Human Exposure I

Contaminants in Swedish human milk. Decreasing levels of organochlorine and increasing levels of organobromine compounds.

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Introduction

Persistent lipophilic organohalogen contaminants accumulate in the human body. They are excreted with milk and transferred to the breast fed infant. The levels are strongly correlated to the fat content of the milk and reflect the accumulated levels in the adipose tissue. Accordingly, milk constitutes a suitable matrix for studies of long term exposure to organohalogen pollutants and for studies of time related trends in environmental contamination. In Sweden the use of several pesticides (DDT, aldrin, dieldrin, chlordane) was prohibited in the early 1970's. The use of polychlorinated biphenyls (PCBs) was restricted in 1972 and all PCB products used in transformers were to be replaced by 1995. For polychlorinated naphthalens (PCNs) and polybrominated diphenyl ethers (PBDEs) no restrictions are applied.

The present report on trend studies of organohalogen compounds in milk includes for most of the compounds the period from 1972 to 1992 or 1997. Although DDT compounds and PCB were determined already in 1967 (1). The early investigations of pesticides and PCBs have by time been completed by congener specific analysis of PCBs, dioxins, polychlorinated naphthalens (PCNs), methylsulphonyl metabolites of DDE and PCBs and polybrominated diphenyl ethers (PBDEs).

Materials and Methods

The milk was from native Swedish mothers living in the Stockholm region. Equal amounts of milk from individual mothers were mixed to pooled samples. In order to get comparable samples, the composition of pools was kept as equal to that of the early sampled milk as possible with 55-65 % of the milk from mothers nursing their first infant. The average age of the mothers was in 1972-1985 27-28 years, in 1988-1994 29-30 years and in 1996-1997 30-31 years. The increase is consistent with the general increase of the age of the mothers giving birth in Sweden (2).

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The analytical procedures have been previously described (3-4). Purification and separation of compounds were made by chromatography on aluminium oxide, silica gel, activated charcoal and gel permeation chromatography on Bio-Beads. Identification and quantification were made by gas chromatography electron capture detection and gas chromatography/mass spectrometry using selected ion recording. Slight modification of the method enabled analysis of PBDEs (5).

Results and Discussion

The present report includes previously reported results (6-8) as well as results from recently sampled milk and reanalysis of samples for retrospective studies of compounds not previously determined in the milk.

Decreasing levels

Due to measures taken to reduce the environmental pollution, the levels of organochlorine compounds have decreased in human milk. For the reported compounds the coarse of decline is exponential with half-lives of 4-15 years, Figures 1 and 2.

DDT compounds. The most consistent decline is noticed for DDT and DDE. The level of DDT was in 1997 only 1 % of the level in 1967. The decrease of its metabolite DDE, originating from food of animal origin and individual metabolism, was not obvious until after 1972, and the half-life is longer. The recently discovered methylsulfonyl metabolites of DDE (MeSO₂-DDE) have decreased in parallel to those of DDE. Fig.1.

PCBs. The concentrations of PCBs have fluctuated during the period from 1967 to 1997. The level of total PCBs was in 1997 about 30% of that in 1972, and the rate of decline is much lower (half-life 14 years) than for DDT (4 years).

PCNs. Recently, PCNs were identified in human milk (8) and the analysis revealed that, as for other organochlorine compounds, the levels were higher in the early 1970s. In 1992 the sum of PCN congeners was 16% of the level in 1972 and the half-life was calculated to 8 years.

TEQs. The toxic equivalents (TEQs) were calculated by using the toxic equivalency factors (TEF) proposed for PCDDs/PCDFs (9) and PCBs (10). The main contribution to TEQs was from 3,3',4,4',5-pentaCB, followed by 2,3,4,7,8-pentaCDF and 2,3,3',4,4',5-hexaCB. The total TEQs have decrease from 100 in 1972 to 39 pg/g lipid in 1972, showing a half-life of 15 years.

Increasing levels

Polybrominated diphenyl ethers (PBDEs). In contrast to organochlorine compounds, the concentration of brominated diphenyl ethers in human milk have increased continuously since 1972. In all time periods, 2,2',4,4'-tetraBDE was the predominant congener (5). For the first time the start of a contamination is demonstrated in breast milk. The levels show the coarse of an exponential increase from 1972 and a doubling time of 5 years, Fig. 2. In 1997 the levels reached a concentration (sum of congeners) of 4 ng/g lipid.

Conclusions

The levels in milk reflect the mothers' life time exposure and the general pollution at the time. The decrease in the levels of organochlorine compounds confirmes the positive effects of prohibitions applied to the usage of the compounds. However, the rate of decline is slow

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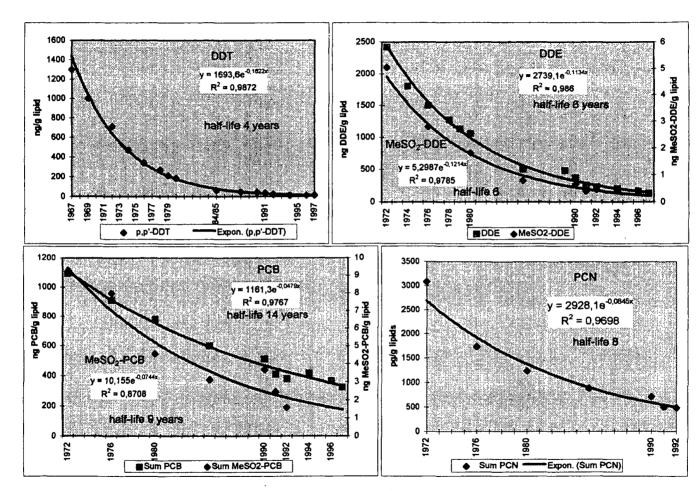


Fig. 1. Concentrations of certain organochlorine contaminants in human milk sampled during different years.

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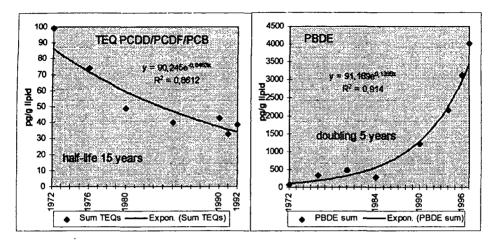


Fig. 2. TEQ levels and sum of the concentrations of PBDE congeners in human milk sampled during different time periods.

for PCBs and dioxin-like compounds, indicating a prolonged contamination by these compounds. For PBDEs no restrictions are applied in Sweden. The exponential increase in the levels in human milk is of great concern and calls for measures to stop the usage of PBDEs.

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