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Long-term biomonitoring of polychlorinated biphenyls and organochlorine pesticides in human milk from mothers living in northern Germany

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Abstract

Polychlorinated biphenyls (PCBs) and organochlorine pesticides are persistent organic pollutants that have a widespread distribution in the environment. Human biomonitoring is a suitable tool to assess the burden of humans with these substances. Over a time span of 8 years, a free analysis of their milk was offered to lactating mothers residing in the state of Lower Saxony, Germany. The human milk was analyzed for a number of organic chemicals including polychlorinated biphenyls (PCBs), 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane (DDT), hexachlorobenzene (HCB) and β-hexachlorocyclohexane (β-HCH). Factors that may influence these levels were investigated using a questionnaire. In total, 4314 samples were collected in the years 1999–2006 and analyzed for their content of these persistent organic pollutants (POPs). A clear downward trend of median total PCB, DDT, β-HCH and HCB values in all participants and also in different selected subgroups could be observed. The median values of calculated total PCB in the year 2006 including all participants was 0.1825 mg/kg lipid, that of DDT 0.0815 mg/kg lipid, β-HCH 0.0116 mg/kg lipid and of HCB 0.0229 mg/kg lipid. There were reductions between 40.9% and 47.1% compared to the year 1999. Among other influencing factors, median concentrations of total PCB, DDT, β-HCH and HCB showed a clear rise with increasing age of mothers whereas an increasing number of breastfed infants per mother led to a decrease. The proportions of other measured substances exceeding limits of quantification were as follows: dieldrin 68.6%, α-HCH 1.3%, γ-HCH 60.1%, heptachlor epoxide 41.5%, musk xylene 15.6%, musk ambrette 0.4%. We conclude that the known declining trend of important xenobiotic substances in human milk of German mothers has continued.

Keywords: Persistent organic pollutants; Human milk; Biomonitoring; Polychlorinated biphenyls; PCB; Organochlorine pesticides; DDT; Lower saxony; Germany

Introduction

Human milk is the natural food for infants having the optimal balance of fats, carbohydrates, and proteins. It provides many immunologic, developmental, psychological, economic, and practical advantages (Pronczuk et al., 2002, 2004; Solomon and Weiss, 2002). On the

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other hand, there is concern about the potential risks posed by the presence of chemical contaminants in human milk. Many nonpharmaceutical xenobiotics, particularly those that are lipophilic such as polyhalogenated chemicals, can occur in breast milk (Solomon and Weiss, 2002; LaKind et al., 2001). Polyhalogenated chemicals tend to degrade slowly in the environment, to bioaccumulate in the food chain, and to have long half-lives in human beings. Different adverse health and reproductive outcomes have been attributed to these chemicals in animals and in also humans (Needham and Wang, 2002; Solomon and Weiss, 2002; Landrigan et al., 2002).

In the following, some basic facts of the most important substances that were measured in this study are described. It should be noted that these anthropogenic substances are ubiquitous contaminants in most environmental and biotic compartments (AMAP, 2004).

PCBs have been used for decades in different applications such as in plasticizers, surface coatings, inks, adhesives, flame retardants and paints. Because of their general chemical inertness and heat stability, they have been used in dielectric fluids in transformers and capacitors (WHO, 2003a; ATSDR, 2000). The environmental concentrations of the organochlorine insecticide DDT (1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane) and its metabolites have been decreasing since the ban on DDT was instituted in the United States and most of the world in 1972. The ban excludes selected uses in some parts of the world especially for controlling malaria vectors. The substances 1,1-dichloro-2,2-bis (*p*-chlorophenyl)ethane (DDD) and 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene (DDE) are both degradation products of DDT. DDD was also manufactured and used as an insecticide (ATSDR, 2002a). Hexachlorocyclohexane (HCH) is a synthetic chemical that exists in eight isomers. γ-HCH (lindane) was produced and used as an insecticide in huge quantities and is still available in prescription medicines to treat ectoparasites. Technical-grade HCH is a mixture of several isomers of HCH and was once also used as an insecticide (ATSDR, 2005). The fully chlorinated hydrocarbon industrial chemical hexachlorobenzene (HCB) was used as a pesticide and to make pyrotechnic materials, and synthetic rubber. It is also an unintentional by-product or a minor part of other chemical products (ATSDR, 2002b). Possible health effects of low-level chronic exposure with these substances were reviewed in detail by the WHO (2003b).

Human biomonitoring can be an efficient and cost-effective tool to evaluate internal exposure of humans with different chemical substances. In many cases it can be an integral part of risk assessment and risk management of a substance (Angerer et al., 2007; Jakubowski and Trzcinka-Ochocka, 2005). Some persistent organic pollutants have been measured in human milk since several decades, but only a few countries have systematic human milk monitoring programs that have tested considerable numbers of women over time using consistent sampling methods (UNEP, 2004; Solomon and Weiss, 2002).

Aim of the human milk monitoring program in Lower Saxony was a biomonitoring of important persistent organic pollutants (POPs) offered to all interested mothers residing in this federal state of Germany. Beside the individual information and assessment for the mothers, the data was collected to monitor the general contamination trend of human milk in Lower Saxony with these substances.

Materials and methods

The human milk monitoring program for mothers living in Lower Saxony, Germany, was based on voluntary donors. Chemical analysis of the milk was free of charge for the mothers. Interested mothers were requested to sign a written declaration of consent. Information about the monitoring program was distributed via local physicians (gynecologists, pediatricians), midwives and obstetrical wards using leaflets. Additionally, in the last years detailed information about the program was posted on the department's homepage. Regularly press announcements of the ongoing project and selected results were made by the Ministry of Social Affairs, Women, Family and Health of Lower Saxony.

Sampling kits, comprising a cleaned 250 ml glass bottle with screw cap (NUK/MAPA, Zeven, Germany), a sampling instruction, a cooling gel package and a questionnaire, were sent to those mothers willing to participate. The return of the package by mail was free for the mothers. Participants were asked to collect at least 100 ml of milk, but lower volumes were also accepted. The mothers were requested to wash their hands thoroughly only using a non-perfumed soap before sampling. The breast should only be rinsed with copious amounts of water. The milk should be expressed either manually or by means of a cleaned breast pump, if possible after finishing with breastfeeding. A questionnaire served to asses factors of interest regarding personal characteristics of the mother and the infant. Additionally, different lifestyle factors (e.g. smoking, eating habits, profession, chemical/industrial plants in proximity to home, home moving) had been asked. Samples were collected between January 1999 and December 2006. Milk samples were stored in their original glass sample bottles at a temperature of $-18\,^{\circ}C$.

The fat content of the breast milk samples was determined by the Gerber method following ISO 488:1983-12. First 10 ml H_2SO_4 (p.a., 91%; Fluka/Riedel-de Haën, Seelze, Germany) were filled into a