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Article

Determination of Total Mercury in Spanish Samples of Baby Food, Fast Food, and Daily Meal

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This study determined the levels of total mercury 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). The total mercury concentrations were determined in thirteen commercial baby foods for infants 6 to 12 months old, six types of fast foods prepared for children, and nine canteen menus prepared for adults. Samples were analyzed using a direct mercury analyzer, and the following concentration ranges were found: baby food (0.57-41.9 μg kg^{-1}), fast food (0.54-68 μg kg^{-1}), and adult menus (0.43-638 μg kg^{-1}). The recovery of different amounts of spiked mercury ranged from 98.6 to 104.9%, and the method's accuracy was checked with an analysis of different certified reference materials. The limits of detection and quantification obtained were 0.1 and 0.3 μg kg^{-1}, respectively, with a relative standard deviation of up to 11%. The contribution of the samples to the TDI varied as follows: baby food (0.3-28%), fast food (0.5-102%), and adult menus (0.3-396%). Therefore, it was concluded that total mercury daily intake does not pose risks for Spanish children and adults if tuna is not included on their menu.

Keywords: infant food, canteen menus, mercury, tolerable daily intake, atomic absorption spectrometry, direct mercury analysis

Introduction

In recent years, information about the concentrations of potentially toxic trace 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.[1-3] Studies[4-6] have shown that food consumption is the primary source of mercury exposure. Excessive intake of Hg may cause damage to the central nervous, cardiovascular, and reproductive systems and affect some physiological functions, such as the kidneys.[7,8]

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 μg kg^{-1} of body weight, that is, a tolerable daily intake (TDI) of 0.57 μg kg^{-1} of body weight, values as recommended by the Joint Food and Agriculture Organization of the United Nations/World Health Organization Expert Committee on Food Additives (FAO/WHO JECFA).[9] Studies[4,10] indicate that children

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are especially vulnerable and may be more exposed to contaminated food than adults. 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 μg of Hg *per* kilogram of body weight (bw) *per* day in the food they eat. However, children between 11 and 14 years presented a Hg dose (0.15 μg Hg *per* kg bw *per* day) about half of this, and adults (0.05 μg *per* kg bw *per* 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.[11]

Studies performed in Spain,[12-14] particularly in the Valencian Region, suggest that the population would be ingesting mercury levels exceeding the limit established by the European Food Safety Agency (EFSA). According to Yusà *et al*.,[12] the results of human biomonitoring projects and programs of food and environmental contaminants made during 2014-2020 show the high exposure to Hg of a relevant part of the child population and mothers, which constitutes a risk to their health. Also, Yusà *et al*.[13] revealed that the content of total mercury in hair among breastfeeding mothers is six times higher than the average internal exposure of mothers from other 17 European countries (0.225 μg g^{-1}), and 27% of mothers exceed the health-based guideline value proposed by EFSA (1.9 μg g^{-1}). Finally, the study conducted by Pérez *et al.*[14] showed that children living in the Valencian Region presented Hg levels in the hair around five times higher than the average in children of 17 other European countries (0.145 μg g^{-1}). About 13% of children had hair mercury levels above the FAO/WHO JECFA guideline of 2.3 μg g^{-1}, and 18% of children had levels above the EFSA health-based guidance.

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.*,[15] 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 (< limit of detection (LOD)), pulses (< LOD), cereals (25 μg kg^{-1}), tubercles (2.5 μg kg^{-1}), fruits (< LOD), fish and shellfish (95 μg kg^{-1}), meat (12 μg kg^{-1}), eggs (10 μg kg^{-1}), dairy products (11 μg kg^{-1}), milk (2.5 μg kg^{-1}), and fats and oils (25 μg kg^{-1}). Exposure to toxic elements such as Hg is hazardous for pregnant women and young children.[16,17]

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,[18,19] China,[20,21] France,[22] Germany,[23] Hong Kong,[24] Iran,[25-27] Italy,[5] Korea,[28] Poland,[29,30] Spain,[31-34] Sub-Saharan Africa,[35] Sweden,[36] and The United Kingdom,[37] 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,[38] such as infant cereals,[6,39] and infant formula.[38,40-42] However, in the literature, few research papers[5,43-47] deal with mercury content in baby food. On the other hand, few studies[5] on the levels of mercury present in fast food samples and canteen menus were found. Despite this, research has already been carried out on the elemental profile of similar foods, as studies published recently determined 12 elements in children's fast food[48] and 14 elements in commercial baby food.[49]

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 various matrices.[19,20,24,34,39,50] Other techniques have also been employed, which include electrothermal atomic absorption spectrometry (ET AAS),[51,52] and inductively coupled plasma-mass spectrometry (ICP-MS).[5,31,33,53] 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 trend makes it essential to develop safe and environmentally friendly methods for accurately determining mercury.

