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Original Research Article

Occurrence and dietary intake of food processing contaminants (FPCs) in Catalonia, Spain

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ABSTRACT

Food processing contaminants (FPCs) form a wide group of chemicals that are generated during different cooking practices. This study was aimed at determining the levels of a number of FPCs (i.e., acrylamide, furans, monochloropropanediols (MCPDs) and glycidyl esters (GEs)) in foodstuffs purchased in Catalonia (Spain), and assessing the human exposure for different population groups. The dietary intake of acrylamide for the adult population was estimated in 2.91 μg/day, while that of furan, 2-methylfurans and 3-methylfurans was 4.32, 7.35 and 0.439 μg/day, respectively. Finally, the exposure to 3-MCPD, 2-MCPD, 3-MCPD esters, 2-MCPD esters and GEs through food consumption was estimated in 0.657, 0.529, 10.7, 5.15 and 8.81 μg/day, respectively. The risk assessment showed that there is a health concern for developing neoplastic effects derived from the intake of acrylamide for all the population groups. In addition, toddlers and infants would exceed the threshold values of 3-MCDP and GEs. The global analysis of these results indicates that a special attention should be paid to the youngest population groups in Catalonia, reinforcing the need of conducting periodical monitoring studies and developing policy measures, especially focused on foodstuffs highly consumed by toddlers, infants and children.

1. Introduction

Food processing contaminants (FPCs) include a wide range of compounds that can be formed during cooking practices (Mogol and Gökmen, 2016). Heating treatment is by far the most widely method for food process in the industry or at home, covering up to 80–90 % of the consumed food (Nerín et al., 2016). This group of chemicals include a variety of toxicants, such as acrylamide, furans and monochloropropanediols (MCPDs), with different physico-chemical and toxicological properties. Their consumption has been also linked to adverse health effects (Kettlitz et al., 2019), although accurate exposure estimates for FPCs may be particularly challenging (Rietjens et al., 2018).

Acrylamide is formed during heating processes above 120 °C, when free asparagines react with reducing sugars, such as glucose and fructose, according to the Maillard reaction (Mesías and Morales, 2016; Abt et al., 2019; Rifai and Saleh, 2020). Acrylamide has been found to be contained in a wide variety of starch-based foods, including potatoes, breads or biscuits, among others (Tareke et al., 2002; Deribew and Woldegiorgis, 2021). The toxicological effects of acrylamide include neurotoxicity, genotoxicity and reproductive toxicity (EFSA, 2015). Therefore, the International Agency for Research on Cancer (IARC) classified acrylamide as a probably carcinogenic to humans (Group 2A) (IARC, 1994).

MCPDs are formed by the reaction of lipids and chlorines at high temperatures, mainly during the deodorization process (Nguyen and Fromberg, 2020). In the late 1970s, 2- and 3-MCPDs were identified in the composition of hydrolyzed vegetable protein (HVP), which is used as a flavoring condiment (EFSA, 2016). However, the presence of MCPDs in foodstuffs was firstly observed in 2004 (Svejkovská et al., 2004), while their occurrence in edible oils and soy/oyster sauces was reported shortly after (Wong et al., 2006; Zelinková et al., 2006). In turn, as the presence of chlorinated compounds is not required, glycidol can be also formed by different routes, but under the same conditions as MCPDs (FAO/WHO, 2017; Nguyen and Fromberg, 2020). The IARC classified 3-MCPD as possibly carcinogenic and GEs as probably carcinogenic to

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humans (Group 2B and 2A, respectively) (IARC, 2013, 2000).

Furan and methylfurans are lipophilic oxygen heterocycles of high volatility (Kettlitz et al., 2019). They are formed by different pathways, such as degradation of carbohydrates, ascorbic acid and its derivatives and thermal oxidation of lipids (Frank et al., 2020). They are detected in a wide range of food products including coffee or canned and jarred foods, as well as baby food containing meat, vegetables, breakfast cereals or cookies (EFSA, 2017). Based on the reported adverse health effects derived from the exposure to furans, the IARC classified them as possibly carcinogenic to humans (Group 2B) (IARC, 1995).

The aim of this study was to determine the levels of several FPCs in food products from the Catalan (Spain) market, where these data are currently inexistent. In addition, health risks derived from the consumption of FPC-containing foodstuffs were assessed for various population groups. The dietary intakes were compared with toxicological endpoints in order to establish the potential health risks for the consumers.

2. Materials and methods

2.1. Sampling

Sampling was carried out in three different periods: July 2015, September 2016 and September 2019, according to the surveillance program established by the Catalan Food Safety Agency (ACSA). Details on the food products purchased in each campaign are given in Table 1. In order to get a good representativeness, samples were bought at different markets, supermarkets and grocery stores across Catalonia.

Food products were selected as those containing high levels of the contaminants assessed in the present study, according to the scientific literature (EFSA, 2017, 2016, 2015). Acrylamide was analyzed in potato chips, potato snacks, sliced bread, toasts and crackers, white bread, cookies, margarine, roasted coffee, instant coffee, baby food, cereal-based baby food and breakfast cereals (wheat-based and others). For MCPDs, their content was determined in potato chips, potato snacks, sliced bread, toasts and crackers, white bread, cookies, margarine, sunflower oil, pomace oil, refined olive oil, infant formula, follow-on formulae, cereals and milk, cereal-based baby food, mayonnaise, soy sauce, salad sauce, bakery products, chocolate spreads, palm oil cookies and frying oil. Finally, furan and methylfurans was determined in ground coffee, coffee beans, instant coffee, coffee substitutes, fruit juices, breakfast cereals, baby food and cereal-based baby food.

Table 1
Food products purchased in the three different sampling periods.

Food product	July 2015 (n = 264)	September 2016 (n = 336)	September 2019 (n = 120)
Potato chips	X		
Potato snacks	X		
Sliced bread	X		
Toasts and crackers	X		
White bread	X		
Cookies	X		X
Margarine	X		
Wheat-based breakfast cereals	X		
Other breakfast cereals	X		X
Roasted coffee	X		
Instant coffee	X		X
Sunflower oil		X	
Pomace oil		X	
Refined olive oil		X	
Infant formula		X	
Follow-on formulae		X	
Cereals and milk		X	
Cereal-based baby food		X	X
Mayonnaise		X	
Soy sauce		X	
Salad sauce		X	
Bakery products (milk bread, puff pastry and muffins)		X	
Chocolate spreads		X	
Palm oil cookies		X	
Frying oil		X	
Ground coffee			X
Coffee beans			X
Coffee substitutes			X
Fruit juices			X
Baby food			X

For determining acrylamide and MCPDs, three composites containing 8 individual units were prepared for each food group. Since furan and methylfurans are very volatile compounds, instead of composites, 8 units of each foodstuff were directly analyzed. Only edible parts of the foods were included. Samples were homogenized and stored at -20 °C until analysis.

2.2. Analytical determination

The analysis of acrylamide was performed after a water extraction and a clean-up using two types of cartridges. One g of each food was extracted with 10 mL of purified water. Samples were subjected for 30 s to vortex, 45 min to orbital shaking and finally 20 min to centrifugation at 12,000 rpm and 10 °C. Two mL of the supernatant were then passed through an Oasis HLB cartridge (Waters, Milford, MA, USA). After drying, 3 mL of methanol were used to elute the acrylamide, which was subsequently cleaned using an Oasis MCX cartridge (Waters, Milford, MA, USA). The elute was collected in a test tube for evaporation, softly dried under nitrogen at 50 °C for 40 min, and finally redissolved with 400 μL of water. The determination of acrylamide was performed by UHPLC coupled to a high-resolution mass spectrometer by using a HRMS-Orbitrap q-Exactive system (Thermofisher Scientific, Waltharm, MA, USA).

