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Dietary intake of metals, metalloids, and persistent organic pollutants in Spanish pregnant women. ECLIPSES study

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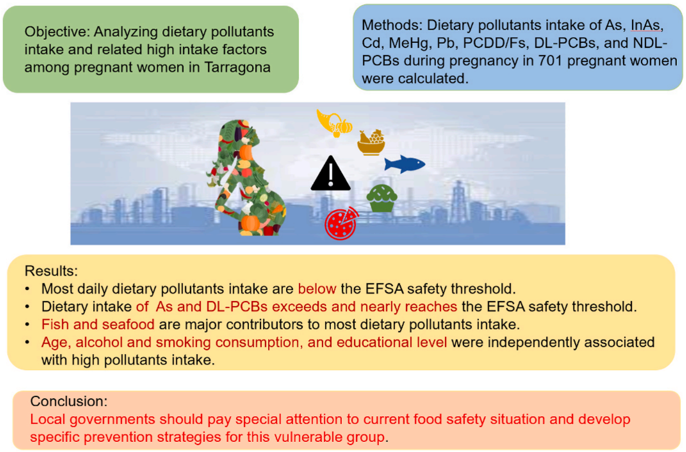
HIGHLIGHTS

- Pregnant women from an industrial area are the target population.
- Dietary pollutants intake among pregnant women are estimated.
- Most dietary pollutants intake are under EFSA safety threshold.
- Dietary As and DL-PCBs intake exceeds and nearly reaches EFSA safety threshold.
- Age, education level, smoking and alcohol are associated with pollutants intake.

GRAPHICAL ABSTRACT



Objective: Analyzing dietary pollutants intake and related high intake factors among pregnant women in Tarragona

Methods: Dietary pollutants intake of As, InAs, Cd, MeHg, Pb, PCDD/Fs, DL-PCBs, and NDL-PCBs during pregnancy in 701 pregnant women were calculated.

Results:
- Most daily dietary pollutants intake are below the EFSA safety threshold.
- Dietary intake of As and DL-PCBs exceeds and nearly reaches the EFSA safety threshold.
- Fish and seafood are major contributors to most dietary pollutants intake.
- Age, alcohol and smoking consumption, and educational level were independently associated with high pollutants intake.

Conclusion:
Local governments should pay special attention to current food safety situation and develop specific prevention strategies for this vulnerable group.

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ABSTRACT

Objective: This study aimed to describe dietary intake and important dietary sources to pollutants as well as to identify maternal socio-economic and lifestyle factors associated with high intake during pregnancy in women residing in a Mediterranean city with heavy industrial activity.
Methods: Dietary intake during pregnancy of As, InAs, Cd, MeHg, Pb, PCDD/Fs, DL-PCBs, and NDL-PCBs in 701 pregnant women participating in the longitudinal ECLIPSES study was calculated based on a 45-item food-

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Total diet study
Risk assessment

frequency questionnaire and a database of pollutants in food of the Catalan Food Safety Agency. Details on socio-economic, lifestyle, and anthropometric variables were also collected.

Results: The mean dietary intake of pollutants per day and the food group that contributed the most (%) was: 286.51 μg of As (71.27% from white fish), 4.14 μg of InAs (70.16% from cereals-tubers), 6.27 μg of Cd (47.51% from seafood), 5.00 μg of MeHg (52.88% from blue fish), 3.32 μg of Pb (30.15% from cereals-tubers), 9.93 pg of PCDD/Fs (from many food categories), 18.39 pg of DL-PCBs (59.74% from blue fish) and 181.00 ng of NDL-PCBs (44.58% from blue fish). Adjusted multivariate analysis revealed that older age was associated with high As intake, higher educational level was related to low InAs, Cd, and DL-PCBs intake, and alcohol use and smoking were linked with high Pb intake.

Conclusion: The dietary intake of pollutants including As and DL-PCBs among pregnant women exceeds or almost reaches the EFSA safety threshold. These findings support the urgent need for local governments to pay special attention to this situation and develop specific prevention strategies for this vulnerable group.

1. Introduction

Diet serves as the primary pathway for human exposure to various chemical and microbiological pollutants (Guo et al., 2019). Pollutants can enter the food chain at any stage, from the farm to the consumer's plate, and pose potential risks for numerous disorders (Toussaint et al., 2019). Among food pollutants, metalloids and metals such as total arsenic (As), cadmium (Cd), lead (Pb), among others are one of the main concerns (Rehman et al., 2018). These elements can change their chemical forms, but they are not degraded or destroyed within the human body, making them highly toxic according to the US Agency for Toxic Substances and Disease Registry (Profiles, 2023). It is widely reported that even a minimal concentration exposure to certain metals can have adverse effects on human health (Fu and Xi, 2020). For instance, methylmercury (MeHg) is a more toxic form of mercury, which accumulates in the food chain and whose exposure can damage the nervous system, resulting in symptoms such as tremors, impaired cognitive function, and developmental delays (Culbreth and Aschner, 2019; Yang et al., 2020). Furthermore, inorganic arsenic (InAs) is a naturally occurring form of arsenic, which can cause skin lesions, cardiovascular disease, and even cancer (Martínez-Castillo et al., 2021; Palma-Lara et al., 2020).

Similarly, persistent organic pollutants (POPs) such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), dioxin like and non-dioxin like polychlorinated biphenyls (DL-PCBs and NDL-PCBs) are highly toxic compounds that are produced intentionally or not by industrial processes, including waste incineration, chemical manufacturing, and paper production (Popli et al., 2022). It has been proved that more than 80% of human exposure to these POPs is through dietary intake, especially from animal sources (Amutova et al., 2021; Li et al., 2019). Over the decades, although persistent organic pollutants are closely monitored by regulatory bodies like European Food Safety Authority (EFSA), only a small margin exists between current exposure levels among European and the levels causing biological effects including reproductive and developmental problems, immune system disorders, endocrine disruption, cancer, etc. (on Contaminants in the Food Chain (CONTAM) et al., 2018; Pasecnaja et al., 2022).

Pregnant women are considered a particularly vulnerable population affected by pollutants, which can have negative effects on both the mother and the developing fetus (Bank-Nielsen et al., 2019; de Figueiredo et al., 2020). Currently, several studies have been conducted to analyze pollutants in blood or urine samples during pregnancy, including studies conducted in Tarragona (Bocca et al., 2019, 2020). However, information regarding dietary intake of pollutants among pregnant women and its association with other socio-economic, lifestyle, and anthropometric factors is much scarce (Caspersen et al., 2013; González et al., 2019, 2021).

