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Dietary exposure to potentially toxic elements through sushi consumption in Catalonia, Spain

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ABSTRACT

Although sushi is considered as a healthy food, it can also be a route of exposure to chemical contaminants such as potentially toxic trace elements. In this study, we analysed the concentration of Cd, I, Ni, Pb and total Hg, as well as iAs and MeHg in sushi samples. Iodine levels were higher in samples containing seaweed, while iAs concentrations were greater in rice-containing sushi. In turn, total Hg and MeHg were significantly higher in sushi samples with tuna. Health risks of sushi consumption were assessed for three population groups: children, adolescents and adults. Considering an average intake of 8 sushi pieces for adults and adolescents, and 3 sushi pieces for children, the estimated exposure to MeHg by adolescents exceeded the tolerable daily intake set by EFSA, while MeHg intake by children and adults was below, but close to that threshold. A relatively high daily exposure of Ni and Pb was also found, especially for adolescents. Since this study focused only on the consumption of sushi, the contribution of other food groups to the overall dietary exposure should not be disregarded. It might lead to an exposure to MeHg and other trace elements above the health-based guideline values.

1. Introduction

Sushi consumption has rapidly increased in the last quarter of the 20th century and in the beginning of the 21st century (Hsin-I Feng, 2012). The number of sushi restaurants worldwide are in constant expansion, and this increase is also expected to continue in the future (Lehel et al., 2020). According to the Spanish Association of Manufacturers and Distributors (AECOC, 2019), 45% of the population reports consuming sushi at least once a month in Spain. The favourite establishments to purchase sushi are supermarkets (44%) and hypermarkets (38%), followed by take away and/or delivery (31%). Finally, 30% of the population also declares going to restaurants to eat sushi at least once a month (AECOC, 2019).

Fish consumption is highly recommended for its well-known nutritional values, being an important source of proteins, essential fatty acids, minerals and micronutrients (Altintzoglou et al., 2016; Sardenne et al., 2020). Moreover, fish consumption has been linked to a reduction

of cardiovascular diseases (Tørris et al., 2018), while also enhances foetal neurodevelopment (Starling et al., 2015) and prevents cognitive diseases, such as Alzheimer or dementia (Cederholm, 2017). However, some seafood species can also be an important source of chemical pollutants, including toxic metals/metalloids, polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs), polychlorinated naphthalenes (PCNs), hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), brominated flame retardants (BFRs), musks and benzothiazoles, contaminants that, in recent years, have been widely analysed in our laboratory (Aznar-Alemany et al., 2017; González et al., 2018, 2019; Martí-Cid et al., 2008; Perelló et al., 2012, 2015a, 2015b; Tralabón et al., 2015, 2017a, 2017b). In most cases, seafood has been the food group showing the greatest contribution to the intake of these chemicals (Domingo et al., 2012; González et al., 2018, 2019; Martí-Cid et al., 2008; Perelló et al., 2015a). Furthermore, the risk-benefit analysis of consuming seafood has been conducted through different approaches, most of them based on the contents of toxic trace elements and nutrients

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(Maulvault et al., 2013; Domingo, 2016; Vilavert et al., 2017; Cressey et al., 2020; Ricketts et al., 2020).

Although in recent years there has been a worldwide rapid expansion of sushi restaurants, there is scarce information regarding the levels of potentially toxic elements in sushi. Therefore, this study was aimed at determining the levels of various metals/metalloids in samples of sushi purchased in different establishments of Catalonia (Spain). The health risks associated to sushi consumption were also assessed for different population groups, according to age.

2. Materials and methods

2.1. Sampling

In February 2020, sushi samples were purchased in supermarkets, hypermarkets, take away food shops and restaurants of Catalonia. In order to consider all consumers' behaviour scenarios, additional samples of sushi were prepared at home with fresh seafood purchased from central markets and other products purchased from supermarkets. Sushi samples were combinations of mostly consumed sushi types (i.e., sashimi, maki and nigiri) and the most popular seafood (i.e., tuna, salmon and eel) (Burger et al., 2014). Three replicates were collected for each combination between the type of sushi and fish. The list of combinations are shown in Supplementary Information (SI; Table S1). In total, 102 samples were analysed. Samples were homogenized with a domestic shredder and kept at -20°C until analysis.

2.2. Chemical analysis

Procedures followed for the analysis of the samples are depicted in Fig. 1.

