ARE POLICY MEASURES REFLECTED IN TIME TRENDS OF HUMAN BIOMONITORING RESULTS IN FLANDERS (BELGIUM)?

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Introduction

In 2003, the Flemish government voted the Decree on Preventive Health Care as a legal recognition of environmental health. The Decree[1] made it mandatory for the Flemish government to perform a human biomonitoring (HBM) program as an important policy signal of preventive public health policy. The first large human biomonitoring survey organized by the Flemish Centre of Expertise for Environment and Health (FLEHS I 2002-2006) and funded by the Flemish government, showed significantly higher concentrations of marker PCBs, dioxin-like substances and chlorinated pesticides in cord blood of newborns and blood of adolescents and adults living in low populated rural communities of East and West Flanders and Flemish Brabant compared to other Flemish regions[2]. Due to the health concern associated with these persistent, bioaccumulative and toxic compounds, the Flemish government decided to follow up their levels in these areas. Human milk was selected as non-invasive matrix. Furthermore, the study results allowed comparison with the Belgian results of the earlier WHO human milk surveys (1989, 1992, 2002 and 2006). Here we present the results of analysis of polychlorinated dibenzodioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), dioxin-like polychlorinated biphenyls (DL-PCBs), marker PCBs and brominated flame retardants (BFRs),eg. hexabromocyclododecane (HBCD) and polybrominated diphenyl ethers (PBDEs) in pooled samples of human breast milk.

Materials and methods

The study area was formed by 24 low-populated rural communities in East and West Flanders. The study aimed to include 100 participants. From May 2009 until June 2010, 84 mothers could be recruited in maternities. To be selected, mothers had to meet inclusion criteria that were based on the 4th WHO human milk survey[6] held in 2006 and some extra inclusion criteria based on FLEHS[2]. Mothers had to sign an informed consent, breastfeed the baby, born in Belgium, between 18 and 35 (included) years old, reside in one of the selected communities, have a normal pregnancy duration (> 36 weeks), HIV negative, give birth to the first or second child, not to give birth to twins, and give birth to a healthy baby.

Participants were asked to donate at least 50 mL of human milk between 2 and 8 weeks after delivery. All participating mothers were given a manual pump and two 50 mL polypropylene containers. After collection, milk samples were refrigerated at 4°C for maximum 72 hours and stored subsequently at -20°C. All samples were labeled with a unique code. All participants completed a questionnaire.

10 mL of each of the 84 individual samples was pooled to prepare one composed sample. Most POPs were analyses on individual samples as well as on the pooled sample. A volume of 400 mL from the pooled sample was sent for analysis to the WHO reference laboratory (State Institute for Chemical an Veterinary Analysis of Food) in Freiburg, Germany. Analytical methods are described elsewhere[3,4,5]. Since only results of pooled samples are discussed (one pooled sample for each study population), no statistical analysis was performed on these results.

Results and discussion

Compared to the population of the Belgian 4th WHO coordinated mother milk suvey[6], the mean age of the rural Flemish population was significantly ($p<0.05$) higher, but the Body Mass Index (BMI) was comparable (table 1). In the rural study, fewer mothers were firstborn themselves, but a comparable number of participants have been breastfed. Because of difficulties in recruiting participants in this rural study, mothers who gave birth to their second child, were also enrolled, resulting in less participants having their first child compared to the Belgian WHO population. Differences were also observed in feeding habits. Compared to the Belgian WHO studypopulation, the rural population consumed more fish ($p<0.05$) and the consumed fish was mainly marine fish ($p<0.05$).

Table 1: Characteristics of the Belgian WHO study population (2006) and the rural study population (2009-2010) expressed as mean values (min-max) or percentages.

