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Long-term time trends in human intake of POPs in the Czech Republic indicate a need for continuous monitoring

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ABSTRACT

Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) from the group of persistent organic pollutants are detected in human tissues years or even decades after their ban. Exposure to PCBs and OCPs can pose risks to human health. In the present study, we calculated the daily intakes of PCBs and OCPs in the Czech population and investigated the long-term trends of human exposure to POPs. Data on POP concentrations from a 16-year period of breast-milk monitoring were used. A toxicokinetic model with consideration of compound-specific elimination half-lives was used to calculate the mothers' daily intake of PCBs and OCPs representing the intake of POPs by all exposure routes. The calculated intakes were compared with dietary intakes calculated by the Czech National Institute of Public Health. The comparison shows good agreement of both intake estimates with decreasing intake trends of POPs in the Czech population in the time period studied. However, several fluctuations with peaks of higher levels were observed in both datasets which are not typical for the period after the ban of use and production of POPs. The available evidence suggests that the increases in chemical concentrations might be caused by food contamination. The calculated intakes of compounds with longer elimination half-lives, such as higher-chlorinated PCBs, were higher in older mothers. This "memory effect" was already observed in other studies and indicates higher exposure in earlier life periods of the mother. Our results suggest that exposure to POPs is still relevant for the Czech population in the period after the ban of the use and production of POPs (post-ban period), especially via food ingestion, though the intake trends are decreasing. Possible food contamination by POPs in the post-ban period requires further assessment.

1. Introduction

Persistent organic pollutants (POPs) have been produced and used for decades all over the world. Although their use was banned or limited several years or even decades ago, they are still present in the environment. They represent risks for environmental and human health (Jepson and Law, 2016; Pumarega et al., 2016). The recognition of the potential threats of POPs led to initiatives on a national and international level, such as the Stockholm Convention (UNEP, 2010) with the mission of regulating the use and production of POPs. Also, several activities are carried out in order to monitor the human exposure to POPs, such as the Global Monitoring Plan (GMP), which has been established to evaluate the effectiveness of the Stockholm Convention (UNEP, 2010; WHO, 2004). Within the GMP, human blood and breast milk have been recommended as core matrices for the human biomonitoring of POPs. Recently, the World Health Organization (WHO) and United Nations Environment Programme (UNEP) global monitoring survey on POPs has reported that the levels of dioxin-like polychlorinated biphenyls (dl-PCBs) have exceed the toxicologically safe levels for breast-fed infants in all of the reported 51 countries (van den Berg et al., 2017). In addition, the levels of indicator PCBs exceed the safety limits for breast-fed children in 46 out of 52 countries reporting

Abbreviations: POPs, persistent organic pollutants; BMI, body mass index; HCB, hexachlorobenzene; HCH, hexachlorocyclohexane; DDT, dichlorodiphenyltrichloroethane; DDE, dichlorodiphenyldichloroethylene; PCBs, polychlorinated biphenyls; OCPs, organochlorine pesticides; CZ-HBM, Czech human biomonitoring; DDX, dichlorodiphenyltrichloroethane and its metabolites; NIPH, National Institute of Public Health; UNEP, United Nations Environment Programme; GMP, Global Monitoring Plan; WHO, World Health Organization; iPCBs, indicator polychlorinated biphenyls; dl-PCBs, dioxine-like polychlorinated biphenyls; EHMS, Environmental Health Monitoring System; SVA, State Veterinary Administration

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indicator polychlorinated biphenyls (iPCBs) and in the Czech and Slovak Republic the highest levels of iPCBs were found (van den Berg et al., 2017). According to the survey, Eastern and Western Europe are the areas with the highest PCB levels measured in human milk. Also, dichlorodiphenyltrichloroethane and its metabolites (DDX) are still present in human milk, especially in tropical countries where they are used for the prevention of malaria. Moreover, DDX have been detected also in countries where they have not been used for decades, including the Czech Republic and Slovakia (van den Berg et al., 2017). DDX in breast milk consumed by breast-fed children may cause early child behavioral problems (Forns et al., 2016; Jönsson et al., 2005).

Studies that monitor human exposure to POPs are generally based on two approaches. Firstly, human internal exposure monitoring studies (human biomonitoring) observe the levels of POPs in human tissue such as breast milk, blood or adipose tissue (Alawi and Al-Tameemi, 2016; Černá et al., 2016; Polder et al., 2008a, 2008b; Zietz et al., 2008). Secondly, external exposure monitoring studies (such as food monitoring) are carried out in order to determine the external doses of POPs to humans (Carlsson et al., 2014; Donat-Vargas et al., 2016; Toms et al., 2016). In the Czech Republic, the levels of POPs in breast milk have been monitored since 1994 within the Czech human biomonitoring (CZ-HBM) program. Also, long-term trends of the food consumption of POPs by the Czech population since 1996 have been monitored (NIPH, 2015). Both monitoring activities are carried out within the Environmental Health Monitoring System (EHMS) executed by the National Institute of Public Health (NIPH) (Czech Government, 1991; Černá et al., 2007, 2016; Mikeš et al., 2012).

Long-term monitoring of POPs in human milk is a useful tool for understanding the time trends of exposure to POPs. Yet, the levels of POPs in human milk by themselves do not inform about the contributions of individual exposure routes to the total or aggregate exposure. In contrast, external exposure monitoring provides information about the external dose for intake through individual exposure routes. Toxicokinetic modeling links external and internal exposure monitoring data and allows for their comparison (Čechová et al., 2016; Gyalpo et al., 2015).

