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Biomonitoring of mercury in hair of breastfeeding mothers living in the Valencian Region (Spain). Levels and predictors of exposure

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HIGHLIGHTS

- Total mercury in hair was determined in 120 samples from breastfeeding women.
- The geometric mean of Hg in hair was of 1.22 μg g^{-1}.
- Mercury concentration in hair was 6 times higher than in other European studies.
- 27% of mothers exceed the EFSA health guidance value of 1.9 μg g^{-1}.
- Older age, smoking and high fish consumption were the major predictors of mercury in hair.

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ABSTRACT

This study focused on the evaluation of the levels of total mercury in hair among 120 breastfeeding mothers aged 20 to 45 -. The concentrations of Hg ranged from 0.07 to 6.87 μg/g with a geometric mean (GM) of 1.22 μg/g. This GM is six times higher than the average internal exposure of mothers from other 17 European countries (0.225 μg/g). Near 70% of mothers presented levels of Hg above the USA EPA internal exposure guideline of 1 μg/g, and 27% exceeded the EFSA health-based guidance value of 1.9 μg/g. The multivariate regression analysis revealed that age, smoking and fish consumption (sword fish, small fat fish, small lean fish) were the major predictors of mercury in hair.

1. Introduction

Human exposure to mercury is a cause of concern owing to the heavy weight of evidence on the adverse health effects produced by this metal (ATSDR, 1999). Mercury in its various forms, but mainly as methylmercury (MeHg) and metallic-mercury (Hg^0) vapours, has been found to cause neurological, nephrological, cardiac and reproductive disorders, as well as genetic damage (Zahir et al., 2005).

Mercury is a ubiquitous heavy metal, naturally present in the earth's crust that can be release in the environment naturally or because of human activities (Pacyna et al., 2006; Chen et al., 2016). Anthropogenic sources have increased its concentrations in the environment about three fold over the last century (Lamborg et al., 2014). Mercury emitted to the atmosphere, primarily as Hg^0, can

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travel long distances before being deposited to the earth surface in dry or wet deposition. When Hg is released into the environment, mainly in aquatic systems (Braune et al., 2015), it suffers a process of transformation to form MeHg, the most toxic form which bioaccumulates in marine organisms and is biomagnified through the trophic chain (EFSA, 2012).

The main sources of Hg exposure for environmentally-exposed individuals are diet (where MeHg predominates) and dental amalgam (which releases inorganic Hg, mainly as metallic Hg^0) (Castaño et al., 2012). Exposure to MeHg is primarily through consumption of fish and seafood (Marin et al., 2017). After ingestion about 95% of MeHg is absorbed in the gastrointestinal tract, while the uptake of inorganic species is much more limited (for $Hg^0 < 1\%$) (Akerstrom et al., 2017).

The human risk assessment to chemicals traditionally follows the classical approach of estimating external exposure (food, air, water), and comparing this intake with health-based guidance values (EPA, 2001). However, there is a growing interest in evaluating exposure to environmental chemicals using biomonitoring (HBM) (National Research Council, 2006). Levels of elements in biological matrices from HBM studies provide information about the dose really taken up from every route and pathway (Yusà and Pardo, 2015). Blood, urine and hair are the biological matrices currently used to measure the internal dose of Hg. The total-blood Hg concentration is mostly due to dietary intake of MeHg, while Hg in urine mainly reflects exposure to inorganic Hg. Total Hg in scalp hair has been widely used as a biomarker to assess long-term MeHg exposure. Hair is widely recommended because i) it is a non-invasive sample, ii) concentrations of Hg are high compared to other matrices and iii) Hg levels correlate to the concentrations in blood, with a frequently cited total blood to hair ratio of 1:250, although large variations exist (EFSA, 2012). Methylmercury usually constitutes at least 80% of the total mercury analysed in hair among fish consumers (McDowell et al., 2004). Therefore, hair mercury is a very good biomarker for MeHg, and is often used to characterise methylmercury exposures.

