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PCB and organochlorine pesticides in home-produced eggs in Belgium

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ABSTRACT

The level of polychlorinated biphenyls (PCB) and persistent organochlorinated pesticides (OC) in home-produced eggs was investigated in Belgium. The concentration of dichlorodiphenytrichloroethane (DDT) is above the norm for 17% of the eggs collected during the spring on 58 different locations. For PCB, aldrin, dieldrin, and chlordane, 3–5% of the samples are above the norm too. These levels are surprisingly high for compounds banned for about 30 years. Higher concentrations in home-produced eggs are expected compared to battery eggs because of contact with the environment and especially the soil. For ten selected locations, the concentration in soils, excreta and feed was measured, but no simple correlation between egg and feed or soil level could be established. Hexachlorohexane, endosulfan, endrin, methoxychlor and nitrofen were not detected in any sample.

1. Introduction

Persistent Organic Pollutants (POPs) are toxic organic compounds resisting biological and chemical degradation. POPs are characterized by low water solubility and high lipid solubility, leading to their bioaccumulation in fatty tissues. They are also semivolatile, enabling them to move long distances in the atmosphere before deposition occurs (UNEP, 1998).

POPs can be classified in two main groups. The first group includes POPs which are formed unintentionally in a wide range of manufacturing and combustion processes, like dioxins or polyaromatic hydrocarbons (PAHs). The second group includes POPs which are or have been produced in large scale like pesticides or polychlorobiphenyls (PCBs) (some of them have been produced in thousands of tonnes per year). Twelve chemicals have been defined as POPs in the Stockholm Convention (2001): aldrin, chlordane, dichlorodiphenyltrichloroethane (DDT), dieldrin, endrin, heptachlor, mirex, toxaphene, PCB, hexachlorobenzene (HCB), dioxins and furans.

The adverse effects of POPs on the environment and human health have been well established and led to a reduction of the emission for the first group (e.g., purification system at the chimney of incinerators) and interdiction of production and use in many countries for most of the compounds belonging to the second group (Moniteur belge, 2006; VITO, 2007). However, the combined properties of POPs make them still a present issue despite the ban of most compounds since the 1970s. High concentrations of POPs in the environment, food and feed or human milk and blood are still observed (Smith, 1999; Schecter and Gasiewicz, 2003; Darnerud et al., 2006; Srogi, 2007; VITO, 2007; Rios et al., 2007; Van den Steen et al., 2008). The World Health Organization (WHO) has proposed Tolerable or Acceptable Daily Intake (TDI

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or ADI) or Tolerable Weekly Intake (TWI) for most of these compounds (JMPR, 1994, 2000, 2002; ATSDR, 2002). ADI/TDI/TWI, properties and year these compounds were banned in Belgium are summarized in Table 1 for compounds investigated in this study.

Because of their high fat content, eggs are known to accumulate POP and can be considered as a good indicator of ambient pollution (Jaspers et al., 2006; Schuler et al., 1997). In this study, the concentrations of several pesticides (most of them defined as POPs) and PCBs in eggs from free-range hens of private owners in Belgium have been determined. The objectives are to assess the level of POPs in a food item which is not under the control of the food chain organized at the national or European level and evaluate the possible impact of eggs' consumption on human health. Since it is well known that, for foraging chickens, soil can be an important source contamination (Schuler et al., 1997; Van Eijkeren et al., 2006), soil and feed samples were analyzed at selected locations to determine the main origin of the contamination. Levels in excreta were investigated as well. Two sets of measurements were performed, one in spring and one in autumn, to evaluate a possible seasonal variation.

This study is a part of a general study on egg contamination in Belgium, called CONTEGG. Details on objectives, sampling and general conclusions can be found in the paper of Van Overmeire et al., accepted for publication in Science of the Total Environment in 2008. The four main aspects of the CONTEGG project are summarized below:

1) determination of the *contamination* of home-produced eggs in Belgium by dioxins and dioxin-like PCB, marker PCBs, pesticides, trace elements, mycotoxins, polyaromatic hydrocarbons, brominated flame retardants such as polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A), PCB metabolites and perfluorinated compounds (PFOA and PFOS);
2) investigation of the *sources of contamination* through analyses of soil samples, as well as of kitchen waste given to the hens;
3) realization of *an assessment of the risks* related to the consumption of home-produced eggs compared to the consumption of commercially produced eggs;
4) suggestion of possible *measures* to reduce contamination levels and the study of their feasibility.

Table 1 – Year banned and ADI or TDI or PTDI of compounds investigated in this study.

Compounds	Year of ban (Law 25 Mai 2005; VITO, 2007)	ADI/TDI/PTDI
PCB (markers)	1986	–
HCH	Still used for veterinary purposes	ADI: 5 μg/kg bw (JMPR, 2002)
HCB	1974	TDI: 0.17 μg/kg bw (ATSDR, 2002)
Heptachlor	1976	PTDI: 0.1 μg/kg bw (heptachlor+ heptachlorepoxide) (JMPR, 1994)
Aldrin/dieldrin	1974 for agricultural use 1976 for all applications	PTDI: 0.1 μg/kg bw (JMPR, 1994)
Chlordane	1981 for agricultural use 1998 for all applications	PTDI: 0.5 μg/kg bw (JMPR, 1994)
Endrin	Never authorized in Belgium	PTDI: 0.1 μg/kg bw (JMPR, 1994)
Methoxychlor	2003	
Nitrofen	1979 in Europe (79/117/EEC)	
DDT	1974 for agricultural use 1976 for all applications	PTDI: 10 μg/kg bw (sum DDT, DDE, DDD) (JMPR, 2000)

ADI: Acceptable Daily Intake; TDI: Tolerable Daily Intake; PTDI: Provisional Tolerable Daily Intake.

2. Materials and methods

2.1. Sampling

Sampling is described in detail in Van Overmeire et al. (2009). Briefly, a set of egg samples belonging to private owners was collected from all over Belgium. Sampling was performed in the autumn 2006 (40 egg samples) and in the spring 2007 (58 egg samples; same location than in spring+18 other locations). Private owners were recruited on a voluntary basis. A set of criteria was used to select the private owners: their hens are living in a free-range system, no commercial egg production occurred, kitchen waste is offered as feed to the hens in addition to commercial feed. The locations where the hen holders lived included densely populated surfaces as well as agricultural areas. Analyses of metals, chlorinated pesticides, marker PCB and dioxin (by Chemically Activated LUciferase gene eXpression [CALUX]) were performed on those samples. Ten locations presenting a special interest were selected for further analysis of dioxins and dioxin-like PCBs by HRGC-HRMS, polybrominated dioxins, polybrominated biphenyls and PCB metabolites in eggs, soil, excreta and feed. A questionnaire was filled out with the help of the hen owners in order to get information on the hens, the egg consumption and the living environment (possible contamination sources, state of the hen house, percentage of soil covered with grass, etc…).

2.2. Analysis

Details of the analysis are given elsewhere (Van Overmeire et al., 2006), and the methods are only briefly described in this paper. All analyses were performed under accreditation, following the 17025 ISO norm.

2.3. Extraction

The extraction conditions are described in detail in the paper of Van Overmeire et al. (2009). The extraction conditions are optimized for the each matrix, as developed during validation of the analytical methods. For the eggs, the fat was extracted with hexane and acetone, the extract was filtrated and dried on a column filled with celite and Na_2SO_4. Kitchen waste was mixed with 20 mL toluene and 4.5 mL MeOH and shaken for