In this study, the levels of PCBs and OCPs in milk from the CZ-HBM breast milk data for the 16-year period from 1996 to 2011 were investigated. In general, the levels show decreasing trends, but the decrease is relatively slow and for most chemicals there are several peaks of higher levels in some years. This makes the time trend different from trends reported for other POPs, which are often exponential (Lignell et al., 2009; Lignell et al., 2012; Nost et al., 2013; Binnington et al., 2016) and are not interrupted by peaks of higher exposure. From the levels in breast milk, we calculated daily intakes of POPs for all mothers and for each year by using a steady-state toxicokinetic model. The calculated intakes were then compared with intakes derived from the food monitoring data in the Czech Republic (NIPH, 2015) for better data interpretation and for a more comprehensive understanding of the current exposure to POPs. The long-term monitoring data of POPs presented here are a valuable source of information that indicates that there is ongoing human exposure to POPs in the Czech Republic although the chemicals were banned many years ago, and that there is a need to identify the sources of this ongoing exposure. The approach and findings of this study extend beyond the Czech Republic; especially countries with similar history and use of POPs can benefit from these findings and application of similar approaches.

2. Methods

2.1. Data characterization and data retrieval

2.1.1. CZ-HBM data for the daily intake calculation

The levels of PCBs and OCPs in breast milk were obtained from the CZ-HBM study. Detailed information of the milk sampling and analysis is provided by Mikeš et al. (2012). In short, integrated sampling of breast milk during the period of 2–8 weeks after birth was performed for each mother. The samples were collected into glass vessels and were stored frozen until analysis. The concentrations of POPs measured in breast milk were normalized for the milk fat content. Detailed information on the monitored population was presented previously (Černá et al., 2007, 2012; Mikeš et al., 2012; NIPH, 2015).

In the present study, concentrations of PCBs and OCPs in milk samples from 4283 mothers obtained in the period from 1996 to 2011 were used to calculate daily intakes of PCBs and OCPs for all mothers. The median number of samples in each year was 286 mothers, with a minimum of 50 and a maximum of 408 mothers. The median age of mothers was 26 years, with the 5th and 95th percentiles being 20 and 34 years, respectively. The median value of the body mass index of the mothers was 24.3 kg m^{-2} with the 5th and 95th percentiles being 19.9 and 31.5 kg m^{-2}, respectively. More of the descriptive statistics of the dataset is provided in Tables S1, S2 and S3 of the Supplementary Material (SM).The original concentrations of PCBs and OCPs in breast milk from which the intake was calculated are presented in Tables S4 and S5 of the SM. Seven iPCBs (PCB 28, 52, 101, 118, 138, 153 and 180), p,p′-dichlorodiphenyltrichloroethane (p,p′-DDT), p,p′-dichlorodiphenyldichloroethylene (p,p′-DDE), hexachlorobenzene (HCB) and two isomers of hexachlorocyclohexane (γ-HCH and β-HCH) were investigated. No data on milk concentrations of any of the compounds are available for the year 2004, as the CZ-HBM was not performed this year. For β-HCH, also data from the year 2001 are missing. For γ-HCH, no data are available for 2001 and 2003.

2.1.2. Food intake data for comparison

The intake data used for comparison were calculated by the Czech NIPH and they are publicly available in the NIPH reports on EHMS (NIPH, 2015). In the reports, detailed information on food analysis and intake calculations can be found. Briefly, a range of typical food commodities were collected from 12 locations in the Czech Republic four times a year. Samples of similar food commodities were combined into composite samples for further analysis (e.g. two types of fermented salami, bread and rolls) and processed, according to standard culinary treatment (cooking, frying, etc.) meeting the typical Czech cuisine (NIPH, 2015). The consumption of specific food items was calculated according to the food guide pyramid, which expresses the dietary recommendations specific for 5 different population subgroups (age and sex dependent) (EFSA/FAO/WHO, 2011; NIPH, 2015). In the present work, the intake calculated for the subgroup of pregnant/lactating women was used for the comparison with the intake of POPs calculated from breast milk concentrations. The Dietary Exposure Monitoring by NIPH has been carried out since 1996 annually, and since 2004 in two-year intervals. For the missing points from the year 2004, the average values of two adjacent years were used for comparison.

2.2. Elimination half-lives

Before the toxicokinetic model was selected, which is introduced further below in Section 3, the time trends in the measured levels in milk were analysed.

In the toxicokinetic model, elimination half-lives of all chemicals are needed. These half-lives should represent the actual elimination from the human body (so-called “intrinsic” elimination half-lives). If half-lives are derived from the longitudinal time trend of levels of a chemical in the body (so-called “apparent” half-lives), these half-lives are often not suitable because the longitudinal time trend may be affected by elimination and continuing intake in parallel and therefore does not describe the actual elimination of the chemical (Shirai and Kissel, 1996; Ritter et al., 2011; Aylward et al., 2014; Russell et al., 2015). The compound-specific human elimination half-lives used for the calculation of intrinsic elimination rate constants were selected from the literature and are listed in Table 1. Half-lives determined from long-term monitoring data were preferred, as suggested by Gyalpo et al.