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Persistenta organiska miljöföroreningar i bröstmjölk från förstföderskor i Uppsala 2002 - 2003

Utfört av
Livsmedelsverket

Programområde
Hälsorelaterad miljöövervakning
Kontrakt nr 215 0210

Sakrapport till Naturvårdsverkets Miljöövervakning:

Persistenta organiska miljöföroreningar i bröstmjölk från förstföderskor i Uppsala, 2002-2003.

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Utförare:	**Livsmedelsverket**
Programområde:	**Hälsorelaterad miljöövervakning**
Delprogram:	**Exponering via livsmedel**
Undersökningar/uppdrag:	**Analys av persistenta organiska föreningar i bröstmjölk – tidsserie:** **1. PCB:er (13 kongener, ej plana)** **2. Klorerade pesticider** **4. PBDE (9 kongener) och HBCD** **5. Plana PCB:er (non-orto, 4 kongener)** *Förseningar av analyserna. Resultaten kommer att redovisas i samband med den kommande tidstrendsrapporten.*

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Report to the Swedish Environmental Protection Agency, 2003-11-05

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Persistent organic pollutants (POP) in breastmilk from primiparae women in Uppsala County, Sweden, 2002-2003.

Background

For risk assessment purposes, the Swedish National Food Administration has made recurrent measurements of levels of selected persistent organic pollutants (POP), chiefly polychlorinated biphenyls (PCBs) and persistent pesticides, in human breast milk. Also polybrominated diphenyl ethers (PBDEs) and HBCD (hexabromocyclododecane), ubiquitously used flame retardants, have been added to this list because of results suggesting the PBDE group as candidate for a new environmental hazard. The ambition is to follow changes in the levels of these environmental contaminants in human breast milk and to establish a time trend. The aim is also to evaluate possible health risks for the mother and in particular for the breastfed infant.

The first sampling of breast milk took part in Uppsala County in 1996-1998. About 300 primiparae participated in this large study that to some parts was financially supported by the Swedish EPA (Environmental Protection Agency). The second sampling took part in Uppsala in 2000-2001 (N=31) and the third sampling was performed in Uppsala in 2002-2003 (N=31). The investigations in 2000-2001 and 2002-2003 were partly financed by the Swedish EPA.

Possible regional trends are also going to be investigated by sampling of breast milk from different parts of Sweden. Samples have been obtained from Gothenburg mothers and sampling activities are proceeding in Lund (southern Sweden) and Lycksele (northern Sweden), also financially supported by the Swedish EPA.

The following report summarises results from the sampling carried out in Uppsala in 2002-2003.

Material and methods

The breast milk was exclusively sampled from primiparas in order to minimise variation. Mothers were recruited among primiparas who were Swedish by birth and delivered at Uppsala University Hospital from March 2002 to February 2003. Women who delivered during the first week in every month and on randomly selected days during this week were asked to participate in the breast milk study. Every month, 2-3 primiparas were recruited and the goal was to recruit totally 30 mothers. The mothers who agreed to participate got instructions on how to collect the milk at home, and they also got a breast milk pump and sampling bottles. The mothers sampled the milk during the third week after delivery (day 14-21 post partum) and they were instructed to sample milk both in the beginning and at the end of the breast-feeding sessions. The goal was to sample a total of 500 ml from each mother during 7 days of sampling. During the sampling week the milk was stored cold, preferably in a freezer. At the end of the sampling week a midwife visited the mother to collect the bottles. A blood sample and a hair sample was also collected from the mother. The blood sample was centrifuged and the serum was stored in a freezer. The midwife helped the mother to answer a questionnaire about lifestyle, medical history and complications during pregnancy, and also

gave the mother a more extensive questionnaire chiefly about dietary habits. The mothers answered this questionnaire at home at a later time-point and sent it to the NFA. After analysis, the remaining parts of the breast milk samples were stored for possible analysis in the future.

All analyses were performed at the NFA. The PCBs and chlorinated pesticides were analysed according to a method for breast milk analysis described in Aune et al. (1999). PBDEs and HBCD were extracted according to Atuma et al. 2000 and analysed using GC-MS with negative chemical ionization (NCI).