A potential alternative is to use a direct mercury analyzer suitable for directly determining Hg in solid samples to provide fast and accurate results.[54] 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 using 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[55] to analyze total mercury concentration in different food items and evaluate human exposure to THg via daily intake. The authors analyzed 58 food items (vegetables, fruits,

fish, meat, viscera, eggs, and rice) in this work. Other works[28,30,38,44,56-58] have also employed the direct mercury analyzer to determine Hg in food samples.

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 TDI, recommended by the FAO/WHO JECFA to assess the health risk.[9,59] 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.

Experimental

Instruments and reagents

To prepare standard solutions containing mercury, we used a Hg^{II} standard stock solution of 1000 μg mL^{-1} (Merck, Darmstadt, Germany) and ultrapure water with a resistivity of 18.2 MΩ cm (Milli-Q, Millipore, Bedford, United States of America (USA)).

The freeze-dried samples of baby food, fast food, and daily meal were analyzed with a Direct Mercury Analyzer (DMA-80, 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 Hg^0. The Hg amalgamator was heated to 850 °C, the Hg^0 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.

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 a piece of about 60 g of bread. Twenty-eight samples were analyzed, and their composition was described in 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).

Sample preparation

The pre-treatment of the samples followed the procedure proposed by Ruiz-de-Cenzano *et al.*,[48] and Mir-Marqués *et al.*,[49,60] briefly described below: the whole mass of the meal samples was 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. Afterward, they were homogenized with a domestic mixer (Braun, Kronberg, Germany) and stored in polyethylene tubes before analysis.

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

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

Food group	Composition[a]	Fresh mass / g	Moisture / (% m m^{-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 1	extra ketchup, extra mustard, yogurt, and cola (soft drink)	498	68
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	pasta 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

[a]The 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.

sample was weighed in a nickel crucible and introduced automatically into the DMA-80. The measurements were performed in three replicates for each sample. The instrument allows for three sequential pre-concentration procedures and has a limit of detection 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 Information section).

Assessment of 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 μg day^{-1}), toddlers from 13 to 24 months (7.4 μg day^{-1}), children from 3 to 7 years (11 μg day^{-1}), children from 7 to 12 years (20 μg day^{-1}), and adults (34 μg day^{-1}).[9] The daily intake (DI) of Hg in μg day^{-1} was calculated as shown in

equation 1, where DCM is the daily consumption of the menu in g day^{-1}, and MMC is the mean Hg concentration in μg kg^{-1} and fresh mass. 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 calculation of DI took into account the Hg content (Table 2), the moisture content, and the fresh mass of the sample (Table 1).

$$DI = DCM \times MMC \times 1000 \quad (1)$$

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 μg kg^{-1} by body weight (according to the age) and divided by 7 days.[59] Mean body weight was considered as follows: 9 kg for infants between 7-12 months; 13 kg for toddlers between 13-24 months;[6] 19 kg for children between 3-7 years; 35 kg for children between 7-12 years, and 60 kg for adults.[61]

Table 2. 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] / (μg kg^{-1})	Daily intake *per* person / (μg day^{-1})	TDI[b] / %	
			7-12 months	13-24 months
Baby food 1 (fish)	21.0 ± 0.2	0.69	14	9.4
Baby food 2 (meat and vegetables)	22 ± 1	0.81	16	11
Baby food 3 (fish and vegetables)	15.9 ± 0.1	0.50	10	6.8
Baby food 4 (meat and vegetables)	2.5 ± 0.2	0.10	2.1	1.4
Baby food 5 (fish and vegetables)	19.2 ± 0.2	0.77	15	10
Baby food 6 (fish and vegetables)	11.5 ± 0.3	0.33	6.6	4.5
Baby food 7 (vegetables)	0.63 ± 0.07	0.02	0.4	0.3
Baby food 8 (vegetables)	0.57 ± 0.01	0.02	0.4	0.3
Baby food 9 (meat and vegetables)	2.85 ± 0.03	0.11	2.2	1.5
Baby food 10 (vegetables)	7.4 ± 0.2	0.25	5.1	3.4
Baby food 11 (fish and vegetables)	39.8 ± 0.4	1.2	24	16
Baby food 12 (fish and vegetables)	18.2 ± 0.2	0.64	13	8.7
Baby food 13 (fish)	41.9 ± 0.8	1.4	28	19
Baby food (mean)	16	0.53	11	7.1
			3-7 years	7-12 years
Fast food 1	13.5 ± 0.1	2.2	20	11
Fast food 2	0.56 ± 0.01	0.09	0.8	0.5
Fast food 3	4.5 ± 0.2	0.72	6.5	3.6
Fast food 4	68 ± 2	11	102	56
Fast food 5	7.1 ± 0.6	1.0	9.5	5.2
Fast food 6	0.54 ± 0.01	0.09	0.9	0.5
Fast food (mean)	16	2.6	23	13
			Adults	
Daily meal 1	14.7 ± 0.7	4.1	12	
Daily meal 2	19.4 ± 0.7	3.1	9.2	
Daily meal 3	3.1 ± 0.1	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	396	
Daily meal 9	13.5 ± 0.2	2.3	6.8	
Daily meal (mean)	77	16	48	

