

Determination of total mercury in Spanish samples of baby food, fast food, and daily meal

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Research Article

Keywords: Infant food, Canteen menus, Toxic element, Tolerable Daily Intake, Atomic absorption spectrometry, Direct mercury analysis

Posted Date: May 5th, 2022

DOI: <https://doi.org/10.21203/rs.3.rs-1614710/v1>

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Abstract

This study investigated the levels of total mercury (THg) in Spanish samples of baby food, fast food, and daily meal, which people of different ages consume, to evaluate potential toxicological risks through the contribution to the Tolerable Daily Intake (TDI) of this element. The total mercury concentrations were determined in thirteen commercial baby foods for infants 6 to 12 months old, six kinds of fast foods prepared for children, and nine canteen menus prepared for adults. Samples were homogenized, freeze-dried, and analyzed using a direct mercury analyzer (DMA). The concentration ranges found were as follows: baby food ($0.57\text{-}41.9 \mu\text{g kg}^{-1}$), fast food ($0.54\text{-}67.7 \mu\text{g kg}^{-1}$), and adult menus ($0.43\text{-}638 \mu\text{g kg}^{-1}$). The recovery of different amounts of spiked mercury ranged from 98.6% to 104.9%, and the accuracy of the method was checked with an analysis of different certified reference materials. The contribution of the samples to the TDI of mercury varied as follows: baby food (0.3-27.8%), fast food (0.5-102.2%), and adult menus (0.3-395.7%). Therefore, it was concluded that total mercury daily intake does not pose risks to Spanish infants, not adults, if not tuna was included on the menu, and it must be taken care of with fast food.

1. Introduction

In recent years, information about the concentrations of toxic elements in foods has become particularly significant, given their potential risk to human health in dietary intake. Mercury (Hg) is a non-essential element to the human body, toxic in low concentrations, and constitutes a potential risk to health due to its classification as a carcinogen, bioaccumulative character, and tendency towards magnification in the food chain (Lavoie et al. 2013; Li et al. 2015; Park and Zheng 2012). Studies have shown that food consumption is the primary source of mercury exposure (Bose-O'Reilly et al. 2010; de Roma et al. 2017; Hernández-Martínez and Navarro-Blasco 2013). Excessive intake of Hg may cause damage to the central nervous, cardiovascular, and reproductive systems and affect some physiological functions, such as the kidneys (Choi and Grandjean 2008; Franco et al. 2007).

The risk of total mercury (THg) intake in the diet of people at different ages is based on the Provisional Tolerable Weekly Intake (PTWI) of THg at $4 \mu\text{g kg}^{-1}$ of body weight, that is, a Tolerable Daily Intake (TDI) of $0.57 \mu\text{g kg}^{-1}$ of body weight, values as recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (WHO 2011). Studies indicate that children are especially vulnerable and may be more exposed to contaminated food than adults (Bose-O'Reilly et al. 2010; WHO 2010). According to the National Health and Nutrition Examination Survey (NHANES), which evaluates the health and nutritional status of adults and children in the United States, children under the age of 10 are exposed to about $0.33 \mu\text{g}$ of Hg per kilogram of body weight per day in the food they eat. However, children between 11 and 14 years presented a Hg dose ($0.15 \mu\text{g Hg/kg bw/day}$) about half of this, and adults ($0.05 \mu\text{g/kg bw/day}$) about six times less when compared to younger children. The daily intake of Hg is higher for children than for adults due to the higher consumption of food by bodyweight of the former to support their growth (Ruggieri et al. 2017).

In a general population, mercury dietary intake mainly comes from fish and shellfish consumption because these have a higher mercury concentration in their bodies, originating from the food chain in aquatic systems. Mercury can also be found in vegetables, seafood, and other foodstuffs in the human diet but in a lower concentration, representing lower exposure to Hg from these foods. Llobet et al. (2003), for example, determined Hg, arsenic (As), cadmium (Cd), and lead (Pb) in different food samples acquired in Catalonia, Spain. These authors found the following mean concentrations of Hg in wet weight of food: vegetables (< LOD), pulses (< LOD), cereals ($25 \mu\text{g kg}^{-1}$), tubercles ($2.5 \mu\text{g kg}^{-1}$), fruits (< LOD), fish and shellfish ($95 \mu\text{g kg}^{-1}$), meat ($12 \mu\text{g kg}^{-1}$), eggs ($10 \mu\text{g kg}^{-1}$), dairy products ($11 \mu\text{g kg}^{-1}$), milk ($2.5 \mu\text{g kg}^{-1}$), and fats and oils ($25 \mu\text{g kg}^{-1}$). Exposure to toxic elements such as Hg is hazardous for pregnant women and young children (Martí-Cid et al. 2007; Ortega-García et al. 2009).