2.2.1. Cd, Ni and Pb

The analyses of Cd, Ni and Pb were performed by a previous microwave digestion treatment with nitric acid (65% Suprapur, E. Merck, Darmstadt, Germany). Briefly, 5 mL of nitric acid were added to 0.5 g of the sample in a hermetic Teflon vessel. Afterwards, samples were incubated for 8 h at room temperature and heated for 8 h at 80°C . Samples were filtered and filled up to 25 mL with distilled water. The concentrations of these elements were finally determined by inductive coupled plasma-mass spectrometry (ICP-MS) (Perkin-Elmer Elan 6000, Woodbridge, ON, Canada). The accuracy of the instrumental methods and analytical procedures was checked by duplication of the samples. Quality control was assured by analysing a certified reference material (*Trace elements in spinach leaves*; NIST-1570a). Recovery rates ranged between 84% and 101%. The limits of detection (LODs) were $0.1\ \mu\text{g/g}$ for Ni and Cd, and $0.05\ \mu\text{g/g}$ for Pb.

2.2.2. Total Hg and MeHg

Methylmercury was extracted from the samples (only those with total Hg levels above $0.100\ \text{mg/kg ww}$) as described by Scerbo and Barghigiani (1998), i.e. freeze-dried samples (approximately, 100 mg) were hydrolyzed in 10 mL of hydrobromic acid (47% wet weight (w/w), E. Merck), followed by MeHg extraction with 35 mL toluene (99.8% w/w, Merck); centrifuged for 10 min at 10,000 rpm and 10°C , and

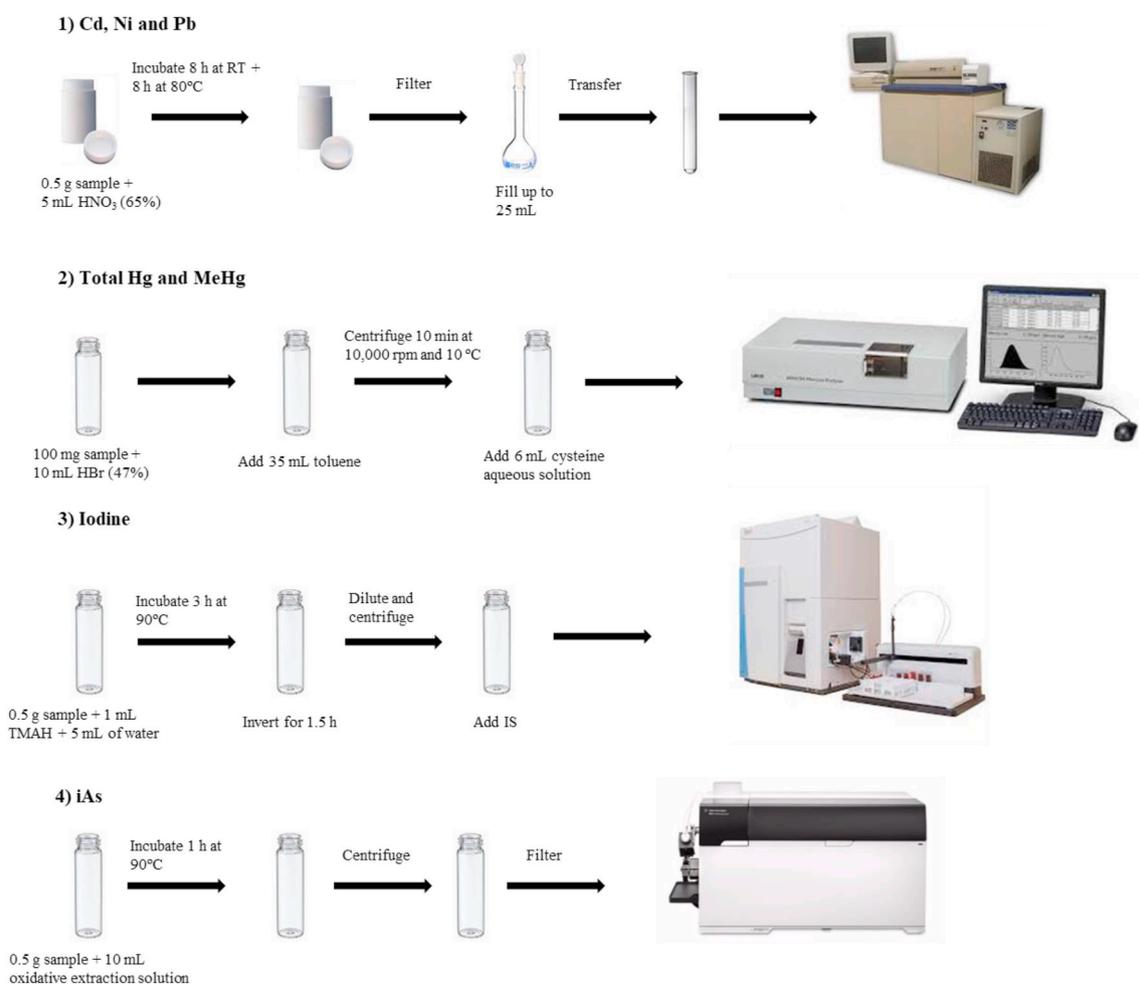


Fig. 1. Diagram of the sample preparation.