The limit of quantification (LOQ) was defined as the concentration of the lowest point of the calibration, being set at 10 μg/kg. A nine-level calibration curve was established by plotting the average response factor against concentration (μg/L) for each standard level. The recovery study for the method validation in the range 10.0−3000 μg/kg of acrylamide showed an accuracy of 101.7 %, with a precision, expressed as percentage of the relative standard deviation (%RSD), of 12.7 %, considering a sample size of N = 58. Quality control/quality assurance of the analytical procedure was ensured by analyzing spiked samples and participating in proficiency testing experiments.

The concentration of free MCPD was determined by gas chromatograph-mass spectrometry (GC–MS) using on a previous method developed by Wenzl et al. (2015). For the determination of 3-MCPD and 2-MCPD, samples were dissolved in n-hexane:acetone Sigma-Aldrich (St. Louis, MO, USA) (1:1 v/v) and spiked with a surrogate standard containing d_5-3-MCPD and d_5-2-MCPD (both from Sigma-Aldrich, St. Louis, MO, USA). The supernatant was collected and water was added, being vortex and the organic phase discarded. The aqueous phase was collected and completely evaporated under vacuum and nitrogen. The residue was dissolved in ethyl acetate and the analytes were derivatised with phenylboronic acid (Sigma-Aldrich, St. Louis, MO, USA). The extract was then evaporated under a nitrogen stream and reconstituted with isooctane. GC–MS was carried out on a 7890 A Agilent gas chromatograph equipped with a 5977B mass spectrometer (Agilent, Santa Clara, CA, USA). A HP 5 MS capillary column (Agilent) 30 m ×0.25 mm, 0.25 μm) was used for separation. Helium with a flow of 1.2 mL/min was used. GC oven temperature was programmed from an initial temperature of 60 °C (for 1 min), with 6 °C/min to 150 °C, and final step up to 300 °C. Characteristic fragment ions of m/z for 3-MCPD and 2-MCPD were 147 and 196, respectively. The quantitation was performed using the quantifier ions for the selected compounds. The ratio of the quantifier / qualifier was similar between standards and samples, with maximum ranges of 20 %. A six-level calibration curve was established by plotting the average response factor against concentration (μg/L) for each standard level. The slope and the intercept were calculated by linear regression for each analyte. The recovery study for the method validation between 10 and 100 μg/kg showed an accuracy of 91.5 % for 3-MCDP (N = 58) and 86.2 % for 2-MCPD (N = 65), while precision results were 12.6 % and 10.5 %, respectively.

Ester bound MCPD and glycidyl esters in food samples were analyzed by means of Gas Chromatography – Triple quadrupole Mass Spectrometry (GC–MS/MS). Fat from infant or nutritional formulas was extracted by liquid-liquid extraction using ethyl acetate. Fat from the rest of the samples was extracted by Accelerated Solvent Extraction (ASE) using tertbutylmethylether (Sigma-Aldrich, St. Louis, MO, USA). The extracted fat and oil samples were analyzed for bound MCPD and Glycidyl esters (GE) following the AOCS (2013) official method CD29a-13. GE were converted to 3-monobromopropanediol (3-MBPD) monoesters in an acid solution containing a bromide salt. 3-MBPD esters, together with 2- and 3-MCPD esters, were then converted into the free (nonesterified) form in an acid methanolic solution. The fatty acid methyl esters generated during the reaction were extracted from the sample, and 2-MCPD, 3-MCPD, and 3-MBPD were derivatized with phenylboronic acid (Sigma-Aldrich, St. Louis, MO, USA) prior to GC-tandem MS (GC–MS/MS) analysis. GC–MS/MS was carried out on a 7890B Agilent gas chromatograph equipped with a 7010B mass spectrometer triple quadrupole detector (Agilent, Santa Clara, CA, USA). A HP 5 MS capillary column (30 m ×0.25 mm, 0.25 μm) (Agilent, Santa Clara, CA, USA) was used for separation. Helium was used as carrier gas with a flow of 16 psi. GC oven temperature was programmed from an initial temperature of 60 °C (for 1 min), 10 °C/min up to 200 °C, and finally at 25 °C/min up to 250 °C. Quantitative analysis was carried out by monitoring the transitions specified in previous studies (Dubois et al., 2019; Wenzl et al., 2015).

The LOQ was set at 2.0 μg/kg for infant formula, and 50 μg/kg for the rest of food commodities. Quantification was performed using the quantifier ions for the selected compounds. A ten-level calibration curve was established by plotting the average response factor against concentration (μg/L) for each standard level. The slope and the intercept were calculated by linear regression for each analyte. The recovery study for the method validation was conducted in the range 2.0−35 μg/kg for infant formula, and between 50 and 1000 μg/kg for the rest of food commodities. Bound 3-MCPD, bound 2-MCPD and glycidil esters showed an accuracy of 97.8 % (N = 16), 96.3 % (N = 16) and 104.8 % (N = 16), respectively, being precision results 11.9 %, 10.3 % and 11.7 % respectively. Quality control/quality assurance of the analytical procedure was ensured by analyzing spiked samples and participating in proficiency testing experiments.

For the furan determination, food samples were analyzed using Head Space (HS) gas chromatography–mass spectrometry (GC–MS) with isotope dilution. The HS method applied was based on the FDA (2004) method. One-half gram of sample was weighted into a 20 mL HS vial, and 10 mL of a 30 % sodium chloride solution were added. For coffee, 0.25 g of sample and 10 mL of water were used. Fifty μL of a d_4-furan solution internal standard (Sigma-Aldrich, St. Louis, MO, USA) was used, being added to the HS vial before capping it. The extraction into the HS was done for 20 min at 50 °C before injecting it the the GC. The determination was done with an Agilent HeadSpace (7697A, Agilent), GC (Agilent 7890) and a MS (Agilent 5977) (Agilent, Santa Clara, CA, USA). The GC column used was a HP-Plot/Q (15 m x0.32 mm; id 20 μm film). Ions for furan (*m/z* 68 and 39) and methylfurans (*m/z* 82 and 53) were monitored, being target compounds quantified with external calibration curves.

The LOQ for baby foods and juices was determined at 5 μg/kg, while that for the remaining foodstuffs was 20 μg/kg. A seven-level calibration curve was established by plotting the average response factor against concentration (in ng per vial) for each standard level. The slope and the intercept were calculated by linear regression for each analyte. The recovery study for the method validation of furan, 2-methylfuran and 3-methylfuan showed an accuracy of 92.4 %, 88.9 % and 92.6 %, respectively, while precision results were 9.3 %, 8.2 % and 15 %, respectively.