The development of reliable methods to determine total mercury from (ultra)trace levels in food samples is gaining importance. Various researchers in different countries have estimated mercury dietary intake, for example, in Chile (Bastías et al. 2010; Muñoz et al. 2005), China (Sun et al. 2011; Wei et al. 2019), France (Leblanc et al. 2005), Germany (Wilhelm et al. 2003), Hong Kong (Chung et al. 2008), Iran (Karami et al. 2021; Karimi et al. 2021; Shariatifar et al. 2020), Italy (de Roma et al. 2017), Korea (Koh et al. 2012), Poland (Jedrzejczak 2002; Koch et al. 2016), Spain (Falcó et al. 2006; González et al. 2019; Martí-Cid et al. 2008; Rubio et al. 2008), Sub-Saharan Africa (Jitaru et al. 2019), Sweden (Jorhem et al. 1998), and The United Kingdom (Rose et al. 2010) conducting their total diet studies. These have demonstrated that mercury exposure is a crucial public health concern.

In the past few years, there has been significant growth in the number of research studies involving the mercury content in foods for infants and toddlers (Guérin et al. 2018), such as infant cereals (Cui et al. 2017; Hernández-Martínez and Navarro-Blasco, 2013), and infant formula (Başaran 2022; Guérin et al. 2018; Mania et al. 2015; Melø et al. 2008). However, in the literature, few research papers deal with the specific question of mercury content in baby food (Carbonell-Barrachina et al. 2012; de Roma et al. 2017; Martins et al. 2013; Pandelova et al. 2012; Tóth et al. 2014; Zand et al. 2012). On the other hand, few studies on the levels of mercury present in fast food samples and canteen menus were found (de Roma et al. 2017). Despite this, research has already been carried out on the mineral profile of similar foods, as studies published recently determined 12 elements in children's fast food (Ruiz-de-Cenzano et al. 2017) and 14 elements in commercial baby food (Mir-Marqués et al. 2015).

Various techniques have been employed to determine mercury levels in food samples based on cold vapor atomic absorption spectrometry (CV AAS) and cold vapor atomic fluorescence spectrometry (CV AFS). These are often employed to determine low mercury levels in food samples and a variety of matrices (Brombach et al. 2017; Chung et al. 2008; Cui et al. 2017; Muñoz et al. 2005; Rubio et al. 2008; Sun et al. 2011). Other techniques have also been employed, which include electrothermal atomic absorption spectrometry (ET AAS) (Malvandi et al. 2020; Sakanupongkul et al. 2019), and inductively coupled plasma-mass spectrometry (ICP-MS) (de Roma et al. 2017; Falcó et al. 2006; Martí-Cid et al. 2008; Sirot et al. 2018). However, most of the techniques generally involve the consumption of reagents in the digestion step sample treatment, generating hazardous and toxic wastes into the environment. This

makes it essential to develop safe and environmentally friendly alternative methods for the accurate determination of Hg.

A potential alternative is to use a direct mercury analyzer suitable for the direct determination of Hg in solid samples to provide fast and accurate results (Panichev and Panicheva 2015). Moreover, the method does not require acid digestion or sample preparation before analysis, thus eliminating the use and generation of substances hazardous to human health and the environment and providing a high sensitivity based on the use of a gold trap. The method is based on total thermal decomposition, gold trap collection of the Hg vapor, and atomic absorption determination. A previous application of the direct mercury analyzer has been reported to analyze total mercury concentration in different food items and evaluate human exposure to THg via daily intake (Cheng et al. 2013). The authors analyzed 58 types of food items (vegetables, fruits, fish, meat, viscera, eggs, and rice) in this work. Other works have also employed the direct mercury analyzer for the determination of Hg in food samples (da Silva et al. 2014; de Paiva et al. 2017; Guérin et al. 2018; Koch et al. 2016; Koh et al. 2012; Kuras et al. 2017; Martins et al. 2013).