Thus, the current research aims to analyze the dietary intake of metalloids (As and InAs), metals (Cd, MeHg and Pb), and POPs (PCDD/Fs, DL-PCBs, and NDL-PCBs) during gestation, as well as to examine the influence of socio-economic, lifestyle, and anthropometric factors in this intake in pregnant women from an industrial area in the north-east of Spain. This information would be highly useful to further develop preventive strategies for this target population.

2. Methods

2.1. Study design and participants

A population-based descriptive longitudinal cohort study of pregnant women who participated in the ECLIPSES study was conducted during pregnancy. A detailed description of ECLIPSES and the inclusion/exclusion criteria has been published elsewhere (Arija et al., 2014). Briefly, healthy women older than 18 years of age, pregnancy at ≤12 weeks, ability to understand the local and official state languages and the characteristics of the study, and signing the informed consent form were included. Meanwhile, women with multiple pregnancies, having taken iron supplements during the months previous to week 12 of pregnancy, hypersensitivity to egg protein, previous severe disease (immunosuppression), or any chronic disease which could affect their nutritional development (cancer, diabetes, malabsorption, or liver disease) were excluded.

In summary, a total of 791 pregnant women were recruited during their first prenatal visit by midwives from 12 sexual and reproductive health care services (ASSIR) of the Catalan Institute of Health in the province of Tarragona (Catalonia, Spain), between 2013 and 2017. For the present analysis, all women who had data regarding dietary intake during pregnancy were included. The total study sample therefore comprised 701 pregnant women. Written informed consent was obtained from each woman. The ECLIPSES study was registered at www.clinicaltrialsregister.eu with identification number EUCTR-2012-005480-28 and at www.clinicaltrials.gov with identification number NCT03196882. The study was approved by the Ethical Committee of the Jordi Gol Institute for Primary Care Research and the Pere Virgili Institute for Health Research and complied with the tenets of the Helsinki Declaration.

2.2. Data collection

Midwives and nutritionists collected participants' information about demographic characteristics (maternal age, socioeconomic information, and education level), health behaviors (physical activity [PA], smoking and diet), as well as anthropometric measurements from the first to third trimester of pregnancy. The socioeconomic level was calculated according to the family's occupational status using the Catalan classification of occupations (CCO-2011) (Catalunya and Catalunya, 2011) and classified as low, middle, and high. Education level was classified as low (primary), medium (high school), and high (university studies or above). Physical activity (PA) was measured using the short version of the International Physical Activity Questionnaire (IPAQ-S) (Fagerström, 1978). From the frequency and duration of different activities, the total metabolic equivalent (MET-min/week) of each participant was estimated, which presented in tertiles (T1: <1125 MET-min/week), T2: 1125–3324 METs-min/week, T3: ≥3324 METs-min/week). The

Fagerström questionnaire (Rodríguez et al., 2008) was used to assess smoking during pregnancy, with women divided into two groups: current or former and never smokers.

Maternal weight (in kg to the nearest 0.1 kg) and height (in cm to the nearest 0.1 cm) were also measured. Early pregnancy body mass index (BMI) was calculated from these measures (weight (kg)/height(m)^2), and women were classified as normal weight (BMI 18.5–24.9 kg/m^2) and overweight and obesity (BMI ≥25 kg/m^2) (World Health Organization WHO, 2006).

Dietary intake was assessed throughout pregnancy (at weeks 12, 24, and 36 of gestation) using a self-administered 45-item or food group food frequency questionnaire (FFQ), validated in our population (deKoning and Anand, 2004). A nutritionist verified that the FFQs administered were complete or had inconsistencies. The FFQ asked for the frequency of weekly or monthly consumption of each item during the previous period of each obstetric interview, and the frequency of consumption per day was calculated. Then, this frequency was transformed into grams per day by applying the average consumption portion of the foods included in each item according to our own data (Rodríguez et al., 2008) and reference data in our population (Javier et al., 2018). Alcohol consumption during pregnancy was also assessed as yes or no.

To estimate the pollutants consumption through diet, the database of pollutants in food of the Catalan Food Safety Agency was used (Agència Catalana de Seguretat Alimentària, 2017). The following 15 food groups were made, according to the similarity between nutritional questionnaires and pollutant database: milk (milk and yogurt), cheese, red meat, white meat (poultry and rabbit), processes meat, white fish, blue fish, seafood, eggs, sweet cereals (biscuits, pies, and pastries), salty cereals (breakfast cereals, bread, pasta, and rice) and tubers, fruits (fruit and preserved fruit), vegetables (salads and vegetables), legumes (peas, beans, lentils, and chickpeas), and nuts. Consumption of each of the 15 food groups (grams/day) was multiplied by the content in metals, metalloids, and POPs expressed in the Pollutant Database, to obtain the dietary pollutants intake for each pregnant woman through the diet. This consumption of pollutants of pregnant women was compared with the tolerable reference levels of the EFSA (on Contaminants in the Food Chain (CONTAM) et al., 2018; on Contaminants in the Food, 2009; (EFSA) et al., 2021; (EFSA) and E.F.S.A, 2009; Panel and Chain, 2012a; on Contaminants in the Food, 2010; (EFSA) and E.F.S.A, 2005).

2.3. *Database of pollutants in food (metalloids and metals and POPs)*

The metalloids and metals database (As, InAs, Cd, MeHg, and Pb) and POPs database (PCDD/Fs, DL-PCBs, and NDL-PCBs) used were carried out following the WHO guidelines. Metalloids and metals and POPs levels were previously reported by Catalan Food Security Agency (Agència Catalana de Seguretat Alimentària, 2017). It comes from 12 cities in the region of Catalonia (Spain), in which the study sample was included (Agència Catalana de Seguretat Alimentària, 2017). The extraction and quantification of metals and metalloids were carried out in the Laboratory of the Public Health Agency of Barcelona through ICP-MS or using an elemental mercury analyzer with amalgam with gold. The extraction of PCDD/Fs and DL-PCB was performed by organic compound and a phase separation by hydrolysis with HCL or by liquid/liquid extraction with oxalate, ethanol ethyl ether and hexane, depending on the type of sample. The extractions of NDL-PCB were analyzed by HRGC-HRMS (minimum resolution 10,000) with Rtx-5MS (PCDD/Fs), TG-5MS (PCBs) chromatographic columns (Agència Catalana de Seguretat Alimentària, 2017). More information regarding methods and results were public avilable at Catalan Food Security Agency report (Agència Catalana de Seguretat Alimentària, 2017) and the main information were translated in Supplementary material.