[a]Mean value ± standard deviation (n = 3); [b]infants: 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. TDI / %: percentage contribution to the tolerable daily intake.

Statistical analysis

In the analysis of variance (ANOVA), all computations were carried out employing the Statistica[62] software, version 10. When a statistically significant difference was found between the means, the Tukey test was applied to group the homogeneous results. A significance level of 5% was used for all data analysis.

Results and Discussion

Instrument calibration

Calibration of the mercury analyzer was performed using standards in aqueous media. Two analytical curves with different mass ranges were used to determine low (0-20 ng) and high (20-1000 ng) content 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 regression equation obtained for the low-level mercury was: absorbance = $0.0629x - 0.0011x^2$, where x is the Hg mass, in ng; while the regression equation for the high-level mercury was: absorbance = $5.454 \times 10^{-4} + 9.180 \times 10^{-4}x - 2.448 \times 10^{-7}x^2$. The calibrations were checked every session employing certified reference material.

Analytical characteristics of the method

For the direct determination of Hg in a 50 mg dry sample mass, the analytical procedure provided an LOD of 0.1 μg kg^{-1} and a limit of quantification (LOQ) of 0.3 μg 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^{-1}) HNO_3, as described by da Silva *et al.*[56]

The relative standard deviation for triplicate samples containing from 0.43 to 638 μg kg^{-1} of Hg varied between 0.8 and 11%, generally lower than 5%. Recovery experiments on samples spiked at 2.5, 10, 25 and 500 μg kg^{-1} were 99 ± 1%, 99 ± 2%, 102 ± 3%, and 105 ± 2%, 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 seen in Table 3, the values obtained in our study generally agreed well with those reported for certified materials. At this stage of method validation, the uncertainty of the measurement was estimated only by the standard deviation (SD) of three replicates.

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 values (UΔ). The value of UΔ was calculated as shown in equation 2, where k is the coverage factor, usually equal to 2, corresponding to a confidence level of approximately 95%, u_m is the uncertainty of the measurement result, and u_{CRM} is the uncertainty of the certified value. The value of u_m, in turn, was calculated as the SD divided by the square root of the number of measurements (n), that is, $u_m = SD / \sqrt{n}$. Finally, u_{CRM} was calculated as the uncertainty of the CRM (u) presented in the certificate of analysis divided by the coverage factor, that is, $u_{CRM} = u / k$.

$$U\Delta = k \times \sqrt{u_m^2 + u_{CRM}^2} \quad (2)$$

If $\Delta m \leq U\Delta$, there is no significant difference between the measurement result and the certified value.[63] 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.[56] Agreement between found and certified values was verified for the eight CRMs used.

Effect of the sample mass

Three typical samples corresponding to each menu type under consideration were analyzed with sample

Table 3. Evaluation of the method's accuracy using a comparison between values found and certified food reference values

Sample	Found value[a] / (μg kg^{-1})	Certified value / (μg kg^{-1})	UΔ (95%) / (μg kg^{-1})
Chicken NCSZC73016	2.9 ± 0.2	3.6 ± 1.5	1.5
Fish protein NRC DORM-3	331 ± 6	382 ± 60	60
Lobster hepatopancreas NRC TORT-2	280 ± 60	270 ± 60	90

[a]Mean value ± standard deviation (n = 3). UΔ (95%): combined uncertainty of certified and measured values, corresponding to a confidence level of 95%.

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 4, the sample size had no acute effects on the concentrations obtained. However, when the Hg content in the sample was around 0.5 μg kg^{-1} (fast food 2), the use of sample mass less than 100 mg produced better results. A mass of 50 mg was selected for method simplicity, which was enough to obtain the appropriate data.

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 μg Hg *per* kg dry weight (d.w.) *per* sample. Table 2 shows that the baby food samples contained between 0.57 and 41.9 μg kg^{-1} d.w. of Hg. The lowest levels corresponded to vegetable purée (0.57-7.38 μg kg^{-1} d.w.), followed by foods containing meat (2.51-21.8 μg kg^{-1} d.w.). The highest concentration corresponded to fish and those with rice or vegetables (11.5-41.9 μg kg^{-1} d.w.).

