

Temporal Changes of PCBs, PCDD/PCDFs and Chlorinated Pesticides in Human Milk from Murmansk, Russia, and Tromsø, Norway.

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Introduction

Breast milk monitoring programs have been performed in several countries for investigating geographical and temporal trends in human exposure to persistent organic pollutants (POPs) such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polychlorinated dibenzo-p-dioxins and /dibenzofurans (PCDDs/PCDFs)¹. In 1998, we reported on high levels of OCPs in human milk from the Kola Peninsula, Russia². As a result of restrictions on production and use of these chemicals, a decline in the human body burden of chlorinated POPs has been observed in most western countries during the last decades^{3, 4, 5}. However little is so far known about the temporal trends in Northern Russian populations. This paper presents the first study on temporal trends of selected POPs in Russia covering a time period from 1993 to 2002. The results are compared with those for a parallel investigation in Tromsø, Norway.



Figure 1. Map of northern Norway and Russia, indicating the locations of Tromsø and Murmansk.

Materials and methods

Sampling and collection: In 1993 and 2000 breast milk from 8 and 14 primiparous mothers were collected within 5 days after delivery in Murmansk on the Kola Peninsula, Russia (Figure 1). During the same years, respectively 10 and 12 breast milk samples of primiparous mothers were collected and pooled in Tromsø within WHO-coordinated studies ^{4, 6}. Samples were collected between 2 weeks and 2 month after delivery. Details of the mothers, such as health status, age, occupation, and dietary habits were obtained. The age of the primiparous mothers was similar for the two time periods with a mean of 22 yrs for Murmansk and 26 yrs for Tromsø, respectively.

Determination of PCBs and OCPs: Concentrations of hexachlorobenzene (HCB), sum hexachlorocyclohexane (α -, β -, γ -HCH), sum chlordanes (oxychlordane, *cis*-chlordane and transnonachlor), sum DDTs (p,p'-DDT, p,p'-DDE and p,p'-DDD), and the sum of 16 PCBs, IUPAC nos.: 28, 52, 74, 99, 101, 105, 118, 128, 138, 153, 156, 157, 170, 180, 187 and 194 were measured at the Norwegian School of Veterinary Science. The extraction, lipid clean-up and GC-ECD analyses were done according to methods described earlier ^{7, 8}. PCBs 29, 112 and 207 were used as internal standards. The lipid concentration of the milk was determined gravimetrically.

Determination of PCDDs/PCDFs and non-ortho PCBs: Determination of PCDDs/PCDFs and non-ortho PCBs was performed with GC-HRMS at the Norwegian Institute of Public Health in pooled samples from each of the two time periods studied, as described earlier ⁹.

Results and discussion

OCPs and PCBs: Except for sum chlordanes, the levels of OCPs were considerably higher in breast milk from Murmansk compared to Tromsø (Table 1). During the studied time period, a decline in concentrations of all the studied OCPs and PCBs in human milk was observed at both sites. Sum HCHs showed the highest percentage of reduction both in Murmansk and Tromsø. The largest difference in decline between the two sites was observed for sum DDTs with 42 % in Murmansk and 26 % in Tromsø. However in 2000 the concentrations of sum DDTs in the Russian town were still about 5 times higher compared to Tromsø. Concentrations of sum chlordanes and sum 16 PCBs were not so much different between the two sites. The levels of sum DDTs and sum HCHs in human milk from Murmansk in 2000 were comparable to corresponding levels in Norway in 1982

¹⁰.

Table 1. Median concentrations of HCB, sum HCHs, sum chlordanes, sum DDTs, sum PCBs and sum OCs ($\mu\text{g/kg}$ milk fat) in human milk from Murmansk and Tromsø in 1993 and 2000.

| | Murmansk 1993 <i>n</i> =8 | Murmansk 2000 <i>n</i> =14 | % decrease | Tromsø 1993 <i>n</i> =10 | Tromsø 2000 <i>n</i> =12 | % decrease |
|-----------------------|---------------------------------|----------------------------------|------------|--------------------------------|--------------------------------|------------|
| <i>HCB</i> | 139 | 65 | 54 | 44 | 24 | 46 |
| Sum HCHs | 1007 | 223 | 78 | 39 | 12 | 70 |
| Sum chlordanes | 45 | 22 | 51 | 43 | 23 | 47 |
| Sum DDTs | 1610 | 928 | 42 | 261 | 195 | 26 |
| Sum 16 PCBs | 519 | 348 | 33 | 390 | 262 | 33 |
| SUM OCs | 3320 | 1585 | 52 | 778 | 514 | 34 |