2.4. *Statistical analysis*

Statistical analyses were performed using SPSS version 25. Descriptive data on dietary intake of As, InAs, Cd, MeHg, Pb, PCDD/Fs and PCBs from food subgroups in pregnant women during pregnancy are expressed as mean ± standard deviation (SD) or percentatge. Based on Kolmogorov–Smirnov test, dietary intake of pollutants were found to follow a normal distribution. Separate multivariate-adjusted logistic models were applied to estimate the odds ratio (OR and 95% CI) for the dietary intake of As, inAs, Cd, Pb, Hg, MeHg, PCDD/Fs and PCBs above the 75th percentile of the study population according to selected sociodemographic and lifestyle characteristics including age (<30 (reference) *vs.* ≥30 years), educational level (low/mèdium (reference) *vs.* high), BMI categories (normal weight (reference) *vs.* overweight/obesity), physical activity (as tertiles (T): T1 and T2 <3336 (reference) *vs.* T3 ≥3336 METs-min/week), alcohol consumption (no (reference) *vs.* yes), and smoking habit (never smoker (reference) *vs.* current/former smoker). All statistical significance was set at $p < 0.05$.

3. Results

3.1. *Descriptive data of the pregnant women*

Table 1 showed the general characteristics of the study subjects. The mean age of women was 30.4 ± 5.2 years with a mean weight of 65.7 ± 12.3 kg. Regarding education level, 33% women had a low level (primary or less), 38% women had medium level (secondary), and 29% women had higher education level (university or more). In terms of occupation, 2% women were students, 73% women were employed, and 25% women were unemployed. Meanwhile, 68% women were never smoker and 32% women were current/former smoker.

Table 1
Characteristics of the study subjects (n = 701).

Maternal characteristics	Summary Statistics
Age (years), mean ± SD	30.4 ± 5.2
Age categories, n (%)	
<30 years	287 (41)
≥30 years	414 (59)
Weight (kg), mean ± SD	65.7 ± 12.3
-BMI (kg/m^2), mean ± SD	25.1 ± 4.5
categories, n (%)	
18.5–24.9 (normal weight)	411 (58)
25.0–29.9 (overweight)	188 (27)
≥30 (obesity)	102 (15)
Educational level, n (%)	
Low (primary or less)	228 (33)
Medium (secondary)	268 (38)
High (university or more)	205 (29)
Social class, n (%)	
Low	110 (16)
Medium	473 (67)
High	118 (17)
Occupation, n (%)	
Students	11 (2)
Employed	515 (73)
Unemployed	175 (25)
Smoking status, n (%)	
Never smoker	477 (68)
Current/former smoker	224 (32)
Alcohol consumption during pregnancy, n (%)	
No	642 (92)
Yes	59 (8)
Physical activity during pregnancy, n (%)	3261.9 ± 4924.7
Tertile 1 (<1125 METs/min/week)	262 (38)
Tertile 2 (1125–3324 METs/min/week)	276 (39)
Tertile 3 (≥3324 METs-min/week)	163 (23)
Energy intake during pregnancy (kcal), mean ± SD	2087.2 ± 603.2

Values are expressed as a mean ± SD (standard deviation) or number (%). Abbreviations: BMI, early pregnancy body mass index; METs, metabolic equivalent of task; MedDiet, Mediterranean diet.

3.2. Dietary intake of pollutants during pregnancy

The mean level of the daily pollutants dietary intake according to daily foodstuff were summarized in Table 2. In brief, the total dietary intake of As was the highest, with a total of 286.51 ± 165.40 µg/day. Total dietary intake of InAs, Cd, MeHg, and Pb was 4.14 ± 1.91, 6.27 ± 2.93, 5.00 ± 2.63, and 3.32 ± 0.93 µg/day, respectively. Regarding POPs, the total dietary intake of PCDD/Fs, DL-PCBs, and NDL-PCBs were 9.93 ± 2.82 pg TEQ/day, 18.39 ± 9.99 pg TEQ/day, and 181.00 ± 90.40 ng TEQ/day, accordingly. Among all pollutants studied, dietary intake of As exceeded its respective EFSA tolerable daily intake level, and the total dietary intake of DL-PCBs almost reached its respective EFSA tolerable daily dietary intake level.

3.3. Contribution of each foodstuff to daily dietary pollutants intake

The percentage of pollutants in daily foodstuff among pregnant women were illustrated in Fig. 1. Specifically, dietary intake of As was mainly due to the consumption of white fish (71.27%), blue fish (17.15%), and seafood (9.75%). Cereal and tubers (70.16%) were the mainly source of InAs. Seafood (47.51%) and cereal and tubers (25.94%) mostly contributed to dietary intake of Cd. MeHg was mainly consumed through blue fish (52.88%) and white fish (27.27%). Pb was remarkable in seafood (13.21%) and cereal and tubers (30.15%). On the other hand, milk (19.57%), blue fish (13.14%), and eggs (11.65%) were the main sources of PCDD/Fs ingestion. DL-PCBs was mainly ingested from blue fish (59.74%), white fish (13.88%), and seafood (7.84%). Regarding NDL-PCBs, blue fish (44.58%) and white fish (32.33%) were main contributors to total daily intake.

3.4. Related risk factors for elevated maternal dietary pollutants intake

Multivariate-adjusted analysis was conducted to elucidate factors associated to elevated (≥75th percentile) pollutants dietary intake. In this analysis, women with age ≥30 years had more risk of elevated dietary intake of As (odd ratio (OR) (95% confidence interval (CI)): 1.48 (1.02, 2.14)). Similarly, alcohol consumption (OR (95% CI): 1.93 (1.09, 3.41)) and smoking habits (OR (95% CI): 1.59 (1.09, 2.24)) were associated to a higher risk of elevated Pb dietary intake. In the contrary, women with high educational level had decreased risk of elevated dietary intake of InAs (OR (95% CI): 0.61 (0.40, 0.92)), Cd (OR (95% CI): 0.63 (0.41, 0.95)), and DL-PCBs (OR (95% CI): 0.65 (0.43, 0.98));

Table 2
Dietary pollutants intake compared to European reference values (EFSA) during pregnancy.

