



## Dietary exposure to metals/metalloids and persistent organic pollutants in Spanish preschool and primary school children

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### ABSTRACT

Diet is a primary source of pollutant exposure. Given children's vulnerability to their effects, this study assessed dietary intake of metals/metalloids and different persistent organic pollutants in children in Tarragona (Spain), compared it with the health-based guideline values (HBGV), and identified the main dietary sources. The analysis included 533 preschoolers and 443 primary school children from the EPINED and ECLIPSES studies. Dietary intake of cadmium (Cd), methylmercury (MeHg), lead (Pb), inorganic arsenic (InAs), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), dioxin-like polychlorinated biphenyls (DL-PCBs), and non-dioxin-like polychlorinated biphenyls (NDL-PCBs) was estimated using validated food frequency questionnaires and a database from the Catalan Agency for Food Safety. Preschoolers and primary school children exceeded the relevant reference value for InAs and DL-PCBs. Only preschoolers exceeded the limit for MeHg and PCDD/Fs. Cd, Pb, and NDL-PCBs intakes did not exceed the limits in either age group. Salted cereals/potatoes were the main source of InAs; fish for MeHg and DL-PCBs; and milk/yogurt for PCDD/Fs, followed by white fish in preschoolers and fatty fish in primary school children. The findings highlight that dietary exposure to some pollutants poses a significant risk to these children, emphasizing the need for public health policies to reduce pollutants in their diets.

### 1. Introduction

Diet is one of the main sources through which humans are exposed to different pollutants in Europe (Papadopoulou et al., 2019). Contamination of food products can occur at any stage of the food chain by direct exposure to the environment, during the food production, processing, and storage, and through contact with food packaging (Rather et al., 2017). Human exposure to harmful pollutants has been a matter of concern in past decades. Among those pollutants of greatest concern, because they pose potential serious adverse effects, are

metals/metalloids (i.e., arsenic [As], cadmium [Cd], lead, [Pb], and methylmercury [MeHg]) and persistent organic pollutants (POPs) (Fu and Xi, 2020; Rehman et al., 2018; Ruzzin, 2012). Metals and metalloids naturally occur in soil and water, but they are also introduced by human derived activities such as the use of metal containing pesticides and fertilizers, industrial production and uses, and mining (Ahmed et al., 2022; Mishra et al., 2023). As consequence, they can easily enter the food chain. On its hand, POPs such as polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), dioxins like polychlorinated biphenyls (DL-PCBs) and non-dioxin like polychlorinated biphenyls (NDL-PCBs) are mostly of anthropogenic origin (derived from waste incineration,

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**Abbreviations:**

BMD	benchmark dose
Bw	body weight
CD	cadmium
DL-PCBs	dioxins like polychlorinated biphenyl
EPINED	The Neurodevelopmental Disorders Epidemiological Research Project
FFQ	food frequency questionnaire;
IARC	International Agency for Research on Cancer
InAs	inorganic arsenic
MeHg	methylmercury
MOE	margin of exposure
NDL-PCBs	non-dioxin like polychlorinated biphenyls
Pb	lead
PCDD/Fs	polychlorinated dibenzo-p-dioxins and dibenzofurans
POPs	persistent organic pollutants
SDQI	Spanish Quality Diet Index
TDI	tolerable daily intake
TDS	total diet study
TWI	tolerable weekly intake
WHO	World Health Organization

industrial processes and agriculture) (Guo et al., 2019). POPs are among the most dangerous compounds ever synthesized and can bioaccumulate and biomagnify, by getting into the food chain (World Health Organization, 2010).

Bearing in mind the above mentioned, from a public health perspective it is important to assess dietary exposure to metals, metalloids and POPs and compare it to health-based guideline values, both in the whole population and in vulnerable subgroups. This is essential to make a judgment about potential risks and to assess the adequacy of current strategies to reduce contaminant levels in food. In this sense, children are among the most vulnerable to the effect of pollutants due to different reasons. Children consume more food in relation to their body weight than adults; their organ systems are still under development and maturation; and they may be unaware of the risks and incapable of making decisions to protect their own health. In addition, due to their early stage of life they have plenty of future years to develop a chronic disease (World Health Organization, 2006, 2017).

Children dietary exposure to metals, metalloids and POPs might vary between geographical areas since it depends not only on the level of food contamination (derived from the amount of natural or anthropogenic contamination of the environment) but also the dietary pattern followed, which is influenced by culture, socioeconomic status and food environment among other factors. Therefore, it is important to assess children's dietary exposure to these pollutants in different geographical areas. Tarragona is a province of Catalonia (Spain) located in the Mediterranean Sea area where one of the largest chemical/petrochemical complex of Southern Europe is located. To our knowledge, only two previous studies have assessed dietary exposure to metals, metalloids and PCDD/Fs and their major dietary contributors in this geographic area, focusing on the entire population (Gonzalez et al., 2018, 2021). Considering the high vulnerability of children to pollutants, the exposure to metals, metalloids and POPs in this subpopulation is of primary importance. Therefore, the objectives of the present study were: (1) to estimate the dietary intake of metals (Cd, MeHg and Pb), metalloids (InAs) and different POPs (PCDD/Fs, DL-PCBs, and NDL-PCBs) in children of two different age groups (preschool and primary school) in Tarragona (Spain); (2) to compare the intake of these pollutants with their corresponding updated health-based guideline values; (3) to identify the foods that contribute most to the total intake of these different pollutants in children.