Sum HCHs = sum α -, β - and γ -HCH

Sum chlordanes = sum Oxychlordane, *cis*-chlordane and *trans*-nonachlore

Sum DDTs = sum *pp*-DDE, *op*-DDD, *pp*-DDD, *op*-DDT, *pp*-DDT

Sum PCBs = sum PCBs, IUPAC nos.: 28, 52, 74, 99, 101, 105, 118, 128, 138, 153, 156, 157, 170, 180, 187 and 194

This study confirms earlier findings that the environmental pollution with organochlorine pesticides HCHs and DDTs is much higher on the Kola Peninsula than in northern Norway⁸. Exposure through contaminated food was suggested as the main route of exposure for the Russian population. High levels of DDTs and HCHs were found in different foodstuffs collected in northern Russia. It was concluded that fish and meat (particularly pork) were the primary sources of DDT, whereas chicken and vegetables were main sources of HCHs to dietary exposure in northern Russia¹¹. Foodstuffs and animal feed have mainly been imported from the southern parts of Russia, the Russian Federation and Ukraine where HCHs and DDTs are still used to a large extent¹².

PCDDs/PCDFs and non-ortho PCBs:

The concentrations of PCDDs/PCDFs and dioxin-like PCBs (non-*ortho* and mono-*ortho* PCBs) in pooled samples were expressed as WHO₁₉₉₈¹³ toxic equivalents (WHO-TEQs) and the percentage of reduction for the studied time periods and areas and are shown in Table 2. The TEQs of the non-*ortho* PCBs showed the most obvious reduction, which was clearest demonstrated in Tromsø compared to Murmansk. TEQ PCDDs/PCDFs and TEQ mono-*ortho* PCBs showed a more moderate decrease in both areas. The level of TEQ mono-*ortho* PCBs in Murmansk was about twice as high as in Tromsø, otherwise only small differences were found between the two geographical areas studied. The contributions of PCDDs/PCDFs, non-*ortho* PCBs and mono-*ortho* PCBs to the total TEQ in 2000 were 20, 19 and 47 % in Murmansk and 18, 21 and 36 % in Tromsø respectively. Levels of PCDDs/PCDFs presented in this study for 2000 were similarly low as those reported in other industrialized countries¹³. The levels of dioxin-like PCBs in Murmansk 2000 were however at the higher end of the range found for European countries.

Table 2. TEQs for PCDD/PCDFs, non-*ortho* and mono-*ortho* PCBs and total TEQs in human milk from Murmansk and Tromsø* measured in 1993 and 2000.

| | Murmansk 1993 <i>n</i> =8 | Murmansk 2000 <i>n</i> =14 | % decrease | Tromsø* 1993 <i>n</i> =10 | Tromsø* 2000 <i>n</i> =12 | % decrease |
|-----------------------------------|---------------------------------|----------------------------------|------------|---------------------------------|---------------------------------|------------|
| <i>TEQ</i> | | | | | | |
| <i>PCDD/PCDFs</i> | 15.8 | 10.0 | 37 | 11.6 | 8.9 | 23 |
| TEQ non-<i>ortho</i> PCBs | 11.7 | 5.6 | 52 | 16.7 | 4.5 | 73 |
| TEQ mono-<i>ortho</i> PCBs | 21.9 | 13.6 | 38 | 12.7 | 7.6 | 40 |
| total TEQ | 49 | 29 | 41 | 41 | 21 | 49 |

* Becher et al. 2002

Conclusion

In conclusion, this first temporal study of POPs in breast milk from a population in northern Russia showed a decrease of concentrations of all the studied chemicals from 1993-2000. This is similar to the findings for northern Norway, however levels for sum HCHs and DDTs are still much higher in breast milk from Murmansk and further measures to reduce human exposure to these POPs is warranted.

Also, the relatively low concentrations of POPs in Norwegian human milk still decreased during the studied time period. This confirms the positive effects of measures to reduce human exposure to POPs.

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