Pollutants Food	As µg/day	InAs µg/day	Cd µg/day	MeHg µg/day	Pb µg/day	PCDD/Fs pg TEQ/day	DL-PCBs pg TEQ/day	NDL-PCBs ng TEQ/day
Milk	0.29 ± 0.15	0.29 ± 0.15	0.29 ± 0.15	0.29 ± 0.15	0.29 ± 0.15	1.94 ± 1,00	1,44 ± 0,73	10.13 ± 4,83
Cheese	0.01 ± 0.01	0.03 ± 0.02	0.03 ± 0.02	0.01 ± 0.01	0.10 ± 0.07	0.57 ± 0.42	0.51 ± 0.38	2.61 ± 1.92
Red meat	0.05 ± 0.03	0.05 ± 0.03	0.03 ± 0.02	0.03 ± 0.02	0.13 ± 0.08	0.16 ± 0.10	0.19 ± 0.11	1.59 ± 0.98
White meat	0.04 ± 0.02	0.04 ± 0.02	0.04 ± 0.02	0.04 ± 0.02	0.08 ± 0.04	0.24 ± 0.12	0.04 ± 0.02	1.85 ± 0.94
Processes meat	0.11 ± 0.05	0.12 ± 0.06	0.04 ± 0.02	0.04 ± 0.02	0.22 ± 0.11	0.39 ± 0.19	0.14 ± 0.07	3.06 ± 1.55
White fish	204.21 ± 142.01	0.02 ± 0.02	0.02 ± 0.02	1.36 ± 0.95	0.22 ± 0.15	1.14 ± 0.80	2.55 ± 1.77	58.51 ± 40.69
Blue fish	49.15 ± 39.25	0.02 ± 0.01	0.20 ± 0.16	2.64 ± 2.11	0.02 ± 0.01	1.30 ± 1.04	10.98 ± 8.77	80.68 ± 64.44
Seafood	27.94 ± 24.66	0.01 ± 0.01	2.98 ± 2.63	0.15 ± 0.13	0.44 ± 0.39	0.72 ± 0.64	1.44 ± 1.27	12.99 ± 11.47
eggs	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	1.16 ± 0.64	0.17 ± 0.09	0.95 ± 0.53
Sweet cereal	0.21 ± 0.16	0.13 ± 0.10	0.46 ± 0.35	0.03 ± 0.02	0.35 ± 0.27	0.25 ± 0.20	0.19 ± 0.15	0.69 ± 0.59
Salty cereal and tubers	3.95 ± 2.60	2.90 ± 1.80	1.63 ± 0.59	0.12 ± 0.04	1.00 ± 0.37	0.61 ± 0.21	0.21 ± 0.07	2.36 ± 1.04
Fruits	0.35 ± 0.21	0.35 ± 0.21	0.17 ± 0.10	0.17 ± 0.10	0.17 ± 0.10	0.87 ± 0.52	0.35 ± 0.21	2.80 ± 1.66
Vegetable	0.14 ± 0.07	0.14 ± 0.07	0.36 ± 0.18	0.08 ± 0.04	0.18 ± 0.08	0.45 ± 0.23	0.08 ± 0.04	1.07 ± 0.51
Pulses	0.05 ± 0.03	0.03 ± 0.02	0.02 ± 0.01	0.02 ± 0.01	0.11 ± 0.07	0.06 ± 0.04	0.03 ± 0.02	0.42 ± 0.27
Nuts	ND	ND	ND	ND	ND	0.06 ± 0.06	0.08 ± 0.09	1.28 ± 1.42
Total contaminant intake	**286.51 ± 165.40**	4.14 ± 1.91	6.27 ± 2.93	5.00 ± 2.63	3.32 ± 0.93	9.93 ± 2.82	**18.39 ± 9.99**	181.00 ± 90.40
Reference values EFSA (TWI/TDI)	15 µg/kg bw/week (E. P. on Contaminants in the Food Chain (CONTAM), 2009)	0.3 µg/kg bw/day (Arcella et al., 2021)	2.5 µg/kg bw/week (E. F. S. A. (EFSA), 2009)	1.3 µg/kg bw/week (Panel and Chain, 2012b)	0.50 µg/kg bw/day (E. P. on Contaminants in the Food Chain (CONTAM), 2010)	2 pg/kg bw/week (Knutsen et al., 2018)*		10 ng/kg bw/day (JECFA, 2016)
Comparable EFSA value in our population[#]	140.59 µg/day	19.71 µg/day	23.65 µg/day	12.22 µg/day	32.85 µg/day	18.77 pg TEQ/day		657 ng/day

Values are expressed in means ± SD (standard deviation); ND: not detected; TWI, tolerable weekly intake; TDI, tolerable daily intake; As, arsenic; InAs, inorganic arsenic; Cd, cadmium; MeHg, methylmercury; Pb, lead; PCDD/Fs, polychlorinated dibenzo-p-dioxins and dibenzofurans; DL-PCBs, dioxin-like polychlorinated biphenyls; NDL-PCBs, non-dioxin-like polychlorinated biphenyls.

The significance of the numbers in bold is that the dietary pollutants intake either exceeds or almost reaches the EFSA safety value.

* 2 pg/kg bw/week is the tolerable weekly intake for both PCDD/Fs and DL-PCBs.