Results

Table 1 gives a summary of basic data for the women participating in the study (age, body mass index before pregnancy, weight reduction from delivery to sampling, weight of the child, smoking habits, complications during pregnancy and breast-feeding).

Table 1. Data on age, BMI (body mass index), weight reduction, birth weight of the child, smoking habits, complications during pregnancy and breast-feeding for the women participating in the study.

	N	**mean**	**median**	**min**	**max**
Age (years)	31	30	30	24	37
BMI (kg/m^2) before pregnancy	31	21.9	21.6	18.5	26.0
Weight reduction from delivery to sampling (kg)	29	10.1	10.0	6	18
Birth weight of the child (g)	31	3461	3470	2650	4255
	N	**percent**			
Smoking habits	30				
Non-smokers	24	80			
Former smokers	2	7			
Smokers	4	13			
Complications/diseases during pregnancy	31				
No	26	84			
Yes*	5	16			
Breast-feeding	24				
Complete, day 0-35 after delivery	21	88			
Incomplete, day 0-35 after delivery	3	12			

*for example high blood pressure

Table 2 shows the results from the analysis of PCBs (N=31) in the breast milk samples, the individual congeners as well as the sum of the congeners (sumPCB). LOD (limit of detection) was 20 pg/g milk, which corresponds to 0,4-1,3 ng/g milk fat. Values below the LOD were set to half the LOD in the calculations of mean and median for the individual PCB congeners and in the calculation of sumPCB. In the individual samples congeners with levels below LOD constitutes 0,2-7% of the sumPCB-levels. The congeners with the highest levels were PCB 118, 138, 153, 170 and 180. These congeners constitutes about 90% of the sumPCB-levels. Mean and median have not been calculated for the congeners 52, 101 and 114 since over 90% of the results were below the LOD, but these congeners were included in the calculations of sumPCB.

Table 3 shows the concentrations of chlorinated pesticides (HCB, beta-HCH, oxychlordan, transnonachlor, different DDT-isomers and the sumDDT) in the breast milk samples (N=31).

LOD (limit of detection) for the DDT-isomers was 40 pg/g milk, which corresponds to 0,9-2,6 ng/g milk fat. Values below the LOD (limit of detection) were set to half the LOD in the calculations of mean, median and sumDDT. In the individual samples congeners with levels below LOD constitutes 0,3-7% of the sumDDT-levels. The dominating congener, p,p-DDE, constitutes 84-96% of the sumDDT-levels. Mean and median have not been calculated for o,p-DDT and p,p-DDD since the levels were below the LOD in 30 and 31 of the 31 samples respectively. o,p-DDT and p,p-DDD were included in the calculations of sumDDT.

Table 2. Concentrations of PCB congeners in breast milk samples (ng/g milk fat) from 31 primiparas. Values below the LOD (limit of detection) were set to ½LOD in the calculations of mean, median and sumPCB. Mean and median have not been calculated for PCB 52, 101 and 114 since over 90% of the results were below the LOD, but these congeners were included in the calculations of sumPCB

	mean	median	min (lowest value >LOD)	max	No. of samples <LOD
PCB 28	1.84	1.33	0.58	8.09	9
PCB 52	-	-	-	-	31
PCB 101	-	-	0.50	1.00	28
PCB 105	1.14	1.15	0.63	3.06	9
PCB 114	-	-	0.73	1.07	29
PCB 118	7.92	7.76	3.83	16.2	0
PCB 138	23.9	23.3	11.1	47.3	0
PCB 153	46.5	43.1	21.8	97.3	0
PCB 156	3.19	2.80	1.44	7.77	1
PCB 157	0.60	0.45	0.58	1.85	22
PCB 167	0.94	0.94	0.51	1.97	9
PCB 170	9.96	9.37	4.90	19.7	0
PCB 180	22.4	20.4	10.9	46.5	0
sumPCB	120	111	56.4	241	-
% fat	2.9	2.9	1.5	4.7	