**2. Material and methods****2.1. Study population**

The present descriptive study included participants from two research projects conducted in the province of Tarragona, Catalonia, Spain: The Neurodevelopmental Disorders Epidemiological Research Project (EPINED) and the ECLIPSES study. Briefly, the EPINED was a two-phase study conducted between 2014 and 2019 to estimate the prevalence of neurodevelopmental disorders (attention-deficit/hyperactivity disorder, autism spectrum) in a representative sample of two scholar age groups (preschoolers and primary school children) in the province of Tarragona. The ECLIPSES study was a randomized controlled trial performed between 2013 and 2017, which aimed to determine the best grade of efficacy of iron supplementation tailored to hemoglobin levels in early pregnancy, which would be optimal for mother-newborn health. More detailed information of EPINED and ECLIPSES studies and the inclusion and exclusion criteria can be found elsewhere (Arija et al., 2014; Canals Sans et al., 2021; Morales Hidalgo et al., 2021).

Among the 781 children participating in the second phase of the EPINED study and the 287 children participating in the ECLIPSES study (assessed at 4 years of age), we excluded 67 children who did not complete the food frequency questionnaire (FFQ) or had implausible data and 25 with missing data on body weight. Hence, 976 participants were finally included in the analyses. All the children included had written informed consent of their parents.

**2.2. Dietary assessment**

Children's dietary intake was assessed using two different FFQs, validated in the Spanish population, completed by their parents with the supervision of the research nutritionists: one FFQ of 41-items for preschool children (Esteban-Figuerola et al., 2020) and another FFQ of 45-items for primary school children (Rodriguez et al., 2008). The FFQs yielded data on the usual frequency of consumption in times per week or month, which were then converted into grams per day considering the reference serving size described for each item and the age group in our population (Agència de Salut Pública de Catalunya, 2005), and in agreement with experts in this field of our research group (Aparicio et al., 2015; Arija et al., 1996a, 1996b; Jardi et al., 2019; Jardi et al., 2019).

The quality of the diet of children was also estimated by means of the Spanish Quality Diet Index (SQDI), which is based on the food recommendations of Spanish Society of Community Nutrition (Norte Navarro and Ortiz Moncada, 2011). This index is composed of 10 different items (nine food groups and one item referring to dietary variety). Participants received a score between 0 and 10 for each item according to their adherence to the recommendations. The final SDQI score is the sum of the scores for each item and ranges from 0 to 100, with higher scores indicating higher quality of diet.

**2.3. Assessment of dietary intake of metals, metalloids and POPs**

We used the database of pollutants in food of the Catalan Agency for Food Safety (see supplementary material) to estimate the dietary intake of metals and metalloids (InAs, Cd, Pb, and MeHg) and POPs (PCDD/Fs, DL-PCBs, and NDL-PCBs) (Bosch-Collet et al., 2020). Firstly, we grouped the different items of the FFQs into 15 food groups, which were made according to the similarities between the database of pollutants and the FFQs. These included dairy products (milk and yogurt), cheese, white meat (poultry and rabbit), red meat (i.e., veal, lamb, pork), processed meat (i.e., hamburger, sausage, cured ham, boiled ham), eggs, blue fish (i.e., sardine, tuna, anchovy, mackerel, swordfish, salmon, red mullet, canned sardine, canned tuna), white fish (i.e., hake, cod, sole, bream), seafood (i.e., cuttlefish, squid, clam, mussel, prawn), salty cereals and

tubers (i.e., bread, breakfast cereals, rice, pasta and potato), sweet cereals (i.e., pies, biscuits and pastries), vegetables (salads [lettuce, tomato, carrot] and other vegetables), fruits (raw fruits and preserved fruits), legumes (lentils, chickpeas, beans and peas), and nuts. Secondly, we multiplied the consumption of these 15 groups, in grams per day or week, by their corresponding content of metals, metalloids and POPs as reported in the database of pollutants. Of note, the toxic equivalents (TEQs) for PCDD/Fs and DL-PCBs were calculated from the WHO-established toxic equivalent factors (WHO-TEF) published in 2006 (Van den Berg et al., 2006). Thirdly, to obtain the total dietary intake per day or week of each metal, metalloid and POP for each child, these intakes were summed and adjusted for body weight (in kg) for enable of comparison with the corresponding health-based guidance values or benchmark dose (BMD) using the following formula:

$$\text{Estimated dietary pollutant intake} = \frac{\sum (\text{Content of pollutant in food}_i \times \text{Food consumption [g/day or week]}_i)}{\text{Body weight [kg]}}$$

More information on the methodology used to quantify the level of metals, metalloids and POPs in foodstuffs can be found elsewhere (Bosch-Collet et al., 2020) and in supplementary material.

#### 2.4. Assessment of other variables

Body weight (bw) (in kg) and height (cm) were measured in face-to-face visits by trained staff and BMI-for-age z score was calculated based on the World Health Organization (WHO) Child Growth Standards using the *zanthro* command in STATA (version 1.0.2 dec2011 (SJ13-2: dm0004\_1)) (Vidmar et al., 2013). Children were classified as having underweight (< -2), normal weight (≥ -2 to ≤ 1), overweight (>1 to ≤3 in children under 5 years of age, and >1 to ≤2 in children over 5 years of age) or obesity (>3 in children under 5 years of age, and >2 in children over 5 years of age) according to the WHO cut-off points (de Onis and Lobstein, 2010).

Socioeconomic status was calculated by the Hollingshead index using data reported by parents on educational level and profession (Hollingshead, 1975).