** = TWI/7 × average weight (65.7 kg) or TDI × average weight (65.7 kg).

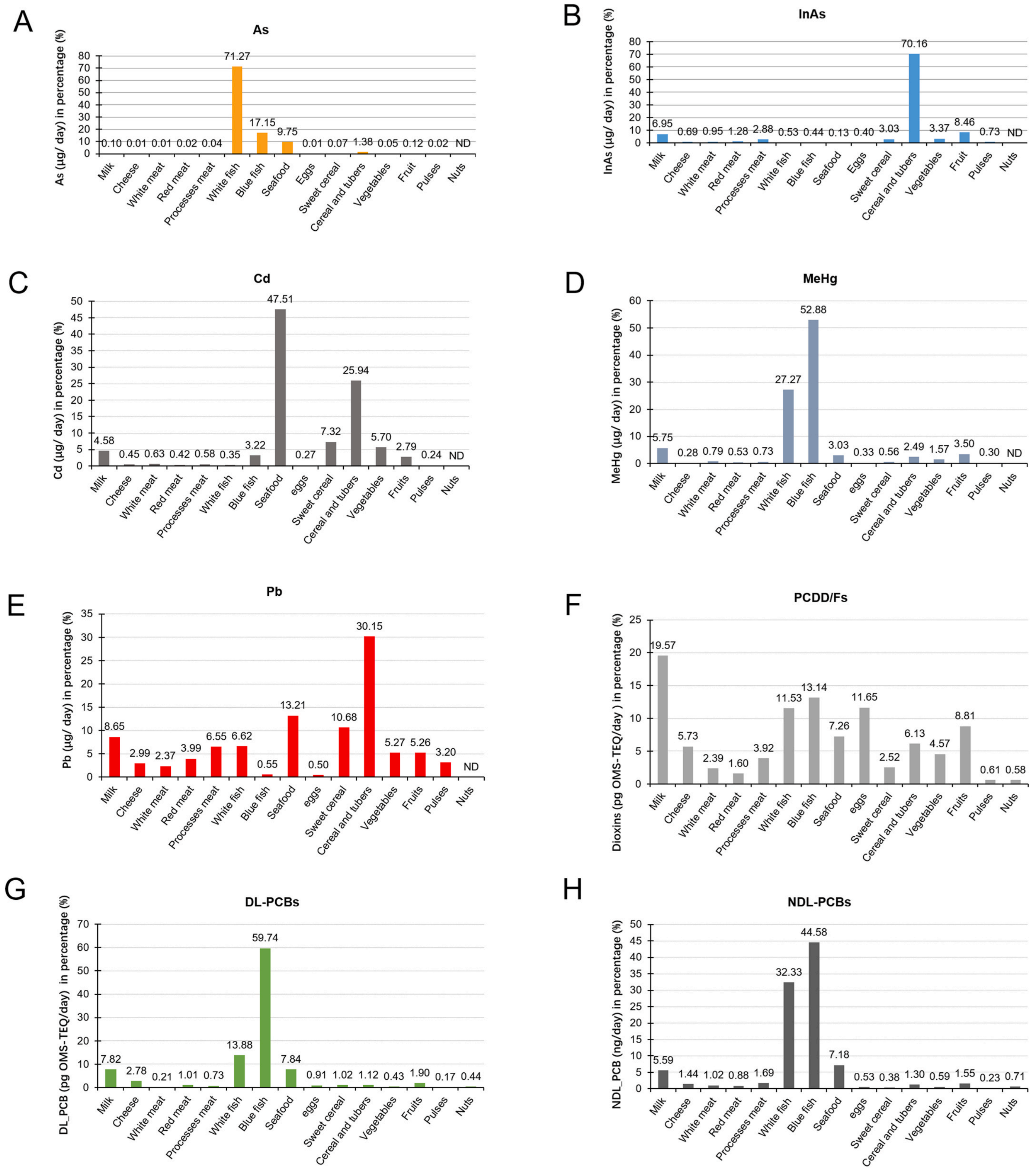

Fig. 1. Food pollutants in different daily foodstuffs among pregnant women
Percentage of As (Fig. 1A), InAs (Fig. 1B), Cd (Fig. 1C), MeHg (Fig. 1D), Pb (Fig. 1E), PCDD/Fs (Fig. 1F), DL-PCBs (Fig. 1G), and NDL-PCBs (Fig. 1H) in daily foodstuff among pregnant women.
As, arsenic; InAs, inorganic arsenic; Cd, cadmium; MeHg, methylmercury; Pb, lead; PCDD/Fs, polychlorinated dibenzo-p-dioxins and dibenzofurans; DL-PCBs, dioxin-like polychlorinated biphenyls; NDL-PCBs, non-dioxin-like polychlorinated biphenyls.

meanwhile, current/former smoker had decreased risk of elevated dietary intake of InAs (OR (95% CI): 0.68 (0.46, 0.99)) (Table 3).

In addition, maternal dietary intake during pregnancy according to above-mentioned related factors (age categories, educational level, alcohol consumption, and smoking status) in pregnant women was presented in Table 4.

4. Discussion

This study explored the dietary pollutants intake including five metals and metalloids spices and three POPs families, as well as the related factors for high dietary pollutants intake in pregnant women. To the best of our knowledge, this was the first large-scale study on this topic being conducted on a cohort of pregnant women in an industrial area in the province of Tarragona, Spain. The present study described the daily dietary pollutants intake, and evidenced that two of them, namely As and DL-PCBs, exceeded and almost reached the safety threshold recommended by EFSA respectively, mainly due to the consumption of fish and seafood. For other pollutants, blue fish (MeHg, DL-PCBs), seafood (Cd), cereals and tubers (InAs, Cd, Pb) or milk (PCDD/Fs) were foods categories that most contributed to daily dietary pollutants intake. Besides, multivariate-adjusted analysis showed that older age, alcohol and smoking consumption were independently associated with high pollutants intake, while higher educational level was independently linked to low pollutants intake.

To date, adverse impact of environmental pollutants on the health of pregnant women and their offspring is well-documented (Bank-Nielsen et al., 2019; Lecorguillé et al., 2022). In the ECLIPSES cohort, we also have evaluated the impact of airborne pollution, including particulate matter, nitrogen dioxide, and ozone, on motor function, and the results suggest a correlation between exposure during pregnancy and lower motor function in their children at around 40 days of age (Iglesias-Vázquez et al., 2022). Since most studies have analyzed the pollutants in blood and urine samples among pregnant women, the evaluation of the dietary pollutants intake is a complementary information with potential for community policies (Bocca et al., 2019, 2020). In the current study, the dietary intake of As (286.51 μg/day) exceeded the EFSA safety threshold (set at 140.59 μg/day) and was also higher

Table 3
Multivariate-adjusted odds ratios of elevated (≥75th percentile) maternal dietary pollutants intake during pregnancy according to selected lifestyle factors in pregnant women form ECLIPSES study (n = 701).