toluene removal with 6 mL cysteine aqueous solution (1% L-cysteinium chloride in 12.5% anhydrous sodium sulfate and 0.775% sodium acetate; E. Merck, Darmstadt). Total Hg and MeHg were then determined in samples (10–15 mg for solids or 100–300 μ L for liquids) by atomic absorption spectrometry (AAS), following the method 7473 of the US EPA (2007), using an automatic Hg analyzer (AMA 254, LECO, St. Joseph, Michigan, USA). Mercury concentrations were calculated from linear calibration (using at least five different standard concentrations), with a Hg (II) nitrate standard solution (1000 mg/L, E. Merck) dissolved in nitric acid (0.5 mol/L, E. Merck). The detection limit was 0.004 mg/kg ww, while the limit of quantification was 0.011 mg/kg ww. Accuracy was checked through the analysis of the certified reference material DORM-4 (fish protein certified reference material for trace metals, National Research Council Canada, Canada). The results obtained in the present study were within the certified range of values (T-Hg: certified value = 0.410 ± 0.055 mg/kg, value obtained in the present study = 0.390 ± 0.025 mg/kg; MeHg: certified value = 0.354 ± 0.031 mg/kg, value obtained in the present study = 0.353 ± 0.062 mg/kg). A minimum of three measurements (replicates) was performed per sample, being the results reported as mg/kg ww, according to sample moisture. Blanks were always tested in the same conditions as the sushi samples. Prior to utilization, all laboratory ware was cleaned with nitric acid (20% v/v) for 24 h and rinsed with ultrapure water to avoid contamination. All standards and reagents were of analytical (pro analysis) or superior grade.

2.2.3. Iodine and iAs analysis

Iodine was determined according to the European standard method EN 17050:2017 (CEN, 2017; Jerse et al., 2020). Briefly, approximately 0.5 g of sample material was used as test portion. Extraction was performed using 1 mL of tetramethylammonium hydroxide (TMAH) (25%) (E. Merck) and 5 mL of water (18.2 M Ω cm at 25 °C, EMD Millipore Corporation, Billerica, MA, USA) at 90 °C in a conventional oven for 3 h. Samples were taken out and inverted at approximately 1.5 h to ensure good contact between sample material and extractant solution. Following extraction, samples were diluted to a concentration of TMAH at 0.5% and centrifuged. If needed, further dilution was then performed prior to analysis of the samples. Internal standard tellurium (Plasma CAL, SCP Science, France) was added to a final concentration of 1 ppb to the diluted samples. Being the samples analysed using ICP-MS (iCAPq, ThermoFischer, Bremen, Germany). Reference materials NIST-3232 Kelp powder (NIST, Gaithersburg, Maryland, USA) and BCR-CD200 Bladderwrack (IRMM, Geel, Belgium) were analysed in every series in order to ensure the quality of the analysis.

Inorganic arsenic was determined according to the European standard method EN16802:2016 (CEN, 2016). Approximately 0.5 g of material was used as test portion. Extraction was performed using 10 mL of oxidative extraction solution (1% nitric acid, 3% hydrogen peroxide in water) (E. Merck, Darmstadt, Germany) at 90 °C in a water bath for 1 h. Samples were inverted every 20 min to ensure complete mixing. Following extraction, samples were centrifuged, and if necessary, further diluted prior to analysis. Samples were filtered and analysed using ion chromatography coupled with ICP-MS (Agilent 8900 ICP-QQQ, Agilent Technologies, Waldbronn, Germany). Calculations were performed using MassHunter software. Reference material NMIJ 7405-b Hijiki Seaweed (NMIJ, Japan) was analysed in every series to ensure the quality of the analysis.

For the chromatography, an Agilent 1200 HPLC system (Agilent Technologies) with a column heater was used. For separation, an IonPac AS-7 (2 \times 250 mm) and a guard column AG-7 (2 \times 50 mm) was used (Thermo Fisher). The eluent was isocratic 50 mM ammonium carbonate in water with 3% methanol. A flow rate of 0.15 mL/min was used.

2.3. Dietary exposure

Each individual piece of sushi was weighted to calculate the dietary

exposure to trace elements. The percentage of seaweed, rice and fish of each type of sushi are summarized in Table 1. These percentages were in accordance to those previously reported by Alves et al. (2017) in Japanese restaurants of Brazil, where the mean contributions of rice, fish and seaweed in sushi were 65%, 30% and 5%, respectively.

