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PCDD/Fs and dioxin-like PCBs in home-produced eggs from Belgium: Levels, contamination sources and health risks

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ABSTRACT

This paper discusses the dioxin TEQ levels as determined by the chemically activated *luciferase* gene expression assay (CALUX) and by HRGC-HRMS in eggs, soils, faeces and kitchen waste samples obtained in the CONTEGG study. The samples were collected in each Belgian province at private homes and in small gardens where chickens are held. The CALUX levels for eggs sampled in autumn were higher than the levels in eggs obtained at the same locations in spring (median values of 5.86 and 4.08 pg CALUX TEQ/g fat, respectively). The total WHO-TEQ levels in eggs, determined by HRGC-HRMS, ranged from 3.29 to 95.35 pg TEQ/g fat in autumn and from 1.50 to 64.79 pg TEQ/g fat in spring. In the soils on which the chickens forage, levels of 2.51–11.35 pg I-TEQ/g in autumn and 2.00–7.86 pg I-TEQ/g in spring were found. The congener pattern of PCDD/Fs in the eggs, soils and faeces was dominated by OCDD, in addition to 1,2,3,4,6,7,8-HeptaCDD, OCDF and 1,2,3,4,6,7,8-HeptaCDF. The predominant dioxin-like PCBs were PCB118, PCB 105 and PCB 156. The dioxin-like PCBs contributed on average 47%, 14% and 20% to the total WHO-TEQ in eggs, soils and faeces, respectively. Kitchen waste samples were very low-contaminated with dioxin-like compounds. The present results showed a good agreement between egg and soil TEQ levels for PCDD/Fs but not for dioxin-like PCBs. This study showed that current soil levels found in some private gardens do not lead to egg levels below the current EU maximal level of 6 pg total TEQ/g fat for dioxins and dioxin-like PCBs. The consumers of the analysed eggs attained 5–79% of the tolerable weekly intake (TWI) of 14 pg TEQ/kg bw for dioxins and dioxin-like PCBs by exposure to their home-produced eggs only.

1. Introduction

Polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs) are ubiquitous environmental contaminants. They are very persistent to chemical degradation and accumulate in the food chain. The contamination of food products with dioxin-like compounds (PCDD/Fs and dioxin-like PCBs (dl-PCBs) is a well studied issue because food is generally considered as the major source of dioxin intake for humans (Fries, 1995). Food products from animal origin like meat, fish and dairy products are the main contributors to dioxin intake. The estimated contribution of eggs to the total dioxin intake from food generally ranges from 2 to 5% in European countries (Finland (Kiviranta et al., 2001), The

