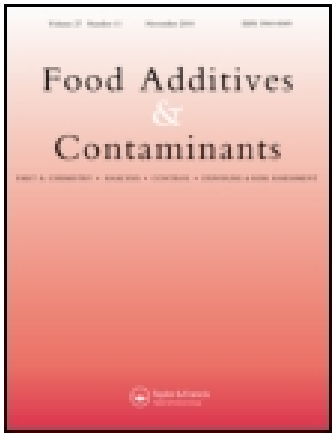

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Exposure assessment to ochratoxin A in Catalonia (Spain) based on the consumption of cereals, nuts, coffee, wine, and beer

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Exposure assessment to ochratoxin A in Catalonia (Spain) based on the consumption of cereals, nuts, coffee, wine, and beer

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Ochratoxin A (OTA) was analysed in composite samples of cereal-based baby foods, beer, breakfast cereals (corn- and rice and wheat-based), loaf bread, peanuts and pistachios. Foodstuffs were collected in hypermarkets and supermarkets from 12 cities in the Spanish region of Catalonia, and composite samples were prepared for analysis involving liquid–liquid extraction, followed by immunoaffinity column clean-up and HPLC with fluorescence detection. Consumption data for the selected foodstuffs were collected by means of a food-frequency questionnaire. The studied population was grouped by age in infants, children, adolescents and adults; and exposure to OTA through the specified foodstuffs, and through wine and coffee, was assessed. Exposure assessment was done through deterministic and probabilistic modelling of the contamination and consumption data. OTA occurrence and mean of positive samples (ng g^{-1} or ng ml^{-1}, for beer) were the following: 8.7% and 0.233 in baby foods; 88.7% and 0.022 in beer; 2.8% and 0.728 in corn-based breakfast cereals; 25% and 0.293 in wheat-based breakfast cereals; 12.9% and 0.283 in loaf bread; 41.7% and 0.241 in peanuts; and 2.9% and 0.228 in pistachios. The median estimated daily intake of OTA through the foodstuffs by each age group were below the latest provisional tolerable daily intakes (PTDIs) of 17 and 14 ng kg^{-1} bw day^{-1} recommended by the European Food Safety Authority (EFSA) in 2006 and the Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 2007, respectively, ranging from 1% and 2% of those values in adolescents and children, to 3% and 11% in adults and infants.

Keywords: chromatography – HPLC; clean-up – affinity columns; exposure assessment; probabilistic modelling; ochratoxin A; baby food; beer; cereals; peanuts

Introduction

Ochratoxin A (OTA) is a fungal secondary metabolite produced by some species of the genera *Aspergillus* and *Penicillium*. Studies on animals have characterised this mycotoxin as nephrotoxic, hepatotoxic, neurotoxic, immunotoxic, teratogenic and carcinogenic. Chronic human exposure to OTA has been related to the development of urinary tract tumours and Balkan Endemic Nephropathy, and the International Agency for Research on Cancer (IARC) classified OTA as possibly carcinogenic to humans (group 2B) (IARC 1993).

Sources of human exposure to OTA are mainly foodstuffs of vegetal origin, and it is possible to find OTA in cereals and derivatives, wines and grape juices, coffee, beer, nuts and dried fruits, spices, and in a minor extent, in animal by-products. Considering such ubiquity and the mentioned toxic effects, international authorities have proposed tolerable daily or weekly intakes for the toxin, which indicate the dose that can be safely consumed daily/weekly over a lifetime without incurring any appreciable adverse health effects (World Health Organization (WHO) 1999). The provisional tolerable daily intakes (PTDIs) of 17 and 14 ng kg^{-1} bw day^{-1} were recommended by the European Food Safety Authority (EFSA) (2006) and the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (2007), respectively.

Control of the presence of mycotoxins in the European Union is based on maximum levels in certain foodstuffs. In the case of OTA, maximum levels were set for unprocessed cereals and derivatives, cereal-based baby foods, coffee, wines, dried vine fruit and grape juices, ranging from 0.5 μg kg^{-1} in foodstuffs intended for babies and infants to 10 μg kg^{-1} in dried vine fruit and soluble coffee (European Commission 2006). More recently, maximum levels for liquorice and some spices have also been set (European Commission 2010).

