Environmental Research 156 (2017) 714–724

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Integrated assessment of infant exposure to persistent organic pollutants and mercury via dietary intake in a central western Mediterranean site (Menorca Island)

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ARTICLE INFO

Keywords:
Organochlorine compounds
Mercury
Food
Dietary exposure
Menorca Island

ABSTRACT

In this research the levels of organochlorine compounds (OCs) and mercury (Hg) in several food items from Menorca Island were presented. The dietary exposure assessment was performed in children population from the island. Finally, body burden of OCs and Hg in these infants were associated with their dietary intakes of the selected food items.

The dietary exposure to persistent pollutants by children population from Menorca Island was assessed. The concentrations of 11 organochlorine pesticides, 6 polychlorinated biphenils (PCBs) and 1 inorganic toxic element, Hg, were determined in 46 food samples that included fish, shellfish, meat, fruit, vegetables, cheese and eggs, which were acquired in local markets and department stores in the Menorca Island. The most contaminated food items were fish and shellfish, followed by meat and cheese products. OC levels were similar or lower than in other previous studies. However, 66% of the analysed fish and shellfish species for Hg exceeded the human consumption safety limits according to the European Union Legislation. Pollutant data from food was combined with the pattern of consumption of these foodstuffs in order to calculate the estimated daily intake (EDI) of these contaminants. According to our results, fish and fruit were the main sources of OCs to the EDIs (contributing to 37% and 29%, respectively) while fish and shellfish were the main sources of Hg (76% and 17%). The estimated EDIs of OCs were well below to the reported FAO/WHO Tolerable Intakes. However, estimated weekly intake of Hg would exceed the Provisional Tolerable Weekly Intake indicated by EFSA in the case that the only fish and seafood source would be from the central western Mediterranean. Direct associations between fish/shellfish consumption and hair concentrations of Hg and fish and meat consumption and 4,4'-DDT concentrations in venous serum in the Menorcan children were observed.

1. Introduction

Anthropogenic release of organochlorine compounds (OCs) and mercury (Hg) into the environment has led to negative effects in human beings and ecosystems (Saeedi and Dehpour, 2016; Lamborg et al., 2014). The OCs are synthetic products widely used since the 1920s for several applications. Some organochlorine pesticides (OCPs) were extensively used in agriculture due to their insecticidal and fungicidal properties. Polychlorinated biphenyls (PCBs) were used for a large number of industrial applications, such as dielectrics in transformers, coatings, paints and insulating fluids (Wurl and Obbard, 2005).

On the other hand, the environmental occurrence of Hg has been enhanced by mining and fossil fuel combustion (Lamborg et al., 2014). Hg is present in many chemical forms, being methylHg more toxic than the original metal. In the aquatic environment, Hg is readily transformed to methylHg by anaerobic bacteria. Most of the metal incorporated into aquatic organisms is in the methylated form (80–90%; Harris et al., 2003).

Human exposure to OCs has been related to several health effects including cancer, reproductive defects, neurobehavioral abnormalities and endocrine and immunological toxicity (Mrema et al., 2013). On the other hand, methylHg is a potent neurotoxic agent that can cause severe neurological damage to humans (Grandjean et al., 1997).

Despite their different origin and structure, OCs and Hg share common properties such as (I) strong chemical stability and environ-

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http://dx.doi.org/10.1016/j.envres.2017.04.030
Received 21 February 2017; Received in revised form 19 April 2017; Accepted 21 April 2017

mental persistence, (II) bioconcentration in living organisms and biomagnification through the food chain due to their hydrophobic character and (III) toxicity for humans and wild animals. Accordingly, the production and use of OCs and Hg has been restricted and/or banned in many countries. However, these contaminants are still found in the environmental compartments (Arellano et al., 2011; Lamborg et al., 2014), foodstuff (Martí-Cid et al., 2010; Olmedo et al., 2013) and human tissues (Vizcaino et al., 2014; Garí et al., 2013).

Dietary intake is a major way of incorporation of these compounds into humans. This is particularly important for children since their organs and metabolism are still under formation and pollutant exposure may have stronger long term effects than in adults (Guxens et al., 2012; Vizcaino et al., 2014).

Assessment of the main routes of exposure to these chemicals in infant populations is needed for implementation of adequate strategies towards minimization of pollutant intake. Previous studies have investigated the main routes of exposure to OCs (Leontjew et al., 2016; Ahmad et al., 2010; Stefanelli et al., 2004) or Hg (Rubio et al., 2008; Olmedo et al., 2013; Millour et al., 2011) but to the best of our knowledge none of them has performed integrated assessments on both types of pollutants in the same cohort. Concerning the OCs, the present study encompasses both PCBs and OCPs.

