "Exposure and Human Health Reassessment of 2,3,7,8-Tetrachlorodibenzo-p-Dioxin and Related Compounds. Draft".
- 2. Food Standards Agency UK (FSA) "Dioxins and PCBs in the UK diet: 1997 Total diet study samples. Information Sheet Number 4/00," 2000.
- 3. Van den Berg, M., Birnbaum, L., Bosveld, A.T., Brunstrom, B., Cook, P., Feeley, M., Giesy, J.P., Hanberg, A., Hasegawa, R., Kennedy, S.W., Kubiak, T., Larsen, J.C., van Leeuwen, F.X., Liem, A.K., Nolt, C., Peterson, R.E., Poellinger, L., Safe, S., Schrenk, D., Tillitt, D., Tysklind, M., Younes, M., Waern, F. and Zacharewski, T. (1998) Environ Health Perspect, 106, 775.
- 4. Liem, A.K., Furst, P. and Rappe, C. (2000) Food Addit Contam, 17, 241.
- 5. Furst, P. and Wilmers, K. (1997) Organohalogen Compounds, 33, 116.
- Kang, H., Watanabe, K., Breen, J., Remmers, J., Stanley, J. and Flicker, M. (1990) "Dioxins and Dibenzofurans in Adipose Tissue of US Vietnam Veterans and Controls" NTIS PB-91167585
- 7. Papke, O. (1998) Environ Health Perspect, 106 Suppl 2, 723.
- 8. Stanley, J.S. "Broad Scan Analysis of the FY82 National Human Adipose Tissue Survey Specimens Volume I Executive Summary," U.S. Environmental Protection Agency, 1986.
- 9. Stanley, J.S. and Orban, J. "Chlorinated dioxins and furans in the general U.S. population: NHATS FY87 Results. Final Report," U.S. Environmental Protection Agency, 1991.
- 10. Arfi, C., Seta, N., Fraisse, D., Revel, A., Escande, J.P. and Momas, I. (2001) Chemosphere, 44, 1347.
- Petreas, M., She, J., McKinney, M., Visita, P., Winkler, J., Mok, M. and Hooper, K. (2001) in ACS Symposium Series 772: Persistent, Bioaccumulative, and Toxic Chemicals I. Fate and Exposure; (Lipnik, R.L., Hermens, J.L.M., Jones, K.C., Muir, D.C.G., Eds.), American Chemical Society, Washington, D.C.
- 12. Graham, M., Hileman, F.D., Orth, R.G., Wendling, J.M. and Wilson, J.D. (1986) Chemosphere, 15, 1595.
- 13. Schecter, A., Ryan, J.J. and Gitlitz, G. (1986) in Chlorinated Dioxins and Dibenzofurans in Perspective; (Rappe, C., Choudhary, G., Keith, L.H., Eds.), Lewis Publishers, Chelsea, MI, pp 51.
- 14. Wittsiepe, J., Schrey, P., Ewers, U., Wilhelm, M. and Selenka, F. (2000) Environ Res, 83, 46.
- 15. Williams, D.T. and LeBel, G.L. (1991) Chemosphere, 22, 1019.
- Shadel, B.N., Evans, R.G., Roberts, D., Clardy, S., Jordan-Izaguirre, D., Patterson, D.G., Jr. and Needham, L.L. (2001) Chemosphere, 43, 967.
- 17. Dewailly, E., Ryan, J.J., Laliberte, C., Bruneau, S., Weber, J.P., Gingras, S. and Carrier, G. (1994) Environ Health Perspect, 102 Suppl 1, 205.
- 18. Longnecker, M., Ryan, J., Gladen, B. and Schecter, A. (2000) Arch Environ Health, 55, 195.
- 19. Patterson, D.G., Jr., Todd, G.D., Turner, W.E., Maggio, V., Alexander, L.R. and Needham, L.L. (1994) Environ Health Perspect, 102 Suppl 1, 195.