The present study aimed to determine the total mercury content in a wide range of human menus (baby food, fast food, and daily meal) by employing a direct mercury analyzer. Also, the estimated dietary mercury intake results were compared with the Tolerable Daily Intake (TDI), recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) to assess the health risk (EFSA 2012; WHO 2011). Thus, we sought to assess how much the analyzed menus contributed to the TDI of Hg, considering values close to or above 100% as alarming since the studied menus do not represent the food intake for a whole day. This work is of great importance as it can serve as a basis for future studies that aim to estimate the risk of exposure of consumers to Hg, considering the frequent intake of baby food, fast food, and daily meal. Finally, there are few reports in the scientific literature on the THg content in these foodstuffs consumed by the population of Spain in different age groups.

2. Materials And Methods

2.1. Instruments and reagents

To prepare standard solutions containing mercury, we used a Hg (II) standard stock solution 1000 $\mu\text{g mL}^{-1}$ Merck standard solution (Darmstadt, Germany) and ultrapure water with a resistivity of 18.2 M Ω cm Milli-Q from Millipore (Bedford, USA).

The freeze-dried samples of baby food, fast food, and daily meal were analyzed with a Direct Mercury Analyzer DMA-80 from Milestone (Sorisole, Italy). The operation of the DMA-80 is as follows: the samples were dried and then thermally decomposed by controlled heating. The decomposed products were then carried to a catalyst by an oxygen flow, where sample oxidation was completed, and halogens and nitrogen/sulfur oxides were trapped. The final products passed through a mercury amalgamator, which collects Hg⁰. The Hg amalgamator was heated to 850 °C, the Hg⁰ accumulated was then released, and

the total mercury content was determined by measuring the absorption at 253.7 nm. The accuracy of the results was controlled by an analysis of certified reference material (CRM) in each calibration range. No reagents were required for sample preparation.

2.2. Samples

Thirteen baby food samples from different brands available in Spain were purchased in local markets and classified according to their meat, fish, or vegetable content. Six children's fast food menus were purchased from different commercial brands in Valencia, Spain's town, and shopping centers. They were composed of beef burgers with cheddar cheese, bread, potato chips, ketchup, mustard, yogurt or milkshake, and a drink. Nine daily menus were purchased in the University of Valencia canteens. They were composed of a starter, a main dish, a dessert, and included a piece of about 60 g of bread. Twenty-eight samples were analyzed, and their composition was described in Table 1.

Table 1

The samples selected for the study had a diversified composition to ensure greater robustness to the results obtained. In TDI studies, we consider that these menus are mainly consumed by the following age groups: baby food (7–24 months), fast food (3–12 years), and daily meal (adults). Baby food and fast food samples represent complementary sources in the diet of the applicable age groups (infants and children, respectively). They may occasionally replace some of the leading daily meals of these individuals. The daily meal samples are composed of varied foods, with sources of carbohydrates, proteins, and lipids distributed in a balanced way on the menu. For this reason, daily meals represent the main meals (lunch or dinner) in the diet of the applicable age group (adults).

An additional three certified reference materials were employed for quality control and to test the method's reliability. Chicken NCSZC73016 was supplied by the China National Analysis Center for Iron and Steel (Beijing, PR China), Fish Protein NRC DORM-3, and Lobster Hepatopancreas NRC TORT-2 were supplied by the National Research Council of Canada (Ottawa, Canada).

2.3. Sample preparation

Samples were crushed, homogenized, and frozen at -20 °C before freeze-drying for a minimum of 48 h at a chamber pressure of 0.05 mBar. Freeze-drying was performed to preserve and pre-concentrate the food samples by eliminating water content. After that, they were homogenized with a domestic Braun mixer (Kronberg, Germany) and stored in polyethylene tubes before analysis.

2.4. Determination of total mercury concentration in freeze-dried samples

To determine the total mercury concentration in baby food, fast food, and daily meal samples, 50 mg of each sample was weighed in a nickel crucible and introduced automatically into the Direct Mercury Analyzer (DMA-80). The measurements were performed in three replicates for each sample. The instrument allows for three sequential pre-concentration procedures and has a detection limit of 0.005 ng

of mercury and a maximum Hg mass of 1000 ng. Thus, two and three pre-concentrations with 50 mg of the sample were tested to study the matrix effect. Oxygen was used as the carrier gas. The operating conditions for DMA-80 are shown in Table S1 (Supplementary Material).

