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Declining levels of PCB, HCB and *p,p′*-DDE in adipose tissue from food producing bovines and swine in Sweden 1991–2004

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ABSTRACT

The official control programme for organochlorine (OC) contaminants in food producing animals in Sweden was used to study temporal and spatial trends of the polychlorinated biphenyl CB 153, hexachlorobenzene (HCB) and *p,p′*-DDE in adipose tissue from bovines and swine 1991–2004. Our results show that efforts to decrease OC contamination of animal feed and the environment have had a positive impact on the contamination of the animal production. OC concentrations declined significantly in almost all studied regions of Sweden. OC temporal trends were slower in bovines (6–8% per year) than in swine (10–12%). Power analyses showed that data from more than 10 years of sampling were needed for a detection of an annual OC level change of 5% in both species in the control programme, due to large within- and between-year variation in OC levels. CB 153 and *p,p′*-DDE levels were higher in southern than in northern Sweden. Levels decreased with age in milk cows, but not in young nulliparous cows (heifers) and bulls. Moreover, milk cows and bulls had significantly lower OC levels than heifers. Levels were not age-dependent among swine, but castrated male swine (barrows) had significantly lower OC levels than young female swine (gilts). Levels of the studied OCs are now in many cases below the LOQ of the analytical method used. Future time trend studies of these OCs thus depend on lowered LOQs in the control programme.

1. Introduction

A steady decline in environmental levels of organochlorines (OCs), such as the group of industrial chemicals polychlorinated biphenyls (PCBs), and the pesticides hexachlorbenzene (HCB), hexachlorocyclohexanes (HCHs) and DDT with its metabolite *p,p′*-DDE, has occurred in the Swedish environment since the early 1970s (Olsson and Reutergårdh, 1986; Bignert et al., 1998). As a result exposures of the human population in Sweden have decreased (Norén and Meironyte, 2000). Nevertheless, PCB is still regarded as a potential health problem in Sweden (NBHW, 2005). The body burdens of PCBs among pregnant women in Sweden are within the range of those found in populations where subtle negative effects on child development have been suggested after pre-natal PCB exposure (Longnecker et al., 2003; Glynn et al., 2007a,b). It is therefore important to continue the efforts to decrease the OC exposure of the general population.

Food of animal origin is currently the largest source of human OC exposure in Sweden (Darnerud et al., 2006). OC contamination of animal feed is an important determinant of contamination of food producing animals, subsequently affecting contamination of dairy products, and meat and meat products. Within the European Union, efforts are made to reduce OC contamination of animal feed (CEU, 2001; Gallian et al., 2004; Commission, 2006).

The Swedish National Food Administration (NFA) has since the early 1970s analysed OCs in bovine and swine adipose tissue in the official control programme for contaminants in food producing animals. In an earlier report, declining trends of CB 153, HCB, α-HCH and *p,p′*-DDE in adipose tissue from bovines and swine between 1991 and 1997 were indicated (Glynn et al., 2000). This suggests that efforts to reduce OC contamination of animal feed and the environment have had positive effects on contamination of animals in food production. The study period was, however, short. In the present study the time period have been extended to cover 1991–2004.

2. Materials and methods

2.1. Sampling

Between 1991 and 2004 sub-cutaneous adipose tissue was randomly sampled from bovines and swine in all major slaughter houses in Sweden. The previous year's production determined

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the sampling frequency. Between 1991 and 1997 approximately 80 samples per year were taken from each animal species, but from 1998 the number of samples decreased to 20–25 per year for each species. In all 161 young nulliparous cows (heifers, age 6–25 months), 359 bulls (6–84 months) and 179 milk cows (28–96 months) were sampled. Among the swine 384 female gilts (4–24 months), 46 male boars (3–8 months) and 295 castrated males (barrows, 4–10 months) were sampled.

2.2. OC analysis

PCB, HCB and *p,p*′-DDE were measured as described in Glynn et al. (2000). In short, between 1991 and 1994 analysis was performed on a GC/ECD using packed columns and the technical product Chlophen A-50 as quantification standard. During 1994 the packed columns were replaced with capillary columns and the quantification of single PCB congeners and pesticides was carried out on a GC with dual capillary columns and dual ECDs using multi-level calibration curves. Internal standards were added before the extraction to correct for analytical losses and to ensure quality control. The PCB congeners included in the analysis were CB 28, CB 52, CB 101, CB 118, CB 138, CB 153 and CB 180. The chlorinated pesticides/metabolites measured were hexachlorocyclohexane (HCH: α- and γ-isomers), hexachlorobenzene (HCB), dieldrine, *p,p*′-DDT, *p,p*′-DDD and *p,p*′-DDE. The compatibility of the two methods were checked on both bovine and swine samples (Glynn et al., 2000), and if necessary the levels detected 1991–1994 were recalculated as described in Glynn et al. (2000). The expanded measurement uncertainty for the individual compounds using the capillary column method is 35% ($k = 2$). For PCB only the results of CB 153 were used, since the levels of the other PCB congeners, in contrast to CB 153, were often below the limit of quantification (LOQ). CB 153 is a good indicator substance for total PCB levels in bovines and swine (Glynn et al., 2000). The ratio between total PCB and CB 153 concentration in bovine adipose tissue was 0.20 (median, range: 0.17–0.23, $N = 10$), and in swine adipose tissue 0.24 (0.19–0.29). Among the pesticides/metabolites levels of HCB (bovines only) and *p,p*′-DDE were above the LOQ frequently enough for statistical analysis to be meaningful. The LOQ for total PCB, using packed columns, was 5 ng g^{-1} lipid (1 ng CB 153 g^{-1} lipid), and for CB 153, using capillary columns, the LOQ was 0.5 ng g^{-1} lipid up to 1997. Thereafter the CB 153 LOQ was increased to 1 ng g^{-1} lipid. For the chlorinated pesticides/metabolites LOQ was 1 ng g^{-1} lipid for both methods.