Table 3. Levels of chlorinated pesticides in breast milk samples (ng/g milk fat) from 31 primiparas. Values below the LOD (limit of detection) were set to ½LOD in the calculations of mean, median and sumDDT. SumDDT is the sum of p,p-DDE, p,p-DDT, o,p-DDT and p,p-DDD. Mean and median have not been calculated for o,p-DDT and p,p-DDD since the levels were below the LOD in 30 and 31 of the 31 samples respectively. o,p-DDT and p,p-DDD were included in the calculations of sumDDT.

	mean	median	min (lowest value >LOD)	max	No. of samples <LOD
HCB	9.43	8.81	6.34	20.6	0
beta-HCH	7.86	7.23	4.61	24.6	0
oxy-chlordan	3.26	3.12	1.74	8.51	0
trans-nonachlor	6.03	5.75	2.45	12.7	0
p,p-DDE	68.6	59.0	24.9	176	0
p,p-DDT	4.49	4.22	2.19	10.7	0
o,p-DDT	-	-	-	1.40	30
p,p-DDD	-	-	-	-	31
sumDDT	74.6	64.8	29.4	184	-
% fat	2.9	2.9	1.5	4.7	

Table 4 shows the results from the analysis of brominated flame retardants in the breast milk samples (N=30). The volume of one sample was too small for the analysis. 9 PBDE congeners (PBDE 28, 47, 66, 99, 100, 138, 153, 154, 183) and hexabromocyclododekan (HBCD) was analysed. LOD (limit of detection) for the PBDE congeners was 0.003 ng/g milk, which corresponds to 0.06-0.19 ng/g milk fat. LOD for HBCD was 0.006 ng/g milk, which corresponds to 0.20-0.37 ng/g milk fat. Values below the LOD were set to half the LOD in the calculations of mean, median and sumPBDE. Because the levels of PBDE 66 and 138 were low (below LOD in 29 and 30 samples respectively) the sumPBDE have been calculated in two different ways. The first calculation included all nine analysed PBDE congeners. In the individual samples, congeners with levels below LOD constitutes 1.2-35% (mean 10%) of the sumPBDE-levels. In the second calculation, PBDE 66 and 138 were excluded. In the individual samples, congeners with levels below LOD constitutes 0-28% (mean 6%) of the sumPBDE-levels. The congener with the highest level in breast milk was PBDE 47. This congener constitutes about 50% of the sumPBDE-levels. Mean and median have not been calculated for PBDE 66 and 138 since over 90% of the results were below the LOD.

Table 4. Levels of nine PBDE (polybrominated diphenylether) congeners, sumPBDE and HBCD (hexabromocyclododekan) (ng/g milk fat) in breast milk from 30 primiparas. Values below the LOD (limit of detection) were set to ½LOD in the calculations of mean, median and sumPBDE. SumPBDE has been calculated in three different ways. Mean and median have not been calculated for PBDE 66 and 138 since the levels were below the LOD in 29 and 30 of the 30 samples respectively.

	mean	median	min (lowest value >LOD)	max	number of samples <LOD
PBDE 28	0.12	0.10	0.07	0.31	13
PBDE 47	1.77	1.30	0.55	6.8	0
PBDE 66	-	-	-	0.06	29
PBDE 99	0.23	0.19	0.09	0.61	2
PBDE 100	0.36	0.28	0.12	1.8	3
PBDE 138	-	-	-	-	30
PBDE 153	0.75	0.66	0.38	2.3	0
PBDE 154	0.06	0.06	0.06	0.14	25
PBDE 183	0.06	0.06	0.07	0.13	26
sumPBDE[1]	3.46	2.93	1.66	12.1	-
sumPBDE[2]	3.35	2.82	1.54	12.0	-
HBCD	0.42	0.35	0.16	1.5	6
% fat	2.96	2.88	1.61	4.69	-

[1]sum of all nine congeners
[2]sum of seven congeners, PBDE 66 and 138 excluded

Former investigations have shown that the concentration of POPs in breast milk is strongly correlated to the age of the mother. In this group of primiparas the differences in age is small. Figure 1-3 show the relationships between concentrations of sumPCB, sumDDT and sumPBDE and age of the mother at the sampling occasion. There is a possible correlation between concentration of sumPCB and sumDDT and age, but no statistic calculations have yet been performed.