2.3. Dietary exposure

Consumption data for the analyzed foodstuffs were obtained from ENALIA and ENALIA2, which are the most recent consumption surveys performed in Spain. ENALIA covers data for infants and adolescents, while ENALIA2 includes data for the adult population and pregnant women (AECOSAN, 2016a, b). These surveys were performed by following the recommendations guide developed by EFSA (2009). They are consumption data studies representative of different Spanish subpopulations, which are based on a random selection of 1862 children and adolescents, 623 adults, 310 seniors, and 157 pregnant women. The method included a 24-h recall in two non-consecutive days (at least 14 days in between) and a food frequency questionnaire. For coffee, a special assumption was made, taking into consideration that three different types of coffee (roasted, ground and beans) were analyzed but only consumption data on "liquid coffee" is available. Therefore, the consumption of each type of coffee was assumed as 1/3 of the total for the "liquid coffee".

The population under study was divided into 7 groups according to age: toddlers (6–11 months), infants (1–3 years), children (3–9 years), adolescents (10–17 years), young adults (18−39 years), adults (40−64 years), seniors (65−74 years). The age bands were selected according to the ENALIA surveys (AECOSAN, 2016a, b). The total exposure to acrylamide, free- and ester-bound-MCPDs, GEs and furan and methylfurans was calculated by multiplying the concentration of each chemical in food and the average consumption of that food. The total dietary intake of each compound was finally obtained by summing the respective intakes from each food group.

Finally, the intake was also estimated according to the respective average body weight (bw) of each population group, whose values were gathered from the scientific literature (Carrascosa et al., 2010; López-Sobaler et al., 2016; Martínez et al., 2017; WHO, 2003). For calculation purposes, mean body weights of 8.4, 12.3, 24, 51, 72, 77, 70.5 and 65 kg were used for toddlers, infants, children, adolescents, adults aged 18−39, adults aged 40−64, seniors, and pregnant women, respectively. When estimating the dietary intake, for those chemicals with levels under their respective limit of detection (LOD), their concentration was assumed equal to one-half of the LOD (medium-bound; MB). The dietary exposure was also calculated under 2 additional scenarios: upper-bound (UB) and lower-bound (LB). To estimate the UB and LB exposures, the values corresponding to the highest and lowest concentrations were used, respectively. When a chemical was under their respective LOD, their concentration was assumed as the LOD, under the UB scenario, or 0, under the LB scenario. Furthermore, for the exposure assessment of acrylamide, furan and methylfurans, specific dilution and loss factors were applied to coffee (Becalski et al., 2016). For acrylamide, a dilution factor of 19.6 was considered because non-brewed coffee was analyzed. In turn, for furan the following reduction (dilution + loss) factors, related to coffee brewing, were applied: 26.8 for furan, 47 for 2-methylfuran and 46 for 3-methylfuran.

2.4. Risk characterization

Two different approaches for the risk characterization were used in the present study. The first one consisted of the comparison of the estimated daily intake (EDI) with the tolerable daily intake (TDI). However, this approach is not valid for carcinogenic substances because there cannot be threshold values associated to cancer effects. In that case, the margin of exposure (MOE) approach was used by applying this equation:

$$MOE = \frac{\mathrm{BMDL10\,or\,T25}}{EDI}$$

Where BMDL_{10} is the 95 % lower confidence limit for this benchmark dose for 10 % extra risk of carcinogenic effects, T25 is the dose corresponding to a 25 % incidence of tumors, and EDI is the estimated daily intake.

3. Results and discussion

3.1. *Concentrations of FPCs*

The mean concentrations of the nine food processing contaminants here analyzed in a number of foodstuffs are summarized in Table 2. Instant coffee showed the highest concentrations of acrylamide (594 μg/kg), followed by potato chips and potato snacks (435 and 243 μg/kg, respectively). Relatively high levels of acrylamide were also noted in cookies and roasted coffee (209 and 202 μg/kg, respectively). None of the foodstuffs exceeded the recommended limits established by the European Commission. (2013). The results found in this study are in accordance to those reported in the literature for a number of countries. For instant coffee, the levels observed in Catalonia are very similar to those reported by Arisseto et al. (2007) in Brazil (582 μg/kg), while values for roasted coffee are also similar to those previously found in the USA (272.4 μg/kg) (Abt et al., 2019). For potato chips, our results are also in the same order of magnitude than those observed in Colombia, USA, Brazil and North Macedonia (635, 498, 591 and 390 μg/kg, respectively) (Abt et al., 2019; Arisseto et al., 2007; Barón Cortés et al., 2021; Dimitrieska-Stojkovikj et al., 2019), but notably lower than values from Lebanon (Hariri et al., 2015). The food levels of acrylamide were also lower than those reported in a number of food products (e.g., salted snacks, biscuits and wafers, breakfast cereals, coffee and toasts and bread) purchased in Slovenia (Mencin et al., 2020). Similarly, mean levels in potato chips and snacks (range: 243−435 μg/kg) were also lower than (but of the same order of magnitude as) the values reported by Mesías et al. (2020) in Spain (664 μg/kg). In a previous study, these researchers found an average acrylamide content of 630 μg/kg in commercial potato crisps (Mesías and Morales, 2015). In addition,

Table 2
Concentrations of FPCs (μg/kg) in different foodstuffs purchased from Catalonia (Spain).