The dietary intakes of each trace element, as well as those of iAs and MeHg, were calculated by multiplying the concentration of each element in a sushi piece with the average consumption by each population group. Adults and adolescents were considered to eat 8 pieces of sushi per meal, while we assumed that children would consume 3 sushi pieces (Burger et al., 2014). The intake of trace elements was estimated for different population groups, considering the following mean body weight: 24 kg for children aged 3–9 years, 51 kg for adolescents aged 10–17 years and 70 kg for adults (18–74 years) (Carrascosa et al., 2010; López-Sobaler et al., 2016).

The European Food Safety Authority (EFSA) has derived a number of health-based guidance values (HBGVs) to limit the exposure to food contaminants and minimize adverse health outcomes. Some examples are the tolerable daily intake (TDI), the lowest-observed-adverse-effect-level (LOAEL), the benchmark dose level (BMDL), the tolerable weekly intake (TWI), and the adequate intake (AI). In the current study, updated information on these HBGVs has been used: 4.3 μ g/kg body weight (bw)/day for acute exposure to Ni (LOAEL), 13 μ g/kg bw/day for chronic exposure to Ni (TDI), 0.50 μ g/kg bw/day for Pb (BMDL₀₁), a range of 0.3–8 μ g/kg bw/day for iAs (BMDL₀₁), 2.5 μ g/kg bw/week for Cd (TWI), 1.3 μ g/kg bw/week for MeHg (TWI), and finally, 150 μ g/day for adults, with a range of 70–130 μ g/day for children, for iodine (AI) (EFSA, 2009a; b, 2010, 2012, 2014, 2020).

For calculations, when the concentration of an element was under its limit of detection (LOD), that value was assumed to be one-half of that limit (ND = 1/2LOD). Exceptionally, in order to avoid an overestimation of the exposure to MeHg, its intake was considered as zero for foods containing levels below its LOD (González et al., 2019).

To elucidate which combinations of sushi pieces are safe for the population, we computed all the possible combinations of up to 8 pieces for adults and up to 3 pieces for children, being computed the total amount of trace elements.

2.4. Statistical analysis

Statistical analysis was performed using the software Statistical Package for the Social Sciences (SPSS v.27) and combinatorial computations were carried out with R statistical package version 4.0. Values with a Z-score above 2.5 and under -2.5 were considered as outliers. A visual inspection of the boxplot was also conducted to verify these outliers. A Kolmogorov-Smirnov test was performed to determine the homogeneity of the groups. If variances were homogenous, ANOVA, followed by the Bonferroni's test was used. When variances were not homogeneous, ANOVA, followed by a T3 Dunnett's test, was used. Statistical significance was set at 0.05 ($p < 0.05$).

Table 1
Percentages (wet weight) of seaweed, rice and fish for each type of sushi.

Sushi	Mean weight \pm SD (g)	% seaweed	% rice	% fish
SS	13 \pm 6	0	0	100
SM	16 \pm 8	8	68	24
SN	35 \pm 7	0	73	27
TS	15 \pm 7	0	0	100
TM	16 \pm 6	5	73	22
TN	35 \pm 6	0	75	25
EN	34 \pm 4	1	75	24
EM	22 \pm 8	5	72	23

SS: Salmon sashimi; SM: Salmon maki; SN: Salmon nigiri; TS: Tuna sashimi; TM: Tuna maki; TN: Tuna nigiri; EN: Eel nigiri; EM: Eel maki; SD: Standard deviation.

3. Results and discussion

3.1. Chemical occurrence

The mean concentrations of iodine, iAs, Ni, Pb, total Hg and MeHg in sushi samples are depicted in Fig. 2. In turn, individual concentrations are given as Supplementary Information (Table S2). Eel maki, salmon maki and tuna nigiri showed remarkably high average levels of iodine (0.466, 0.434 and 0.393 $\mu\text{g/g}$, respectively), while other kinds of sushi samples presented levels well below 0.200 $\mu\text{g/g}$ (Fig. 2a). The high contents of iodine in maki samples are likely related to the presence of seaweed, which may have large levels of iodine (Filippini et al., 2021; Smyth, 2021).

Most sushi sample, excepting sashimi, showed similar mean concentrations of iAs, with values ranging between 0.022 and 0.025 $\mu\text{g/g}$ (Fig. 2b). On the other hand, salmon sashimi and tuna sashimi presented significant lower concentrations of iAs when compared to nigiri and maki, which should be due to the presence of rice. Despite both, nigiri and maki, contain rice, iAs levels in sushi were lower than those recently found in rice and rice-based products (González et al., 2020). It would be associated to the fact that the rice used for sushi preparation is rinsed with water several times and then boiled, thus decreasing the content of iAs up to 60% (Althobiti et al., 2018; Gray et al., 2015; Jitaru et al., 2016).

