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Probabilistic mercury multimedia exposure assessment in small children and risk assessment

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ABSTRACT

Objectives: Emissions of mercury in the environment have been decreasing for several years. However, mercury species are still found in different media (food, water, air and breast-milk). Due to mercury toxicity and typical behaviour in children, we have conducted a mercury exposure assessment in French babies, and small children aged 0 to 36 months.

Method: Consumption and mercury concentration data were chosen for the exposure assessment. The Monte Carlo technique has been used to calculate the weekly exposure dose in order to integrate inter-individual variability and parameter uncertainty. Exposure values have been compared to toxicological reference values for health risk assessment.

Results: Inorganic mercury median exposure levels ranged from 0.160 to 1.649 μg/kg of body weight per week (95th percentile (P95): 0.298–2.027 μg/kg bw/week); elemental mercury median exposure level in children was 0.11 ng/kg bw/week (P95: 28 ng/kg bw/week); and methylmercury median exposure level ranged from 0.247 to 0.273 μg/kg bw/week (P95: 0.425–0.463 μg/kg bw/week). Only elemental mercury by inhalation route (indoor air) and methylmercury by ingestion (fish and breast-milk) seem to lead to a health risk in small children.

Conclusions: These results confirm the importance of assessing total mercury concentration in media like breast-milk, indoor air and dust and methylmercury level in food, other than fish and seafood. In this way, informed monitoring plan and risk assessment in an at-risk sub-population can be set.

1. Introduction

Mercury occurs in the environment in various forms: elemental mercury (Hg0), inorganic mercurial salts (e.g. mercuric chloride (HgCl2)) and organic mercury (e.g. methylmercury (MeHg)) (EFSA, 2012a). Mercury emissions into the air have steadily decreased in recent years (−82.5% throughout the 1990–2010 period), particularly since the ban in batteries and medical thermometers and the improvements in waste incineration performance (CITEPA, 2012). In France, chlorine and soda production industries caused mercury emissions predominantly in the Alsace region (Rémy et al., 2003). Concerning French soil, three anomalies were discovered by Ottesen et al. (2013): Paris, Verdun and the Vosges department. Waste incinerators and industries are probably the cause of these mercury contaminations in soil. About French seacoasts, in 2011, the IFREMER (French Research Institute for Exploitation of the Sea — Institut Français de Recherche pour l'Exploitation de la Mer) has revealed mercury concentrations in mussels and in oysters from the Mediterranean Sea (Harbours of Toulon and Marseille) and the English Chanel ("Pays de Caux") that were two to four times higher than the French median mercury level (0.12 mg/kg) (Belin et al., 2012). Freshwater from the Seine estuary, the North and the North-East of France is the most contaminated freshwater due to high mercury concentrations in sediments (Eau-Evolution, 2013).

However, the environmental cycle of mercury is very complex and this metal and its compounds are still found in the air, soil and water, and can contaminate food and indoor dust (Thomassin et al., 2003). In fact, mercury can be turned into numerous mercurial forms in the environment. In the air, elemental mercury can be absorbed on particles or oxidised into inorganic mercury (UNEP Chemicals, 2002). Its compounds are deposited in the soil and are immobilized in the first 10 cm of soil (Thomassin et al., 2003). However, a portion of mercury in soil is rapidly volatilised to the air due to the reduction of inorganic mercury to elemental mercury (Selin, 2009). Organic mercury is generated by micro-organisms and/or natural chemical processes from inorganic mercury (methylation). These reactions are essentially done in water to produce MeHg, which bio-accumulates and biomagnifies in edible fish and marine mammals (UNEP Chemicals, 2002).

Health hazards of mercury have been known for centuries, essentially due to observations in Minamata (Japan) and Iraqi epidemics (Counter and Buchanan, 2004). Mercury is highly toxic to human kidney or central nervous system, according to mercury species. Foetuses and small children are more sensitive to this metal, especially to MeHg, due to their

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intense neurological and physiological development (Moya et al., 2004; WHO, 2009).

Furthermore, infants (0–11 months) and toddlers (12–36 months) are an at-risk sub-population because of their typical behaviour (hand-to-mouth activity, outside activities, breast-milk consumption…) and physiological differences (body weight, inhalation rates…). Also, their exposure to mercury was considered to be higher than other sub-populations for these previous reasons (Moya et al., 2004).

Generally, risk assessments have been based on food exposure to MeHg and more particularly on fish intake. In France, we could cite the National Food Safety Agency (AFSSA, now renamed ANSES), which worked on mercury exposure from food in children over 3 years old and adults. A recommendation on fish consumption was estimated and published for young children (1–30 months): no more than 60 g of fish a week (AFSSA, 2004). More recently, the EFSA conducted a risk assessment of inorganic mercury and MeHg based, among others, on food and breast-milk consumption in European infants and toddlers (EFSA, 2012a).

However, little work has been done to understand multimedia exposure to various forms of mercury in infants and toddlers in France (from birth to 36 months old). It was demonstrated that breast-feeding was considered like a major source of mercury exposure in the first six months of life (Chien et al., 2006).

Moreover, children can swallow soil and dust due to the hand-to-mouth behaviour and outside games. It is essentially inorganic mercury that is found in soil and dust, which is less toxic than MeHg. Knowing that mercury is immobilized in the first 10 cm of soil (Thomassin et al., 2003), and therefore reachable for children, this exposure route cannot be dismissed for this at-risk subpopulation.