Food samples	Acrylamide	Furan	2-methylfurans	3-methylfurans	3-MCPD	2-MCPD	3-MCPD ester	2-MCPD ester	GE
Potato chips (n = 24)	**435 ± 110**	n.a.	n.a.	n.a.	<10	<10	**102 ± 40**	**50.7 ± 25**	**68 ± 5.3**
Potato snacks (n = 24)	**243 ± 140**	n.a.	n.a.	n.a.	<10	<10	**389 ± 190**	**152 ± 69**	**138 ± 44**
Sliced bread (n = 24)	**14.7 ± 4.2**	n.a.	n.a.	n.a.	**15.1 ± 1.5**	<10	**21 ± 9.2**	**5 ± 2**	**1 ± 1.7**
Toasts and crackers (n = 24)	**43.4 ± 17**	n.a.	n.a.	n.a.	**4.67 ± 0.58**	<10	**68.3 ± 37**	**24.3 ± 24**	**61.7 ± 31**
White bread (n = 24)	**9.6 ± 8**	n.a.	n.a.	n.a.	**4.87 ± 0.23**	<10	**0.67 ± 1.2**	**1 ± 1**	**3 ± 0**
Breakfast cereals (wheat) (n = 24)	**86 ± 21**	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Breakfast cereals (other) (n = 24)	**59.7 ± 6**	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Roasted coffee (n = 24)	**202 ± 76**	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Instant coffee*	**594 ± 160**	**380 ± 330**	**869 ± 640**	**62.1 ± 40**	n.a.	n.a.	n.a.	n.a.	n.a.
Margarine (n = 24)	<40	n.a.	n.a.	n.a.	<20	<20	**287 ± 38**	**174 ± 32**	**294 ± 160**
Cereal-based baby food*	**11.3 ± 3.1**	**4.70 ± 6.5**	**2.65 ± 1.4**	<1	<10	<10	**2.33 ± 1.5**	**1.17 ± 0.76**	<1
Cookies (n = 24)	**209 ± 49**	n.a.	n.a.	n.a.	**4.47 ± 0.7**	<10	**191 ± 95**	**91.3 ± 42**	**305 ± 110**
Baby food*	**9.27 ± 6.9**	**1.13 ± 0.36**	<1	<1	n.a.	n.a.	n.a.	n.a.	n.a.
Ground coffee (n = 8)	n.a.	**1670 ± 170**	**5210 ± 1100**	**261 ± 32**	n.a.	n.a.	n.a.	n.a.	n.a.
Coffee beans (n = 8)	n.a.	**2410 ± 260**	**7130 ± 1100**	**373 ± 29**	n.a.	n.a.	n.a.	n.a.	n.a.
Coffee substitutes (n = 8)	n.a.	**257 ± 190**	**380 ± 230**	**12.6 ± 8.5**	n.a.	n.a.	n.a.	n.a.	n.a.
Fruit juices (n = 8)	n.a.	<1	<1	<1	n.a.	n.a.	n.a.	n.a.	n.a.
Breakfast cereals (n = 8)	n.a.	**21.6 ± 29**	**8.13 ± 9.6**	**1.26 ± 0.73**	n.a.	n.a.	n.a.	n.a.	n.a.
Sunflower oil (n = 24)	n.a.	n.a.	n.a.	n.a.	<10	<10	**617 ± 190**	**321 ± 100**	**407 ± 140**
Pomace oil (n = 24)	n.a.	n.a.	n.a.	n.a.	<10	<10	**2270 ± 210**	**733 ± 220**	**737 ± 400**
Refined olive oil (n = 24)	n.a.	n.a.	n.a.	n.a.	<10	<10	**397 ± 210**	**198 ± 130**	**267 ± 29**
Infant formula (n = 24)	n.a.	n.a.	n.a.	n.a.	<10	<10	**304 ± 180**	**101 ± 61**	<68
Follow-on formulae (n = 24)	n.a.	n.a.	n.a.	n.a.	<10	<10	**171 ± 120**	**73 ± 36**	<54
Cereals and milk (n = 24)	n.a.	n.a.	n.a.	n.a.	<20	<20	<6	**1.83 ± 1.3**	<7
Mayonnaise (n = 24)	n.a.	n.a.	n.a.	n.a.	**34.1 ± 20**	**5.7 ± 0.58**	**245 ± 96**	**102 ± 47**	**246 ± 37**
Soy sauce (n = 24)	n.a.	n.a.	n.a.	n.a.	**18.8 ± 7.6**	**46 ± 71**	n.a.	n.a.	n.a.
Salad sauce (n = 24)	n.a.	n.a.	n.a.	n.a.	**12.3 ± 1.2**	**12.3 ± 1.2**	**229 ± 130**	**97.3 ± 58**	**95.3 ± 21**
Bakery products (n = 24)	n.a.	n.a.	n.a.	n.a.	**7.23 ± 3.9**	<10	**388 ± 310**	**107 ± 110**	**265 ± 380**
Chocolate spreads (n = 24)	n.a.	n.a.	n.a.	n.a.	<10	<10	**457 ± 160**	**207 ± 49**	**617 ± 140**
Palm oil cookies (n = 24)	n.a.	n.a.	n.a.	n.a.	**4.50 ± 0.87**	<10	**297 ± 15**	**114 ± 13**	**343 ± 15**
Frying oil (mixture) (n = 24)	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	**727 ± 58**	**313 ± 32**	**700 ± 53**

n.a.: not analyzed; *different number of analyzed samples, depending on the target chemicals: n = 24 for acrylamide, 3-MCPD, 2-MCPD, 3-MCPD ester, 2-MCPD ester and GE, and n = 8 for furan, 2-methylfutran and 3-methylfuran.

cookies contained comparable levels of acrylamide to those reported in the USA or North Macedonia (181.8 and 231 μg/kg, respectively) (Abt et al., 2019; Dimitrieska-Stojkovikj et al., 2019), but lower levels in cookies marketed in Spain (423 μg/kg) (Rufian-Henares et al., 2007). In 2015, EFSA released an opinion on acrylamide in food, indicating that higher levels are found in coffee and potato-based products, as well as biscuits and cereals, which could also contain notable levels of acrylamide. Moreover, EFSA (2015) reported low levels in baby food, in agreement to the findings here presented.

The highest levels of furan, 2-methylfurans and 3-methylfurans were found in coffee beans (2410, 7130 and 373 μg/kg, respectively). In general, the levels of 2-methylfuran in all the foodstuffs were higher than those of furan and 3-methylfuran. Kettlitz et al. (2019) had previously observed that 2-methylfuran was the contaminant with the highest concentrations in ground coffee (18,300 μg/kg), being followed by furan and 3-methyfuran (3140 and 690 μg/kg, respectively). Moreover, instant coffee followed the same pattern (4030, 730 and 210 μg/kg for 2-methylfuran, furan and 3-methylfuran, respectively). Finally, the levels of furans in breakfast cereals from Catalonia are similar to those previously found by Kettlitz et al. (2019) (21.6 vs. 27 μg/kg). However, our results are, in general, notably lower than those typically found in the scientific literature. Thus, furan mean levels reported by EFSA (2017) were higher in coffee beans (4580 vs 2410 μg/kg) and ground coffee (2360 vs 1670 μg/kg), but similar in instant coffee (310 vs 380 μg/kg). In turn, lower concentrations (range: 20–57 μg/kg) were found in other types of food, such as breakfast cereals, ready-to-eat meals for infants or bread and rolls, among others (EFSA, 2017).

Mayonnaise showed the highest concentration of 3-MCPD (34.1 μg/kg), while salad sauce presented the greatest levels of 2-MCPD (12.3 μg/kg). Regarding ester-bound MCPDs, pomace oil was the food item showing the highest levels of 3-MCPD and 2-MCPD esters (2270 and 733 μg/kg, respectively). Accordingly, pomace oil also showed the highest concentrations of GEs (737 μg/kg). Infant and follow-on formulae also contained notable levels of 3-MCPD and 2-MCPD esters. Results for esters in refined oils are lower than those found in commercialized samples of extra virgin olive oils, olive oils and oil blends (Kamikata et al., 2018), where levels of 3-MCPD, 2-MCPD esters and glycidyl ester were as high as 3770, 1910 and 1600 μg/kg, respectively. In another study conducted in China, Zhang et al. (2020) analyzed the presence of 3- and 2-MCPD esters in vegetable oils and related products. Vegetable oils contained 3-MCPD and 2-MCPD esters values of 760 and 330 μg/kg, respectively, being similar to those found in the present study for sunflower oil (617 and 321 μg/kg, respectively). In samples of the USA, Beekman et al. (2019) found higher concentrations of 3-MCPD esters and GEs in refined olive oil (560 and 480 μg/kg, respectively), and similar levels in sunflower oil (550 and 390 μg/kg, respectively) than the concentrations observed here.

In contrast to other foodstuffs, the levels of MCDPs and GEs in infant formulas from Catalonia were higher than those found in Denmark, where concentration ranges were the following: 2-MCPD esters: 1.3–29 μg/kg; 3-MCPD esters: 8.6–65 μg/kg; GEs: 3.1–32 μg/kg (Nguyen and Fromberg, 2020). They were also higher than the concentrations of 2-MCPD and 3-MCPD in infant formula from the Canadian market, where maximum values of 47 and 89 μg/kg, respectively, were reported in samples collected in 2013 (Becalski et al., 2015). However, the concentrations are within the same range of magnitude than those found in other countries, such as the USA (3-MCPD esters: 93–620 μg/kg; GE: 8–360 μg/kg) (Beekman et al., 2020) and the Czech Republic (3-MCPD esters: 62–588 μg/kg) (Zelinková et al., 2009).