Exposure to OTA by humans can therefore be assessed by the detection of the toxin in possibly contaminated foodstuffs, as well as by the evaluation of the dietary habits of a population, especially of the consumption of those foodstuffs. Thus, the evaluation

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of the exposure of the inhabitants of the Spanish region of Catalonia to OTA was the aim of this study. For this, certain possibly contaminated foodstuffs were collected in several localities of the region and OTA levels were therein determined. The following step was to estimate the intake of OTA due to the consumption of the analysed foodstuffs. Therefore, consumption data of Catalan individuals were used together with the contamination data to estimate quantitatively the daily intake of OTA. Two methodologies were used to perform this estimation: an analysis by simple distributions (deterministic analysis) and a probabilistic analysis. The first method employs distributions of food consumption, but uses a fixed value for the concentration variables; whereas probabilistic analysis utilises distributions of both the food consumption and contamination data, and simulates dietary exposure by drawing random values from each input distribution (Kroes et al. 2002).

Materials and methods

Sampling

Samples of breakfast cereals made of corn and of wheat and rice, cereal-based baby foods (multicereals), beer, loaf bread (white and wholemeal), peanuts, and pistachios were purchased in June–November 2008 in hypermarkets and supermarkets of 12 cities of the region of Catalonia, Spain: Barcelona, Girona, L'Hospitalet de Llobregat, Lleida, Manresa, Mataró, Reus, Sabadell, Tarragona, Terrasa, Tortosa and Vilanova i la Geltrú. These cities account for 72% of the total population of Catalonia.

Samples were purchased in six stores per city, and three samples of each foodstuff, when available, were randomly picked in each store. Samples corresponding to each store were pooled to obtain a composite sample per store. For this, 50 g were taken from the package of each sample, and thus the total weight of the composite sample was 150 g. The number of composites per foodstuff is listed in Table 2.

OTA chemical analysis

Preparation of food samples

- *Breakfast cereals and loaf bread:* breakfast cereals were crushed (Moulinex crusher DPA139). Loaf bread was dried and afterwards crushed.
- *Beer:* samples were degassed by ultrasound treatment for 40 min; pH was adjusted to 7.2 by adding 2 M NaOH.
- *Peanuts:* most of the samples were purchased unshelled. If peanuts were shelled, shells were removed and the nuts were milled (Moulinex crusher DPA139).
- *Pistachios:* whole pistachios (shells and nuts) were milled (FOSS 1093 Cyclotec^{TM} Sample Mill).

Extraction of OTA

A total of 5 g of sample was mixed with 20 ml (peanuts and pistachios) or 25 ml (breakfast cereals, loaf and cereal-based baby food) of 60% acetonitrile in an amber flask. The mixture was blended for 10 min in the capped flask by means of a magnetic stirrer and afterwards filtered (Whatman No. 1 filter).

Clean-up of samples by immunoaffinity chromatography columns (IACs)

This step consisted on mixing certain volumes of the filtered liquid extract of a foodstuff (except beer) with a certain volume of phosphate buffered saline (PBS) solution, and then on loading this mixture onto the IAC (Ochraprep, R-Biopharm Rhône Ltd, Glasgow, UK). The mixture was allowed to pass by gravity. PBS was prepared by dissolving in 1 L of water the following: 0.2 g potassium chloride, 0.2 g potassium di-hydrogen phosphate, 1.2 g di-sodium hydrogen phosphate anhydrous and 8 g sodium chloride (Panreac, Barcelona, Spain). The pH was adjusted to 7.4 with sodium hydroxide. For each foodstuff, the volumes of extract and PBS were as follows:

- *Breakfast cereals, loaf bread, and cereal-based baby food:* 2 ml of filtrate were diluted with 22 ml PBS.
- *Peanuts and pistachios:* 4 ml of filtrate were diluted with 44 ml PBS.
- *Beer:* samples were not mixed with PBS; 150 ml of the sample (adjusted to pH 7.2) was allowed to pass through the IAC.