#### 2.5. Risk characterization

The risk characterization was performed by comparing the mean intake estimates of the study population with the tolerable daily intake (TDI), tolerable weekly intake (TWI) or lower limit of BMD (BMDL) established by the European Food Safety Authority (EFSA). For MeHg and Cd, the TWI of 1.3 µg/kg bw/week and 2.5 µg/kg bw/week, respectively was used (EFSA Panel on Contaminants in the Food Chain (CONTAM), 2011, 2012). For Pb, we used the BMDL<sub>01</sub> of 0.5 µg/kg bw/day defined for neurotoxic effects in young children (European Food Safety Authority (EFSA), 2012). For inAs, EFSA has recently set up a BMDL of 0.06 µg/kg bw/day for 5% excess risk of skin cancer (BMDL<sub>05</sub>), which should also be considered to cover other cancers (lung and bladder) and chronic kidney disease, skin lesions, ischemic heart disease, respiratory disease, stillbirth, spontaneous abortion, infant mortality and neurodevelopmental effects (Schrenk et al., 2024).

Regarding POPs, we used the TWI of 2 pg TEQ/kg bw/week for both PCDD/Fs and DL-PCBs established in 2018 (Knutsen et al., 2018). For NDL-PCBs, EFSA has concluded that it is not possible to establish a TWI, TDI or BMDL (EFSA Panel on Contaminants in the Food Chain, 2005). Hence, for the risk characterization, we have used the TDI of 10 ng/kg bw/day established by the French Food Safety Agency (Agence

Française De Sécurité Sanitaire Dels Aliments, 2007) for six congeners of NDL-PCBs (28, 52, 101, 138, 153 and 180), which has also been used in other previously health risk assessment studies (Hulin et al., 2020; Lee et al., 2021; Morales-Suarez-Varela et al., 2018).

The percentage of children exceeding the health-based guidance values (i.e., TWI, TDI) or BMDL was calculated. For pollutants (i.e., inAs and Pb) with available BMDL rather than health-based guidance values, the margin of exposure (MOE) was also calculated from the mean exposure of each age group by dividing the BMDL value by the estimated dietary intake of the pollutant (European Food Safety Authority (EFSA), 2023). An MOE ≤ 1 corresponds to a dietary intake of pollutant that could be associated to an increased risk of skin cancer (for inAs) or increased risk of developmental neurotoxicity (for Pb). It should be noted that in the last EFSA update on the risk assessment of inAs in food,

the Panel concluded that it was not possible to determine a value for an MOE of low concern since there is no precedents in the EFSA for the identification of an MOE of low concern when using a BMDL obtained from data on human cancer (Schrenk et al., 2024). Therefore, one should be cautious when interpreting an MOE >1 for this pollutant, as a risk cannot be ruled out.

#### 2.6. Statistical analysis

Descriptive data are presented as mean ± standard deviation for quantitative variables and percentage for qualitative variables. Characteristics of the study population across age groups were compared using the *t*-test for independent samples or the chi-squared test, as appropriate. The *t*-test for independent samples was used to test whether there were statistically significant differences in the mean intake per

**Table 1**  
Characteristics of the study population.

Characteristics	Total study population (n = 976)	Preschoolers (n = 533)	Primary school children (n = 443)	P-value <sup>a</sup>
Sex, boy, n (%)	568 (58.2)	294 (55.2)	274 (61.9)	0.04
Age, years	7.6 ± 3.2	4.8 ± 0.6	11.1 ± 0.5	<0.01
Weight, kg	30.0 ± 14.7	18.9 ± 3.8	43.4 ± 11.3	<0.01
Height, cm	126.0 ± 20.7	108.4 ± 6.5	147.3 ± 8.3	<0.01
z-score BMI	0.55 ± 1.35	0.35 ± 1.30	0.78 ± 1.36	<0.01
Categories, n (%)				<0.01
Underweight	27 (2.8)	14 (2.6)	13 (2.9)	
Normal weight	596 (61.1)	383 (71.9)	213 (48.08)	
Overweight	238 (24.4)	106 (19.9)	132 (29.8)	
Obese	115 (11.8)	30 (5.6)	85 (19.2)	
Socioeconomic status, n (%)				0.21
Low	156 (15.9)	76 (14.3)	80 (18.1)	
Medium	629 (64.6)	346 (64.9)	283 (63.9)	
High	191 (19.6)	111 (20.8)	80 (18.1)	
Total energy intake (kcal/day)	1508 ± 417	1474 ± 391	1550 ± 444	<0.01
SDQI score	59.4 ± 7.2	61.0 ± 7.0	57.4 ± 6.9	<0.01

Data expressed as mean ± standard deviation for continuous variables and number and percentage for categorical variables.

Abbreviations: BMI, body mass index; SDQI, Spanish Diet Quality Index.

<sup>a</sup> P-value for differences between pre-schoolers and primary school children by *t*-test or chi-square test, as appropriate.

body weight of different food groups between preschoolers and primary school children. Significant level was set at  $p$ -value  $< 0.05$ . All statistical analyses were performed using STATA statistical software version 15.0 (StataCorp LP, Tx. USA).