2.5. Assessment of Tolerable Daily Intake (TDI) of mercury from baby food, fast food, and daily meal consumption.

The risk of exposure of consumers to the Hg present in the menus was calculated as the percentage contribution to the TDI of this element. The Hg TDI values for the different age groups studied are as follows: infants from 7 to 12 months ($5 \mu\text{g day}^{-1}$), toddlers from 13 to 24 months ($7.4 \mu\text{g day}^{-1}$), children from 3 to 7 years ($11 \mu\text{g day}^{-1}$), children from 7 to 12 years ($20 \mu\text{g day}^{-1}$), and adults ($34 \mu\text{g day}^{-1}$) (WHO 2011). The daily intake (DI) of Hg, in $\mu\text{g day}^{-1}$, was calculated as follows: $\text{DI} = \text{DCM} \times \text{MMC} \times 1000$; where DCM is the daily consumption of the menu, in g day^{-1} , and MMC is the mean Hg concentration, in $\mu\text{g kg}^{-1}$. The studied menus did not represent the food intake for a whole day; therefore, safe values of Hg intake would be those considerably lower than 100% of the TDI.

The percentage of TDI was calculated as micrograms of Hg that each menu contained, divided by the values of TDI for each age, and multiplied by 100. The TDI for each age was calculated as micrograms of Hg per day, multiplying $4 \mu\text{g kg}^{-1}$ by body weight (according to the age) and divided by 7 days (EFSA 2012). Mean body weight was considered as follows: 9 kg for infants between 7–12 months; 13 kg for toddlers between 13–24 months (Hernández-Martínez et al. 2013); 19 kg for children between 3–7 years; 35 kg for children between 7–12 years, and 60 kg for adults (de Lara et al. 2010).

3. Results And Discussion

3.1. Instrument calibration

Calibration of the mercury analyzer was performed using standards in aqueous media. Two analytical curves with different concentration ranges were used to determine low (0–20 ng) and high (20–1000 ng) concentrations of Hg in food samples, employing cells with different optical path lengths, with the coefficient of determination (r^2) values higher than 0.99. The calibrations were checked every session employing certified reference material.

3.2. Analytical characteristics of the method

For the direct determination of Hg in a 50 mg dry sample mass, the analytical procedure provided a limit of detection (LOD) of $0.1 \mu\text{g kg}^{-1}$ and a limit of quantification (LOQ) of $0.3 \mu\text{g kg}^{-1}$, both based on the variations of ten independent blank measurements. A mass of 50 mg of wheat flour free of Hg was used as blank since the samples evaluated in this work were solid. The decontamination procedure of the direct mercury analyzer was carried out by analyzing the same wheat flour, followed by the analysis of 5% (v/v) HNO_3 , as described by da Silva et al. (2014).

The relative standard deviation for triplicate samples containing from 0.43 to 638 $\mu\text{g kg}^{-1}$ varied between 0.8 and 11%, generally lower than 5%. Recovery experiments on samples spiked at 2.5, 10, 25 and 500 $\mu\text{g kg}^{-1}$ were $98.6 \pm 0.9\%$, $99.2 \pm 2.0\%$, $102 \pm 3\%$, and $104.9 \pm 1.9\%$, respectively. The good recoveries obtained in all cases confirmed the lack of THg losses for these samples in a wide range of concentrations, indicating the accuracy of the developed methodology. Additionally, as can be seen in Table 2, the values obtained in our study generally agreed well with those reported for certified materials.

Table 2

According to the recommendations of the European Commission-Institute for Reference Materials and Measurements (IRMM), the difference between the certified and measured values (Δm) should be compared with the combined uncertainty of certified and measured value ($U\Delta$): If $\Delta m \leq U\Delta$, then there is no significant difference between the measurement result and the certified value (Linsinger 2010). According to the combined uncertainty obtained, the measured mean value was not significantly different from the certified value for all CRMs used (Chicken, Fish Protein, and Lobster Hepatopancreas). This analytical method using the same equipment (DMA-80) has already been evaluated for accuracy by employing other CRMs (Fucus IAEA 140-TM, Coal Fly Ash NIST 1633b, NIES Rice 10-a, 10-b, and 10-c) in previous work carried out by our research group (da Silva et al. 2014). Agreement between found and certified values was verified for the five CRMs used.