	Elevated (≥75th percentile) maternal dietary intake of environmental contaminants							
	As		InAs		Cd		Pb	
Characteristic	n (%)	OR (95% CI)	n (%)	OR (95% CI)	n (%)	OR (95% CI)	n (%)	OR (95% CI)
Age categories (years)								
<30	62 (22)	1 Reference	74 (26)	1 Reference	72 (25)	1 Reference	80 (28)	1 Reference
≥30	114 (28)	**1.48 (1.02, 2.14)***	102 (25)	1.04 (0.72, 1.49)	104 (25)	1.09 (0.76, 1.57)	96 (23)	0.81 (0.56, 1.16)
BMI categories (kg/m^2)								
<25	103 (25)	1 Reference	103 (25)	1 Reference	101 (25)	1 Reference	110 (27)	1 Reference
≥25	73 (25)	0.93 (0.65, 1.32)	73 (25)	0.92 (0.64, 1.31)	75 (26)	1.02 (0.71, 1.45)	66 (23)	0.80 (0.56, 1.15)
Educational level								
Low/medium	12 9 (26)	1 Reference	136 (27)	1 Reference	137 (28)	1 Reference	133 (27)	1 Reference
High	47 (23)	0.74 (0.50, 1.11)	40 (20)	**0.61 (0.40, 0.92)***	39 (19)	**0.63 (0.41, 0.95)***	43 (21)	0.78 (0.52, 1.18)
PA (METs-min/week)								
T1/T2	137 (25)	1 Reference	136 (25)	1 Reference	144 (27)	1 Reference	137 (25)	1 Reference
T3	39 (24)	0.95 (0.63, 1.44)	40 (25)	0.98 (0.65, 1.48)	32 (20)	0.66 (0.43, 1.03)	39 (24)	0.86 (0.57, 1.30)
Alcohol consumption								
No	162 (25)	1 Reference	160 (25)	1 Reference	160 (25)	1 Reference	154 (24)	1 Reference
Yes	14 (24)	0.89 (0.47, 1.68)	16 (27)	1.15 (0.62, 2.12)	16 (27)	1.12 (0.61, 2.07)	22 (37)	**1.93 (1.09, 3.41)***
Smoking status								
Never smoker	125 (26)	1 Reference	129 (27)	1 Reference	109 (23)	1 Reference	105 (22)	1 Reference
Current/former smoker	51 (23)	0.81 (0.56, 1.19)	47 (21)	**0.68 (0.46, 0.99)***	67 (30)	1.41 (0.98, 2.02)	71 (32)	**1.59 (1.09, 2.24)***
Characteristic	**MeHg n (%)**	**OR (95% CI)**	**PCDD/Fs n (%)**	**OR (95% CI)**	**DL-PCBs n (%)**	**OR (95% CI)**	**NDL-PCBs n (%)**	**OR (95% CI)**
Age categories (years)								
<30	65 (23)	1 Reference	64 (22)	1 Reference	67 (23)	1 Reference	66 (23)	1 Reference
≥30	111 (27)	1.33 (0.92, 1.92)	112 (27)	1.34 (0.93, 1.94)	109 (26)	1.28 (0.88, 1.84)	110 (27)	1.26 (0880, 1.82)
BMI categories (kg/m^2)								
<25	93 (23)	1 Reference	97 (24)	1 Reference	92 (22)	1 Reference	93 (23)	1 Reference
≥25	83 (29)	1.32 (0.93, 1.88)	79 (27)	1.18 (0.82, 1.68)	84 (29)	1.34 (0.94, 1.90)	83 (29)	1.31 (0.92, 1.86)
Educational level								
Low/medium	131 (26)	1 Reference	128 (26)	1 Reference	135 (27)	1 Reference	130 (26)	1 Reference
High	45 (22)	0.75 (0.50, 1.12)	48 (23)	0.84 (0.56, 1.24)	41 (20)	**0.65 (0.43, 0.98)***	46 (22)	0.79 (0.53, 1.18)
PA (METs-min/week)								
T1/T2	132 (25)	1 Reference	132 (25)	1 Reference	134 (25)	1 Reference	134 (25)	1 Reference
T3	44 (27)	1.21 (0.81, 1.80)	44 (27)	1.19 (0.79, 1.76)	42 (26)	1.11 (0.74, 1.67)	42 (26)	1.10 (0.73, 1.65)
Alcohol consumption								
No	163 (25)	1 Reference	161 (25)	1 Reference	163 (25)	1 Reference	162 (25)	1 Reference
Yes	13 (22)	0.78 (0.41, 1.48)	15 (25)	0.96 (0.52, 1.77)	13 (22)	0.79 (0.41, 1.51)	14 (24)	0.88 (0.47, 1.67)
Smoking status								
Never smoker	119 (25)	1 Reference	118 (25)	1 Reference	120 (25)	1 Reference	123 (26)	1 Reference
Current/former smoker	57 (25)	1.03 (0.71, 1.50)	58 (26)	1.06 (0.74, 1.54)	56 (25)	0.99 (0.68, 1.43)	53 (24)	0.90 (0.62, 1.31)

Logistic regression models were used to calculate the OR and 95% confidence interval (95% CI). The models were mutually adjusted for age categories (<30 (ref.), ≥30 years), early pregnancy BMI categories (<25 (ref.), ≥25 kg/m^2), educational level (low/medium (primary or secondary) (ref.), high (university students), physical activity tertile (T1 and T2:<3324 (ref.), T3: ≥3324 METs-min/week), alcohol consumption (no (ref.), yes), and smoking status (never smoker (ref.), current/former smoker). The significance of the numbers in bold as p-value <0.05. *p value < 0.05 compared with the reference category. Abbreviations: BMI, body mass index; PA, physical activity; As, arsenic; InAs, inorganic arsenic; Cd, cadmium; MeHg, methylmercury; Pb, lead; PCDD/Fs, polychlorinated dibenzo-p-dioxins and dibenzofurans; DL-PCBs, dioxin-like polychlorinated biphenyls; NDL-PCBs, non-dioxin-like polychlorinated biphenyls.

Table 4

Maternal dietary intake during pregnancy according to lifestyle factors in pregnant women form ECLIPSES study (n = 701).