### 3. Results

#### 3.1. Description of the study population

Table 1 depicts the general characteristics of the study population by age group. Briefly, in the preschoolers, 55.2% were boys, and the mean age was 4.8 years, with a mean BMI z-score of 0.35. The majority of preschoolers (71.9%) were of normal weight, while 25.5% had overweight or obesity. The vast majority (64.9%) had a medium socioeconomic level. In the primary school children, 61.9% were boys, the mean age was 11.1 years, with a mean BMI z-score of 0.78. Forty-eight percent of primary school children had normal weight, whereas 49% had overweight or obesity. Most of these children were of middle socioeconomic status. Preschoolers had better adherence to the SDQI index than the primary school children (mean score: 61.0 vs 57.4, respectively).

#### 3.2. Dietary intakes of metals, metalloids and POPs and main food groups contributing to intake

The mean dietary intake of each pollutant by age group (preschool and primary school children) is presented in Table 2. Preschoolers showed higher intake per kg of body weight of all pollutants than primary school children. Briefly, preschoolers showed a mean dietary intake of 0.16  $\mu\text{g}/\text{kg}$  bw/day of InAs, 1.75  $\mu\text{g}/\text{kg}$  bw/week of Cd, 1.81  $\mu\text{g}/\text{kg}$  bw/week of MeHg, 0.26  $\mu\text{g}/\text{kg}$  bw/day of Pb, 3.72 and 6.17  $\text{pg}/\text{kg}$  bw/week of PCDD/Fs and DL-PCBs, respectively, and 9.36  $\text{ng}/\text{kg}$  bw/day of NDL-PCBs. Primary school children showed a mean dietary intake of 0.10  $\mu\text{g}/\text{kg}$  bw/day of InAs, 1.06  $\mu\text{g}/\text{kg}$  bw/week of Cd, 0.88  $\mu\text{g}/\text{kg}$  bw/week of MeHg, 0.13  $\mu\text{g}/\text{kg}$  bw/day of Pb, 1.78 and 3.11  $\text{pg}/\text{kg}$  bw/week of PCDD/Fs and DL-PCBs, respectively, and 4.51  $\text{ng}/\text{kg}$  bw/day of NDL-PCBs.

Figs. 1 and 2 show the percentage contribution of the different food groups to the total dietary intake of metals/metalloids and POPs, respectively, in preschool and primary school children. The percentage of pollutants ingested through specific food groups were distributed very similarly between the two age groups, with small variations in the proportion and in the ranking of contributors. In both preschool and primary school children, salted cereals and potatoes contributed most to the InAs intake; Cd was predominantly ingested from salted cereals and potatoes and seafood; most MeHg came from fatty fish and white fish; and cereals and potatoes and sweet cereals were the main source of Pb. In terms of POPs, many food groups contributed to PCDD/Fs intake,

although milk and yogurt were the main food source, followed by white fish in preschoolers and fatty fish in primary school children. In relation to DL-PCBs, fatty fish contributed the most to the total intake. Finally, fish was the main food source of NDL-PCBs, being white fish in preschoolers and fatty fish in primary school children.

Table 3 shows that preschoolers consumed more grams of all food groups per kg of body weight than primary school children.

#### 3.3. Risk characterization

To assess any potential risk, mean estimated dietary intakes of InAs, Cd, Pb, MeHg, PCDD/Fs, DL-PCBs and NDL-PCBs were compared with their corresponding TDI, TWI or BMDL (Table 2). The estimated intakes of Cd, Pb and NDL-PCBs did not exceed the reference values in any age group. Estimated intakes of InAs and DL-PCBs exceeded the TDI and TWI, respectively, in both age groups, whereas estimated intakes of MeHg, and PCDD/Fs exceeded the TWI in preschoolers but not in primary school children.

The percentage of children exceeding the reference values was also calculated for each pollutant (Table 2). For preschoolers, more than 90% exceeded the InAs, PCDD/Fs and DL-PCBs limits, and 64.35% exceeded the MeHg reference value. In primary school children, the 83.30% and 62.98% exceed the InAs and DL-PCBs limits, respectively.

For InAs and Pb, an MOE was calculated based on the estimated mean intake and the corresponding BMDL. The MOE of InAs was 0.38 and 0.60 for preschoolers and primary school children, respectively. The MOE of Pb was 1.92 for preschoolers and 3.84 for primary school children.

### 4. Discussion

In the present study, the dietary intake of six different pollutants in two different age groups, preschoolers and primary school children, in the province of Tarragona, Catalonia (Spain), was evaluated and compared with the relevant reference values established by EFSA. In addition, the main food groups contributing to the total dietary intake of each pollutant were identified. The findings showed that both preschoolers and primary school children exceeded the relevant reference value for InAs and DL-PCBs, whereas only preschoolers exceeded the limit for MeHg and PCDD/Fs. Intakes of Cd, Pb and NDL-PCBs did not exceed the limits in either age group. This highlights the fact that dietary exposure to the above-mentioned pollutants exceeding EFSA limits poses a potential health risk for children in this geographical area.

Although pollutants impact all human beings, children are among the most vulnerable to their effects mainly because their food intake relative to their body weight is higher than that of adults and their immune, endocrine, reproductive, digestive and central nervous systems

**Table 2**

Estimated dietary intake of metals, metalloids and POPs in the study population by age group compared to relevant reference values.