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Netherlands (Baars et al., 2004) and Spain (Llobet et al., 2003; Bordajandi et al., 2004; Fernandez et al., 2004; Bocio and Domingo, 2005). In Belgium, the contribution of eggs to the total dietary intake of PCDD/Fs and non-*ortho*-PCBs was estimated to be 8%. The analysed eggs in the Belgian study included home-produced and commercially available eggs (Focant et al., 2002). Bilau et al. (2008) found a contribution of 3–5% by eggs to the intake of dioxin-like substances in the Flemish population. Higher contributions by eggs to the dioxin intake were reported in Japan (15%) by Tsutsumi et al. (2001) and recently in Spain (17%) (Marti-Cid et al., 2008). These are low values compared to the contributions of fish and meat (Domingo et al., 1999; Schecter et al., 2001; Focant et al., 2002; Baars et al., 2004; Llobet et al., 2003). The contribution by eggs is generally calculated from intake levels of eggs produced by hens without soil contact or eggs that are commercialised. Recent data on TEQ levels in eggs range from 0.6 to 4.1 pg WHO-TEQ/g fat for dioxins and dl-PCBs in different European countries (Finland (Kiviranta et al., 2001), Spain (Eljarrat et al., 2002; Bordajandi et al., 2004; Fernandez et al., 2004), Belgium (Focant et al., 2002), The Netherlands (Baars et al., 2004). In contrast to these levels, it is known that eggs from free-range and organic farms contain higher levels of dioxins (Schuler et al., 1997; Traag et al., 2002; Tlustos et al., 2004; De Vries et al., 2006; Schoeters and Hoogenboom, 2006). In Belgian home-produced eggs, elevated levels up to 9.9 pg PCDD/F TEQ/g fat were reported (Pussemier et al., 2004). The contact with the outdoor environment is believed to cause the increase of the contamination levels. It has been suggested that soil and soil organisms contribute to the egg contamination of free-range hens (Schuler et al., 1997; Harnly et al., 2000; Kijlstra, 2004). Stephens et al. (1995) studied the transfer of dioxins and furans from soil to hens. Other studies focused on the uptake of dioxins by hens through contaminated feed (Pirard and De Pauw, 2005a,b; Hoogenboom et al., 2006a; Traag et al., 2006). Comparisons between dioxin levels in free-range eggs and in soils on which the chickens forage are limited and a good correlation has not always been found (Schuler et al., 1997; Traag et al., 2002; Pirard et al., 2004, 2005). For home-produced eggs in Belgium such data are lacking. The present paper discusses the results of the dioxin analyses performed in the frame of the CONTEGG study. The background of this study and the sampling are described in detail in an overview paper by Van Overmeire et al. (2009-this volume). In brief, during The Contegg study a number of eggs, soils, faeces and kitchen waste samples were analysed to investigate their contamination with various compounds (dioxins, PCBs, organochlorine pesticides, brominated flame retardants, trace elements and mycotoxins). Multiple locations in Belgium were included in the sampling and samples were collected in two seasons to investigate possible seasonal differences in contaminant levels. More specifically for dioxins there could be seasonal differences in egg and/or soil levels related to dioxin deposition on the soil or to the soil covering by vegetation which could influence soil uptake by the chickens. In addition to eggs and soils we also focused on food waste products (offered as additional feed to the hens) to investigate whether this could constitute an important source of dioxin intake by the chickens. Faecal matter was collected and analysed to compare the found levels of contaminants with levels in soil and eggs and to investigate amount of soil intake (determination of ash content in faeces might serve as a measure for the amount of soil intake). The present paper discusses the PCDD/F and dl-PCB data in selected samples of soil, eggs, faeces and kitchen waste, obtained from identical locations and in two seasons. In addition, an assessment of the TEQ intake by the consumers of these eggs is made and compared to the intake by commercially available eggs.

2. Materials and methods

2.1. *Samples*

The origin of the samples sample and their pre-treatment are described by Van Overmeire et al. (2009-this volume). In summary, egg samples, soil samples, faeces samples and kitchen waste samples were collected from private owners of hens in Belgium during autumn 2006 and spring 2007. Soils, faeces, and kitchen waste samples were analysed for 10 selected locations in Belgium. Sample selection occurred based on relevant data (both elevated and low) for one or more of these contaminants in 40 eggs samples taken in autumn 2006: dioxins (analysed by CALUX bioassay), pesticides (DDTs), trace elements and marker PCBs. For more information about the origin of the 10 samples the reader is referred to Van Overmeire et al. (2009-this volume). CALUX analyses were performed on eggs (n=98), soils (n=20), faeces (n=20) and on kitchen waste samples (n=11). Analyses of PCDD/Fs and dioxin-like PCBs by HRGC-HRMS occurred on eggs (n=20), soils, (n=20), faeces (n=20) and kitchen waste samples (n=11).

2.2. *CALUX analyses*

2.2.1. *Reagents*

All used solvents (acetone, n-hexane, toluene, etyl acetate) are pestigrade quality (Merck or Fluka) and are checked for the absence of compounds, interfering with the cell response (Windal et al., 2005).

2.3. *Extractions and purifications*

2.3.1. *Eggs*

30 g of egg homogenate was dissolved in 30 ml of hexane and 15 ml of acetone and 30 g anhydrous Na_2SO_4 (Baker) were added. The mixture was thoroughly shaken for 10 min. A glass column (25 ml, 13 mm .i.d.) was filled with 0.6 g Celite 545 (Sigma) and 15.5 g anhydrous Na_2SO_4 and rinsed with 40 ml hexane. The clear (fat containing) supernatant was passed over this column and collected in a pre-weighted tube. The extraction was repeated two more times and the fat was gravimetrically determined after drying under nitrogen and it was divided for the analyses of dioxins, marker PCBs, organochlorine pesticides (OC) and polyaromatic hydrocarbons (PAHs) (Windal et al., 2009-this volume). The fat was purified on an acidified silicagel column in combination with a carbon column as previously described (Van Overmeire et al., 2006). Determination of dioxins by exposure of CALUX cells (mouse cell line from Xenobiotic Detection Systems Inc., USA (XDS)) to purified extracts was performed as described (Van Overmeire et al., 2004, 2006).