	Age categories			Educational level			Alcohol consumption			Smoking status		
dietary intake	<30 years	≥30 years	*p*-value	Low/ medium	High	*p*-value	No	Yes	*p*-value	Never smoker	Current/ former smoker	*p*-value
	Means ± SD	means ± SD		means ± SD	means ± SD		means ± SD	means ± SD		means ± SD	means ± SD	
Milk	280.2 ± 144.9	291.0 ± 147.0	0.336	289.9 ± 153.8	278.6 ± 125.5	0.352	289.0 ± 148.0	262.8 ± 123.0	0.188	275.2 ± 131.2	310.9 ± 171.4	**0.002**
Cheese	11.7 ± 9.3	15.9 ± 10.6	**<0.001**	13.2 ± 10.0	16.4 ± 10.6	**<0.001**	13.9 ± 10.1	17.2 ± 11.3	**0.008**	13.8 ± 10.1	14.9 ± 10.6	0.202
Red meat	29.5 ± 18.3	25 ± 14.5	**<0.001**	28.0 ± 16.9	23.9 ± 14.2	**0.002**	26.9 ± 16.4	26.8 ± 15.8	0.489	26.0 ± 15.9	28.7 ± 17.0	**0.036**
White meat	40.4 ± 20.0	38.5 ± 19.1	0.193	39.0 ± 19.2	39.9 ± 20.3	0.571	39.4 ± 19.6	38.6 ± 19.1	0.767	39.2 ± 19.6	39.5 ± 19.5	0.822
Processes meat	38.4 ± 20.0	34.7 ± 16.2	**0.007**	36.9 ± 18.5	34.8 ± 16.3	0.163	36.0 ± 18.0	39.9 ± 17.2	0.115	34.8 ± 17.8	39.4 ± 17.7	**0.001**
White fish	19.5 ± 14.2	22.3 ± 14.4	**0.009**	20.7 ± 15.0	22.4 ± 12.7	0.146	21.3 ± 14.2	21.4 ± 14.7	0.925	21.9 ± 15.1	19.6 ± 12.6	0.052
Blue fish	17.7 ± 14.3	20.2 ± 14.5	**0.028**	19.5 ± 15.5	18.2 ± 11.4	0.277	19.4 ± 14.6	18.3 ± 12.2	0.589	19.5 ± 14.7	18.5 ± 14.0	0.413
Seafood	5.2 ± 5.1	5.5 ± 4.6	0.452	5.6 ± 5.1	5.0 ± 3.9	0.173	5.3 ± 4.7	6.8 ± 5.8	**0.019**	5.2 ± 4.8	6.0 ± 4.7	**0.041**
eggs	16.8 ± 10.1	17 ± 8.6	0.801	16.9 ± 9.6	16.8 ± 8.2	0.853	16.9 ± 9.3	15.7 ± 7.3	0.352	17.3 ± 9.0	16.0 ± 9.6	0.069
Sweet cereal	31.0 ± 25.0	25.6 ± 19.9	**0.002**	29.7 ± 23.2	23.4 ± 19.2	**<0.001**	27.9 ± 22.7	28.4 ± 18.0	0.862	27.0 ± 22.7	29.5 ± 21.2	0.167
Salty cereal and tubers	128.2 ± 44.7	120.8 ± 41.9	**0.027**	125.8 ± 44.2	118.9 ± 40.3	**0.048**	123.7 ± 43.6	123.8 ± 40.9	0.988	124.5 ± 43.9	122.4 ± 41.8	0.554
Fruits	163.7 ± 102.3	182.4 ± 104.8	**0.019**	172.2 ± 108.1	180.9 ± 93.8	0.320	173.2 ± 105.8	186.8 ± 86.7	0.340	183.0 ± 103.7	157.3 ± 103.2	**0.002**
Vegetable	69.1 ± 36.2	84 ± 38.8	**<0.001**	73.2 ± 37.0	89.4 ± 39.5	**<0.001**	76.8 ± 38.2	88.5 ± 38.8	**0.025**	77.6 ± 39.2	78.7 ± 36.7	0.714
Pulses	15.0 ± 9.8	15 ± 8.4	0.910	15.2 ± 9.4	14.5 ± 7.9	0.329	14.9 ± 8.9	15.0 ± 9.2	0.986	15.0 ± 9.1	15.0 ± 8.7	0.915
Nuts	2.4 ± 2.8	3.2 ± 3.4	**<0.001**	2.4 ± 2.8	3.9 ± 3.7	**<0.001**	2.8 ± 3.2	3.1 ± 3.1	0.482	2.9 ± 3.2	2.7 ± 3.1	0.357

Values are expressed in means ± SD (standard deviation). The significance of the numbers in bold as p-value <0.05.

than the average intake of the adult population in Catalonia, (98.2 μg/day) (González et al., 2019; on Contaminants in the Food, 2009), which indicated the presence of high levels of environmental pollution in the province of Tarragona. The food groups we identified as the major contributors to dietary As intake (fish, blue fish, and seafood) were similar to those previously found in other studies conducted in Catalonia (González et al., 2021; Martí-Cid et al., 2009). Furthermore, dietary intake of InAs (4.14 μg/day) among the studied population was lower than EFSA safety threshold level (19.71 μg/day) and also lower than that in French pregnant women (14.88 μg/day), while it was higher than that of the general population of Catalonia (2.58 μg/day); meanwhile, cereal and tubers as the main source of InAs were also in consist with local diet survey (González et al., 2019; (EFSA) et al., 2021; Chan-Hon-Tong et al., 2013). Regarding Cd, MeHg, and Pb, the dietary intake in our study was below the EFSA safety threshold and similar to the intake for the general population of Catalonia (González et al., 2019; (EFSA) and E.F.S.A, 2009; Panel and Chain, 2012a; on Contaminants in the Food, 2010); meanwhile, the consumption of MeHg in our population (5.00 μg/day) was higher than that in French pregnant women (1.19 μg/day) but lower than that in Spanish pregant women in Murcia religion (9.38 μg/day) (Chan-Hon-Tong et al., 2013; Ortega-García et al., 2009). As expected, fish and seafood were the main sources of Cd and MeHg, due to the higher consumption of fish and seafood as part of the Mediterranean Diet and the fact that these food groups contented high levels of these pollutants (Bosch et al., 2016). A lot of food categories contributed to dietary Pb intake, among which cereal and tubers were the most important contributors accounting for the total. Generally, food contributors fo dietary pollutants intake in the current study were similar to those for pollutants reported in blood or urine of pregnant women living in the same area (Bocca et al., 2020).

The emission of POPs in chemical complex areas sometimes is extremely high (González et al., 2018). It is worth noting that a number of toxic POPs in breast milk are detected and high concentration of several POPs in metarnal serum sample are observed among pregnant women living in Tarragona, which can directly trigger adverse effect to their offsprings and further bring a huge social burden (Rovira et al., 2022; Junqué et al., 2020). Although several diet surveys have shown that the dietary intake of PCDD/Fs among the general population in Tarragona presents a declining trend year by year, dietary intake of PCDD/Fs among pregnant women in the current study (9.93 pg TEQ/-day) was still higher than general population (8.54 pg TEQ/day) (González et al., 2018). This could be partly explained by that pregnant women might have more diverse dietary pattern compared to the general local population and a variety of foodstuffs were sources of PCDD/Fs exposure (above-mentioned). Even though the dietary intake of PCDD/Fs among the studied population was below the EFSA safety threshold (18.77 pg TEQ/day), they could accumulate in adipose tissues and become more concentrated over time (on Contaminants in the Food Chain (CONTAM) et al., 2018; Dioxins-and-Their--Effects-on-Human-Health). In our study, the main food contributors to PCDD/Fs intake were animal-based foods including milk, blue fish, white fish, and eggs, which was accorded with WHO published evidence (Dioxins-and-Their-Effects-on-Human-Health). Notably, the dietary intake of DL-PCBs in our research was almost reached EFSA safety threshold (DL-PCBs: 18.39 vs. 18.77 pg TEQ/day) (on Contaminants in the Food Chain (CONTAM) et al., 2018); meanwhile, dietary intake of NDL-PCBs among pregnant women in our study living in the industrial zone of Tarragona was also exceeded that in Norwegian pregnant women (2.75 vs.0.81 ng/kg bw/day) (Caspersen et al., 2013). The main food contributors for DL-PCBs intake were similar to those for NDL-PCBs intake in this study, including blue fish, followed by white fish, and then seafood. Fish and seafood could be a valuable part of a healthy diet due to their nutritional profile, while several guidelines recommend that pregnant women should consume them in moderation and limit their intake of large fish due to the presence of pollutants (Marushka et al., 2021; Advice-about-Eating-Fish); meanwhile, fish and seafood were also the main contributors to dietary pollutants intake based on the current study. Therefore, greater attention should be paid to fish and seafood intake during this critical stage of gestation.