Characteristics	Belgian study population WHO 2006	Rural study population 2009-2010
N	197	84
Age	26.1 (18-30)	28.5 (20.6-35.6)
Body Mass Index (kg/cm²)	23 (16-38)	23 (17.7-45.6)
Mother was firstborn	51%	40%
Mother received breastfeeding	52%	48%
Parity: first child	100%	57%
Baby's age at first sampling (weeks)	3 (0-9)	4 (2-8)

In the WHO human milk surveys (1989-2006) a declining time trend can be observed (figure 1) for the Belgian levels of PCDDs/PCDFs, DL-PCBs and sum of 6 marker PCBs (PCB28, PCB52, PCB101, PCB138, PCB153 and PCB180). The levels of these classic POPs measured in the pooled sample of the rural population in 2009-2010 followed the ongoing declining trend. Despite the declining trend, Belgian dioxin levels are still higher than reported results from other European countries[7] participating the 4th WHO human milk survey.

Historically (municipal) waste incinerators and industrial activities were the main sources of PCDDs/PCDFs in Flanders and Europe. Due to implementation of several guidelines and limit values, dioxin emissions from these activities were considerably reduced. According to the European Directive 94/67/EC concerning incineration of hazardous waste, the Directive 2000/76/EC for emissions of dioxins and furans for all waste incineration, and the Flemish environmental legislation, Vlarem, emissions of PCDDs/Fs are limited to 0.1 ng I-TEQ/m³ for all incinerators, 0.1 ng I-TEQ/m³ for refineries, 1.0 ng I-TEQ/Nm³ for existing installations and 0.1 ng I-TEQ/Nm³ for new installations in ferrous industry, 2.5 ng I-TEQ/Nm³ for existing installations and 0.5 ng I-TEQ/Nm³ for new installations in shredders and 0.1 ng I-TEQ/m³ for crematoria. Production of PCBs and PCB containing applications are no longer allowed in Belgium since 1986. Implementation of Directive 96/59/EC in the Flemish legislation set the date for cleaning and/or destruction of the inventoried applications at December 31st, 2005 (instead of December 31st, 2010 in the EC directive). Accidental contamination of cattle feed with PCBs and dioxins in Belgium resulted in several laws preventing contamination of feed and food and limit values for PCBs in cattle feed and in food for human consumption. Because of this legislation, emission of dioxins and furans from industrial sources and waste incineration installations and emissions of PCBs decreased considerably. This is reflected in measured levels of PCDDs/PCDFs in Belgian human milk over time.

Brominated flame retardants were included in the WHO coordinated human milk surveys for the first time in the 4th WHO human milk survey. Compared to the overall Belgian levels in the 4th WHO human milk survey (year 2006), the PBDE levels (sum of BDE28, BDE47, BDE99, BDE100, BDE153, BDE154, BDE183 and BDE209) decreased, but HBCD levels more than doubled, in the Flemish low-populated areas (2009-2010). Comparing HBCD and PBDE levels in Belgian human milk samples of 2006 with results from other European countries of the same time period (2005-2007) shows Belgian HBCD levels comparable with mean levels reported for human milk in Norway[8], higher than levels in French human milk samples[9], but much lower than results reported in a Spanish study[10]. Belgian results for the sum of 7 most abundant PBDEs (28, 47, 99, 100, 153, 154 and 183) in human milk samples from 2006 are comparable with levels reported for human milk samples from France[9] and

Norway[8] in 2005-2007, but much lower than levels in human milk samples from the United States and Canada in 2003[11].

BFRs are widely used in several consumer products/equipment to make them more resistant to fire. BFR legislation differs between countries and continents. In Europe, the use of Penta- and Octa-BDEs in electric and electronic equipment was banned since 2006. Deca-BDE could be used in Europe until June 2008. To replace PBDEs, other flame retardants such as tetrabromobisphenol A (TBBPA) and HBCD, have been increasingly used. Our results show a 20% decrease of PBDEs and a factor 2 increase of HBCD levels in human milk sampled in the current study population (2009-2010), compared to levels measured in the Belgian WHO population (2006), 3-4 years earlier. This might reflect policy measures with respect to BFRs. Since they are good measurable in breast milk, human milk appears a good matrix for follow-up of the BFRs policy.