3.2. Dietary intake of FPC

The estimated mean dietary intake of acrylamide, furan, 2-methylfuran, 3-methylfuran, 3-MCPD, 2-MCPD, ester-bound 3- and 2-MCPDs and GEs by the adult population (18–74 years) of Catalonia is summarized in Tables 3–5. The mean dietary exposure of acrylamide was estimated in 2.91 μg/day (0.042 μg/kg bw/day, considering a body weight of 70 kg), being cookies the main contributor (53 %). Because of its high consumption (62.3 g/day), the second most important contribution corresponded to white bread (11 %) (Table 3). The mean dietary intakes of furan, 2-methylfurans and 3-methylfurans by the adults was calculated in 4.32, 7.36 and 0.439 μg/day (0.0617, 0.105 and 0.0063 μg/kg bw/day), respectively. For the three contaminants, coffee (beans + ground) was the main contributor, with exposure percentage ranges between 85 % and 97 % of the total exposure to furans (Table 4). In relation to MCPDs, the mean dietary exposure was found to be 0.657, 0.529, 10.7, 5.15 and 8.81 μg/day for 3-MCPD, 2-MCPD, 3-MCPD esters, 2-MCPD esters and GEs, respectively (Table 5). Considering a body weight of 70 kg, these values would mean an exposure of 0.0094, 0.0076, 0.153, 0.074 and 0.126 μg/kg bw/day, respectively. White bread was the greatest contributor to the intake of free 3- and 2-MCPDs (46 % and 58 %, respectively), due to the high consumption associated to this food item (62.3 g/day). In turn, the greatest contribution of the ester-bound MCPDs and GEs corresponded to refined olive oil (range: 48 %–60 %), which is a highly consumed food item in the Mediterranean diet (15.7 g/day).

Data on the exposure assessment of the current study were compared to those reported in the literature. For acrylamide, our results were lower than those observed in Japan (Kito et al., 2020), where an exposure for the adult population of 0.15 μg/kg bw/day was reported. Moreover, higher intakes were also found in the USA (0.36 μg/kg

Table 3
Dietary intake of acrylamide by the adult population of Catalonia (Spain).

Food samples	Consumption (g)	Exposure (μg/day)
Potato chips	0.46	0.198 (7)
Potato snacks	0.46	0.111 (4)
Sliced bread	11.4	0.167 (6)
Toasts and crackers	0.64	0.028 (1)
White bread	62.3	0.311 (11)
Breakfast cereals (wheat)	1.31	0.113 (4)
Breakfast cereals (other)	0.98	0.058 (2)
Roasted coffee	27.2	0.280 (10)
Soluble coffee	0.17	0.101 (3)
Margarine	0.85	0.017 (1)
Baby cereals	n.d	n.d
Cookies	7.31	1.53 (53)
Baby food	n.d	n.d
TOTAL	**113**	**2.91**

n.d.: not determined. In brackets, contribution percentage of each food sample to total exposure.

Table 4
Dietary intake of furan, 2-methylfurans and 3-methylfurans by the adult population of Catalonia (Spain).

Food samples	Consumption (g)	Exposure (μg/day)		
		Furan	2-methylfurans	3-methylfurans
Soluble coffee	0.17	0.065 (2)	0.148 (2)	0.011 (3)
Cereal-based baby food	n.d	n.d	n.d	n.d
Baby food	n.d	n.d	n.d	n.d
Ground coffee	27.2	1.69 (39)	3.01 (41)	0.154 (35)
Coffee beans	27.2	2.44 (57)	4.12 (56)	0.220 (50)
Coffee substitutes	n.d.	n.d	n.d	n.d
Fruit juices	49.6	0.05 (1)	0.050 (1)	0.050 (11)
Breakfast cereals	3.26	0.07 (2)	0.027 (0)	0.004 (1)
TOTAL	**107**	**4.32**	**7.36**	**0.439**

n.d.: not determined. In brackets, contribution percentage of each food sample to total exposure.

Table 5
Dietary intake of 3-MCPD, 2-MCPD, 3-MCPD and 2-MCPD esters and GEs by the adult population of Catalonia (Spain).

Food samples	Consumption (g)	Exposure (µg/day)				
		3-MCPD	2-MCPD	3-MCPD esters	2-MCPD esters	GE
Potato chips	0.46	0.00228 (0.4)	0.00228 (0.4)	0.0464 (0.43)	0.0230 (0.4)	0.0309 (0.4)
Potato snacks	0.46	0.00228 (0.4)	0.00228 (0.4)	0.177 (2)	0.0690 (1.3)	0.0628 (0.7)
Sliced bread	11.4	0.172 (26)	0.0570 (11)	0.239 (2.22)	0.0570 (1.1)	0.0114 (0.1)
Toasts and crackers	0.64	0.00299 (0.5)	0.00320 (0.6)	0.0437 (0.4)	0.0156 (0.3)	0.0395 (0.4)
White bread	62.3	0.303 (46)	0.311 (58)	0.0415 (0.4)	0.0623 (1.2)	0.187 (2)
Margarine	0.85	0.00850 (1.3)	0.00850 (1.6)	0.244 (2.3)	0.148 (3)	0.250 (3)
Cereal-based baby food	n.d	n.d	n.d	n.d	n.d	n.d
Cookies	7.31	0.0327 (5)	0.0366 (6.9)	1.39 (13)	0.668 (13)	2.23 (25)
Sunflower oil	1.82	0.00910 (1.4)	0.00910 (2)	1.12 (10)	0.585 (11)	0.740 (8.4)
Pomace oil	n.d.	n.d	n.d	n.d	n.d	n.d
Refined olive oil	15.7	0.0785 (12)	0.0785 (15)	6.22 (58)	3.11 (60)	4.18 (48)
Infant formula	n.d.	n.d	n.d	n.d	n.d	n.d
Follow-on formulae	n.d.	n.d	n.d	n.d	n.d	n.d
Cereals and milk	n.d.	n.d	n.d	n.d	n.d	n.d
Mayonnaise	n.d.	0.0273 (4.2)	0.00456 (0.85)	0.196 (1.83)	0.0819 (2)	0.197 (2)
Soy sauce	0.8	0.00128 (0.2)	0.00313 (0.59)	n.d	n.d	n.d
Salad sauce	0.068	0.00037 (0.1)	0.00037 (0.06)	0.00687 (0.1)	0.00292 (0.1)	0.00286 (0.03)
Bakery products	0.03	0.0132 (2)	0.00910 (2)	0.707 (6.6)	0.195 (4)	0.482 (5.5)
Chocolate spreads	1.82	0.00320 (0.5)	0.00320 (0.6)	0.292 (2.72)	0.132 (2.6)	0.395 (4.5)
Palm oil cookies	0.64	n.d	n.d	n.d	n.d	n.d
Frying oil (mixture)	n.d.	n.d	n.d	n.d	n.d	n.d
TOTAL	**104**	**0.657**	**0.529**	**10.7**	**5.15**	**8.81**

n.d.: not determined. In brackets, contribution percentage of each food sample to total exposure.

bw/day) and in North Macedonia (0.643 µg/kg bw/day) (Abt et al., 2019; Dimitrieska-Stojkovikj et al., 2019). The estimated intake of acrylamide by the Catalan adult population (2.91 µg/day) was notably lower than the estimated values reported in other Spanish regions (range: 15.3–18.7 µg/day), according to Freisling et al. (2013). Moreover, it was lower than the dietary intake by Spanish women (0.3 µg/kg bw/day) given by Obón-Santacana et al. (2014), who estimated a value of 0.3 µg/kg bw/day (equivalent to 18 µg/day for a 60 kg-body weight woman). Finally, the intake was similar to the values found by Mesías and Morales (2015), considering only the consumption of potatoes (0.035 µg/kg bw/day).