Toxicant	Relevant reference value	Contaminant intake in our study population		% of participants exceeding the reference value		
		Pre-schoolers (n = 533)	Primary school children (n = 424)	Pre-schoolers (n = 533)	Primary school children (n = 424)	
InAs	BMDL <sub>05</sub>	0.06 $\mu\text{g}/\text{kg}$ bw/d	<b>0.16 <math>\pm</math> 0.06</b>	<b>0.10 <math>\pm</math> 0.04</b>	98.31	83.30
Cd	TWI	2.5 $\mu\text{g}/\text{kg}$ bw/week	1.75 $\pm$ 0.84	1.06 $\pm$ 0.56	14.26	2.71
MeHg	TWI	1.3 $\mu\text{g}/\text{kg}$ bw/week	<b>1.81 <math>\pm</math> 1.10</b>	0.88 $\pm$ 0.61	64.35	19.19
Pb	BMDL <sub>01</sub>	0.50 $\mu\text{g}/\text{kg}$ bw/d	0.26 $\pm$ 0.16	0.13 $\pm$ 0.09	7.69	0.68
PCDD/Fs	TWI	2 $\text{pg}/\text{kg}$ bw/week <sup>a</sup>	<b>3.72 <math>\pm</math> 1.24</b>	1.78 $\pm$ 0.71	93.62	30.02
DL-PCBs			<b>6.17 <math>\pm</math> 4.02</b>	<b>3.11 <math>\pm</math> 2.27</b>	90.99	62.98
NDL-PCBs	TDI	10 $\text{ng}/\text{kg}$ bw/d	9.36 $\pm$ 5.38	4.51 $\pm$ 3.01	35.83	6.09

Values are expressed as means  $\pm$  SD (standard deviation).

The numbers in bold indicate that the dietary pollutant intake exceeds the established reference value (TWI or BMDL).

Abbreviations: As, arsenic; BMDL, Benchmark dose response modelling; Cd, Cadmium; CONTAM, Panel on Contaminants in the Food Chain; DL-PCBs, dioxin-like polychlorinated biphenyls; EFSA, European Food Safety Authority; InAs, Inorganic arsenic; MeHg, methylmercury; NDL-PCBs, non-dioxin-like polychlorinated biphenyls; Pb, lead; PCDD/Fs, polychlorinated dibenzo-p-dioxins and dibenzofurans; POPs, persistent organic pollutants; TDI, tolerable daily intake; TWI, tolerable weekly intake.

<sup>a</sup> Tolerable weekly intake is 2  $\text{pg}/\text{kg}$  bw/week for both DL-PCBs and NDL-PCBs.

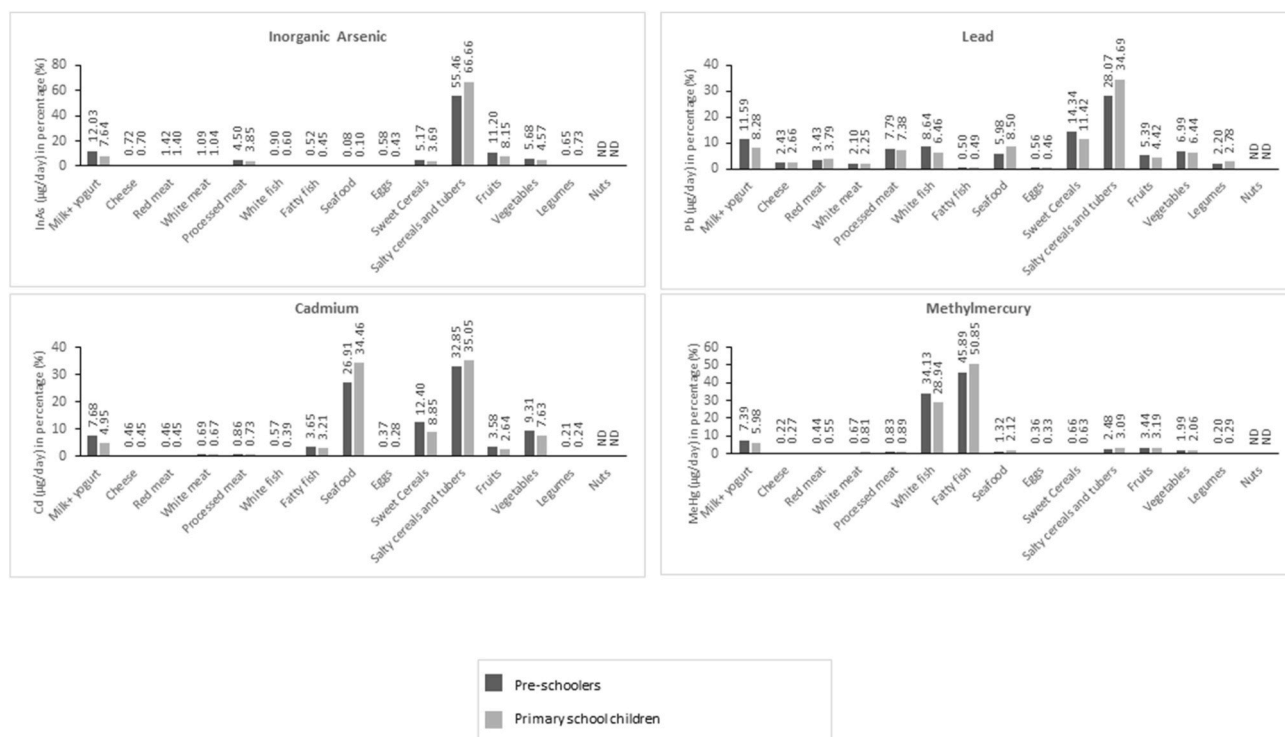


Fig. 1. Percentage contribution of different food groups to the total dietary exposure intake of inorganic arsenic (InAs), lead (Pb), cadmium (Cd), and methylmercury (MeHg) by age group.

