

Concentrations of polybrominated diphenyl ethers, organochlorine compounds and nitro musks in mother's milk from Germany (Bavaria)

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Abstract

The aim of this study was to determine a new spectrum of substances that will be selected for future breast milk monitoring in Bavaria, Germany. Up to now, the analysis of breast milk in Bavaria was limited to selected organochlorine pesticides (OCP) and polychlorinated biphenyls (PCB). Information on background levels of toxicologically interesting substances, such as dioxins and dioxin-like polychlorinated biphenyls (dl-PCB) or on flame retardants, such as polybrominated diphenyl ethers (PBDE) are very limited or not available for Bavaria. We present here levels on OCP, some nitro musks, indicator PCB, polychlorinated dibenzo-*p*-dioxins (PCDD), polychlorinated dibenzofurans (PCDF) and dl-PCB concentrations in breast milk collected at 12 weeks post-partum of 43 primiparous mothers living in Bavaria. The average concentrations of PCDD, PCDF and dl-PCB were 4.98, 4.93 and 9.92 pg WHO-TEQ g⁻¹ lipid, respectively. The mean contribution of PCDD, PCDF, non-ortho and mono-ortho PCB to the total WHO-TEQ is consistently about 25% each.

Furthermore the concentration on PBDE in breast milk at two sampling points, 12 weeks and 16 weeks after delivery, were determined. Overall, 19 PBDE congeners were analysed, however the level of 12 PBDE congeners were below the limit of detection. BDE-153 and BDE-47 were the predominant congeners accounting for about 66% of the total PBDE. The means of the total concentrations of PBDE (five congeners) at the first and second sampling point were 1.90 and 2.03 ng g⁻¹ lipid, respectively.

Based on our results the overall concentrations of the analysed substances in milk samples from Bavaria are consistent with the levels of breast milk samples of other European countries reflecting the low background body burden of these compounds.

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1. Introduction

Breast milk is a convenient medium for monitoring the body burden of persistent, lipophilic chemicals. They are ubiquitously present in the environment and due to their lipophilic characteristics and low biodegradability these

compounds tend to concentrate in the food chain (Hoover, 1999; Boersma and Lanting, 2000). Through dietary intake, particularly from fish, meat and milk, these compounds bioaccumulate in human adipose tissue and during pregnancy they may be transferred to foetus and postnatal via breastfeeding to the infant (Schechter, 1998; DeKoning and Karmaus, 2000; Wang et al., 2004; Alawi et al., 2006). Epidemiological studies addressing health effects on children related to prenatal or postnatal exposure to substances like PCB, dioxin and organochlorine pesticides

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are conflicting. Some studies found significant association between prenatal exposure to these substances and adverse effects on immunological parameters, reproduction and neurodevelopment (Weisglas-Kuperus, 1998; Patandin et al., 1999; Dewailly et al., 2000; Walkowiak et al., 2001; Longnecker et al., 2003; Nagayama et al., 2007).

Breast milk monitoring programs have been implemented in many countries to assess the human exposure to organohalogen compounds and to predict the infants' daily intake. Since more than 20 years the Bavarian Health and Food Safety Authority carries out breast milk analyses of persistent pesticides and selected organohalogen compounds. On request by the mother breast milk samples were analysed after 4 month of breastfeeding. Since the spectrum of analyzed substances does not comprise chemicals of public concern, the demand on breast milk analyses declined strongly, up to less than 20 samples per year. Hence, given the unreliable data on background levels for the Bavarian population as well as the limited spectrum of analyzed substances, mainly chlororganic pesticides, a new concept for the monitoring of breast milk has been established in Bavaria as part of public health prevention. Details on the new Bavarian breast milk monitoring have been published recently (Raab et al., 2007).

In contrast to data on organochlorine pesticides (OCP) and polychlorinated biphenyls (PCB) only little is known on background levels on dioxins and no information is available on levels of dl-PCB and PBDE in breast milk samples from Bavaria. In the present study we report on OCP, indicator PCB levels and for the first time current PCDD/F, dl-PCB and PBDE levels in breast milk samples from primiparous women living in Bavaria, Germany. Results of perfluorinated compounds in breast milk were previously published by Völkel et al. (2007).