Between 1991 and 1995 the analysis was performed at the NFA, but during 1995 the analysis was commissioned to a commercial laboratory (AnalyCen AB, Lidköping, Sweden). Interlaboratory comparisons showed that the analytical performance of the new laboratory was satisfactory. The commercial laboratory is accredited according to SS-EN ISO/IEC 17025 (accreditation number ID-number 1977) for the analyses and control samples produced by the NFA is regularly analysed (blinded) with satisfactory results.

2.3. Calculations and statistical analysis

Multiple regression (MINITAB® For Windows, 12.22) was used to describe the associations between the dependent variable "OC concentration" (y) and the independent variables "age at slaughter" (months), "year at slaughter", "sex" (male, female), and "region of slaughter". The variable "OC concentration" was ln-transformed in order to stabilise the variance. The observations with standard residuals $\geqslant 3$ (outliers) in the regression analyses were excluded.

For bovines, the associations between the dependent variable "OC concentration" were first analysed with the independent variables "year at slaughter", "sex" and "region of slaughter" in the model. "Sex" was categorized into heifers (nulliparous females $\leqslant$27 months of age), bulls (males $\leqslant$36 months) and milk cows (females > 27 months). Bulls of an age > 36 months were excluded due to few observations with a large impact on the analysis ($N = 3$). In this analysis the variable "age at slaughter" was not included, since preliminary analysis showed that the levels of OCs markedly decreased with age for milk cows but not for heifers and bulls. The variable "region of slaughter" was categorized into 6 regions (Supplementary material, Fig. SM1), starting with region 1 in the south of Sweden including the counties of Skåne and Blekinge. Region 2 consisted of the counties Halland, Småland and Gotland; region 3 Västergötland and Dalsland; region 4 Östergötland, Södermanland and Närke; region 5 Uppland, Västmanland and Värmland; and region 6 of counties north of region 5.

In separate analyses the association between age and OC concentrations were studied using two regression models, one model for heifers and bulls with the independent variables "year of slaughter", "age at slaughter", "sex" and "region of slaughter", and another model for milk cows with the independent variables "year of slaughter", "age at slaughter" and "region of slaughter".

In the analyses of the results for swine the independent variables "year of slaughter", "age at slaughter", "sex" (gilt, boars, barrows), and "region of slaughter" were included.

The partial regression coefficients (b) for the independent variables "year of slaughter" and "age at slaughter" were used in the estimation of % change in concentrations per year of slaughter (Eq. (1)) and % decline of concentrations per year of age of the animals (Eq. (2)):

$$\%\text{change}_{\text{year}} = (1 - \exp(b_{\text{year}})) \times 100 \quad (1)$$

$$\%\text{change}_{\text{age}} = (1 - \exp(b_{\text{age}})) \times 100 \quad (2)$$

The procedure general linear model (GLM) (MINITAB® For Windows, 12.22) was used to estimate adjusted means of the categorized independent variables ´region of slaughter‘ and ´sex‘. OC levels below LOQ were set to 1/2 the LOQ in the statistical analysis. The level of significance was set to $p \leqslant 0.05$ in all tests.

Using the annual geometrical means obtained from the whole dataset we estimated the minimum annual change in OC level that was likely to be detected after a monitoring period of 10 years at a statistical power of 80% and a statistical significance level of $p = 0.05$. We also estimated the number of years required to detect an annual trend of at least 5% at a statistical significance of $p = 0.05$ and a statistical power of 80%.

3. Results

The concentrations of OCs varied considerably during the study period (Table 1). Swine had slightly lower median concentrations of CB 153 than bovines. The levels of the studied OCs seemed to decline in both bovines and swine during the study period between 1991 and 2004 (Table 1). In bovines median levels were above LOQ during the whole study period. In swine the median levels of CB 153 were below the LOQ already in year 2000, whereas median levels of *p,p*′-DDE reached the LOQ in 2003.

Regression analysis revealed that the levels of CB 153, HCB and *p,p*′-DDE in bovines and swine declined on average 6–8% per year and 10–12% per year, respectively, from 1991 to 2004 (Table 2). The levels declined significantly in all regions, except for CB 153 in bovines from region 3, *p,p*′-DDE in bovines from region 2, and CB 153 in swine from region 4 (Supplementary material: Table SM1).

Using the whole data set, the minimum annual change in OC levels likely to be detected after a monitoring period of 10 years ranged between 8.5% and 11% for the three studied OCs in bovines, and 5.6–8.6% in swine (Supplementary material: Table SM2). The