are still maturing and developing. Despite all of this, to the best of our knowledge no previously published study has performed a risk evaluation of both metal/metalloids and different POPs specifically in this subpopulation in the province of Tarragona, Catalonia (Spain). The two previous studies conducted in this geographical area have evaluated dietary exposure to some metals/metalloids (such as As, Cd, Pb and Hg) (Gonzalez et al., 2021) and PCDD/Fs (Gonzalez et al., 2018) in various age groups (including children and adolescents), but the main food contributors to the total intake of these pollutants were not assessed in children. Moreover, although the authors included in their evaluation As and Hg, they did not include data on inAs and MeHg which are much more toxic (Balali-Mood et al., 2021). In addition, neither DL-PCBs nor NDL-PCBs were evaluated. Hence, the present study is the first one performing a risk evaluation of inAs, Cd, Pb, MeHg, PCDD/Fs, DL-PCBs and NDL-PCBs as well as the identification of the main food groups contributing to their intake specifically in preschoolers and primary school children from the province of Tarragona, Catalonia (Spain).

Direct comparison of the average intake of the different contaminants included in the present study and other studies conducted in other regions of the world is a challenge, since dietary exposure to contaminants depends not only on the concentration levels of pollutants present in food but also on the dietary habits of the population, which could vary significantly between countries and regions. This is why in the following discussion section we have mainly compared our results with other studies conducted in Catalonia or other regions of Spain.

In the current study, dietary intake of inAs in both age groups exceeded the new updated BMDL<sub>05</sub> of 0.06 µg/kg bw/day established by EFSA based on its effects on skin cancer. Moreover, the MOE was smaller for preschoolers (0.38) than primary school children (0.60), reflecting that younger children have higher dietary exposure to inAs relative to their body weight. These small MOEs highlight that certain risk cannot be ruled out. These results are in line with the fifth total diet study (TDS) conducted in Catalonia, where children aged 3–9 years and adolescents aged 10–17 years showed inAs intakes higher than the new BMDL<sub>05</sub> (0.13 and 0.07 µg/kg bw/day, respectively) (Bosch-Collet et al., 2020).

Our findings related to inAs have serious implications since this pollutant is considered a category 1 carcinogen by the International Agency for Research on Cancer (IARC) (International Agency for Research on Cancer, 2024). Moreover, inAs has also shown to have adverse effects on children neurodevelopment (Bellinger, 2013). The level of inAs in each of the food groups included in the present study shows that rice has the highest concentration (0.108 µg/g) followed by breakfast cereals (0.021 µg/g) and pasta (0.018 µg/g), although it does not exceed the maximum legal values. Therefore, it is not surprising that the main contributor to the total inAs intake in both preschoolers and primary school children is salted cereals and potatoes group as it is one of the food groups with the highest daily intake per kg of body weight in both age groups ( $6.44 \pm 2.93$  g/kg bw/day in preschoolers and  $3.87 \pm 1.83$  g/kg bw/day in primary school children). In fact, cereal products have been reported as important contributors to the daily exposure to inAs in the general European population (Arcella et al., 2021).

Regarding DL-PCBs, also the daily intake in both preschoolers (6.17 pg/kg bw/week) and primary school children (3.11 pg/kg bw/week) exceed the EFSA limit value of 2 pg/kg bw/week set by EFSA in 2018 based on their critical effect on sperm quality (Knutsen et al., 2018). These results are very similar with those of the population of Catalonia (Spain) where the mean weekly intake exceed the reference value in children (6.16 pg/kg bw/week) and adolescents (2.97 pg/kg bw/week) (Bosch-Collet et al., 2020). Of note, DL-PCBs intake was much higher than that of young people aged 6–15 years (PCDD/Fs + DL-PCBs intake = 3.4 pg/kg bw/week using the worst-case scenario) from Valencia (Spain) (Lacomba et al., 2024). DL-PCBs have lipophilic properties and are poorly degraded, therefore they easily accumulate in the food chain, mainly in animal products (fish, dairy and meat). In fact, in agreement with our results, fish was found to be the main dietary source of DL-PCBs in both aforementioned studies. Considering this, it has been suggested in the Codex Alimentarius (CXC 62–2006 – revised in 2018) that good agricultural, manufacturing and animal feeding practices will contribute to the reduction of POPs (including dioxins and PCB) levels in food (Food and Agriculture Organization of the United Nations, World Health

