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GRAPHICAL ABSTRACT

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- medians of indicator PCBs
- ng/g lipids
- Legend: Eastern Siberia (orange), Malsich, van Leeuwen, 2003 (blue)
- | Country | Eastern Siberia (ng/g lipids) | Malsich, van Leeuwen, 2003 (ng/g lipids) |
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| Brazil | 10 | 10 |
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| Hungary | 20 | 20 |
| New Zealand | 25 | 25 |
| China | 30 | 30 |
| Malaysia | 35 | 35 |
| Colombia | 40 | 40 |
| Belgium | 45 | 45 |
| Italy | 50 | 50 |
| France | 55 | 55 |
| Spain | 60 | 60 |
| Sweden | 65 | 65 |
| Finland | 70 | 70 |
| Croatia | 75 | 75 |
| Ukraine | 80 | 80 |
| Belarus | 85 | 85 |
| Belgium | 90 | 90 |
| U.S. Alaska | 95 | 95 |
| The Netherlands | 100 | 100 |
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| Luxembourg | 110 | 110 |
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ABSTRACT

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The aim of our study is to investigate the spatial distribution of polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB), dichlorodiphenyltrichloroethane (*p,p'*-DDT) and its metabolites, α - and γ -isomers of hexachlorocyclohexane (HCH) in 155 samples of human milk (HM) from Eastern Siberia (six towns and seven villages in Irkutsk Region, one village of the Republic of Buryatia and one town in Zabaikalsk Region, Russia), and to examine the dietary and social factors influencing the human exposure to the organochlorines. The median and range of the concentration of six indicator PCBs in HM in 14 localities in Eastern Siberia (114 (19–655) ng g⁻¹ lipids respectively) are similar to levels in the majority of European countries. However, in one village, Onguren, the median and range of levels of six indicator PCBs (1390 (300–3725) ng g⁻¹ lipids) were comparable to levels measured in highly contaminated populations. The Lake Baikal seals are highly exposed to persistent organic pollutants (POPs) and could be a potential source of PCB and DDT exposure in the Onguren cohort via the consumption of the Lake Baikal seal tissue. The location of food production in areas exposed to the emissions of local POP sources can also significantly influence POP levels in HM samples from industrialized areas. Estimated daily intakes (EDI) of HCH and HCB for infants are considerably lower or close to acceptable daily intake (ADI). The EDI of total DDTs and total PCBs are higher than ADI.

1. Introduction

Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) are among the most troublesome organic environmental contaminants. Due to their physical and chemical properties they

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are widely distributed in the environment and accumulated through the food web up to and including humans and influence human health (AMAP, 1998). Food contributes the main part of non-occupational exposure in humans (AMAP, 1998; Hicks, 1996), and populations with a high consumption of contaminated fish and consumption of marine mammals are likely to be highly exposed (Hicks, 1996; Fångström et al., 2005; Grandjean et al., 1995).

The first investigations of persistent organic pollutants (POPs) in the Lake Baikal region started in 1981 within the framework of the Soviet-Swedish cooperation by Bobovnikova et al. (1988, 2001) and later were continued by different teams of scientists from USA, Germany, Canada, Japan and Russia (Schechter et al., 1990; Kucklick et al., 1996; Nakata et al., 1995; Mamontov et al., 2000; Tarasova et al., 1997; Mamontova et al., 1999, 2005, 2007). As a result of those studies elevated levels of POPs including polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), PCBs and OCPs were found in fat of Lake Baikal seal and in some fish from Lake Baikal and the River Angara (Tarasova et al., 1997; Kucklick et al., 1996; Nakata et al., 1995; Mamontov et al., 2000; Bobovnikova et al., 2001; Mamontova et al., 2006). First data on hexachlorocyclohexane (HCH) and dichlorodiphenyltrichloroethane (DDT) levels in human milk (HM) from Irkutsk Region were obtained in the early 1980s (Bobovnikova et al., 1987). In 2000s elevated PCDD/F and PCB levels were found in adipose tissue and HM of residents of Usol'e-Sibirskoe and Onguren (Mamontova et al., 1998, 2005). The investigation of PCDD/Fs and PCBs in soil from the Lake Baikal region confirms that PCDD/F and PCB contamination of soil results from atmospheric transport from a source located close to Usol'e-Sibirskoe, the home to one of the largest chemical complexes producing organochlorines, paints etc. in the former Soviet Union (Mamontov et al., 2000).

The aim of this study is to investigate the spatial distribution of PCBs and OCPs in HM in Eastern Siberia and to examine the dietary and social factors influencing human exposure to PCBs and OCPs.