Regarding furans, the estimated exposure in the current study was well below that reported in Austria (range: 0.312 – 0.498 µg/kg bw/day) (Mayerhofer et al., 2019) and Belgium (0.38 µg/kg bw/day) (Scholl et al., 2012). In Canada, the average furan and total furan (including 2- and 3-methylfuran) intakes by adults (≤20 years) were estimated at approximately 0.37 and 0.71 µg/kg bw/day (Becalski et al., 2010).

In China, Zhang et al. (2020) analyzed the content of MCPDs in vegetable oils and related products from Zhejiang market. They estimated a dietary exposure of 3-MCPD esters and 2-MCPD esters of 0.62 and 0.26 µg/kg bw/day, respectively, which are notably higher than those here found. Also in China, Li et al. (2015) estimated the 3-MCPD esters exposure levels for the adult population (18–49 years), which was estimated in 0.71 µg/kg bw/day. Our results are also lower than those found by Chung et al. (2013) in Hong Kong (range: 0.20−0.53 µg/kg bw/day). As for free MCPDs, our levels are notably lower than those found in Brazil (range: 0.06−0.51 µg/kg bw/day) and in the Netherlands (0.674 µg/kg bw/day) (Arisseto et al., 2013; Boon and Te Biesebeek, 2016).

The dietary intake of acrylamide, furans and MCPDs for different population groups are depicted in Figs. 1–3. Children aged 3–9 years presented the highest exposure level of acrylamide (5.41 µg/day). However, taking into account the body weight, toddlers (6–11 months) were the most exposed group (0.405 µg/kg bw/day). For all the age groups, the highest contribution to acrylamide corresponded to cookies, with the only exception of toddlers, whose main contributor was breakfast cereals.

The adult population (40–64 years) was the group showing the highest exposure to furan, 2-methylfurans and 3-methylfurans (5.33, 15.9 and 0.856 µg/day, respectively). For the youngest population groups (toddlers, infants, children and adolescents), among these three contaminants, furan was the most important contributor in comparison to 2-methylfurans and 3-methylfurans, with ranges of 45 %–63 %. In turn, adults and senior people presented a higher intake of 2-methylfurans than that of furan and 3-methylfurans (Fig. 2). For the latter population groups, 2-methylfurans meant 72 % of the total exposure to furans. Breakfast cereals were the main contributor to furan exposure for the young population groups (toddlers, infants, children and

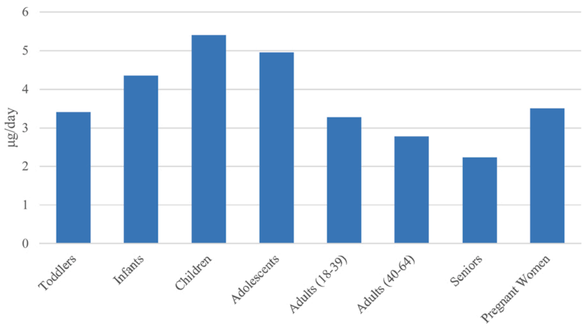

Fig. 1. Dietary intake of acrylamide for several population groups living in Catalonia (Spain).

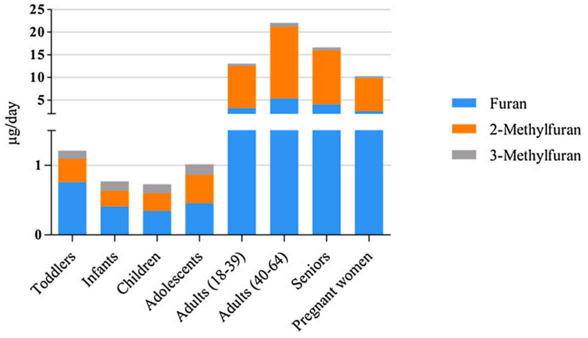

Fig. 2. Dietary intake of furan, 2-methylfurans and 3-methylfurans for several population groups living in Catalonia (Spain).

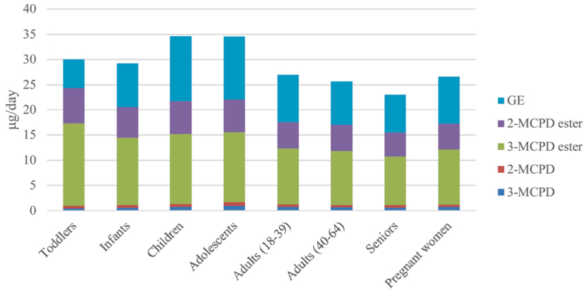

Fig. 3. Dietary intake of 3-MCPD, 2-MCPD, 3-MCPD and 2-MCPD esters and GEs for several population groups living in Catalonia (Spain).

adolescents), while the greatest intake of furan by adults and seniors corresponded to coffee beans. Regarding 2-methylfurans, the intake pattern was different according to the age. The foodstuffs with the highest contribution were breakfast cereals in toddlers and infants, fruit juices in children and adolescents, and coffee beans in adults, seniors and pregnant women. The food pattern was more varied for 3-methylfurans. Toddlers were primarily exposed to this contaminant through baby food consumption, while fruit juices were the main intake of 3-methylfurans in infants, children and adolescents. As for most FPCs, coffee beans were the main contributors to exposure of 3-methylfurans for adult groups.

Adolescents (10–17 years old) and children (3–9 years old) were the most exposed groups for the MCPDs and GEs, respectively (34.6 μg/day for both). The chemical profile was dominated by 3-MCPD, with a contribution range to total exposure of 40 %–55 %. In contrast, free 3- and 2-MCPDs were the contaminants showing the lowest levels of exposure (Fig. 3). For free MCPDs, follow-on formula was the food item mainly contributing to their exposure in toddlers and infants, due to their high consumption. On the other hand, white bread was the main contributor for adolescents, adults and seniors, as well as pregnant women. Ester-bound MCPDs followed a similar pattern in young population groups, with the main contribution corresponding to follow-on formula consumed by toddlers and infants. By contrast, refined olive oil was the food product with the highest contribution to exposure of ester-bound MCPDs in the remaining population groups. Finally, toddlers, young adults, adults, seniors and pregnant women, were mainly exposed to GEs through refined olive oil consumption, while infants, children and adolescents were mostly exposed through cookies consumption.

