Decrease of serum concentrations of dioxins and PCBs in Belgium between 2000 and 2003

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Introduction

A continuous decline in human dioxin and PCB body burden has been observed in most industrialized countries during the last decades.¹⁻³ This decline is explained by restrictions on production and use of these chemicals. In Belgium, comparison between the first (1988) and second (1993) round of the WHO-coordinated exposure study on human milk showed significant decreases in dioxin and PCB concentrations.³ Apart from this WHO survey, the only study having investigated the temporal evolution of dioxin body burden in Belgium was the survey on blood donors which compared dioxin levels in serum before and after the 1999 PCB/dioxin food contamination incident.⁴ This study did not reveal any clear impact of the incident. On the other hand, conclusions about the temporal variations are difficult to be drawn, both because of the short time interval and due to the ongoing accumulation with age, since comparison was based on the same subjects. We report here the temporal changes of serum concentrations of dioxins and PCBs observed when recruiting different groups of the general population in Belgium over a period of more than three years.

Materials and Methods

The study was approved by the Ethical Committee of the Université catholique de Louvain. A total of 325 fasting serum samples were collected in order to assess the exposure to dioxins and PCBs of different groups of the Belgian population. The description of five of the eight groups was already given in a previous paper.⁵ These groups were: (i) 51 residents living near the municipal solid waste incinerator (MSWI) of Thumaide, recruited between February and April 2000; (ii) 33 residents living near the MSWI of pont-de-Loup, recruited on April and May 2000; (iii) 52 residents living near a dumping site, recruited on April 2001; (iv) 58 residents living near sinter plants from the iron and steel industry, recruited on December 2000 and January 2002 and (v) 63 subjects from a rural area away from industrial sources of dioxins, recruited on March and April 2000 and May 2001. Three other groups that were not previously presented are: (i) 15 bus drivers from Namur recruited on December 2001; (ii) 15 bus drivers from Brussels recruited on June 2003 and (iii) 38 residents living near the incinerator of Brussels, recruited on June and July 2003. The time elapsed between the blood sampling of the first subject and the last subject of this population is thus of forty months.

In order to evaluate the dioxin body burden, the seventeen 2,3,7,8 -substituted polychlorinated dibenzodioxin/dibenzofuran congeners (PCDD/Fs) and the four dioxin-like non-ortho-PCBs (coplanar PCBs or cPCBs; IUPAC no. 77, 81, 126 and 169) were quantified by GC-HRMS on the lipid fraction of serum.^{6,7} The results were expressed per gram fat as equivalents of TCDD using the WHO-TEFs (1998). Twelve PCB markers (ortho-PCBs: IUPAC n^{os} 3, 8, 28, 52, 101, 118, 138, 153, 180, 194, 206 and 209) were also quantified in the study groups, with the exception of the two groups recruited in Brussels. The statistical analysis was done using the SAS software version 9.1 (Enterprise Guide, release 3.0).

Results and Discussion

The mean concentrations of PCDD/Fs (total TEQ of the 17 congeners), coplanar PCBs (total TEQ of the 4 congeners) and ortho-PCBs (sum of the 12 congeners) of the total population were 23.5 pg TEQ/g fat, 6.6 pg TEQ/g fat and 402 ng/g fat, respectively.

Multiple linear regression analyses were used to identify factors influencing the accumulation of PCDD/Fs, coplanar PCBs or ortho-PCBs (analyzed separately as dependent variables) in the studied population. We tested the

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following independent variables in the different models: age, gender, BMI (log-transformed), membership to each of the studied groups (yes/no), animal fat intake (log-transformed), fish consumption (< or > 300 g/week), weight loss (> 5 kg during last year, yes/no), diabetes (yes/no), time factor (time elapsed since the start of the study, in months) and smoking status according to gender (women current smokers, yes/no; men current smokers, yes/no).

Figure 1. Time trend for PCDD/Fs (A), cPCBs (B) and ortho-PCBs (C).

The time elapsed since the start of the study appears to be a highly significant negative determinant of the serum levels of PCDD/Fs (r²=0.03; p=0.0002), coplanar PCBs (r²=0.12; p<0.0001) and ortho-PCBs (r²=0.02; p=0.0085). In order to further analyze the relationships between the time factor and PCDD/Fs, coplanar PCB and ortho-PCB concentrations, values were adjusted for other covariates identified by the regression analyses. These covariates were, for PCDD/Fs: age, BMI, fat intake, diabetes, residence near the MSWI of Thumaide, gender, current smoking

(women) and weight loss. For coplanar PCBs, covariates were age, BMI, diabetes, fish consumption and residence near the MSWI of Thumaide. For ortho-PCBs, the covariates were age, gender, fat intake, diabetes, fish consumption and current smoking (men). Figure 1 illustrates the highly significant decline of PCDD/F, coplanar PCB and ortho-PCB concentrations (adjusted for other determinants) observed between 2000 and 2003.

Values are adjusted for other determinants than time factor. The equations of the regression lines are: 'Log adjusted PCDD/Fs $=$ $-$ 0.003322*month + 1.428' ; 'Log adjusted cPCBs $=$ $-$ 0.005613*month + 0.845' and 'Log adjusted ortho-PCBs = -0.003040 *month + 2.576'.

From the regression equations of Figure 1, the mean annual decrease of PCDD/F concentrations in serum, adjusted for other covariates, can be estimated at 8.8%. An almost identical decrease (8.1% per year) was observed in the serum levels of ortho-PCBs. In the same time, the mean annual decrease of coplanar PCBs in serum was almost double, reaching 14.4%. The decreases observed with PCDD/Fs and ortho-PCBs were very similar to that reported in 1996 by WHO for human milk samples from Belgium (around 8%).³

Although these observations have been made over a relatively short period of time and concern groups living in different areas of Belgium, the time trend observed in our study, after adjustment for all known determinants of dioxin and PCB accumulation, suggests that human exposure to dioxins and PCBs in Belgium continues to decline at a similar rate as estimated in the early 90s.

Acknowledgements

This study was partly supported by the Ministry of Environment of the Walloon Region, Belgium. S Fierens is Research Fellow of the Brussels-Capital Region (Prospective Research for Brussels) and A Bernard is Research Director of the National Fund for Scientific Research, Belgium.

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