3.3. Effect of the sample mass

Three typical samples corresponding to each menu type under consideration were analyzed with sample masses from 50 to 150 mg, used in different proportions. A comparison was made between the values found at one step of the analysis and the amalgamation approach with two or three portions of 50 mg. As shown in Table 3, the amount of sample used had no acute effects on the concentrations obtained. However, when the Hg content in the sample was around 0.5 $\mu\text{g kg}^{-1}$ (Fast Food 2), the use of sample mass less than 100 mg produced better results. For method simplicity, a mass of 50 mg was selected, which was enough to obtain the appropriate data.

Table 3

3.4. Total mercury concentration in baby food, fast food, and daily meal samples

The total Hg content was determined in the twenty-eight menu samples employed in this study. The results are expressed in $\mu\text{g Hg}$ per kg of dry weight (d.w.) per sample. As Table 4 shows, the baby food samples contained between 0.57 and 41.9 $\mu\text{g kg}^{-1}$ d.w. of Hg. The lowest levels corresponded to vegetable purée (0.57–7.38 $\mu\text{g kg}^{-1}$ d.w.), followed by foods containing meat (2.51–21.8 $\mu\text{g kg}^{-1}$ d.w.). The highest concentration corresponded to fish and those with rice or vegetables (11.5–41.9 $\mu\text{g kg}^{-1}$ d.w.).

Table 4

On considering fast food menus aimed at children, the values found varied between 0.54 and 67.7 $\mu\text{g kg}^{-1}$ d.w. It must be noticed that there was no significant difference in the menu composition of these samples (Table 1). Also, variations in the mercury concentration could not be assigned to any of the components, and no relation with the brand was found. Regarding canteen menus, a mercury range from 0.43 to 19.4 $\mu\text{g kg}^{-1}$ d.w. was found, except for Daily Meal 8, where there was a high level of 638 $\mu\text{g kg}^{-1}$ d.w. of Hg. Once again, the level of Hg seems to be related to the presence of fish on the menu (13.5–19.4 $\mu\text{g kg}^{-1}$ d.w.), specifically the presence of tuna (638 $\mu\text{g kg}^{-1}$ d.w.), and rice with seafood products (3.14 $\mu\text{g kg}^{-1}$ d.w.).

According to Cheng et al. (2013), the results for THg ranged from 0.16 to 171 $\mu\text{g kg}^{-1}$, with the levels of Hg in fish significantly higher than in other food groups, such as vegetables, fruits, meat, viscera, eggs, and rice. Moreover, among the foods studied by these authors, rice and fish contributed most to the total daily intake of mercury. Other researchers have also studied the level of mercury in various types of food, including fish and shellfish.

Martorell et al. (2011) studied the dietary intake of Hg for 12 food groups, including meat, fish and seafood, vegetables, tubers, fruits, eggs, milk, and cereals. Among the analyzed foods, the tuna presented the highest concentration of Hg (222–776 $\mu\text{g kg}^{-1}$), only behind swordfish (869 $\mu\text{g kg}^{-1}$).

De Roma et al. (2017), in turn, evaluated the occurrence of toxic elements (As, Cd, Hg, and Pb) in different meals (baby food, fast food, vegetarian meal, canteen meal, and restaurant meal) in Italy. The Hg concentrations determined were relatively low (< 1.5–3.27 $\mu\text{g kg}^{-1}$), except for a restaurant meal sample (14.9 $\mu\text{g kg}^{-1}$). These authors related the higher level of Hg to the presence of seafood and cereals on this menu, placing these two foodstuffs among the most significant contributors to THg intake.

González et al. (2019), finally, investigated the presence of As, Cd, Hg, and Pb in foodstuffs (meat and meat products, fish and seafood, vegetables, eggs, milk and derivatives, bread and cereals, oils, industrial bakery, sauces, chocolates, and infant food) widely consumed in Catalonia, Spain. The Hg levels were below the LOD (< 2 $\mu\text{g kg}^{-1}$) of the method for most of the analyzed foods, except for fish and seafood, which showed a mean concentration of 152 $\mu\text{g kg}^{-1}$. The detailed study of this foodstuff revealed Hg concentrations ranging from 3 $\mu\text{g kg}^{-1}$ (panga) to 856 $\mu\text{g kg}^{-1}$ (swordfish), which confirms the tendency of this metal to accumulate in fish and seafood.

From the data above, given the percentages of moisture and the average mass of sample consumed in fresh form, the daily intake of Hg per person could be calculated (Table 4). Data found indicated ranges from 0.02 to 1.4 μg in the case of baby food, from 0.09 to 11.2 μg for fast food, and from 0.09 to 4.1 μg for adult canteen menus, except for the Daily Meal 8, which provided 134 μg . Table 4 also shows the percentage of the tolerable daily intake (TDI) provided by each menu analyzed.