The 95th percentile (P95) of the FPC dietary intake by the Catalan population is summarized in Table 6. The estimated exposure of acrylamide for toddlers, infants, children and adolescents ranged between 0.65 and 1.70 μg/kg bw/day, while for young adults, adults, seniors and pregnant women, the range was between 0.15 and 0.23 μg/kg bw/day. For furan, the exposure of the youngest population (toddlers, infants, children and adolescents) was estimated in 0.034−0.32 μg/kg bw/day, while the intake for the adults was between 0.17 and 0.22 μg/kg bw/day. The daily exposure to 2-methylfuran and 3-methylfuran for the youngest people ranged 0.018−0.17 and 0.010−0.089 μg/kg bw/day, respectively, while for the older groups, these values were within the range 0.048−0.63 μg/kg bw/day, for 2-methylfuran, and 0.030−0.036 μg/kg bw/day, for 3-methylfuran. Finally, the exposure ranges of 3-MCPD, 2-MCPD, 3-MCPD ester, 2-MCPD ester, and GE were 0.078−0.15, 0.057−0.14, 1.40−4.42, 0.64−2.06, and 1.40–2.81 μg/kg bw/day, respectively, for the youngest groups, and 0.029−0.040, 0.022−0.028, 0.50−0.59, 0.24−0.28, and 0.42−0.50 μg/kg bw/day, respectively, for the adults. In all the cases, the P95 of the dietary intake of FPC were lower than typical intake data reported by EFSA (2015, 2016, 2017).

3.3. Health risk assessment

In order to be compared with threshold values, either MOE or TDI (Table 7), data on dietary intake according to the body weight were considered. For acrylamide, the risk characterization for neoplastic effects was derived from the $BMDL_{10}$ of 170 μg/kg bw/day, taken from data on Harderian gland adenomas and adenocarcinomas in mice. The CONTAM Panel established that a MOE of 10,000 or higher would be of low concern for the public health. On the other hand, the $BMDL_{10}$ value of 430 μg/kg bw/day was used to perform the risk characterization for non-neoplastic effects (neurotoxicity). In this case, a MOE of 100 or higher is considered sufficient to conclude that there is no health concern (EFSA, 2015). In the present study, none of the population groups was at risk of suffering non-neoplastic effects, being all MOEs above 100 for the three scenarios considered (UB, MB and LB). However, the MOEs for all the population groups (except seniors and adults between 40 and 64 years) were below 10,000 for the neoplastic effects; therefore, there would be a health concern for that type of effects derived from the exposure to acrylamide. In addition, under the UB, all the population groups were below 10,000, indicating a health risk for neoplastic effects. In fact, some studies have observed an increased risk of suffering some specific types of cancer (endometrial and ovarian), when the exposure to acrylamide overcomes 20 μg/day, while no association is found with some of the common types of cancers (Adani et al., 2020; Pelucchi et al., 2014).

Regarding furan, the CONTAM Panel selected the $BMDL_{10}$ of 64 μg/

Table 6
Mean and P95 exposure to acrylamide, furans and MCPDs by selected groups of population of Catalonia (Spain).

Compound	Exposure (μg/kg bw/day)	Adults (18−74 y)	Toddlers	Infants	Children	Adolescents	Adults (18−39 y)	Adults (40−64 y)	Seniors (65−74 y)	Pregnant women
Acrylamide	Mean	0.042	0.41	0.35	0.23	0.097	0.045	0.036	0.032	0.054
	P95	0.17	1.70	1.52	1.30	0.65	0.23	0.15	0.15	0.19
Furan	Mean	0.062	0.090	0.033	0.014	0.0089	0.045	0.069	0.057	0.039
	P95	0.22	0.32	0.17	0.062	0.034	0.19	0.22	0.22	0.17
2-MF	Mean	0.11	0.040	0.019	0.011	0.008	0.13	0.21	0.17	0.11
	P95	0.63	0.17	0.10	0.033	0.018	0.54	0.63	0.63	0.48
3-MF	Mean	0.006	0.014	0.011	0.0055	0.0030	0.0076	0.011	0.0093	0.0071
	P95	0.036	0.089	0.057	0.019	0.010	0.031	0.035	0.036	0.030
3-MCPD	Mean	0.0094	0.054	0.047	0.031	0.019	0.0099	0.0081	0.0081	0.011
	P95	0.034	0.13	0.15	0.12	0.078	0.040	0.030	0.029	0.047
2-MCPD	Mean	0.0076	0.054	0.044	0.024	0.015	0.0074	0.0069	0.0068	0.0078
	P95	0.025	0.13	0.14	0.095	0.057	0.027	0.022	0.023	0.028
3-MCPD ester	Mean	0.153	1.96	1.08	0.58	0.27	0.15	0.14	0.14	0.17
	P95	0.55	4.42	3.78	2.72	1.40	0.59	0.51	0.50	0.54
2-MCPD ester	Mean	0.074	0.83	0.49	0.27	0.13	0.073	0.067	0.067	0.080
	P95	0.26	2.06	1.75	1.25	0.64	0.28	0.25	0.24	0.26
GE	Mean	0.126	0.68	0.71	0.54	0.25	0.13	0.11	0.11	0.14
	P95	0.48	2.31	2.47	2.81	1.40	0.50	0.44	0.42	0.49

Table 7
Risk assessment of acrylamide, furan and 3-MCPD (free + esters) for several population groups living in Catalonia (Spain).

MOE or TDI		Exposure	Toddlers	Infants	Children	Adolescents	Adults (18–39 y)	Adults (40–64 y)	Seniors	Pregnant women
Acrylamide										
Neoplastic effects	170 µg/kg/day	UB	338	368	548	1176	2507	2997	3339	2187
		MB	419	481	755	1747	3740	4709	5371	3154
		LB	600	694	1059	2572	5512	7157	8360	4604
No neoplastic effects	430 µg/kg/day	UB	856	930	1386	2975	6341	7580	8445	5531
		MB	1061	1217	1909	4418	9459	11,912	13,587	7977
		LB	1518	1756	2678	6506	13,492	18,104	21,145	11,645
Furan										
Neoplastic effects	1310 µg/kg/day	UB	3670	11,073	27,552	45,435	22,696	15,488	17,972	25,560
		MB	14,527	39,730	91,046	146,987	29,403	18,929	22,802	33,556
		LB	-[a]	- [a]	20,993,490	1,712,126	37,691	23,377	28,752	43,648
No neoplastic effects	64 µg/kg/day	UB	179	541	1346	2220	1109	757	878	1249
		MB	710	1941	4448	7181	1436	925	1114	1639
		LB	- [a]	- [a]	102,277	83,646	1841	1142	1405	2132
3-MCPD (free + esters)[*]										
TDI	0.8 µg/kg/day	UB	3.37	1.82	0.914	0.440	0.247	0.228	0.225	0.271
		MB	2.01	1.13	0.610	0.290	0.164	0.147	0.146	0.179
		LB	1.00	0.607	0.335	0.152	0.086	0.075	0.074	0.095
GEs										
Neoplastic effects	10,200 µg/kg/day	UB	10,527	10,441	14,125	31,336	60,435	69,911	74,799	55,467
		MB	15,042	14,367	18,944	41,577	78,380	90,730	95,188	71,120
		LB	29,464	22,908	27,599	57,950	105,126	117,824	120,928	93,342

MOE: Margin of exposure; TDI: Tolerable daily intake; UB: Upper-bound; MB: Medium-bound; LB: Lower-bound.

[*] Results are given in µg/kg/day; [a]the estimated exposure was zero; therefore, a MOE could not be calculated.

kg bw/day as a reference point for the risk characterization of non-neoplastic effects. In turn, the risk characterization for neoplastic effects was derived from the BMDL$_{10}$ of 1310 µg/kg bw/day. Again, a MOE of 100 or higher and a MOE of 10,000 or higher for non-neoplastic and neoplastic effects, respectively, would be enough to conclude that there is no health concern (EFSA, 2017). The calculated MOEs showed that no group is at risk of developing non-neoplastic effects, being the exposure levels for all the population groups above 100, for the three scenarios considered. With the calculation of the MOE for the neoplastic effects, it can be concluded that none of the groups are at risk of suffering neoplastic effects, since the MOE was above 10,000, for LB and MB estimates. However, for the UB, toddlers are at risk for neoplastic effects, as the calculated MOE was less than 10,000. Because of the lack of information, the health risks associated to the dietary intake of 2-methylfurans and 3-methylfurans could not be characterized.