So, a multimedia analysis was necessary to achieve a comprehensive and accurate assessment of mercury exposure in French small children. In this effort, two sources of exposure were chosen: ingestion (food, breast-milk, soil and dust) and inhalation (indoor and outdoor air). Dermal contact with water, during shower, has not been studied in this work. In fact, Chien et al. (2006) have considered this type of exposure to be negligible. Moreover, numerous forms of mercury require an exposure and a risk assessment for each mercurial compound found in the children's environment: elemental mercury, inorganic mercury and methylmercury. The purpose of this study was to present an exposure assessment to mercury in French infants and toddlers for a chronic health risk. In this effort, the Monte Carlo technique was used to integrate inter-individual variability and parameter uncertainty (US EPA, 2001a). The contribution of all various media has been also detailed for small children. Finally, the health risk was evaluated by the determination of hazard quotients (HQs) and the percentage of exposed population over different toxicological reference values (TRVs).

2. Material and methods

2.1. Contamination and consumption data origins

2.1.1. Breast-milk

Unfortunately, no studies have dealt on breast-milk consumption and mercury concentration in France. So, foreign data have to be necessarily studied. Breast-milk consumption data came from the study of Butte et al. (2002). It concerned 1397 mother and infant pairs in Australia, USA, Canada and Europe (Finland, UK, Sweden…). The consumption of exclusively breast-milk consumers only was described. 24-hour or 48-hour test-weighing was mostly used to evaluate breast-milk intake. This variable followed a normal distribution (769 ± 54 mL/d) (Butte et al., 2002).

The EFSA (2012a) has reviewed some European surveys about breast-milk contamination. We chose a recent study conducted from 2007 to 2009 on the Mediterranean area (Italy, Slovenia, Croatia and Greece) (Miklavcic et al., 2013). Italian concentrations were chosen for exposure assessment because Italy is close to France and the number of breast-milk samples was high ($N = 605$). P5 (5th percentile), P50 (50th percentile) and P95 (95th percentile) (0.05, 0.2 and 0.8 μg/kg) were used to set a lognormal distribution.

2.1.2. Infant formula, standard and babies' food

The survey of Fantino and Gourmet (2008) furnished a nutritional data of food items. It was conducted in 2005 on children aged 0 to 36 months. Overall, 706 French infants and toddlers, not breastfed, were included in the study. The sample representativeness of this French sub-population was based on region size, children's age, mothers' professional activity and family's socio-economic group. Parents recorded food quantities taken by children with an individual three-day report. Unfortunately, raw data for food consumption were not available. In fact, this study was commissioned by a private organisation (i.e. the French Syndicate of Children's Food — Syndicat Français des Aliments de l'Enfance). So, food intake was calculated by the following equation:

$$\mathrm{FI} = (\%\mathrm{EI}_f \times \mathrm{DEI})/\mathrm{E}_f$$

where FI: food intake (g/d); $\%\mathrm{EI}_f$: percentage of energy intake attributable to food item f; DEI: total daily energy intake (kJ/d) (Fantino and Gourmet, 2008) and E_f: energy of food item i (kJ/g). Nutritional values of foodstuffs were provided from products sold in French markets.

For milk-based formula, infants aged 0 to 5 months consumed, on average, 106 g of "starting" infant formula powder per day (g/d). For toddlers (6–36 months), the consumption of "follow-on" formula decreased with age from 75.4 to 3.8 g/d. Mean consumption values for each age group (6, 7, 8–9, 10–12, 13–18, 19–24, 25–30 and 31–36 months) were calculated. Then, a normal distribution was adjusted on the global mean consumption level. Mean and standard deviation (SD) (39 ± 3 g/d) were obtained from the simulated distribution.

For infant food, seven groups were defined: ultra-fresh dairies, cereals, soups and vegetable-based purees, fruit juices and ready-to-eat food: vegetable-based, meat or fish-based and fruit-based. Concerning standard food, twenty categories were detailed for consumption data: milk and ultra-fresh dairies, cheese, cereals, vegetables, potato based-products, starchy, meat, delicatessen, fish and seafood, eggs, cooked dishes, fruits, fruit juices, sweeteners, chocolate, bread and rusk, cakes and biscuits, soft drinks, animal fat, vegetable fat and condiments.

The EFSA reported the statistical description of total mercury (THg) concentrations in infant food group and milk-based formula taken in the European market (EFSA, 2012a). The CONTAM panel of the EFSA followed WHO's recommendations:

- LB (lower bound) assumption: if concentration is inferior to LD (limit of detection, i.e. 5 ng/g), it is replaced by 0 and if value is between LD and LQ (limit of quantification, i.e. 10 ng/g), it is replaced by LD.
- UB (upper bound) assumption: LD is kept if concentration is inferior to LD and LQ replaces concentration if this one is included in LD and LQ.

Concerning standard food, contamination data from EAT 2 (TDS – Total Diet Study – in France) from ANSES was used (ANSES, 2011; Arnich et al., 2012). Analyses were conducted in 2006 and total mercury was analysed in 1319 samples of food usually consumed by the French population. The French mercury levels were used for infants' foodstuffs with a low number of samples (ready-to-eat fruit-based, fruit juices and yogurt, cheese and milk-based dessert) in the EFSA report (EFSA, 2012a).

Total mercury mean for LB and UB assumptions for each food group were used for exposure calculation (Table 1).

2.1.3. Drinking water

The level of mercury in drinking water was obtained by the EFSA's review in European foodstuffs (EFSA, 2012a). Analyses were conducted