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LEVELS AND PATTERNS OF ORGANOCHLORINE PESTICIDES, POLYCHLORINATED BIPHENYLS, POLYBROMINATED DIPHENYL ETHERS AND HEXABROMOCYCLODODECANE IN SELECTED FOODS FROM NORTH-WEST RUSSIA AND IMPLICATIONS FOR DIETARY EXPOSURE

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Introduction

Due to their chemical persistence and lipophilicity persistent organic pollutants (POPs) accumulate and biomagnify in the food chains. It is assumed that > 90% of POP levels in humans have their origin in consumption of contaminated food[1]. Investigation of potential human health consequences through the ingestion of contaminated food is a matter of concern worldwide and many nations have developed monitoring food contaminants programs. In EU, control of contaminants in food and feed is regulated through Council Regulation 315/93/EEC. In Russia, after our knowledge, no such monitoring program existed by 2002. Earlier investigations of persistent organic pollutants (POPs) in human breast milk from North-West Russia (1993-1996) showed that levels of certain organochlorine pesticides (OCPs) were 2-10 times higher and varied much more than corresponding levels in Norway[2]. Food was suggested as the main cause of these differences. In many countries levels of organochlorines (OCs) declined after bans and restrictions were put into force. However, the presence of "new pollutants" like polybrominated diphenyl ethers (PBDEs) in the environment have become of increasing concern[3]. PBDEs were detected in low levels in Russian breast milk in 2000-2002[4], and are thus present in the Russian environment. The objectives of the present study were to elucidate the levels and patterns of POPs, PBDEs and HBCD in selected foods from North-West Russia and estimate the impact of these contaminants on human dietary exposure in this geographic area.

Figure 1. Map of the Barents region and sampling locations in Russia

Materials and methods

Sampling and collection:
During 1998-2002, different foods were purchased from supermarkets, shops, local markets and/or from local citizens in the cities and surroundings of Murmansk, Arkhangelsk, Narjan-Mar and Kargopol in North-West Russia (Figure 1). Some foods were staple foods of indigenous people. The samples were frozen at -20°C and transported to Norway for chemical analyses.

Chemical analyses, separation and determination:
Concentrations of hexachlorobenzene (HCB), α-, β-, γ-hexachlorocyclohexane (∑HCHs), oxychlordane, *cis*-chlordane and *trans*-nonachlor (∑CHLs), *bis*-2, 2-(4-chlorophenyl)-1,1,1-trichloroethane (*p,p'*-DDT) and its metabolites *bis*-2, 2-(4-chlorophenyl)-1,1-dichloroethane (*p,p'*-DDD) and *bis*-2, 2-(4-chlorophenyl)-1,1-dichloroethylene (*p,p'*-DDE) (∑DDTs), Mirex, polychlorinated biphenyls (∑PCBs), IUPAC nos.: CB-28, -52, -74, -99, -101, -105, -118, -128, -138, -153, -156, -157, -170, -180, -187 and -194, polybrominated diphenylethers (∑PBDEs) BDE-28, -47, -99, -100, -153, -154 and -209 and hexabromocyclododecane (HBCD) were measured in the Russian food samples at the Laboratory of Environmental Toxicology at The Norwegian School of Veterinary Science, Oslo, Norway. The laboratory is accredited by Norwegian Accreditation for testing the analysed chemicals in biological material according to the requirements of the NS-EN ISO/IEC 17025 (TEST 137). The analytical quality of the laboratory is frequently approved in intercalibration tests. The analytical method includes in short homogenization of the sample before weighing, extraction with cyclohexane and acetone (3:2) using an ultrasonic homogenizer, gravimetrical lipid determination and lipid removal by concentrated ultra-clean H_2SO_4. Separation and determination of OCs was performed on a GC-ECD, and of PBDEs and HBCD on a GC-MS. Blank values of PBDEs were subtracted. Further details on the analytical method, separation, determination, calculation and quality control were described earlier[4].

