

## ORGANOCHLORINE PESTICIDES IN DANISH MILK

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### Introduction

Organochlorine pesticides and PCB congeners are persistent organic compounds found in the environment. Because of their stability and tendency to accumulate in fat they can enter the food chain and therefore be found in cow milk. As these chemicals are banned in most countries, it is expected that the compounds can still be found in cow milk due to environmental contamination of the feed. The content of the compounds are low in milk but due to the high consumption of milk the intake from milk products is an important source.

Levels of organochlorine pesticides and PCB congeners were determined in cow milk from Danish milk producers. Results from 1999 to 2002 are presented looking at differences between organic and non-organic milk.

### Methods and Materials

*Compounds:* The samples were analysed for their content of the following organochlorine pesticides:  $\alpha$ -HCH (hexachlorocyclohexane),  $\beta$ -HCH,  $\gamma$ -HCH (lindane), HCB (hexachlorobenzene), heptachlor, heptachlorepoxyd (cis), aldrin, dieldrin, endrin, isodrin,  $\alpha$ -chlordan,  $\gamma$ -chlordan, oxy-chlordan, trans-nonachlor,  $\alpha$ -endosulfan, *p,p'*-DDD, *p,p'*-DDE, *o,p'*-DDT and *p,p'*-DDT. The following PCB congeners (IUPAC no.) were analysed PCB-28, PCB-52, PCB-101, PCB-105, PCB-118, PCB-138, PCB-153, PCB-156, PCB-170 and PCB-180.

*Samples:* 213 samples of raw cow milk from Danish milk livestock were collected from 1999 to 2002. Samples were collected by two different sampling strategies. From 1999 to 2000 the samples were taken as pool samples at dairy plans silos where milk from a number of milk producers are collected. From 2001 to 2002 the samples were taken directly at different individual livestock silos.

*Sample clean-up:* Milk was mixed with potassium oxalate, ethanol, diethylether and pentane in a separation funnel. The organic phase was shaken with water and the water phase was extracted twice with diethylether/pentane. The assembled organic phase was cleaned twice with sodium chloride and filtered through sodium sulphate after which the solvent was evaporated. 0.6 g fat was added to a Florisil column deactivated by water and eluted with dichloromethane:*n*-pentane (1:4). The eluate was carefully evaporated and the sample dissolved in isooctane. The final sample was analysed by gas chromatography using two different columns and electron capture detectors.

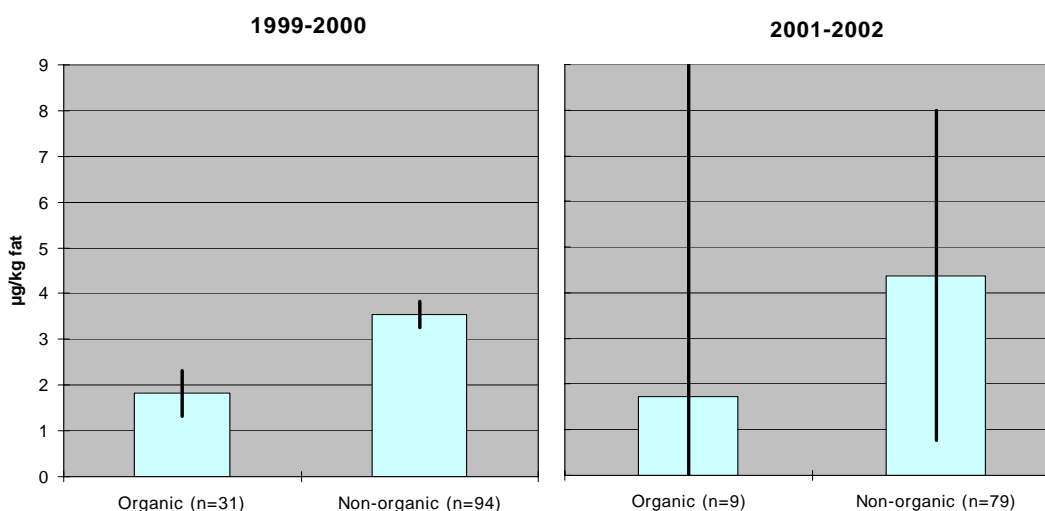
*GC-ECD parameters:* Perkin Elmer autosystem gas chromatograph. Column: 50 m CP-Sil-5CB (Chrompack) and 60 m DB-17 (J&W), 0.25 mm i.d., 0.25  $\mu$ m film thickness. Carrier gas: Helium,

15 psi (CP-Sil-5CB) or 37 psi (DB-17). 2 µl injected splitless, splitless time 2.5 min. Injector held at 220°C. Temperature programme: 90°C for 1 min., 30°C/min. to 180°C in 10 min., 2°C/min. to 240°C, 10°C/min. to 280°C in 20 min. (CP-Sil-5CB) or 30 min. (DB-17). Detector temperature 320°C. PCB congeners and organochlorine pesticides were quantified by comparing responses with those of standard mixtures. Limits of detection for organochlorine pesticides and PCB congeners were 1 to 4 µg/kg fat.