The percentage contribution of samples to the TDI for baby food varied from 0.3 to 18.8% or 27.8% depending on the age (Table 4). This is considered not alarming, taking into account that it represents the

daily main meal and the percentages were lower than 30%. In the case of children's fast food, the percentage of TDI varied from 0.5 to 56.2% or 102.2%, depending on the body weight considered, in both cases, higher than 50%, posing a potential problem for children's health. Regarding the canteen menus, which were the main meal, the percentage of TDI, except for Daily Meal 8, varied from 0.3 to 12.1%, causing no human health problems. For Daily Meal 8, the percentage was 395.7% of TDI for adults and half of the Provisional Tolerable Weekly Intake (134 µg/person in front 240 µg/person/week) in a single meal, which confirms the recommendation from the European Food Safety Authority (EFSA) (Dietetic Products, Nutrition, and Allergies 2014) that the consumption of fish/seafood species with a high content of mercury in the daily diet should be limited to only a few of servings (< 1–2) per week.

A comparison of the concentrations of Hg found in baby food from different countries is shown in Table 5. The Hg concentration was studied in European countries and the levels varied between < 0.10 and 29.9 µg kg⁻¹. The lowest mean was obtained for the samples of baby food from Portugal (0.40 µg kg⁻¹) and the shortest range in the samples from the Czech Republic, Republic of Hungary, and Slovak Republic (0.3 to 10.2 µg kg⁻¹). A comparison of the results obtained in the present study for samples of baby food 11 and 13 with those found in the literature shows that they agree with the observations of Tóth et al. (2014). The latter reported the highest level of Hg in the sample with the addition of the tuna, with a value of 10.2 µg kg⁻¹. Martins et al. (2013) also reported the highest value of Hg in a baby food containing fish (19.6 µg kg⁻¹). A possible explanation for this is that mercury concentrations are found mainly in muscle tissues, the liver, and the kidneys of marine fish (Kasper et al. 2009).

Table 5

4. Conclusions

The studies mentioned here verify the general safety of baby foods commercialized in Spain regarding their Hg content and give evidence of the significant Hg contents in fish. Additionally, it seems that the contribution of the sample ingredients to the total Hg content in children's fast food was not significant, and the percentages of TDI were lower than 20%, except in a single case where it was between 56–102% for children's fast food. On the other hand, the presence of tuna fish in one of the adult canteen menus provided a high content of Hg and was four times the maximum tolerable daily intake. However, the percentages were up to 12% on the rest of the canteen menus. The study indicated that the presence of Hg in the foods studied did not represent toxic levels for the most part. Nevertheless, the TDI values referred to a single meal (baby food, fast food, or daily meal), which means dietary exposure to Hg may be higher. Therefore, future studies are needed to assess dietary exposure to Hg from the three main daily meals (breakfast, lunch, and dinner), considering the typical Spanish menus for these meals.

Declarations

Acknowledgments

The English text of this paper has been revised by Sidney Pratt, Canadian, MAT (The Johns Hopkins University), RSAdip - TESL (Cambridge University). Financial support for this was given by Núcleo de Química Analítica Avançada de Pernambuco (NUQAAPE/FACEPE) (grant number APQ-0346-1.06/14).

Author contribution

Maria José da Silva: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data Curation, Writing - Original Draft, Visualization; **Ana Paula S. Paim:** Data Curation, Writing - Review & Editing, Visualization; **Iago J. S. da Silva:** Data Curation, Writing - Review & Editing, Visualization; **Maria Fernanda Pimentel:** Data Curation, Writing - Review & Editing, Funding acquisition; **M. Luisa Cervera:** Conceptualization, Methodology, Validation, Formal analysis, Resources, Data Curation, Writing - Original Draft, Visualization, Supervision, Project administration, Funding acquisition; **Miguel de la Guardia:** Resources, Data Curation, Writing - Review & Editing, Supervision, Funding acquisition.

Funding

This study was funded by the Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) project Cooperation International CAPES/DGU 225/2010, and Ministerio de Ciencia, Innovación y Universidades - Agencia Estatal de Investigación - FEDER (EU) Project CTQ2016-78053-R.

Availability of data and material

Data will be made available on request.

Code availability

Not applicable.

Ethics approval

This article does not contain any studies with human participants or animals performed by any of the authors.