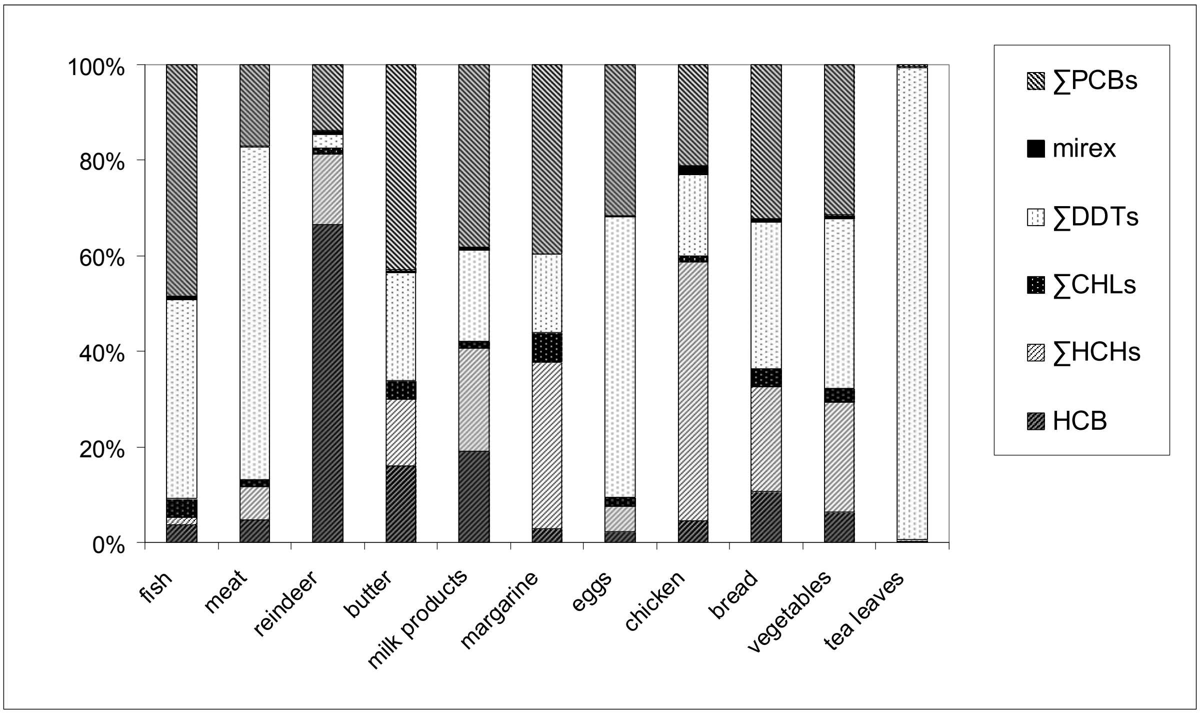

Figure 2. Relative contributions of HCB, ∑HCHs, ∑CHLs, ∑DDTs, mirex and ∑PCBs to ∑OCs in different foods from Russia.

Results and discussion

OCPs and PCBs:

Highest median concentrations of ∑OCs (sum of HCB, ∑HCHs, ∑CHLs, ∑DDTs, mirex and ∑PCBs) in foods were measured in fish (8.7 ng/g wet weight (ww)) and butter (8.7 ng/g ww), followed by meat (6.4 ng/g ww), chicken (5.7 ng/g ww), eggs (5.6 ng/g ww), and margarine (4.1 ng/g ww). Median ∑OC levels in milk products, reindeer, bread and vegetables were 2.8, 1.7, 0.5 and 0.3 ng/g ww, respectively. POP levels varied sometimes considerably within the food groups. In butter, for example, levels of ∑DDTs ranged from <LOD-16 ng/g ww. In Denmark, ∑DDTs ranged from 2 (Danish) to 11 ng/g ww (foreign), showing country dependent variations[5]. Levels of ∑DDTs and ∑PCBs varied also considerably between and within the meat products, as in pork fat (range of ∑DDTs: 0.3-130 and for ∑PCBs 1-24 ng/g ww). The range of ∑DDTs was wider in pork fat from Russia compared to Denmark. Variations of PCB levels in pork fat were however similar in Russia and Denmark[5]. Investigations of POPs in feed for domestic animals in Russia could contribute to understand some of the differences found in this study. Tea leaves originating from Georgia showed elevated levels of ∑DDTs (298 ng/g ww), exceeding maximum residue limits (MRL) of 0.1 mg/kg. In most of the foods DDTs and/or PCBs were the major OCs. However, HCB dominated the OC pattern in reindeer and HCHs dominated the OC pattern in chicken meat (Figure 2). ∑OCs were in general 1-2 times higher in Russian foods compared to Norwegian food[6].

In order to obtain information on levels of non-dioxin-like PCBs (NDL-PCBs) in the Russian foods, the sum of 6 indicator PCBs (∑NDL-PCBs$_6$: CB-28, -52, -101, -138, -153, and -180) and the contribution of ∑NDL-PCBs$_6$ to ∑PCBs were calculated. This information is relevant for the establishing of maximum levels for non-dioxin-like PCBs (NDL-PCBs) in food, which is planned by The European Commission. The European Food Safety Authority (EFSA) has published a risk assessment on non-dioxin-like (ndl) PCBs in food and feed, followed by similar risk assessments done in several other countries as Norway[7]. In the Russian foods, ∑NDL-PCBs$_6$ contributed 38-60% to ∑PCBs. This is in the same range (38-59%) as reported in European and Norwegian foods[8]. In Norwegian foods, PCB-153 is the main contributor to ∑NDL-PCBs$_6$ (15-50%). Corresponding percentages were 19-49% in the Russian foods, and thus in the same range.