Nigiri was the type of sushi with the highest mean levels of Ni, with a range between 1.10 and 1.74 $\mu\text{g/g}$ (Fig. 2c). Regarding Pb, tuna sashimi

and eel nigiri were the sushi items showing the highest average concentrations (0.120 and 0.091 $\mu\text{g/g}$, respectively) (Fig. 2d). However, no significant differences were observed in the levels of Ni and Pb between the different types of sushi.

Sushi samples containing tuna had significantly higher levels of total Hg and MeHg than those samples based on salmon or eel, with tuna sashimi showing the highest average concentrations (0.680 and 0.427 $\mu\text{g/g}$ for total Hg and MeHg, respectively) (Fig. 2e and f). Tuna has been identified as one of the fish species containing more MeHg, mainly because it bioaccumulates in larger specimens, while salmon and eel usually present lower levels of MeHg (de Paiva et al., 2017; González et al., 2019; Perelló et al., 2015b; Sadhu et al., 2015; Barone et al., 2021). Finally, no traces of Cd were detected in any sushi sample.

When assessing Japanese restaurants in Brazil, Alves et al. (2017) reported very similar concentrations of total Hg in tuna and salmon sashimi than those observed in the current study: 0.589 vs 0.680 $\mu\text{g/g}$ for tuna sashimi, and 0.0113 vs 0.018 $\mu\text{g/g}$ for salmon sashimi. Moreover, similar values were also reported for tuna and salmon sashimi in the USA (Burger et al., 2014). The content of total Hg in tuna sashimi was 0.675 $\mu\text{g/g}$, which is close to the value here found (0.680 $\mu\text{g/g}$), while the total Hg concentration in salmon sashimi was 0.021 $\mu\text{g/g}$, also very similar to the result of the present survey (0.018 $\mu\text{g/g}$). However, the mean concentrations of Hg in tuna maki and salmon maki were 0.460 and 0.040 $\mu\text{g/g}$, respectively. These levels are higher than those found in the current survey (0.109 and 0.00435 $\mu\text{g/g}$, respectively), which is very probably due to the different origin of the fish. In turn, the