Informed consent

Not applicable.

Conflict of Interest

The authors have no competing interests to declare that are relevant to the content of this article.

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Tables

Table 1 Food composition of the baby food, fast food, and daily meal analyzed samples.

Food group	Composition ^a	Fresh mass (g)	Moisture (% w w ⁻¹)
Baby food 1	Sole with white sauce	250	87
Baby food 2	Lamb stew with vegetables	250	85
Baby food 3	Hake with rice	200	84
Baby food 4	Beef stew with vegetables	250	83
Baby food 5	Selected vegetables and monkfish	250	84
Baby food 6	Whiting with vegetables in cream	200	86
Baby food 7	Mixed vegetables	250	87
Baby food 8	Cream of green beans with potatoes	230	85
Baby food 9	Chicken with vegetables	250	84
Baby food 10	Carrots with rice in chicken broth	250	86
Baby food 11	Selected vegetables and sea bass	200	85
Baby food 12	Mashed peas and rice with hake	200	82
Baby food 13	Hake and white sauce	200	83
Fast Food	Extra ketchup, extra mustard, yogurt, and cola (soft drink)	498	68

1				
Fast Food 2	Extra ketchup, extra mustard, yogurt, and orangeade (soft drink)	580	71	
Fast Food 3	Extra ketchup, yogurt, and lemonade (soft drink)	521	69	
Fast Food 4	Extra ketchup, yogurt, and cola (soft drink)	529	69	
Fast Food 5	Yogurt and lemon tea (soft drink)	566	74	
Fast Food 6	Extra ketchup, milkshake, and cola (soft drink)	696	75	
Daily Meal 1	Seafood paella (rice, mussels, grouper, and squids rings); salmon with chips; and pear	653	57	
Daily Meal 2	Salad (lettuce, carrots, ham, soy, cheese, and mayonnaise); cod with carrots, parsley, and other vegetables; and orange	671	76	
Daily Meal 3	Rice with squid, cuttlefish, and prawns; pork loin with carrots, peas, and chips; and lemon yogurt	630	58	
Daily Meal 4	Salad (lettuce, tomato, corn, carrots, eggs, cucumber, pepper, soy, and olives); mixed spinach and mushrooms with steamed potatoes; and orange	843	79	
Daily Meal 5	Macaroon with tomato; ham, bacon, and sausage grilled with potatoes; and apple	896	68	
Daily Meal 6	Beans; meatballs with sauce and chips; and orange gelatin	796	64	
Daily Meal 7	Beans with ham; zucchini gratin with béchamel sauce, cheese, and bacon; strawberry and orange juice	672	78	
Daily Meal 8	Soup with bread, garlic, egg, and onion; grilled tuna with steamed potatoes; and lemon yogurt	794	73	
Daily Meal 9	Vegetable pie with tomato sauce (carrots, tomatoes, peppers, beets, and zucchini); tuna omelet with mashed potatoes; home-made crème caramel	525	68	

^aThe fast food menus were made up of a hamburger bun, a beef hamburger, cheddar cheese, gherkin, ketchup, mustard, a regular portion of French fries, yogurt or milkshake, and soft drink. Daily meals were composed of a starter, a main dish, a dessert, and a 50-70 g piece of bread.

Table 2 Evaluation of the method's accuracy using a comparison between values found and certified food reference values.

Sample	Found value ^a ($\mu\text{g kg}^{-1}$)	Certified value ($\mu\text{g kg}^{-1}$)	U Δ (95%) ($\mu\text{g kg}^{-1}$)
Chicken NCSZC73016	2.94 \pm 0.18	3.6 \pm 1.5	1.5
Fish Protein NRC DORM-3	331 \pm 6	382 \pm 60	60
Lobster Hepatopancreas NRC TORT-2	280 \pm 58	270 \pm 60	90

^aMean value \pm SD (n = 3).

Table 3 Effect of sample mass on direct Hg determination.

Sample mass (mg)	Hg concentration ^a ($\mu\text{g kg}^{-1}$)		
	Baby Food 1	Fast Food 2	Daily Meal 1
50	20.98 \pm 0.19	0.56 \pm 0.01	14.7 \pm 0.4
50 + 50	19.76 \pm 0.16	0.54 \pm 0.02	14.55 \pm 0.20
100	19.5 \pm 0.4	0.58 \pm 0.04	13.9 \pm 0.4
50 + 50 + 50	19.86 \pm 0.05	0.46 \pm 0.06	14.6 \pm 0.4
150	19.45 \pm 0.17	0.40 \pm 0.07	13.77 \pm 0.14

^aMean value \pm SD (n = 3).