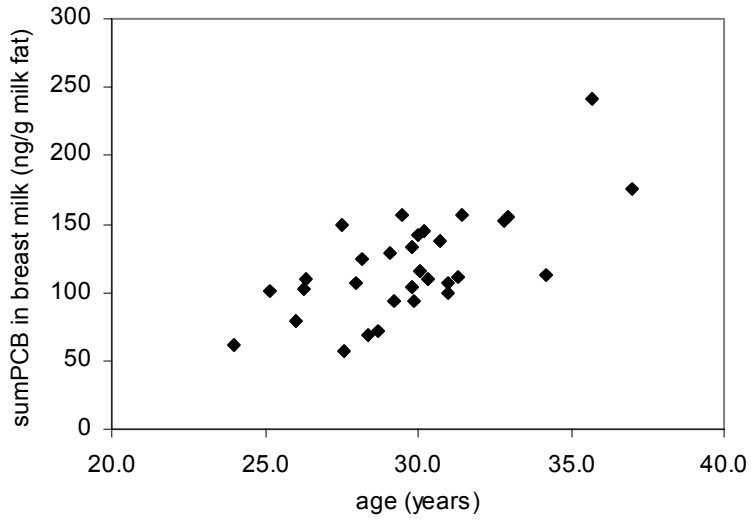

Figure 1. Relationship between concentration of sumPCB in breast milk and the age of the mother at the sampling occasion (N=31).

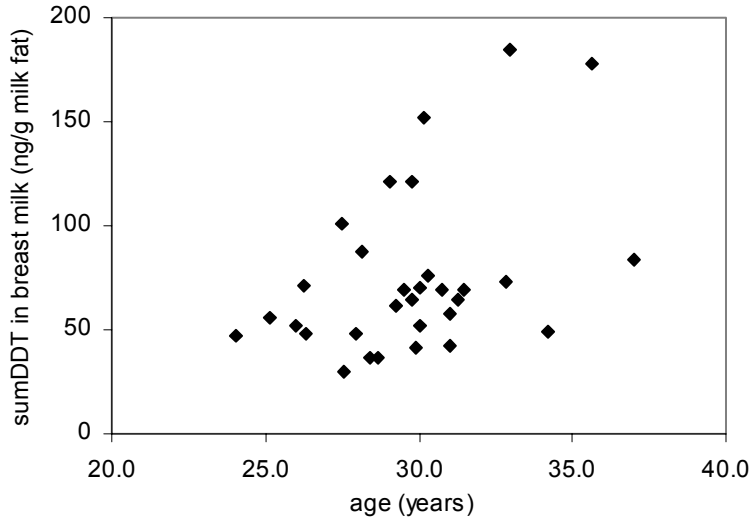

Figure 2. Relationship between concentration of sumDDT in breast milk and the age of the mother at the sampling occasion (N=31).

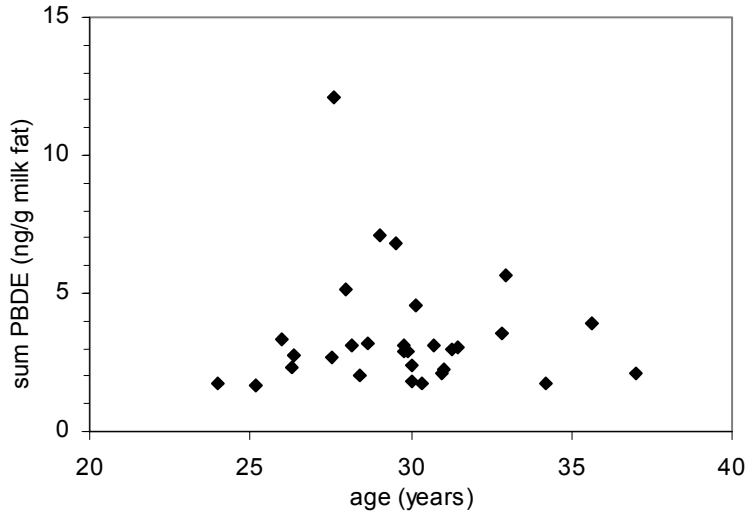

Figure 3. Relationship between concentration of sumPBDE (sum of all nine analysed congeners) in breast milk and the age of the mother at the sampling occasion (N=30).

