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Levels of polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans and polychlorinated biphenyls in human milk from different regions of France

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HIGHLIGHTS

- ► Levels of PCDD/Fs and PCBs in French maternal milk are reported.
- ► Levels are compared to an earlier study.
- Infant intake is estimated.
- Levels are located at the upper range of European countries.

ARTICLE INFO

Article history: Received 6 December 2012 Received in revised form 5 February 2013 Accepted 5 February 2013 Available online 15 March 2013

Keywords:

Polychlorinated dibenzo-*p*-dioxins (PCDDs) Polychlorinated dibenzofurans (PCDFs) Polychlorinated biphenyls (PCBs) Human milk Intake Time trend

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We report on the pilot study carried out before the start of the Elfe project (French longitudinal study from childhood). A total of 44 samples of mature human milk were collected at home 8 weeks after delivery. A total of 7 polychlorinated dibenzo-p-dioxins (PCDDs), 10 polychlorinated dibenzofurans (PCDFs), 12 dioxin-like (DL) polychlorinated biphenyls (PCBs), and 6 non dioxin-like (NDL)-PCBs were measured. For total TEQ (PCDD/Fs and DL-PCBs), the geometric mean concentration was 17.81 pg TEQ_{WH005}/g lipids. Relative PCDD, PCDF, and DL-PCB contributions to the arithmetic mean TEQ_{WH005} were 38%, 18%, and 44%, respectively. The use of TEF_{WH005} instead of TEF_{WH098} resulted in a 27% reduction of the total TEQ value. Although PCDD levels did not significantly change (less than 0.5% increase), PCDF and DL-PCB levels both decreased by 35% and 38%, respectively. Levels have been compared to data obtained during a previous non-reported national study conducted in 1998 (TEF_{WH098}) in French lactaria (n = 244). The mean of PCDD/Fs has decreased about 39.4% (18.8 pg TEQ_{WH098}/g lipids in 1998 vs 11.4 pg TEQ_{WH098}/g lipids in pilot study), respectively 41.5% for PCDDs (10.6 pg TEQ_{wH098}/g lipids in 1998 vs 6.2 pg TEQ_{wH098}/g lipids in pilot study) and 36.7% for PCDFs (7.9 pg TEQ_{wH098}/g lipids in 1998 vs 5.0 pg TEQ_{WH098}/g lipids in pilot study). For the sum of the 6 NDL-PCBs, the 2007 geometric mean concentration in milk was 176.3 ng/g lipids. The arithmetic mean lipid concentration in 2007 breast milk was 26.4 g/l (range from 6.0 to 46.7 g/l). A PCDD/F and DL-PCB daily intake was estimated to be 62.3 pg TEQ_{wH005}/kg body weight per day (85.0 pg TEQ_{wH098}/kg body weight per day) for a baby of 5 kg of body weight fed daily with 700 ml of maternal milk containing 25 g/l of lipids.

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1. Introduction

Estimating human exposure to dioxins and polychlorinated biphenyls (PCBs) remains of prime importance nowadays as these compounds are amongst the most toxic persistent organic pollutants (POPs) included in the Stockholm Convention (2001). Despite the fact that their levels in human have continuously decreased for the last couple of decades for most populations (WHO, World Health Organization. ENHIS, European Environment and Health Information System, 2009), a continuous monitoring of general populations is of interest to ensure health protection as well as to estimate the impact of actions taken at the environmental level. Moreover, infants are proportionally more exposed to these toxicants than adults. At their early stage of life, they are often still exposed at levels significantly above those generally defined as acceptable (Ulaszewska et al., 2011; JECFA, 2001). Because mother's milk is, in most cases, the major route of intake during the early months of feeding, studies focusing on human milk are important to estimate the exposure of breast-fed infants (LaKind et al., 2000). Surprisingly, very little information is available in the literature for the French population. No data were neither collected for France during the different rounds of the WHO/UNEP-coordinated exposure studies on levels of PCBs,

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^{0048-9697/\$ -} see front matter © 2013 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.scitotenv.2013.02.057

polychlorinated dibenzo-*p*-dioxins (PCDDs), and polychlorinated dibenzofurans (PCDFs) in human milk (WHO, World Health Organization, 2007). The unique set of data available so far issued from a French national study conducted in lactaria in France in 1998 (Fréry et al., 2000a) but not reported in the literature.

Recently, a longitudinal study project was started in France, the Elfe project (French longitudinal study from childhood). The purpose of the Elfe project (is to conduct a nationally representative cohort of 20,000 children followed from birth to adulthood under a multidisciplinary approach (Vandentorren et al., 2009). Because of the complexity of such a large scale study, a pilot study was set up in east of France in fall 2007 to validate the content of the questionnaires, the efficiency of recruitment, the interview procedures, and more generally, to test survey acceptability at the maternity hospital and at the end of the maternity leave. The pilot also served to test all analytical aspects such as biological sampling, sample storage, and sample transfer in real situation. Among samples that were targeted during this pilot study, mature mother's milk specimens were collected at home between weeks 6 and 8 after delivery.

Because a descriptive analysis of the preliminary concentration data was foreseen to gather information on the distribution of some variables in the population, as well as regarding current background levels, the collected milk samples were actually analyzed. The set of tested compounds consisted in 35 compounds: seven 2,3,7,8-substituted PCDDs, ten 2,3,7,8-substituted PCDFs, 12 dioxin-like (DL) PCBs (four non-*ortho*-PCBs, eight mono-*ortho*-PCBs) and six non dioxin-like (NDL)-PCBs. Levels measured in these samples have been compared to levels recorded during the previous unpublished national study conducted in 1998, and put into perspective against foreign data.

2. Methods

2.1. Study Population

Pilot counties were chosen for regional diversities in terms of population and urbanization. There were five different counties: Seine-Saint Denis (county code, 93), Ardèche (07), Isère (38), Loire (42), and Savoie (73) (Fig. 1). A total of 38 public and private maternities were concerned. Criteria of inclusion were: babies born on the 1st, 2nd, 3rd and 4th of October 2007; at least 22 weeks of amenorrhea; maximum of two living twins delivered (no multiple births greater than 2). From all births fulfilling these requirements, 301 mothers accepted to be part of the pilot study. Some 126 mothers breastfed their babies at the maternity. In practice, only a subset of 44 mothers actually sent a milk sample back for analysis after reaching the mature state (from 1 to 2 months). The contribution of each county in terms of samples was 4 for Seine-Saint Denis, 1 for Ardèche, 14 for Isère, 16 for Loire, and 9 for Savoie.

2.2. Questionnaire

The data collection included a questionnaire filled in during a one hour face-to-face interview with the midwife; a collection of medical data using the newborn's and mother's medical files to obtain information about pregnancy, the perinatal period and the mother's and newborn's health at delivery, basic demographic (age, gender, education, marital status), questions about height, weight, recent weight change, smoking status, childbearing and breastfeeding. The data collection also included a self-administered questionnaire for the mother to determine food habits and environmental patterns. Food intake (fish consumption) was quantified by a validated food frequency and portions questionnaire detailed for the food vectors of animal lipids, which are also the food vectors of dioxins.

2.3. Sample Collection and Storage

The recruitment took place at home using a specific glass bottle given to participants at the maternity. All participants were contacted by phone one month after leaving the maternity to receive relevant information on how to properly collect and ship breast milk specimen for storage in a centralized biobank before analysis. The expected



Fig. 1. Map of France showing counties where mature milk samples were collected for the pilot study in fall 2007.

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