Regarding fast food menus aimed at children, the values varied between 0.54 and 67.7 μg 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 μg kg^{-1} d.w. was found, except for daily meal 8, where there was a high level of 638 μg 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 μg kg^{-1} d.w.), specifically the presence of tuna (638 μg kg^{-1} d.w.), and rice with seafood products (3.14 μg kg^{-1} d.w.).

According to Cheng *et al*.,[55] the results for THg ranged from 0.16 to 171 μg 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 mercury level in various types of food, including fish and shellfish.

Martorell *et al*.[64] 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, tuna presented the highest concentration of Hg (222-776 μg kg^{-1}), only behind swordfish (869 μg kg^{-1}).

De Roma *et al*.,[5] 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 μg kg^{-1}), except for a restaurant meal sample (14.9 μg 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.

Finally, González *et al*.[32] 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 μg kg^{-1}) of the method for most of the analyzed foods, except for fish and seafood, which showed a mean concentration of 152 μg kg^{-1}. The detailed study of this foodstuff revealed Hg concentrations ranging from 3 μg kg^{-1} (panga) to 856 μg 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 2). Data found indicated ranges from 0.02 to 1.4 μg in the case of baby food, from 0.09 to 11 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 2 also shows the percentage of the TDI provided by each menu analyzed.

The percentage contribution of samples to the TDI for baby food varied from 0.3 to 19% or 28%, depending on

Table 4. Effect of sample mass on direct Hg determination

Sample mass / mg	Hg concentration[a] / (μg kg^{-1})		
	Baby food 1	Fast food 2	Daily meal 1
50	21.0 ± 0.2 (A)	0.56 ± 0.01 (A)	14.7 ± 0.4 (A)
50 + 50	19.8 ± 0.2 (B)	0.54 ± 0.02 (A)	14.6 ± 0.2 (AB)
100	19.5 ± 0.4 (B)	0.58 ± 0.04 (A)	13.9 ± 0.4 (AB)
50 + 50 + 50	19.86 ± 0.05 (B)	0.46 ± 0.06 (AB)	14.6 ± 0.4 (AB)
150	19.4 ± 0.2 (B)	0.40 ± 0.07 (B)	13.8 ± 0.1 (B)

[a]Mean value ± standard deviation (n = 3). Mean values with different letters in the same column differ significantly at a significance level of 5%.

the age (Table 2). This result 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 or 102%, 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%, causing no human health problems. For daily meal 8, the percentage was 396% of TDI for adults and about half of the provisional tolerable weekly intake (134 μg *per* person of 240 μg *per* person *per* week) in a single meal, which confirms the recommendation from the EFSA[65] that the consumption of fish/seafood species with a high content of mercury in the daily diet should be limited to only a few 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.*[46] 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.*[44] also reported the highest value of Hg in a sample of baby food containing fish (19.6 μg kg^{-1}). A possible explanation for this is that mercury concentrations are found mainly in marine fish muscle tissues, liver, and kidneys.[66] This article was preprinted[67] by the Research Square repository.

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 to 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.

Supplementary Information

Supplementary information on the operating conditions of the DMA-80 (Milestone) is available free of charge at http://jbcs.sbq.org.br as a PDF file.

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

Country	Range of concentration Hg / (μg kg^{-1})	Mean / (μg kg^{-1})	Reference
Italy	–	1.69	5
China, Spain, UK, and USA	China (< 4-15), Spain (< 4-21), the UK (< 4-10), and the USA (< 4-6)	China (< 4), Spain (5), the UK (5), and the USA (< 4)	43
Portugal	< 0.10-19.6	0.40	44
Italy, Slovakia, Spain, and Sweden	2.7-29.9	9.2	45
The Czech Republic, Republic of Hungary, and the Slovak Republic	0.3-10.2	1.82	46
Spain	0.57-41.9	15.5	this work

UK: United Kingdom; USA: United States of America.

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Author Contributions

Maria José da Silva was responsible for the conceptualization, methodology, validation, formal analysis, investigation, data curation, writing - original draft, visualization; Ana Paula S. Paim for the data curation, writing - review and editing, visualization; Iago J. S. da Silva for the data curation, writing - review and editing, visualization; Maria Fernanda Pimentel for the data curation, writing - review and editing, funding acquisition; Maria Luisa Cervera for the conceptualization, methodology, validation, formal analysis, resources, data curation, writing - original draft, visualization, supervision, project administration, funding acquisition; Miguel de la Guardia for the resources, data curation, writing - review and editing, supervision, funding acquisition.

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