There is a general recommendation that pregnant women, children and women of childbearing age should avoid mercury exposure as much as possible (EFSA, 2015). Consequently, it is important to know the actual exposure in the general population and particularly in these groups of special risk, in order to evaluate the success of the recommendations and programmes of control and risk management.

The present manuscript reports results for mercury in hair of breastfeeding mothers participating in a biomonitoring programme (BETTERMILK project) in the Valencian Region (Spain). Exposure results were compared with national and international studies and with health-based guidelines in order to perform a risk assessment. We obtained information on personal characteristics, diet and lifestyle of the participating mothers, and the correlation of hair mercury with physical and socio-demographic variables were examined. Likewise, multiple linear regression analyses were carried out to identify predictors of exposure to organic mercury.

2. Materials and methods

2.1. Study area and population

University and Polytechnic Hospital “La Fe” (Valencia, Spain) was used as study site. Lactating mothers who had given birth from June to November 2015 were asked to participate in the study. The selection criteria for joining the programme were: i) a normal pregnancy and delivery with healthy mother and child, ii) mother having lived in the Valencian Region for at least 10 years and iii) having decided to breastfeed.

A total of 120 lactating mothers aged between 20 and 45 volunteered to join the study. The sample size met the recommendation of the International Federation of Clinical Chemists (IFCC) on the calculation and application of coverage intervals for biological reference values (Poulsen et al., 1997).

2.2. Samples and data collection

Hair was collected between 2 and 8 weeks after birth. In order to harmonise sampling with other European Biomonitoring studies, hair samples were collected according to the procedure used in the COPHES/DEMOCOPHES project (Esteban et al., 2015). Briefly, hair was sampled in the occipital region using scissors that had been previously cleaned with ethanol 96°. The number of hair tufts (10 mm width) sampled varied according to the hair length (see Table S1). Samples were stored until analysis at room temperature in a dry and dark place.

Questionnaires with detailed information on socio-demographic characteristics, lifestyles and diet (Food Frequency Questionnaire) were administered to the studied population. The variables studied are presented in Tables 1 and S2. Food consumption frequency by groups was converted to semi-quantitative intakes ($g \cdot month^{-1}$, or $mL \cdot month^{-1}$ in drinks). The calculation was achieved adding the monthly intake of individual foods of the same group. Monthly intake of individual foods was estimated according to the following formula:

Food intake ($g \cdot month^{-1}$, or $mL \cdot month^{-1}$ in drinks) = [weight in g (volume in mL in drinks) of the portion size] x [Intake

Table 1
Studied population characteristics.

Mother Age (years) (n = 120)	33 (20–45)[a]
Diet during pregnancy	n (%)
Yes	17 (14.41)
No	101 (85.59)
Mother Country of birth	
Spain	104 (88.14)
Foreign	14 (11.86)
Mother Occupational status	
Employed	100 (84.75)
Unemployed	18 (15.25)
Smoker (mother)	
Yes	9 (7.5)
Ex-smoker	48 (40)
Never	63 (52.5)
Child gender	
Boy	47 (40.17)
Girl	70 (59.83)
Mother Place of residence	
Urban	85 (80.19)
Rural	21 (19.81)
Mother was Breastfed	
Yes	81 (70.43)
No	34 (29.57)
Consumption of Fish products	**g month^{-1}**
Fish products	4242.5 (1040–15640)[a]
Fish[b]	2925 (660–11950)[a]
Seafood[b]	1070 (380–5280)[a]
Bivalve molluscs[c]	675 (270–4400)[a]
Lean fish[d]	1200 (200–8800)[a]
Fat fish[d]	1310 (390–7010)[a]
Small fat fish[e]	575 (200–5150)[a]
Big fat fish[e]	535 (190–4560)[a]

n = number of participants. More variables are presented in Table S2.
[a] Values expressed as median (minimum – maximum).
[b] Subdivision of the group Fish products.
[c] Subdivision of the group Seafood.
[d] Subdivision of the group Fish.
[e] Subdivision of the group Fat fish.