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Estimation of the daily intake of hexachlorobenzene from food consumption by the population of Catalonia, Spain: Health risks

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ABSTRACT

In 2000 and 2006, we determined the dietary intake of hexachlorobenzene (HCB) by the population of Catalonia, Spain. In order to establish the temporal trend in the levels of HCB in foodstuffs, as well as in the dietary exposure to that environmental pollutant, the concentrations were again analyzed by HRGC/HRMS in 65 composite food samples widely consumed by the Catalan population. Food samples were randomly purchased in November–December 2008 in local markets, small stores, supermarkets, and large grocery stores from 12 representative cities from Catalonia. The daily intake of HCB associated with this consumption was estimated for four age groups of the population of Catalonia: children, teenagers, adults and seniors, which were in turn divided according to sex. The highest mean HCB levels in food were detected in oils and fats (0.297 ng/g fw), dairy products (0.225 ng/g fw), and fish and seafood (0.170 ng/g fw). In the 2000 and 2006 surveys, total dietary intakes of HCB were 166.2 and 71.6 ng/day, respectively (or 2.4 and 1.0 ng/kg of body weight per day). In the current study, it was 37.7 ng/day (or 0.54 ng/kg of body weight per day), which means considerable decreases with respect to the previous intakes. According to recommendations of international regulatory organisms, the current dietary intake of HCB should not mean any significant health risk (carcinogenic and non-carcinogenic) for any of the age/gender groups of population here assessed.

1. Introduction

Hexachlorobenzene (HCB) is a lipophilic organochlorine compound that was used as a seed dressing to prevent fungal disease for several crops from the late 1940s to the early 1970s (ATSDR, 2002; Reed, Buchner, & Tchounwou, 2007. HCB is one of the 12 persistent organic pollutants (POPs) listed under the Stockholm Convention (Aylward, Hays, Gagné, Nong, & Krishnan, 2010). It has characteristics of persistence, bioaccumulation, toxicity, and long-range environment transport, being extremely stable and globally distributed (Meijer et al., 2003). The US EPA has classified HCB as a probable human carcinogen (Group B2), while the US Department of Health and Human Services determined that HCB might be reasonably expected to be a carcinogen (ATSDR, 2002). In turn, the International Agency for Research on Cancer (IARC) classified HCB as possibly carcinogenic to humans (group 2B). In addition to cancer, the human health effects associated with HCB exposure involve systemic impairment (thyroid, liver, bone, skin), as well as damage to the kidneys, blood cells, and the immune, endocrine, developmental and nervous systems (Reed et al., 2007). A consistent downward trend in the environmental HCB levels has been noted over the past 25–30 years (Barber, Sweetman, van Wijk, & Jones, 2005). However, this trend seems not be quite general yet. For example, in the limited studies on temporal trends of HCB levels in China, HCB concentrations in air, sediment, fish and human milk did not show a consistent decreasing trend (Wang et al., 2010).

In order to prevent human exposure to microbiological and chemical contaminants, dietary exposure studies are of great interest (Brera et al., 2011; Coronel, Marin, Cano, Ramos, & Sanchis, 2011; Domingo & Bocio, 2007; Iñigo-Núñez, Herreros, Encinas, & González-Bulnes, 2010; Martí-Cid et al., 2010; Martorell et al., 2010; Shundo, Navas, Lamardo, Ruvieri, & Sabino, 2009). Although exposure to HCB can occur through the inhalation of HCB-contaminated air, by dermal contact, or through in utero exposure and breast milk, as for many other organic contaminants, non-occupational exposure to HCB is primarily due to eating low levels of this compound in contaminated food (ATSDR, 2002; Martí-Cid, Llobet, Castell, & Domingo, 2008; Reed et al., 2007). In 2000, we

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initiated in Catalonia (Spain) a wide surveillance program focused on measuring the levels of a number of chemical contaminants in various groups of foodstuffs. Dietary intake of the pollutants, including HCB was estimated for various age and sex groups of the population of Catalonia (Falcó et al., 2004). In order to establish the temporal trend in the total dietary intake of HCB, food items belonging to the same groups assessed in the 2000 survey were again collected and analyzed in 2006. For a standard male adult of 70-kg body weight, total dietary intake of HCB was found to be 166.2 ng/day (2.4 ng/kg of body weight per day) in the 2000 study, while in the 2006 survey the intake was 71.6 ng/day (1.0 ng/kg of body weight per day) (Martí-Cid et al., 2008). On a body-weight basis, it meant a decrease of 57%, which was due to the important reductions in the concentrations of HCB in dairy products (mainly cheese), as well as those in meat and meat products, and fish and seafood. We here present the concentrations of HCB in a number of foodstuffs corresponding to a recent survey (sampling performed in 2008), as well as data on the human dietary exposure to HCB. To estimate the potential changes in the total dietary intake of HCB by the Catalan population, we have collected food items belonging to the same food groups of our 2000 and 2006 surveys, and analyzed for HCB concentrations. Human dietary exposure to HCB was estimated and compared with the results obtained in our 2000 (Falcó et al., 2004) and 2006 (Falcó, Llobet, Bocio, & Domingo, 2008; Martí-Cid et al., 2008) studies.