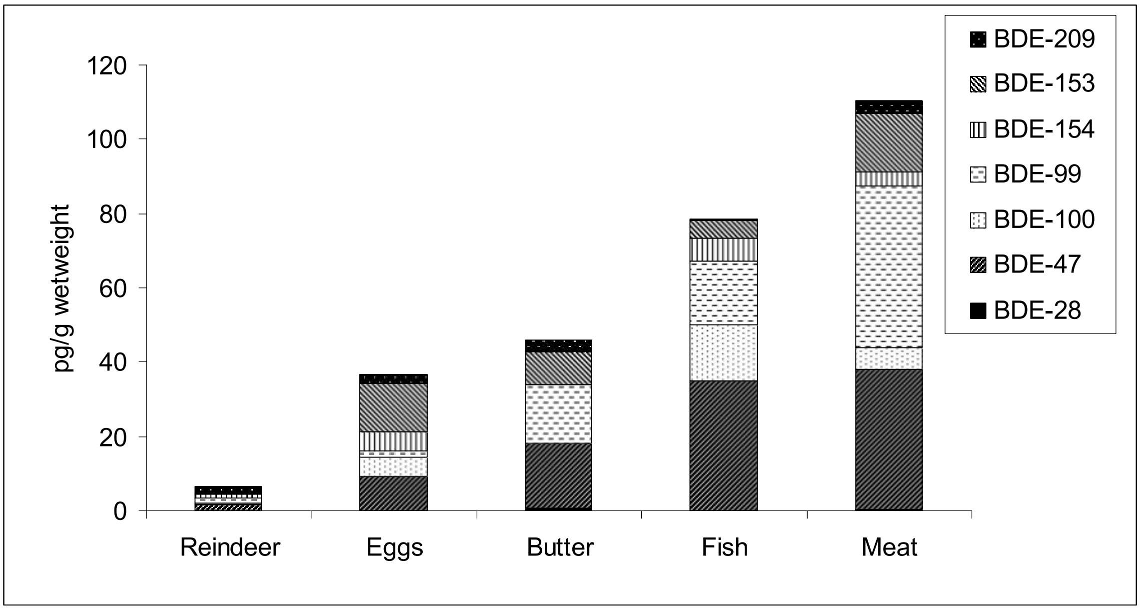

Figure 3. Concentrations and composition of ∑PBDEs and BDE-congeners in Russian foods

PBDEs and HBCD

PBDEs were analysed in meat products (n=6), fish (n=2), butter (n=3), eggs (n=1) and reindeer (n=1). Highest levels were measured in meat (111 pg/g ww) > fish (79 pg/g ww) > butter (46 pg/g ww) > eggs (37 pg/g ww) > reindeer (7 pg/g ww) (Figure 3). The BDE-pattern differed between the foods (Figure 3). In meat products, the major congener was BDE-99 contributing 39% to ∑PBDEs, followed by BDE-47, contributing with 34%. In fish and butter BDE-47 was the dominating congener, contributing 45 and 38% to ∑PBDEs, respectively. In eggs, the major congener was BDE-153 (35%) and secondly BDE-47 (26%). In reindeer the pattern was dominated by BDE-209 (34%), and secondly by BDE-47 (26%). HBCD was only measured in fish (66 pg/g ww). PBDEs levels in Russian fish and butter were 6-fold lower than in Belgium[9]. However, PBDE levels in Russian meat were 3 times higher than in Belgium. Levels of PBDEs in Russian eggs were comparable and 2-fold lower to findings in Belgium and USA, respectively. In general, PBDE levels in Russian foods were far much lower compared to findings in United States foods[10].

Dietary exposure of POPs

The daily intake was calculated based on information from personal interviews on dietary habits and a few Russian unpublished reports. Therefore, the interpretation of the presented results should be done with caution. It seemed that 50% of the daily intake of HCB was derived from milk products, 34% of HCH from chicken, and 21 and 34% of CHLs from bread and fish. Possible usage of margarine or fat based on marine oil products in the production of bread might explain the high contribution of CHLs. For ∑DDTs, 22, 28 and 32% of the daily intake was derived from milk products, fish and meat, respectively. For PCB-153, corresponding percentages were 25, 41 and 21%, and for ∑PCBs, 26, 38 and 13%, respectively. Compared to the Danish food monitoring study, it seemed that the contribution of milk products to the daily intake of HCB was higher in Russia than in Denmark, that fish contributed less to intake of ∑DDTs but similarly to intake of PCB-153 in Russia and Denmark.

Conclusion

This study showed that ∑OC levels in Russian foods were 1-2 times higher than corresponding levels in Norway. PCBs and DDTs were major contributors to ∑OCs in most of the foods, but not in reindeer and chicken. PBDE levels in the Russian food were comparable with other European countries or lower. BDE-209 was found present in all the analysed foods. Although there was limited information on dietary habits this study provided important results on the major food sources for human exposure to POPs.

Acknowledgements

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