Potential anthropometric and lifestyle factors associated with the higher dietary pollutants intake during pregnancy were also investigated in the current study. Firstly, age was positively related to higher As intake, which was consistent with previous findings in the literature (Sidhu et al., 2015). This could be explained by the fact that older pregnant women might be more likely to adhere to dietary advice. Fish intake during pregnancy is recommened due to its benefit for child's brain development, albeit with certain restrictions regarding fish type in guildlines (Advice-about-Eating-Fish). However, as mentioned before, fish was also the main contributor to dietary As intake. Secondly, higher education level was associated with lower dietary intakes of InAs, Cd, and DL-PCBs. This finding could be due to pregnant women with higher education levels were more aware about nutrition and health (such as information about safe food handling practices, food labeling, and government regulations). On the contrary, women with higher educational levels had lower consumption of cereals and tubers, the latter contained high levels of InAs and Cd pollutants. Thirdly, alcohol consumption and smoking were associated with increased dietary Pb intake, which was caused by that pregnant women with these habits consumed higher amounts of milk, cheese, red meat, processed meat, seafood, or vegetable, which were the main food contributors to Pb intake. The current situation of dietary pollutant intake during pregnancy necessitates the implementation of preventive strategies concerning both the identified risk factors of high pollutants intake and food items with high pollutants. Pollutants can infiltrate food through various routes: (1) air, soil, and water pollution: Human activities such as mining, industrial waste disposal, and the use of fertilizers and pesticides could increase the levels of these pollutants in the environment, leading to their accumulation in crops and water sources and ultimately consumed by humans or animals (Schleiffer and Speiser, 2022). (2) Food processing: Toxic metals could be present in food packaging materials, especially those that were made of metal or had a metal coating (Li et al., 2021).

The reasons for prioritizing metals, metalloids and POPs in this dietary intake assessment were as follows: 1. These substances had been studied extensively, and their potential risks to human health are better understood compared to emerging organic compounds (Culbreth and Aschner, 2019; Yang et al., 2020). 2. There was often more data available on the presence of these substances in food items and the environment compared to emerging organic compounds (Amutova et al., 2021; Malisch and Kotz, 2014). This availability of data made it more feasible to conduct comprehensive dietary intake assessments for these contaminants. 3. Many countries and international regulatory bodies had established permissible exposure limits and guidelines for these substances due to their known health risks ((EFSA) and E.F.S.A, 2005; on Contaminants in the Food, 2012). Monitoring and assessing dietary intake of these substances was crucial for ensuring compliance with regulatory standards and protecting public health. 4. Metals, metalloids, and POPs were pollutants longer monitored in the main foodstuff in the region where the cohorts were placed (González et al., 2019; (EFSA) and E.F.S.A, 2009; Panel and Chain, 2012a; on Contaminants in the Food, 2010). Our study had several strenghts: the large sample size of this study ensured statistical power and accounted for potential confounding factors; the use of pollutant datasets from local products provided persuasive information for the local population; the study covered a wide range of socioeconomic status representative of the population. Nevertheless, some limitations still could not be ignored. First, the accuracy of exposure assessments might be limited by the sensitivity and specificity of the methods used to measure pollutants in food. Second, assessing individual dietary intake relied on FFQ methods might be subject to recall bias and consequently affect the accuracy of data collection. Third, no correlation between age and the consumption of PCBs and PCDD/Fs was found among pregnant women, although those women over 30 years old also consumed more fish (the main food source of PCBs and PCDD/Fs), more studies were still needed to assess it. Fourth, the evaluation of the congener profile of PCBs and PCDD/Fs could be conducted in the further study.

In the current research, the majority of the daily dietary pollutant intake is below the safe levels established by EFSA. However, the dietary intake of As and DL-PCBs is concerning, as it exceeds or nearly reaches the EFSA safety threshold. This exceedance of the safety threshold, combined with cumulative nature of some pollutants, poses a risk to the health of pregnant women. Moreover, there is also concern regarding the potential impact on the developing fetus, which remains to be determined. Overall, these findings highlight that fish and seafood are major contributors to dietary pollutants intake, particularly for those pollutants that exceed or approach the EFSA safety threshold among pregnant women. This underscores the need for awareness of these food pollutants not only in our area but also in other highly industrialized areas and similar food consumption patterns.

Entities and participants in the ECLIPSES study

Research Group in Nutrition and Mental Health (NUTRISAM), Universitat Rovira i Virgili, Reus, Spain (Victoria Arija, Josefa Canals, Carmen Hernández-Martínez, Núria Voltas). Center for Environmental, Food and Toxicological Technology (TECNATOX). Universitat Rovira i Virgili, Reus, Spain (Monica Bulló, Joaquim Rovira Solano). Sexual and Reproductive Health Care Services (ASSIR) of Tarragona, Spain (Francesc Fargas, Francisca Ruiz, Gemma March, Susana Abajo) and the team of midwives who recruited the study participants (Susana Abajo, Irene Aguilar, Sònia Aguiles, Rosa Alzúria, Judit Bertrán, Carmen Burgos, Elisabet Bru, Montserrat Carreras, Beatriz Fernández, Carme Fonollosa, María Leiva, Gemma March, Demetria Patricio, Teresa Pinto, María Ramírez, Eusebia Romano, Inés Sombreo). Research Support Unit-Tarragona (Josep Basora, Meritxell Pallejà) and Central Unit-Barcelona (Rosa Morros, Helena Pere, Anna García Sangenís) from the Jordi Gol University Institute for Primary Care Research, Institut Català de la Salut.

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Ethics approval

The study was designed in agreement with the Declaration of Helsinki/Tokyo. All procedures involving human subjects were approved by Clinical Research Ethics Committee of the Jordi Gol University Institute for Primary Care Research [Institut d' Investigació en Atenció Primària; IDIAP], the Pere Virgili Health Research Institute [Institut d'Investigació Sanitària Pere Virgili; IISPV] and of the Spanish Agency for Medicines and Medical Devices [Agencia Española del Medicamento y Productos Sanitarios; AEMPS]. Signed informed consent was obtained from all women participating in the study.

Author contribution

Xiruo Kou: Formal analysis, Investigation, Writing-Original Draft, Writing-Review&Editing. Monica Bulló Conceptualization, Methodology, Investigation. Joaquim Rovira Solano Conceptualization, Methodology, Investigation, Supervision. Andrés Díaz: Formal analysis, Investigation, Supervision. Victoria Arija: Data curation, Conceptualization, Funding, Formal analysis, investigation, Project administration, Resources. All author: writing review & editing.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2023.140319.

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