All together, these results will be taken into account to conduct an ongoing breast milk monitoring (BAMBI, Bavarian Monitoring of Breast Milk) that will be adapted regularly to record data on chemicals suspected to be of concern. At the same time, the new monitoring may act as control mechanism to revise legal requirements on regulations, restrictions or bans on certain persistent environmental contaminants as well as support of risk assessment.

2. Materials and methods

2.1. Collection of samples

Eighty-five breast milk samples were collected from primiparous mothers, who delivered their baby in Department for Neonatology of the Children's Hospital at the Department of Gynecology and Obstetrics in Munich during May 2005 and August 2005. The ethic committee of the Bavarian Chamber of Physicians approved the study and written informed consent of all participants was obtained. Detailed informations about the procedures regarding the sample collection together with glass bottles cleaned with acetone and manual suction pumps were sent to the participating

mothers. The mothers collected 12 weeks (1st sampling period, $n = 43$) and 16 weeks (2nd sampling period, $n = 42$) after delivery up to 150 ml of breast milk. The samples were stored at +4 °C during the transport to the laboratory and subsequently stored at -20 °C until analysis. All participants completed a detailed questionnaire on age, weight, height, possible exposure through occupational contact, dietary habits, smoking habits, living area, and other data.

2.2. Statistical methods

Statistical analyses were carried out using SPSS program (version 12.0). Spearman's rank correlation coefficient was used to determine the strength of the association between mother's age and the concentrations of OCP, PCB, PBDE, PCDD/PCDF and dl-PCB. Statistical significance was based on the level of $p < 0.05$. The Mann-Whitney U test was employed to detect differences of concentrations of the analysed substances in breast milk of mothers with different dietary habits. The concentrations of the analysed substances were calculated assuming values below LOQ as half of the LOQ.

2.3. Analytical methods

2.3.1. Determination of OCPs, nitro musks and PCBs

Sample clean-up was performed following the official German procedure developed for the determination of pesticides and PCBs in fatty animal food (*Amtliche Sammlung von Untersuchungsverfahren nach § 35 LMBG (1998), 00.00, 38/2 u. 38/3*). For this reason, 20 g of milk were centrifugated and the separated lipid-phase was ground with water-free Na₂SO₄ to obtain dry and homogeneous material. The dry powder was filled into a micro glass column and eluted with n-hexane/acetone (2:1, v/v). After evaporation of the solvent, 0.5 g of the resulting milk lipid were dissolved in ethyl acetate/cyclohexane (1:1, v:v) and were subjected to gel permeation chromatography (GPC) with bio-beads S-X3 as stationary phase and ethyl acetate/cyclohexane (1:1, v:v) as eluent. After GPC, the solvent was changed to isooctane and the volume reduced to 4 ml.

The samples were analysed for the OCP hexachlorobenzene (HCB), α -, β - and γ -hexachlorocyclohexane (HCHs), oxychlordane, *cis*-chlordane, 1,1-dichloro-2,2-bis(4-chlorophenyl) ethylene (p,p'-DDE), 1,1-dichloro-2,2-bis(4-chlorophenyl) ethane (p,p'-DDD), 1,1,1-trichloro-2,2-bis(4-chlorophenyl) ethane (p,p'-DDT), *cis*-heptachloroepoxide (HCEP) and dieldrin. Furthermore nitro musks (musk xylene, musk ketone and musk ambrette) and six PCB congeners were analyzed. Total PCB was calculated as the sum of PCB 153, PCB 138 and PCB 180 multiplied by 1.64, Total DDT was calculated as the sum of p,p'-DDE and p,p'-DDT multiplied by 1.11.

Sample extracts were measured using a HRGC/ECD-system consisting of a Hewlett-Packard 6890 gas chromatograph equipped with an auto sampler and a

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