Table 4 Mercury content in samples in dry weight, daily intake per person, and its contribution to the Tolerable Daily Intake (TDI).

Food group	Total Hg content ^a ($\mu\text{g kg}^{-1}$)	Daily intake per person ($\mu\text{g day}^{-1}$)	% TDI ^b	
			7-12 months	13-24 months
Baby Food 1 (fish)	20.98 \pm 0.19	0.69	13.8	9.4
Baby Food 2 (meat and vegetables)	21.8 \pm 1.1	0.81	16.2	11.0
Baby Food 3 (fish and vegetables)	15.89 \pm 0.14	0.50	10.0	6.8
Baby Food 4 (meat and vegetables)	2.51 \pm 0.20	0.10	2.1	1.4
Baby Food 5 (fish and vegetables)	19.22 \pm 0.16	0.77	15.4	10.4
Baby Food 6 (fish and vegetables)	11.5 \pm 0.3	0.33	6.6	4.5
Baby Food 7 (vegetables)	0.63 \pm 0.07	0.02	0.4	0.3
Baby Food 8 (vegetables)	0.57 \pm 0.01	0.02	0.4	0.3
Baby Food 9 (meat and vegetables)	2.85 \pm 0.03	0.11	2.2	1.5
Baby Food 10 (vegetables)	7.38 \pm 0.24	0.25	5.1	3.4
Baby Food 11 (fish and vegetables)	39.8 \pm 0.4	1.21	24.2	16.4
Baby Food 12 (fish and vegetables)	18.23 \pm 0.22	0.64	12.8	8.7
Baby Food 13 (fish)	41.9 \pm 0.8	1.39	27.8	18.8
Baby Food (mean)	15.5	0.53	10.6	7.1
			3-7 years	7-12 years
Fast Food 1	13.51 \pm 0.14	2.18	19.8	10.9
Fast Food 2	0.56 \pm 0.01	0.09	0.8	0.5
Fast Food 3	4.46 \pm 0.20	0.72	6.5	3.6
Fast Food 4	67.7 \pm 2.4	11.24	102.2	56.2
Fast Food 5	7.1 \pm 0.6	1.05	9.5	5.2
Fast Food 6	0.54 \pm 0.01	0.09	0.9	0.5
Fast Food (mean)	15.6	2.56	23.3	12.8

			Adults
Daily Meal 1	14.7 ± 0.7	4.11	12.1
Daily Meal 2	19.4 ± 0.7	3.11	9.2
Daily Meal 3	3.14 ± 0.12	0.84	2.5
Daily Meal 4	0.95 ± 0.09	0.17	0.5
Daily Meal 5	0.48 ± 0.01	0.14	0.4
Daily Meal 6	0.43 ± 0.02	0.12	0.4
Daily Meal 7	0.63 ± 0.04	0.09	0.3
Daily Meal 8	638 ± 63	134.5	395.7
Daily Meal 9	13.5 ± 0.2	2.31	6.8
Daily Meal (mean)	76.9	16.16	47.5

^aMean value ± SD (n = 3).

^bInfants: 7-12 months (9 kg body weight); Toddlers: 13-24 months (13 kg body weight); Children: 3-7 years (19 kg body weight); Children: 7-12 years (35 kg body weight); and Adults: 60 kg body weight.

Table 5 Comparison of Hg levels found in baby food from different countries.

Countries	Range of concentration Hg ($\mu\text{g kg}^{-1}$)	Mean ($\mu\text{g kg}^{-1}$)	Reference
China, Spain, UK, and USA	China (< 4 - 15), Spain (< 4 - 21), UK (< 4 - 10), and USA (< 4 - 6)	China (< 4), Spain (5), UK (5), and USA (< 4)	Carbonell-Barrachina et al. (2012)
Italy, Slovakia, Spain, and Sweden	2.7 - 29.9	9.2	Pandelova et al. (2012)
Portugal	< 0.10 - 19.6	0.40	Martins et al. (2013)
Czech Republic, Republic of Hungary, and Slovak Republic	0.3 - 10.2	1.82	Tóth et al. (2014)
Italy	-	1.69	de Roma et al. (2017)
Spain	0.57 - 41.9	15.5	This work

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