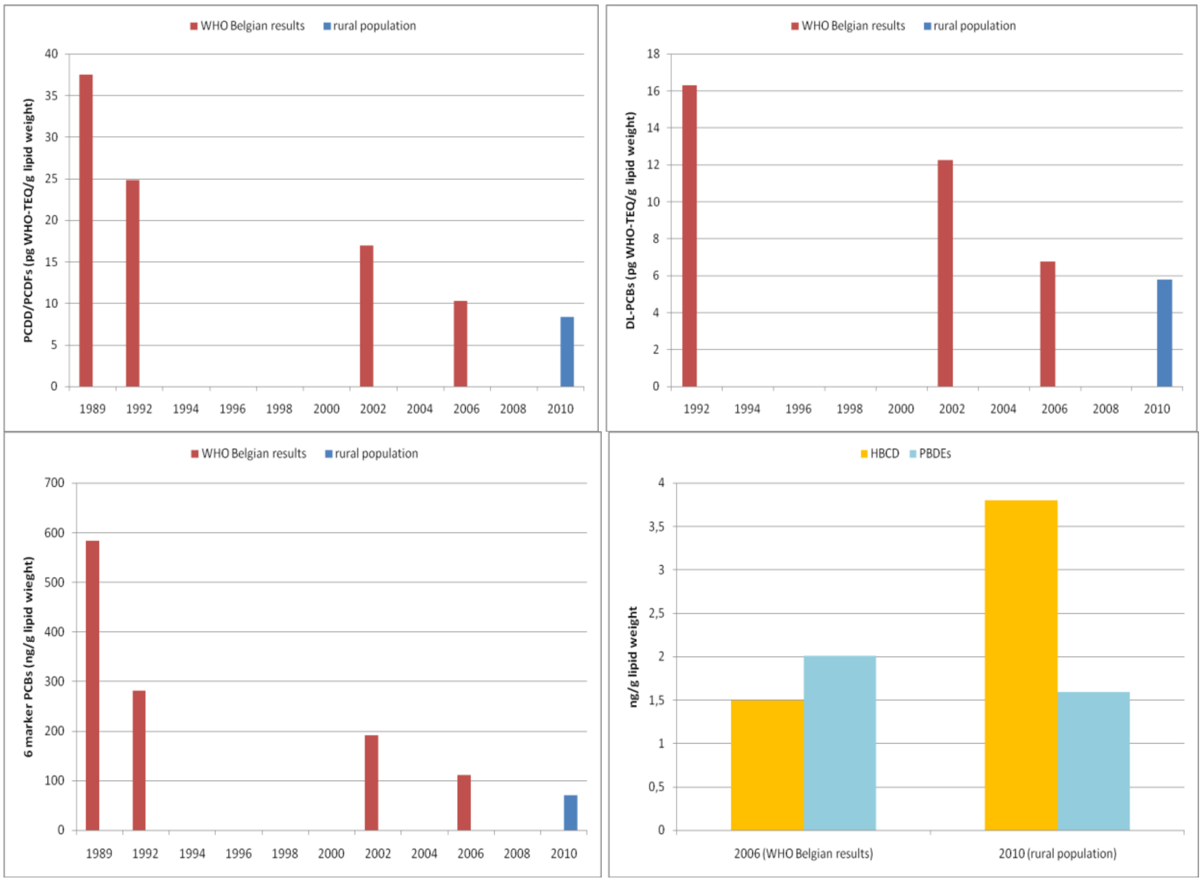

Figure 1: Temporal trends in levels of PCDDs, PCDFs, DL-PCBs (in pg WHO-TEQ/g lipid weight), sum of 6 marker PCBs (PCB28, PCB52, PCB101, PCB 138, PCB153, PCB180), sum PBDEs (BDE28, BDE47, BDE99, BDE100, BDE153, BDE154, BDE183, BDE209) and sum HBCD (in ng/g lipid weight) for the Belgian WHO human milk surveys and levels measured in pooled human milk of the rural study population of 2010.

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