In all cases, after the diluted extracts passed through the IACs, the columns were washed with 20 ml PBS, then air was passed through and the wash liquid was discarded. The final step of the clean-up procedure was the elution of OTA into an amber vial. For that, 1.5 ml desorption solution (methanol:acetic acid, 98:2) was loaded onto the IAC. During elution, back-flushing (reversing the flow in the IAC) was performed three times. Finally, 1.5 ml Milli-Q water were passed and a final volume of 3 ml was obtained. Air was passed to collect the last drops of eluate.

HPLC analysis

Analysis of the clean-up final extracts was by HPLC using a Waters 2695 Separations Module (Alliance) coupled to the Waters 2475 Multi λ

fluorescence detector. Waters Spherisorb ODS2 C18 column (5 µm, 4.6 × 150 mm) equipped with a Waters Spherisorb ODS2 guard column (5 µm, 4.6 × 10 mm) (Waters Corporation, Ireland) was used. The integration software used to manage the chromatographic data was Empower 2 (2006 Waters Corporation, Database Version 6.10.00.00). The mobile phase consisted of acetonitrile : Milli-Q water : acetic acid (51 : 47 : 2). Flow rate was 1 ml min^{-1}; injection volume was 100 µl. Excitation and emission wavelengths were 333 and 443 nm, respectively. The temperature of the column and guard column was maintained at 40°C. The OTA retention time was 5.4 min.

Validation of the analytical methods

Validation of the methods of analysis of the different foodstuffs was performed by the evaluation of their linearity, recovery, repeatability and limit of detection (LOD). LOD was calculated using a signal-to-noise ratio of 3. A calibration curve was built for the analysis of each foodstuff by serial dilution of a stock solution in the range 0.012–12.5 ng ml^{-1} and it was linear in that range. The coefficient of determination (R^2) > 0.998 for all cases. Recovery rates were evaluated by spiking samples ($n = 3$) with certain amounts of OTA standard solutions. Results were not corrected by recovery. Inter-day repeatability was evaluated on 3 different days for a certain concentration in each foodstuff. the results of the validation assays are shown in Table 1.

Consumption data

Data of consumption of the listed foods were obtained by means of a food-frequency questionnaire (FFQ), which included 33 foodstuffs possibly contaminated by OTA (Coronel et al. 2009). The survey was administered by trained interviewers from January 2008 to February 2009 to inhabitants of several localities in the Catalan province of Lleida. Gender, age and weight of the participants were also recorded. The population was classified according to their age as infants (0–3 years old, $n = 164$), children (4–9 years old, $n = 68$), adolescents (10–17 years old, $n = 211$), and adults (18–65 years old, $n = 905$). Parents were interviewed for infants' responses. The number of participants classified by gender was, in the adolescent group, 89 males and 122 females; and in the adult group, 396 males and 509 females. Food consumption of infants and children was assumed to be equal for both genders.

For calculation purposes, individual consumption data (g foodstuff $person^{-1}$ day^{-1}) obtained from the FFQs was normalised by dividing them by the corresponding individual body weight (g foodstuff kg^{-1} body weight day^{-1}).

Table 1. Results for recovery in the different spiking levels, repeatability (intra- and inter-day), and LOD for each foodstuff.

Foodstuff	Spiking level (ng g^{-1})[a]	Recovery rate (%)	RSD intra-day (%)	RSD inter-day (%)	LOD (ng g^{-1})[a]
Baby foods	0.3	101.67	7.90		0.180
	0.5	102.00	1.47		
	0.8	99.06	2.38		
	1	93.25	0.46	4.27	
Beer	0.05	89.40	4.06		0.003
	0.2	85.73	2.95		
	0.5	89.91	12.59	6.30	
Breakfast	0.8	90.94	2.82		0.098
cereals	1.5	106.83	2.93	2.59	
	3	100.42	3.96		
	5	100.45	2.65		
Loaf bread	0.5	103.90	4.32		0.139
	0.8	104.00	0.72		
	1.5	110.13	2.27		
	3	99.65	1.02	2.24	
Peanuts	0.5	71.48	7.79		0.072
	0.8	99.30	4.98		
	1	93.94	1.37	4.19	
Pistachios	0.5	100.96	0.73		0.129
	1	88.76	1.39		
	1.5	96.08	1.85	7.45	
	2	97.84	6.23		

Note: [a]ng ml^{-1} in the case of beer.