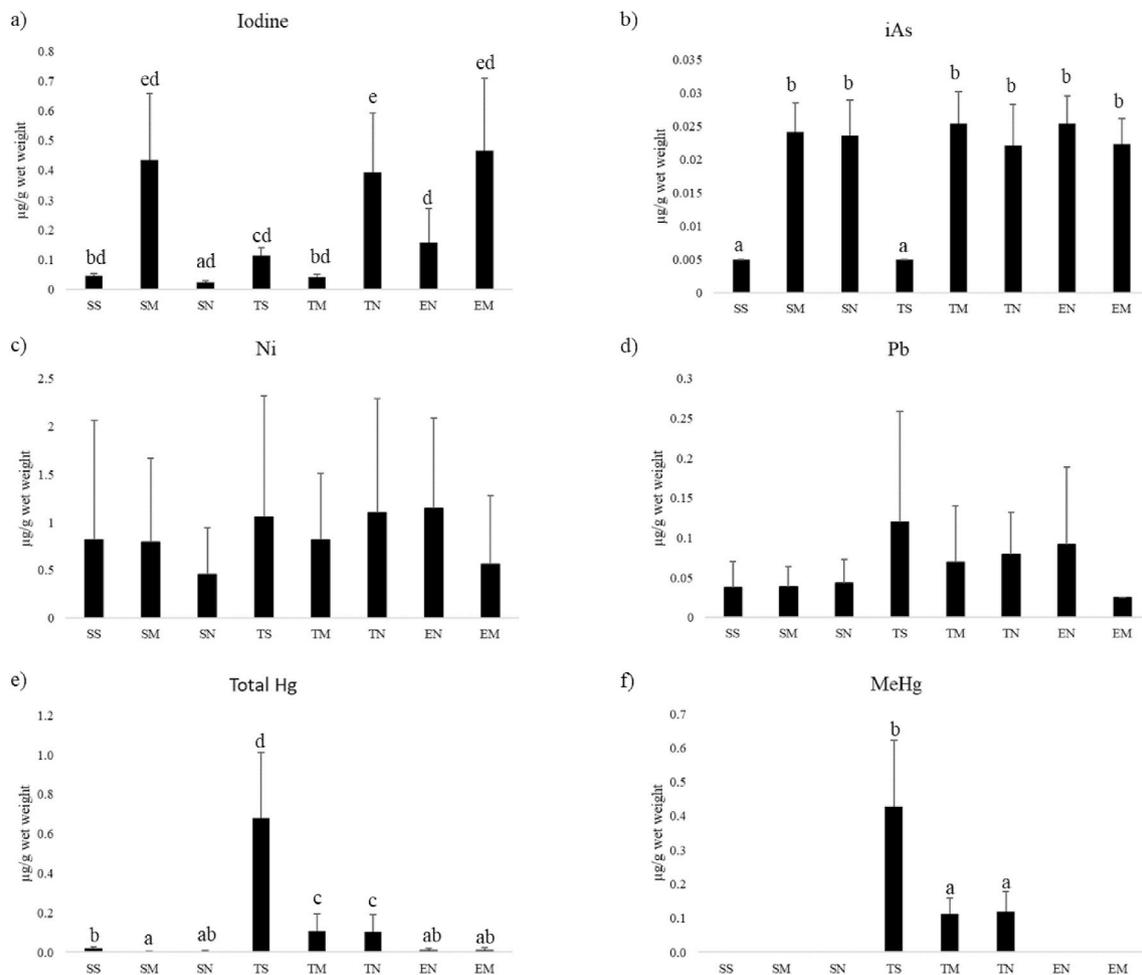


Fig. 2. Mean concentrations of iodine (a), iAs (b), Ni (c), Pb (d), Total Hg (e) and MeHg (f) in sushi samples. Data are given as means \pm standard deviations. Different letters mean significant differences among sushi types ($p < 0.05$). SS: Salmon sashimi; SM: Salmon maki; SN: Salmon nigiri; TS: Tuna sashimi; TM: Tuna maki; TN: Tuna nigiri; EN: Eel nigiri; EM: Eel maki.

average level of Hg in eel was 0.063 µg/g, a value that is also higher when compared to those of eel maki (0.0126 µg/g) and eel nigiri (0.0144 µg/g) (Burger et al., 2014). Morgano et al. (2015) reported similar results to our study for MeHg in 30 tuna sushi samples from Japanese restaurants and supermarkets in Campinas, São Paulo State, Brazil. The mean concentrations of MeHg and total Hg were 0.195 and 0.279 µg/g, respectively. In a previous study, Lowenstein et al. (2010) had tested the Hg content of 100 tuna sushi samples from 54 restaurants and 15 supermarkets of New York, New Jersey and Colorado (USA), which were collected between October 2007 and December 2009. The mean Hg concentrations of all samples exceeded the maximum concentration permitted in Japan, with bigeye tuna as the species with the highest Hg levels (0.65 and 2.254 µg/g). Finally, and in contrast to the current results, Kulawik et al. (2018) reported detectable Cd levels in sushi samples (range: 0.023–0.133 µg/g), which were dependent on the amount of nori seaweed used for preparing the sushi.

In 2006, the European Commission (EC) set maximum levels for certain potentially toxic trace elements -among other chemical contaminants-in several foodstuffs (European Commission, 2006). For Pb, the maximum level was set at 0.30 µg/g for any kind of fish. In contrast, different thresholds were established for Cd according to the species, with maximum values at 0.10 µg/g for tuna, and 0.050 µg/g for eel and salmon. With respect to Hg, maximum levels in tuna and eel were set at 1.0 µg/g, with 0.50 µg/g for salmon. Finally, the highest allowed concentration of iAs in white rice was established at 0.20 µg/g. In the present survey, one individual sample of tuna sashimi (TS5) was above the maximum concentration for Pb (0.469 µg/g), whereas another sample of tuna sashimi (TS10) was above the limit for Hg (1.02 µg/g). However, none of the samples was above the maximum threshold for Cd and iAs. It must be highlighted that sushi is a complex mixture of rice, fish and seaweed, which is also prepared in a specific way. Therefore, the comparison with guidance values must be carried out with caution.

3.2. Dietary intake

Considering a standard portion of 8 pieces -consisting in one piece of each type of sushi for adults and adolescents, and a portion of 3 pieces for children-the estimated dietary exposure of metals/metalloids for children and adolescents is summarized in Table 2. In addition, the percentage of contribution to the threshold limits for the adult population is depicted in Fig. 3.

Among the three analysed age groups of population, and in agreement with previous findings (González et al., 2021), teenagers was the group showing the highest dietary intake of trace elements. The estimated exposure to MeHg in the adolescent population was well above

Table 2
Dietary exposure through sushi consumption to Ni, Pb, MeHg, iAs, Cd and iodine for adolescents and children in Catalonia, Spain.