With respect to free- and ester-bound 3-MCPD, the CONTAM Panel established a TDI of 0.8 µg/kg bw/day. No TDI could be established for 2-MCPD and its fatty esters due to the lack of toxicological information (EFSA, 2016). The mean exposure levels estimated for different population groups living in Catalonia showed that toddlers and infants intakes are above the TDI (2.01 and 1.13 µg/kg bw/day, respectively), while those of the remaining population groups are below that threshold (range: 0.146−0.61 µg/kg bw/day). Under the UB scenario, children, along with toddlers and infants, also exceeded the TDI (0.914, 3.37 and 1.82 µg/kg bw/day, respectively). Differently, under the LB exposure scenario, only toddlers are above the established TDI (1.00 µg/kg bw/day), meaning that toddlers, even at a low exposure scenario, are at risk for the potential effects derived from MCPD exposure.

Since glycidol is genotoxic and carcinogenic, the CONTAM Panel considered that it was not appropriate to establish a TDI for glycidol. Thus, the MOE approach was used to characterize the risk. Moreover, it was assumed that the complete hydrolysis of the esters to free glycidol is achieved upon ingestion. In this case, a T25 approach (25 % increase in incidence of a specific tumor above background incidence in the lifespan of the species) was used, instead the BMD. The reference point used for glycidol was 10,200 µg/kg bw/day. Moreover, the CONTAM Panel considered that a MOE of 25,000 or higher was enough to conclude that there was not a health concern (EFSA, 2016). Toddlers, infants and children had MOEs below 25,000, under the UB and MB exposure scenarios, which means that they may be at risk for the genotoxic and carcinogenic effects derived from glycidyl esters exposure. In turn, under the LB exposure scenario, none of the groups was above a MOE of 25,000.

When the MOE according to the P95 dietary exposure was calculated, it was concluded that all the population groups were at risk for neoplastic effects derived from acrylamide exposure for the three scenarios considered (MOE < 10,000), while no risk was determined for the non-neoplastic effects (MOE > 100) (Table 8). For furan, most of the groups were at risk of suffering neoplastic effects in the three scenarios, with the exception of adolescents (UB and MB), children (MB) and pregnant women (LB). For the non-neoplastic effects, only toddlers were at risk, with a MOE of 54 (Table 8). In addition, the TDI set for 3-MCPD (free + esters) was exceeded by most of the population groups in the UB, with the exception of the seniors, while at the MB and LB, only the youngest population groups (aged <17) surpassed the threshold value. Finally, the MOE calculated for the GE was below 25,000 for all the population groups in the UB and MB scenarios, while in the LB, only the youngest groups were at risk of suffering neoplastic effects (Table 8).

4. Conclusions

To the best of our knowledge, this is the first study reporting the occurrence and the exposure and risk assessment of several FPCs in Spain. The highest concentrations of acrylamide and furan were found in different types of coffee (roasted, ground and beans), while the greatest levels of MCPDs were found in edible oils, including sunflower, pomace and frying oils. The exposure assessment identified cookies, coffee, bread and refined olive oil as the main contributors to the exposure of acrylamide, furan, MCPDs and GEs, respectively. Moreover, this pattern of contribution was similar irrespective of the population group. However, some specific foodstuffs, such as breakfast cereals or infant formulas, showed a remarkable contribution in the dietary intake of some FPCs by toddlers and infants. According to risk assessment calculations, all the population groups are currently at risk of suffering neoplastic effects derived from the exposure to acrylamide. Moreover, the human exposure to total 3-MCPD (free + ester-bound) and GEs by toddlers and infants exceeds the threshold values set by international organizations. In contrast, the current exposure to furans is not associated to health risks, for both neoplastic and non-neoplastic effects. Our findings clearly indicate that the current dietary exposure to FPCs in Catalonia (Spain)

Table 8
Risk assessment of acrylamide, furan and 3-MCPD (free + esters) for several population groups living in Catalonia (Spain) in the P95 dietary exposure.

MOE or TDI		Exposure	Toddlers	Infants	Children	Adolescents	Adults (18–39 y)	Adults (40–64 y)	Seniors	Pregnant women
Acrylamide										
		UB	76	84	97	192	510	789	779	621
Neoplastic effects	170 µg/kg/day	MB	100	112	130	263	726	1141	1134	875
		LB	176	168	179	366	1042	1678	1693	1269
		UB	192	211	246	486	1291	1996	1970	1570
No neoplastic effects	430 µg/kg/day	MB	253	283	330	664	1837	2885	2869	2213
		LB	446	425	453	927	2637	4244	4283	3209
Furan										
		UB	1110	2153	5879	10,586	5251	4595	4245	5557
Neoplastic effects	1310 µg/kg/day	MB	4050	7681	21,226	38,742	6970	6071	5930	7629
		LB	-[a]	-[a]	-[a]	-[a]	8956	7844	7912	10,106
		UB	54	105	287	517	257	224	207	271
No neoplastic effects	64 µg/kg/day	MB	198	375	1037	1893	341	297	290	373
		LB	-[a]	-[a]	-[a]	-[a]	438	383	387	494
3-MCPD (free + esters)*										
		UB	7.38	6.23	4.20	2.20	0.94	0.81	0.78	0.86
TDI	0.8 µg/kg/day	MB	4.54	3.93	2.85	1.48	0.63	0.54	0.53	0.58
		LB	2.55	2.17	1.58	0.80	0.35	0.30	0.29	0.35
GEs										
		UB	3279	2983	2663	5328	14,821	16,917	17,581	16,217
Neoplastic effects	10,200 µg/kg/day	MB	4410	4124	3635	7268	20,217	23,095	24,134	20,790
		LB	6958	6495	5509	10,809	29,673	33,580	35,663	28,852

MOE: Margin of exposure; TDI: Tolerable daily intake; UB: Upper-bound; MB: Medium-bound; LB: Lower-bound.
* Results are given in µg/kg/day; [a]the estimated exposure was zero; therefore, a MOE could not be calculated.

may be a health concern for some groups of the population, especially toddlers, infants and children, who are the most vulnerable. Therefore, food monitoring studies should be performed periodically to see the trends of FPCs and to assure that their exposure by young people follows a decreasing trend. In parallel, specific policy measures should be adopted to minimize the intake of FPCs in Catalonia.

CRediT authorship contribution statement

Neus González: Formal analysis, Writing - original draft. **Montse Marquès:** Investigation, Writing - review & editing. **Josep Calderón:** Methodology, Investigation, Writing - original draft. **Roger Collantes:** Methodology, Investigation. **Lidia Corraliza:** Methodology, Investigation. **Isabel Timoner:** Conceptualization, Validation. **Jaume Bosch:** Conceptualization, Validation. **Victòria Castell:** Conceptualization, Supervision. **José L. Domingo:** Writing - review & editing, Funding acquisition. **Martí Nadal:** Resources, Writing - original draft, Supervision.

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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