2. Materials and methods

2.1. Sampling

In November–December 2008, 65 food samples were randomly purchased in local markets, small stores, supermarkets, and large grocery stores from 12 representative cities from Catalonia (Barcelona, Girona, Hospitalet del Llobregat, Lleida, Manresa, Mataró, Reus, Sabadell, Tarragona, Terrassa, Tortosa and Vilanova i la Geltrú). The collected food samples corresponded to the most consumed foodstuffs in Catalonia according to Serra-Majem et al. (2003). That information had been already used in our 2006 survey, while for the 2000 study, selection of foodstuffs was based on data from Capdevila et al. (2000). Foods included: meat (veal steak, hamburger, loin of pork, pork sausage, chicken breast, and steak and rib of lamb) and meat products (boiled ham, "Frankfurt" sausages, salami, and cured ham); fish and seafood (sardine, canned sardine, tuna, canned tuna, anchovy, mackerel, swordfish, salmon, hake, red mullet, sole, cuttlefish, squid, clam, mussel, and shrimp); vegetables (lettuce, tomato, cauliflower, string bean, onion, pepper, carrot, and eggplant); tubers (potato); fruits (apple, orange, pear, banana, mandarin, strawberry, and peach); eggs; milk (whole, and semi-skimmed); dairy products (1 composite for yoghurt, 3 composites for different types of cheese, and 1 composite for pudding–custard–cream); cereals (French bread, sandwich bread, rice, and pasta); pulses (lentils, haricot beans, chickpeas, and peas); oils and fats (olive oil, sunflower oil, margarine, and butter), and industrial bakery (croissant, cookie, and fairy cake) (Table 1). Unless otherwise stated, two composite samples were prepared for each food item. Each composite sample consisted of 24 individual units of each food item. Only edible parts of each food were selected for chemical analysis.

2.2. Analytical procedure

HCB concentrations in the different food samples were determined following the US EPA procedure 1625 (semivolatile organic compounds by isotope dilution gas chromatography) and the California Environmental Protection Agency Air Resources Board method 429 (1997) (Falcó et al., 2004; Martí-Cid et al., 2008). Briefly, appropriate isotope-labeled extraction standards ($^{13}C_6$-HCB) were added to the homogenized sample in order to control the entire sample preparation procedure. HCB was extracted from the samples by solid–liquid extraction using hexane-acetone as extraction solvent, and then concentrated for HCB determination. The clean-up and fractionation of the crude extracts was performed using size-exclusion chromatography. The cleaned extracts were then analyzed by high-resolution gas chromatography (Agilent gas chromatographer; 5890 and 6890) coupled to high-resolution mass spectrometry (Waters Autospec Ultima) (HRGC/HRMS) with selected ions at resolution 10,000. Samples were injected into non-polar DB5MS-type gas chromatography columns. The quantification was performed using the corresponding isotope-labeled compound as internal standard. Three replicates were analyzed for each food sample. The detection limit (LOD) was 1.0 ng/kg.

2.3. Estimation of dietary intakes and data analysis

Consumption data by the general population of Catalonia of the analyzed foodstuffs were obtained from Serra-Majem et al. (2003). Twenty-four hours recall questionnaires were also used. Total dietary intake of HCB for each food group was calculated by summing the results of multiplying the concentration in each specific food item by the amount (proportionally estimated) consumed of that food. Finally, total dietary intake was obtained by summing the respective intakes from each food group. For calculations, when the concentration of HCB in a determined food item was under the LOD, that value was assumed to be equal to one-half of the LOD (ND = 1/2 LOD). Data were evaluated using the statistical software SPSS 17.0. Given the non-parametric distribution of the samples, the Kruskal–Wallis or the U-Mann Whitney tests were used to assess the statistical significance of differences between food groups or age of the consumers, respectively. A probability of 0.05 or lower ($P \leq 0.05$) was considered as statistically significant.

A probabilistic assessment was also performed by means of a Montecarlo method, taking into account the variability associated to food intakes per unit of body weight (simulated by non-parametric distributions derived from the dietary intake data) and the uncertainty of the mean concentration of individual residues in each food (simulated by lognormal distributions). For risk assessment, the commercially available software package Crystal Ball (Version 4.0) was used. Crystal Ball propagates the uncertainty and variability of the parameters throughout the calculation of the risk. This propagation results in a distribution function for the risk estimated. Crystal Ball uses a Monte-Carlo simulation in order to propagate the distributions. The Monte-Carlo simulation calculates the risk several thousand times by drawing parameter values randomly from the distribution functions (Ferré-Huguet, Nadal, Schuhmacher, & Domingo, 2009; Pasuello, Mari, Nadal, Schuhmacher, & Domingo, 2010). The simulations were run in an Excel© spreadsheet. In the probabilistic approach, we used the raw data from the frequency-quantity questionnaires of the dietary survey (Serra-Majem et al., 2003), while in the deterministic estimation we used the data from 24-h recall questionnaires. Therefore, some differences might be expected in the results because of the particular biases of both types of dietary estimation methodologies. Frequency survey data for children were not available. Consequently, this population group was not included in the probabilistic assessment.

3. Results and discussion

The concentrations of HCB in a number of food samples purchased from Catalonia are shown in Table 1. The highest mean