Since the Swedish MLs (maximum limits) for PCB in foods are expressed as the concentration of the congener PCB 153, it is interesting to compare the concentration of PCB 153 with the concentration of sumPCB in breast milk. Figure 4 shows that there is a correlation between levels of PCB 153 and sumPCB, but no statistic calculations have yet been performed.

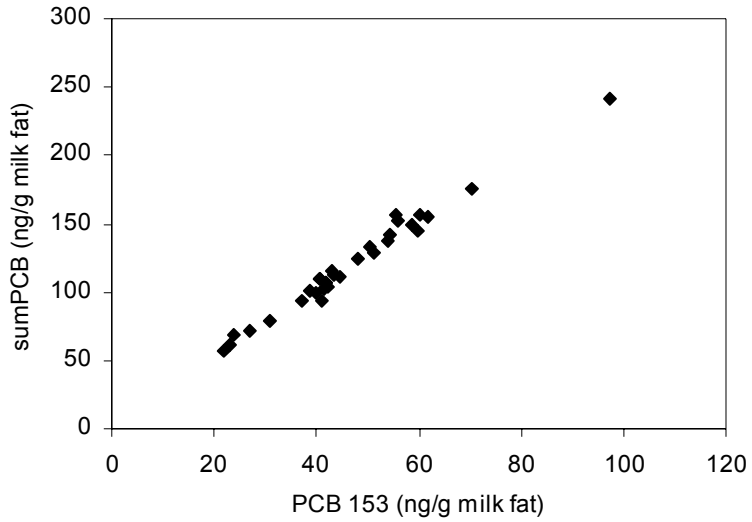

Figure 4. Relationship between levels of sumPCB and PCB 153 in breast milk (N=31).

It is interesting to investigate possible relationships between different kinds of POPs in breast milk since such relationships might indicate that the substances originate from the same source. The levels of p,p-DDE and PCB 153 are compared in figure 5 and the levels of trans-nonachlor and PCB 153 are compared in figure 6. A correlation is indicated in both figures, but no statistic calculations have yet been performed. The breast milk sample with the highest level of PCB 153 (97.3 ng/g milk fat) did also contain the highest level of transnonachlor (12.7 ng/g milk fat) and the second highest level of p,p-DDE (166 ng/g milk fat).

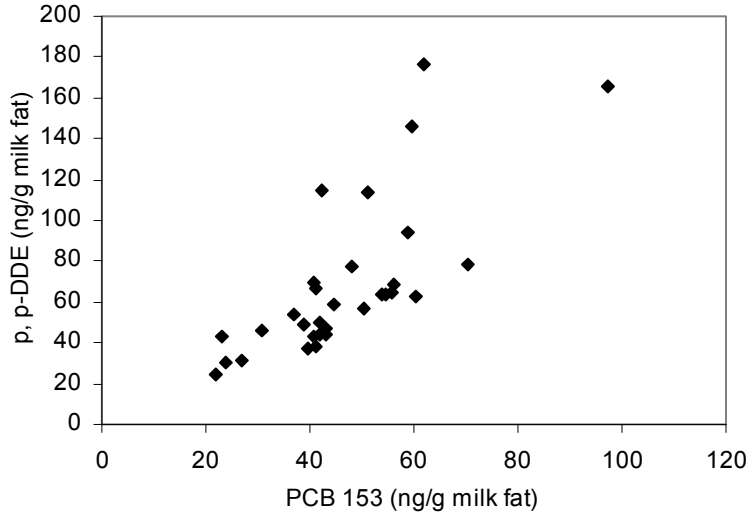

Figure 5. Relationship between levels of p,p-DDE and PCB 153 in breast milk (N=31).