Trace element	Exposure (µg/kg bw/day)		EFSA values (µg/kg bw/day)	Reference
	Adolescents	Children		
I	0.782	0.554	2.94 (adolescents) ^a 2.92 (children) ^a	EFSA (2014)
iAs	0.0765	0.0542	0.3	EFSA, 2009
Ni	3.99	2.83	4.3 (acute exposure) 13 (chronic exposure)	EFSA (2020)
Pb	0.282	0.200	0.5	EFSA (2010)
MeHg	0.242	0.172	0.186 ^b	EFSA (2012)

^a These daily values were calculated from the acceptable intake (AI) of 150 µg/day for adults and adolescents and 70 µg/day for children. The AI was divided between the respective body weights.

^b This daily intake value was calculated from the TWI of 1.3 µg/kg bw/week set by the EFSA.

the threshold for MeHg (0.242 vs. 0.186 µg/kg bw/day), while the MeHg intake by adults and children was below -although close-to that limit (0.176 and 0.172 µg/kg bw/day, respectively). With respect to Ni, exposure was estimated in 2.83 (children), 2.91 (adults) and 3.99 µg/kg bw/day (adolescents), which means 66%, 68% and 93% of the LOAEL set for acute exposure to this element. Adolescents also showed the highest exposure to Pb (0.282 µg/kg bw/day), followed by adults and children (0.206, and 0.200 µg/kg bw/day, respectively). In relation to iAs and iodine, sushi consumption contributed less than 30% of the HBGVs.

The scarce literature regarding risk assessment related to sushi consumption is mainly focused on MeHg, due to the health issues associated to this metal species. The exposure to MeHg for the adult population here estimated (0.176 µg/kg bw/day) is almost one-half of that reported by Burger et al. (2014), who estimated a value of 0.34 µg/kg bw/day considering a daily consumption of 38.6 g of sushi. On the other hand, Alves et al. (2017) estimated exposure to MeHg considering 1 to 7 portions/week of sushi and sashimi. The estimated weekly intake of MeHg through the consumption of tuna sushi and tuna sashimi ranged between 0.333 and 9.278 µg/kg bw/week (equivalent to 0.048–1.33 µg/kg bw/day). For an adult population, considering a consumption of 4 portions/week of tuna sushi, the estimated MeHg exposure is already higher than that found in the present study (0.190 µg/kg bw/day). For children, the range was between 0.444 and 12.371 µg/kg bw/week, corresponding to 0.063 and 1.77 µg/kg bw/day. In this case, assuming a consumption of 3 portions/week of tuna sushi, the estimated exposure was 0.190 µg/kg bw/day, which is higher than the current value. Regarding tuna sashimi, only with 1 portion/week of 150 g, the estimated exposure for adults and children is already higher than that calculated in the present study (0.189 and 0.252 µg/kg bw/day, respectively) (Alves et al., 2017). The same researchers (de Paiva et al., 2017) determined the contents of MeHg and total Hg in twelve fish species used in the preparation of sashimi in Japanese restaurants of Brazil (de Paiva et al., 2017). They also estimated the exposure to MeHg, highlighting that the ingestion of only two portions of sushi made of needlefish and tuna, already exceeded the established HBGVs by 100%. In contrast, the intake of MeHg was much lower for mullet and salmon sashimi. It was concluded that frequent consumers (>once a week), and especially those belonging to sensitive population groups (e.g., pregnant women, lactating mothers and children) should diversify their diet, in terms of consumption of marine species.

A specific statistical treatment of the results was conducted to elucidate which combinations of sushi pieces, limited to a maximum of 8 pieces for adults and adolescents, and 3 for children, were safe; that is to say, how many combinations did not exceed the HBGVs. The statistical analysis was conducted for Pb, Ni and MeHg as target elements, since the contribution of their exposures to the HBGVs was more remarkable than for other parameters (Table 2; Fig. 3). Out of the 12,869 possible combinations, 62% were found to be safe for adults, while for adolescents only 35% was associated to an exposure lower than the threshold. For both adults and adolescents, the combination corresponding to the consumption of a standard meal consisting of eight different sushi items did not exceed the safety limit for Pb, Ni and MeHg. Regarding children, 164 combinations of three or less pieces were made, being 51% safe according to the current standards (EFSA, 2010, 2012, 2020). Safe combinations were mostly based on salmon sushi (either sashimi, maki or nigiri) and/or eel maki, which were associated to a low exposure to Pb, Ni and MeHg. In contrast, combinations with a higher contribution of tuna-containing sushi were not safe, taking into account that this sushi contains high concentrations of MeHg.