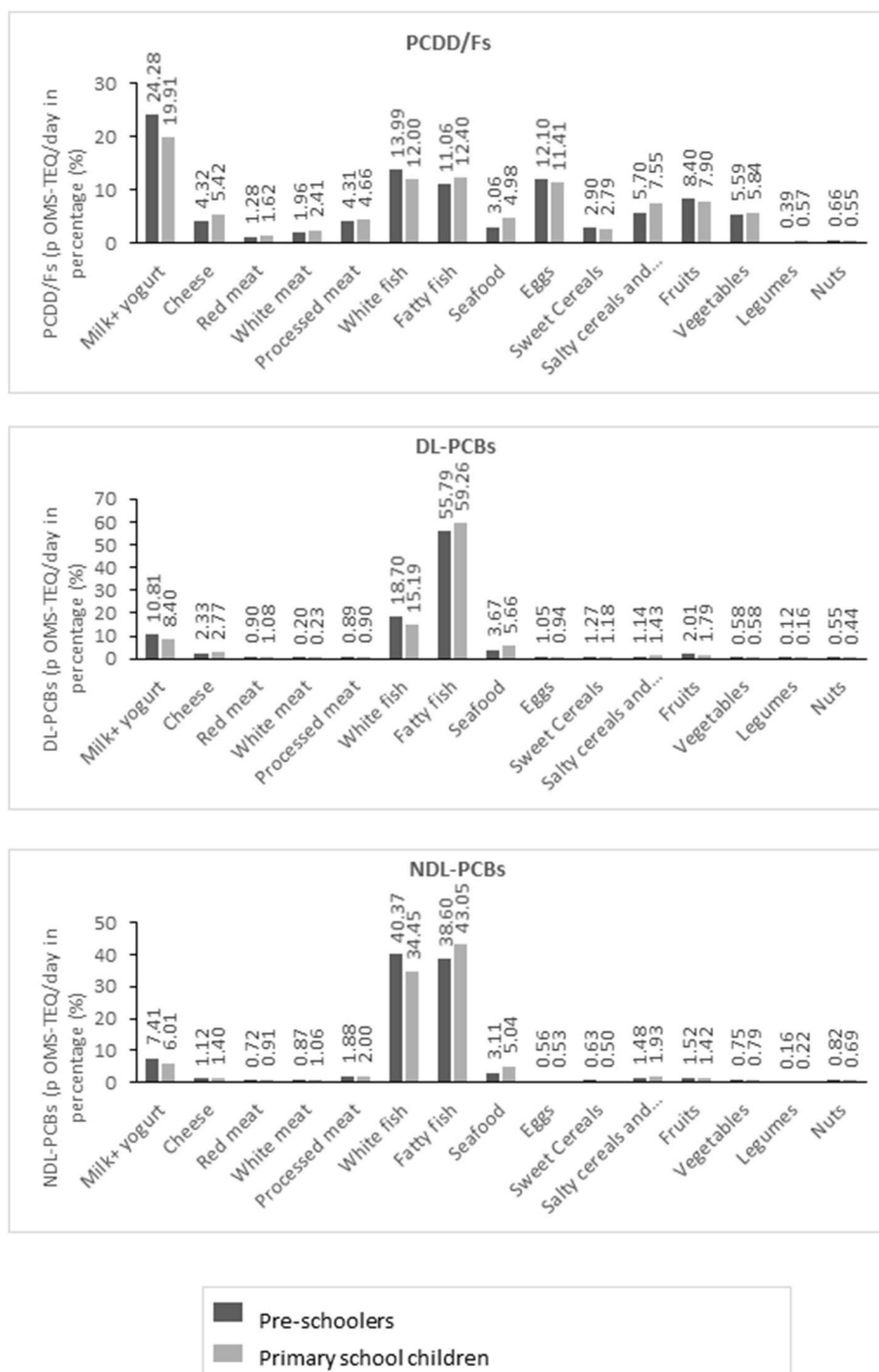


Fig. 2. Percentage contribution of different food groups to the total dietary exposure intake of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), dioxins like polychlorinated biphenyls (DL-PCBs) and non-dioxin like polychlorinated biphenyls (NDL-PCBs) by age group.

Organization, 2006). These measures, along with specific dietary recommendations (such as controlling the amount and type of fish consumed) are of extreme importance considering that, in the current study, children exceeded the health-based guideline values of DL-PCBs.

Our findings also showed that only preschoolers exceeded the limit for MeHg (1.81 µg/kg/bw/week) and PCDD/Fs (3.72 pg/kg bw/week). In contrast to our results, in the fifth TDS performed in Catalonia neither children nor adolescents exceeded the relevant EFSA values of MeHg. However, their results mirrored ours in terms of PCDD/Fs, as only children, but not adolescents exceeded the established limit (Bosch-Collet et al., 2020). In the two previous studies carried out in the

Tarragona County, MeHg was not evaluated, but the daily intake of PCDD/Fs was assessed according to different age groups. The results showed that children (3–9 years) had an intake of about 0.40 pg/kg bw/day and adolescents (10–17 years) about 0.25 pg/kg bw/day, equivalent to 2.8 pg/kg bw/week and 1.75 pg/kg bw/week, respectively (Gonzalez et al., 2018). Therefore, the results of this study are similar to ours, since children but not adolescents exceeded the new updated EFSA limit of 2 pg/kg bw/week established in 2018. Dietary intake of MeHg above the EFSA limit of 1.3 µg/kg/bw/week is of concern, as this pollutant may induce neurotoxic effects in children (whose brain is still developing) (EFSA Scientific Committee, 2015). Likewise, PCDD/Fs

**Table 3**

Dietary intake (g/kg body weight/day) by age group.

Food group	Preschoolers (n = 533)	Primary school children (n = 443)	P-value <sup>a</sup>
Milk and yogurt	19.28 ± 9.28	7.55 ± 4.09	<0.01
Cheese	0.58 ± 0.55	0.34 ± 0.32	<0.01
Red meat	1.13 ± 0.843	0.69 ± 0.50	<0.01
White meat	1.739 ± 0.94	1.00 ± 0.52	<0.01
Processed meat	2.13 ± 1.08	1.11 ± 0.54	<0.01
White fish	1.42 ± 1.02	0.59 ± 0.47	<0.01
Fatty fish	0.82 ± 0.84	0.44 ± 0.46	<0.01
Seafood	0.12 ± 0.17	0.09 ± 0.11	<0.01
Eggs	0.93 ± 0.57	0.42 ± 0.26	<0.01
Sweet cereals	2.20 ± 1.55	1.11 ± 0.80	<0.01
Salty cereals and tubers	6.44 ± 2.93	3.87 ± 1.83	<0.01
Fruits	9.02 ± 6.74	4.006 ± 3.052	<0.01
Vegetables	5.16 ± 2.77	2.56 ± 1.63	<0.01
Legumes	0.52 ± 0.34	0.36 ± 0.25	<0.01
Nuts	0.18 ± 0.23	0.07 ± 0.10	<0.01

Values expressed as mean ± standard deviation.