2. Materials and methods

2.1. Sampling of breast milk

HM samples from 155 mothers were collected between 1997 and 2009 in six towns (Irkutsk, Usol'e-Sibirskoe, Schelekhovo, Ust'-Ilimsk, Baikal'sk, Bratsk) and seven villages (Balagansk, Maloye and Bolshoye Goloustnoe, Kachug, Elantsy, Onguren, Khuzhyr) in Irkutsk Region, one village (Tankhoy) in the Buryatia Republic and one town (Petrovsk-Zabaikal'skiy) in Zabaikal'sk Region (Fig. 1). Six of the towns have at present or had in the past at least one major chemical industry (Usol'e-Sibirskoe – former chemical plant with production of organochlorine compounds; Ust'-Ilimsk – wood industry including a pulp and paper mill (UI-LPK); Baikal'sk – pulp and paper mill; Schelekhovo – cable (using polyvinylchloride), aluminum and silicon plants; Bratsk – wood industry with pulp and paper mill and aluminum plant; Petrovsk-Zabaikal'skiy – former plant of ferrous metallurgy). Irkutsk is the administrative center in Irkutsk Region with 597,846 inhabitants. Kachug, Elantsy and Balagansk are small district centers located at the headstream of the River Lena, on the shore of Lake Baikal and on the shore of the River Angara, respectively. Onguren, Khuzhyr, Tankhoy and Maloye Goloustnoe and Bolshoye Goloustnoe are small villages located near Lake Baikal. So the populations of the localities have their particularities relating to POP exposure as a result of POPs sources and sources of food production including waterbodies where local fish is caught. The Buryat population of settlements located on the shore of Lake Baikal traditionally hunts for the Lake Baikal seal (nerpa) and consume its fat and meat (Petrov, 2009). For example, the settlement of Onguren is a settlement where the residents eat

the meat and blubber of nerpa pups seasonally (in late spring and at the beginning of summer) and use melted seal fat as a food additive and medical treatment the whole year round. A detailed characteristics of residence, lifestyle and dietary habits of women involved in the investigation are presented in Table 1S and Supplementary Information (S.1.).

HM were collected with the assistance of medical staff in pediatric hospitals located in towns and district centers in Irkutsk and in Zabaikal'sk Regions serving the settlements and all small villages surrounding these settlements. The investigation was approved by the Irkutsk Regional Medical Department as well as by the administrations of regional hospitals. The mothers were asked to take part in the investigation and they gave their consent to take part in it. Only healthy mothers with healthy children were involved in the investigation. The mothers breastfed one child and reside in the settlement concerned for at least 5 years. Most HM samples were collected within 2 weeks–2 months of delivery. In total 155 HM samples were collected including 115 from primiparous mothers and 40 from multiparous mothers. Mothers filled in a questionnaire about their diet, anthropometric measurements, smoking, place of employment and income level. The details of lifestyle and dietary habits of mothers are presented in Supplementary Information (S1). Most of the mothers were Caucasian, except in Elantsy and Onguren, where 7 of the 16 and 7 of the 9 mothers respectively belonged to the Buryat population.

HM samples were collected by mothers by expulsion into chemically clean glass bottles after the babies had been fed. Mothers were instructed to wash their hands and breasts with water and soap before collecting the sample. At least 50 mL of milk was collected by each mother. In one case in the town of Ust'-Ilimsk a pooled milk sample of women working in the wood processing industry was combined from ten individual HM samples of 10 mL each. The remaining HM samples were analyzed individually. The milk collected was frozen, transported to the laboratory in Irkutsk and stored at -18°C until analysis.

2.2. Analytical methods

PCB and OCP analyses were performed in the laboratory of the Institute of Geochemistry in Irkutsk using the method described in Mamontova et al. (2007) and in Supplementary information. Briefly, the milk samples (30–50 mL) were thawed and a surrogate and internal standards containing two PCB congeners (21.5 ng PCB-14 and 8.3 ng PCB-65) were added. The milk was freeze-dried, the residue was taken up in *n*-hexane:acetone (1:1), transferred to a pre-weighed flask, the solvent was evaporated, and the lipid weight was determined gravimetrically. The samples were cleaned using a gel permeation chromatography column filled with bio-bead S-X3 and a chromatographic column containing silica gel (3 g), aluminum oxide (3 g), and Na_2SO_4 (3 g). The fraction containing the PCBs and OCPs was evaporated to 30 μL under purified nitrogen.

The GC/ECD analyses were performed on a HP 5890A Series II gas chromatograph using a DB-5 capillary column (J&W Scientific, 0.25 μm film thickness, 0.25 mm inner diameter, 60 m long). All samples were analyzed for 26 congeners of PCBs (IUPAC no., listed in order of elution): 28, 52, 49, 44, 74, 70, 95/66, 101/90, 99, 97, 87/115, 110, 118, 153, 132/105, 138, 158, 187, 183, 180, and 190/170; HCB, *p,p'*-DDT, *p,p'*-DDD, *p,p'*-DDE, α - and γ -isomers of HCH.

Individual PCB congeners, PCB mixture and OCP mixture standards were purchased from the Dr. Ehrenstorfer Laboratory (Augsburg, Germany).

Method recoveries were determined using spiked samples. They lay between 80% and 120% for most compounds. Blanks were run with each batch of 10–12 samples. Only samples in which the analyzable compound level exceeded the level in the blank 3.5

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