It is important to note that the present study was targeted to a specific food item, with a relatively low consumption among the Catalan population. The contribution of other foodstuffs to the total intake of the metals/metalloids here analysed might mean an overexposure to these elements for the population, especially for the most sensitive groups like children. Nickel is a metal of special concern due to its widespread

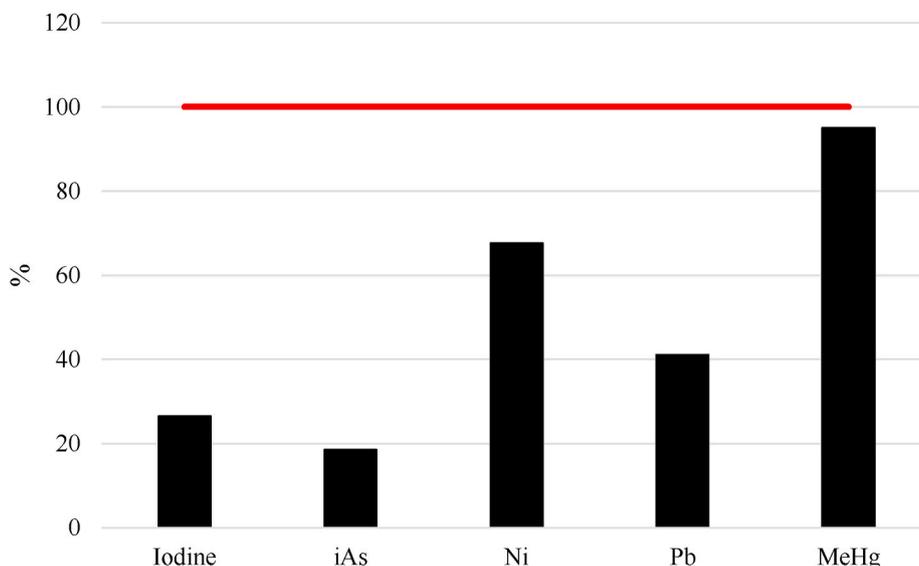


Fig. 3. Contribution percentage of the exposure to a number of trace elements through sushi consumption with respect to the corresponding HBGVs (AI for iodine, BMDL₁₀ for iAs and Pb, LOAEL for Ni, and TWI for MeHg) by the adult population.

occurrence in diverse food products. Therefore, the consumption of a single meal based on sushi, in addition to the intake of other meals during the day, might be associated to an exceedance of threshold Ni values not only for children, but also for the other population groups, including adults (Fig. 3). Other food products that contain significant levels of Ni include vegetables, cocoa-products, legumes and grains (Babaahmadifooladi and Jacxsens, 2020; Cubadda et al., 2020; EFSA, 2015). In a very recent total diet study that we performed in the same area of study, Ni was pointed out as a target chemical by food safety authorities, because of the increasing dietary intake by the population, being suggested its inclusion in future Total Diet Studies (González et al., 2021).

On the other hand, rice and fish are not foodstuffs with the largest contribution to the intake of Pb. Exposure to Pb mostly comes through consumption of bread, meat and meat products (EFSA, 2010; González et al., 2019). Therefore, special attention must be paid to other meals and foodstuffs in order not overcoming the HBGVs. Because of their widespread presence in food other than fish and rice, this recommendation is extendable to other elements in addition to Ni and Pb.

4. Conclusions

Iodine was significantly higher in sushi samples containing seaweed, especially maki sushi, while iAs concentrations were notably higher in sushi with a higher content of rice, namely nigiri and maki. As expected, concentrations of total Hg and MeHg were more abundant in sushi samples made of tuna, with sashimi showing the highest values. On the other hand, Ni and Pb levels were similar among all types of sushi, with no specific trends according to the sushi composition. Furthermore, Cd was not detected in any sushi sample. The estimated exposure to MeHg was identified as of great concern, especially for the adolescent population. The daily MeHg intake by this group exceeded the current HBGV established by the EFSA. In turn, exposure to Ni and Pb through sushi consumption for all population groups was below the HBGVs. Contributions were around 65% and 40% of the threshold value for Ni and Pb, respectively, with the exception of exposure to Ni by adolescents, which resulted in a 93% of the total HBGV. However, the risk to overcome the HBGVs would be reduced if the population -logically- diversify the types of sushi and the type of seafood that they consume, especially focusing on salmon or eel, with lower levels of Pb, Ni and MeHg. The remaining elements here assessed were not considered as a potential risk for human health, given the low exposure through sushi consumption. We would

like to conclude highlighting that although the present study was only focused on a single food item, sushi, other foodstuffs can contribute to an overexposure to the same potentially toxic elements.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.fct.2021.112285>.

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