### Results and discussion

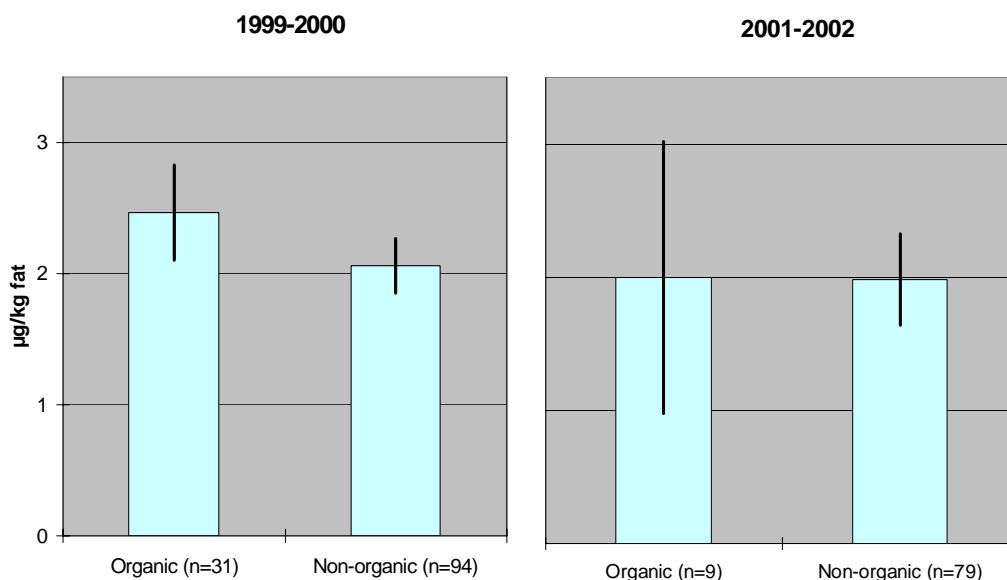
Many of the compounds investigated were not detected in any of the samples and compounds detected were found in less than 10% of the samples except for dieldrin found in 30% of the samples, HCB in 93% of the samples and *p,p'*-DDE in 82% of the samples. As a conservative estimate for the content in samples where HCB and *p,p'*-DDE were not detected, half of the limit of detection was used in the calculations below (LOD for HCB and DDE were 1 µg/kg fat).

**Figure 1. *p,p'*-DDE in Danish cow milk**



In Figure 1 result for *p,p'*-DDE in Danish cow milk from 1999-2002 are displayed. For samples collected at dairy (1999-2000) the mean levels for organic produced and non-organic produced milk are significantly different ( $P = 3.3 \times 10^{-8}$ ) where the lowest mean level is found in the organic produced milk samples. When samples are collected directly at livestock silos (2001-2002), as expected major variations are observed. Mean levels were at the same levels as observed for the period 1999-2000 but due to the major variation no conclusions can be drawn concerning difference between the means.

In Figure 2 results for HCB in cow milk is given. There is not a significantly different between the two mean, although the P value is 0.054 for the samples collected at dairy plans silos (1999-2000). In contrast to the results for *p,p'*-DDE, HCB mean level for the organic samples is the highest.

**Figure 2. HCB in Danish cow milk**

The results do not reveal why there is difference in the levels for the organochlorine pesticides in organic and non-organic milk, but it is expected to be due to differences in the feed used at organic and non-organic livestock. The levels of the organochlorine contaminants found in this study is in good agreement with previously reported results from Germany and Austria<sup>1,2</sup>. *p,p'*-DDE concentrations found in Danish milk is, however around 10 times lower than concentrations found in Spain, Mexico, India and China, whereas the concentration for HCB is just below the level found in Germany, Austria and Mexico<sup>3,4,5,6,7</sup>. HCH is found in less than 4% of the Danish samples whereas it is often found in the other countries.

In Table 1 the intake of *p,p'*-DDE and HCB is calculated. Calculations are based on Danish average consumption of 35 g of fat from cow milk<sup>8</sup>.

**Table 1: Mean concentration, daily intake and percent of TDI of DDE and HCB from milk**

	<i>p,p'</i> -DDE			HCB		
	Content [µg/kg fat]	Intake [µg/day]	Percent of TDI [%]	Content [µg/kg fat]	Intake [µg/day]	Percent of TDI [%]
Organic	1.8	0.06	0.2	2.4	0.08	0.7
Non-organic	3.5	0.14	0.4	2.0	0.07	0.8

The intake is compared with TDI using a body weight estimate of 70 kg. The Danish Veterinary and Food Administration has found that a TDI of 0.5 µg/kg bodyweight/day for the sum of DDT, DDE and DDD is relevant and a TDI of 0.16 µg/kg bodyweight/day for HCB<sup>9</sup>. The intake for HCB and *p,p'*-DDE is compared with these TDIs in the table.

### Conclusion

Out of 213 samples only HCB and *p,p'*-DDE are found in a majority of the samples. Significantly difference between the levels in organic and non-organic cow milk were only demonstrated for *p,p'*-DDE with the highest level found in non-organic milk.

*p,p'*-DDE concentrations found in Danish milk is at the same level as found in Germany and Austria, but around 10 times lower than concentrations found in Spain, Mexico, India and China, whereas the concentration for HCB is just below the levels found in Germany, Austria and Mexico.

From the results in this study it can be concluded, that the content of organochlorine pesticides and PCB congeners is low in all the Danish milk samples and the Danish intake of HCB and *p,p'*-DDE through milk is less than one percent of TDI.

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