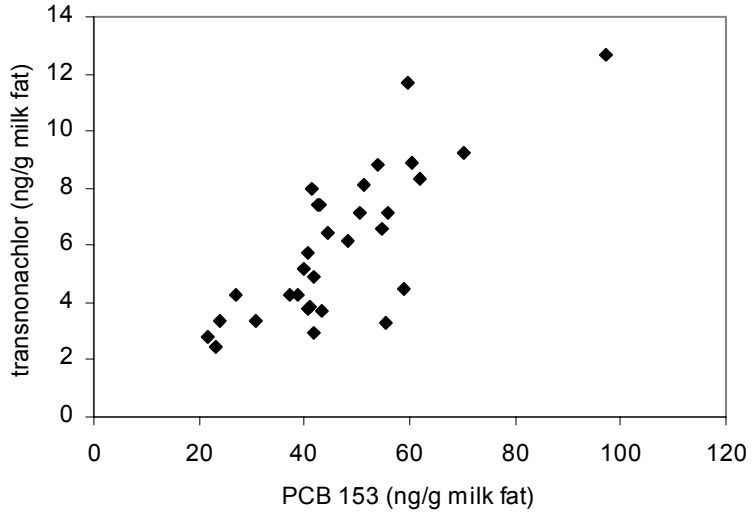

Figure 6. Relationship between levels of transnonachlor and PCB 153 in breast milk (N=31).

Conclusions

In earlier investigations (Aune et al. 1999, Darnerud et al. 2002, Darnerud 2001), POPs have been analysed in breast milk from primiparas from Uppsala County, Sweden. Results from these studies as well as from the study presented in this report are compared in table 5. Note that the comparison is approximate since the data not has been corrected for the age of the mothers, the number of analysed congeners and other factors that might influence the POP levels.

Table 5. Comparison between levels of POPs in breast milk (ng/g milk fat) sampled at different occasions, Uppsala 1996-98, 1996-99, 2000-2001 and 2002-03.

Study	**N**	**sumPCB**		**sumDDT**		**sumPBDE**		**Reference**
		mean	**min-max**	**mean**	**min-max**	**mean**	**min-max**	
Uppsala 1996-98	187	156[1]	51-402	157[3]	27-870	-	-	Aune et al. 1999
Uppsala 1996-99	93	-	-	-	-	4.0[6]	0.91-28	Lind et al. 2003
Uppsala 2000-01	31	148[2]	63-296	122[4]	34-903	3.2[7]	1.3-8.0	Darnerud 2001
Uppsala 2002-03	31	120[2]	56-241	75[5]	29-184	3.4[8]	1.5-12	this study

[1]sum of 10 PCB congeners [2]sum of 13 PCB congeners [3]sum of 3 DDT congeners [4]sum of 2 DDT congeners
[5]sum of 4 DDT congeners [6]sum of 5 PBDE congeners [7]sum of 6 PBDE congeners [8]sum of 7 PBDE congeners

The level of sumPCB is comparable with the levels reported earlier. Note that in Aune et al. (1999) 10 PCB congeners were analysed, but in the later investigations 13 PCB congeners were included in the sumPCB. The level of sumPBDE is also comparable with the levels reported earlier. Note that 5 PBDE congeners were included in the sumPBDE in Lind et al. (2003), 6 congeners were included in Darnerud (2001) and 7 congeners were included in this study. The sumDDT-level in this study is slightly lower than in earlier investigations (Aune et al. 1999, Darnerud et al. 2001). This might indicate a downward time trend, but no statistic calculations have yet been performed. Note that in Aune et al. (1999) 3 DDT-congeners were included in the sumDDT, in Darnerud 2001 2 congeners were included and in this study 4 congeners were included. Possible time trends in the levels of POPs in breast milk will be more thoroughly investigated in a coming report.

A positive correlation between the age of the mother and the level of sumPCB and sumDDT in breast milk is indicated. An explanation to the age-related uptake in milk could, apart from the fact that the compounds are accumulated in the body fat with time, also be caused by a continuous decrease in organochlorine levels in the environment. Like reported earlier (Aune et al. 1999, Darnerud 2001), PCB 153 seems to be a good indicator substance for total PCB in breast milk, and both trans-nonachlor and p,p-DDE show a correlation with PCB 153.

Note that the discussion above is preliminary, and no statistic calculations have been performed.

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