<sup>a</sup> P-value for differences between pre-schoolers and Primary school children by *t*-test.

intakes above the health-based guideline value are worrying, as these are highly toxic compounds that can cause damage to the immune system, alter neurodevelopment, and have adverse reproductive and developmental effects (Gonzalez and Domingo, 2021).

It should be highlighted that our study is the first to evaluate the food groups that contributed most to the total intake of MeHg and PCDD/Fs in children in Tarragona. Fish was found to be the main dietary source of MeHg, which is in line with results observed in the European region (EFSA Scientific Committee, 2015). In addition, fish is an important source of beneficial nutrients, particularly proteins, polyunsaturated fatty acids (omega-3), selenium, iodine, calcium and vitamin D (EFSA NDA Panel, 2014), and a core component of many healthy dietary patterns, such as the Mediterranean Diet. Therefore, according to the EFSA, fish species with a high content of mercury should be limited in diet of vulnerable populations, such as children, to protect against MeHg neurotoxicity and to obtain the health benefits of fish consumption which has been observed at 1–4 servings of fish/week (EFSA Scientific Committee, 2015). With regards to PCDD/Fs, we observed that dairy products, followed by fish, were the main contributors to their total intake in children, which is somehow in line with a recent study conducted in children from Italy (Barone et al., 2021).

In the present study, intakes of Cd, Pb and NDL-PCBs did not exceed the limits in either age group. These results are completely in line with those reported in the fifth TDS conducted in Catalonia, where none of the exposed population groups (children aged 3–9 years and 10–17 years) exceeded the EFSA limit values for Cd and Pb (Bosch-Collet et al., 2020). However, although NDL-PCBs were also assessed, exact results of dietary intake in ng/kg bw/day for the different age groups were not reported. Our results are also quite consistent with those of a previous study conducted in the Tarragona County where neither children nor adolescents exceeded the TWI of 2.5 µg/kg bw/week for Cd. However, their results did show that dietary intake of Pb in children (0.88 µg/kg bw/week), but not adolescents, was above the BMDL01 of 0.50 µg/kg bw/day established for neurotoxicological development (Gonzalez et al., 2021).

The present study has some limitations that need to be highlighted. First, the sensitivity and specificity of the methods used to assess pollutants in food may have affected the accuracy of the exposure (metals/metalloids and POPs) assessment. Second, dietary intake was assessed by means of an FFQ, therefore measurement errors are inevitable. However, the FFQs used in the present study were validated for this specific population. Third, we did not include drinking water as a source of pollutants in our analysis, which could result in an underestimation of inAs intake. However, in Spain there is a regulation limiting the

maximum arsenic content in drinking water to 10 µg/l (Gobierno de España, 2023), being the mean concentration in 2017–2022 between 0.92 and 1.03 µg/l (Ministerio de Sanidad, 2022). Therefore, we do not expect to have this an important impact in our results, since relatively low levels of inAs in drinking water has been reported in Spain. The present study also has some strengths that deserve to be mentioned, such as the relatively large sample size, the use of validated FFQs, the use of a composition table on pollutants derived from local foodstuff, the inclusion of inAs and MeHg in the assessment, and the measurement of body weight by trained staff (previously studies used the mean weight reported in the literature for their risk characterization assessment).

## 5. Conclusions

The present study showed that children in the Tarragona area (Spain), mainly preschoolers, exceeded EFSA limits for different metals/metalloids and POPs. Therefore, efforts are still needed in this geographical area to reduce dietary exposure to pollutants in children. In view of the large presence of pollutants in food and their serious health risks, the reduction of food contamination needs to be considered a major public health priority. In addition, considering that pollutants intake depends not only on the level of food contamination but also on the type of dietary followed, specific dietary recommendations for children should be developed, weighting the health risk and benefits of dietary exposure to pollutants and nutrients present in food.

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## Ethical statement

The studies were designed in agreement with the Declaration of Helsinki and the Tokyo Update. The protocol of the EPINED study was approved by the Ethics Committee at the Sant Joan University Hospital (13-10-31/10proj5) and the protocol of the ECLIPSES study by the Ethical Committee of the Jordi Gol Institute for Primary Care Research and the Pere Virgili Institute for Health Research.

## Data availability

The data that has been used is confidential. The datasets generated and/or analyzed in the current study are available from the corresponding author upon reasonable request.

## CRediT authorship contribution statement

**Nerea Becerra-Tomás:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis. **Victoria Arja:** Writing – review & editing, Resources, Project administration, Investigation, Funding acquisition, Data curation, Conceptualization. **Cristina Jardí:** Writing – review & editing, Investigation, Data curation. **Lucía Riggioni-Saborío:** Writing – review & editing, Investigation. **Cristina Bedmar:** Writing – review & editing, Investigation, Data curation. **Josefa Canals-Sans:** Writing – review & editing, Resources, Project

administration, Investigation, Funding acquisition, Data curation, Conceptualization.

## 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.

## Data availability

The data that has been used is confidential.

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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.2024.115030>.

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