Menorca Island is located in the central Western Mediterranean (39°47' −40°05'N, 3 °47' −4 °19'E) and has an extension of 702 Km^2. This small island represents a relatively isolated Mediterranean environment in which a large proportion of the food consumed by the inhabitants is generated locally. Furthermore, most of the fish consumption depends on the captures by local fishermen. The island has not any industry involving Hg use or production of OCs. Until the emergence of tourism, the main economic activity was agriculture. This island constitutes a representative case of a population living in a Mediterranean environment. High Hg levels are observed in the food-web of this semi-enclosed sea (Cossa et al., 1997; Gagnon et al., 1997; Pirrone et al., 2003; Heimburger et al., 2010; Žagar et al., 2007) despite its anti-estuarine circulation pattern that transfers nutrients and pollutants to the Atlantic Ocean. Identification of the routes of transport of this metal and other pollutants into the population living in sites under Mediterranean influence is important. Menorca can be taken as a model ecosystem of communities living in marine Mediterranean environments in which the main routes of dietary pollutant incorporation of OC and Hg can be assessed.

2. Materials and methods

2.1. Sampling

Food samples were acquired in local markets and department stores from the main two cities: Maó and Ciutadella (Fig. 1; December 2014, n = 46). The selection was based on Food Frequency Questionnaires (FFQs) from 4 year old children of the Menorca cohort (see next subsection below) and interviews with local experts.

The food samples included fish species of several trophic levels and fat contents (Table 1) and shellfish (Table 2), all them captured in the island surroundings. These fish species were representative of the local consumption, encompassing angler (*Lophius piscatorius*), European hake (*Merluccius merluccius*), black seabream (*Spondyliosoma cantharus*), common dentex (*Dentex dentex*), common pandora (*Pagellus erythrinus*), common seabream (*Pagrus pagrus*), dusky grouper (*Epinephelus marginatus*), forkbeard (*Phycis phycis*), Mediterranean moray (*Muraena helena*), red scorpionfish (*Scorpaena scrofa*), small-spotted catshark (*Scyliorhinus canicula*), thornback ray (*Raja clavata*), red mullet (*Mullus barbatus*), red sea bream (*Pagellus acarne*) and surmullet (*Mullus surmuletus*). The selected shellfish species were mussel (*Mytilus galloprovincialis*), squid (*Loligo vulgaris*), scampi (*Nephrops norvegicus*) and shrimp (*Aristeus antennatus*).

The food samples examined also included meat, e.g. beef, chicken and lamb (Table 2), fruits and vegetables (Table 3), cheese (Table 3) and chicken eggs (Table 3), all them produced locally.

After collection, each food item was dissected in two composite samples for OC and Hg analyses. The former were wrapped in aluminium foil and the second were sealed in plastic bags. The samples were then frozen at −23 °C until further analysis in the laboratory. Only the edible parts of each food item were analysed.

2.2. Study population

The Menorca cohort is part of the INMA (Spanish Children's Health and Environment) project, a research network that focuses on the effects of environmental contaminants during pregnancy and on fetal and child development (Ribas-Fito et al., 2006; Guxens et al., 2012).

Between 2001 and 2002, 4-year old children from this cohort provided serum samples (n = 285) and hair samples (n = 302) for the determination of OCs and Hg, respectively. These data have been described in Carrizo et al. (2006) and Garí et al. (2013), respectively. Children's dietary intakes were assessed through FFQs (Vioque and Gonzalez, 1991; Willet et al., 1985). Only the dietary intakes which concurred with the analysed food items were selected for study, as follows: fish, shellfish, meat (beef and chicken), fruit, vegetables, cheese (semi-skimmed cheese) and eggs. The rest of food items (e.g. dairy products, cereals, fats) were not included in the analyses.

All child's parents gave written permission for participation in the study, which was approved by the Ethics Committee of the Institut Municipal d'Investigació Mèdica (Barcelona).

2.3. Chemicals and standards

Solvents for residue analysis, *n*-hexane, dichloromethane, isooctane, acetone, concentrated 95–97% sulphuric acid, 65% nitric acid, 30%

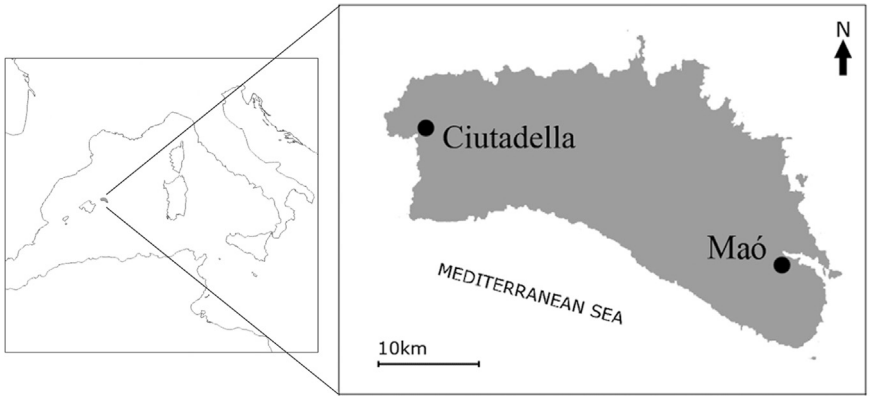

Fig. 1. Map of the Menorca Island.