Estimation of the daily intake of OTA

Estimation of the OTA daily intake (ng OTA kg^{-1} bw day^{-1}) was carried out by deterministic and probabilistic procedures, taking into account the contamination data of OTA in the analysed foodstuffs (ng OTA g^{-1} foodstuff) and the normalised consumption data (g foodstuff kg^{-1} bw day^{-1}) of the surveyed population. Results of the intake estimations were listed as descriptive statistics such as means, median and high quantiles of the obtained distributions. Measures of asymmetry of the distributions (skewness and kurtosis) were also shown.

Data of the contamination distribution in coffee were taken from a previous publication (Coronel et al. 2011), in which the sampling procedure was the same as in this work. Additional contamination data for red and dessert wine were also included: distribution data of OTA presence in samples previously collected in Spain (Bellí et al. 2004; Valero et al. 2008) were incorporated in this work in order to complete the analysis of the exposure to OTA, as wine consumption is considered to be an important source of OTA in the diet.

Treatment of contamination-censored data: alternatives for the values below the limit of detection of the method of analysis

Contamination-censored data (values < LOD or non-detectable results (ND)) were treated as advised

by GEMS/Food-EURO (1995) to obtain a simple estimate of the mean. The alternatives depend on the proportion of results below the LOD:

- If the proportion is ≤60%, LOD/2 should be used for the results less than LOD.
- If the proportion is >60% but ≤80%, and with at least 25 results quantified, two estimates should be produced: using zero and LOD for the results less than LOD.
- If the proportion is >80%, two estimates should be produced: using zero and LOD for the results less than LOD and the estimation of other descriptive statistics will not be practicable.

Thus, values for mean contamination could be one value if ND data were replaced by LOD/2, or two if ND data were replaced by zero and LOD.

Deterministic approach

The deterministic estimation of the intake was achieved by the analysis of simple distributions, which consider the average value of contamination of a foodstuff, and the individual values of consumption of the study population (Kroes et al. 2002). Contamination mean values were calculated according to the above-mentioned criteria. The estimation of the daily intake was performed by multiplying the individual consumption data of each foodstuff by its mean contamination obtained after the treatment of the ND. Total OTA daily intake was the sum of the individual OTA intakes through the different foodstuffs, and therefore two exposure scenarios were obtained (ND replaced by zero and ND replaced by the LOD). The values replaced by the LOD/2 were included in both estimations.

Probabilistic approach

The probabilistic or stochastic procedure used was based on the mixed parametric–parametric (MP-P) method reported extensively in Gauchi and Leblanc (2002) and Cano-Sancho et al. (2011). Advantages against non-parametric methods were hence elucidated, the MP-P method leading to more reliable estimations, especially of the high quartiles.

In this methodology, a mixed probability density function (pdf) was fitted to each food consumption, and a parametric pdf was fitted to each food contamination (Gauchi and Leblanc 2002).

The appearance of the consumption histograms was irregular (Figure 3), especially for those foods whose consumption is seasonal or sporadic, such as dessert wine, pistachios or peanuts. Data in the histograms could be divided in two: the numbers of non-consumers and consumers. Thus, a mixed distribution was fitted as follows:

$$U_{\pi_0,j}^{[D]} = \left\{ \left[U\left(0, c_{i_{\min(\pi_0),j}}\right) \right] j, h; \left[\Gamma(r, \lambda, \theta)\right]_{\pi_{0j},} (1-h) \right\} \quad (1)$$

where $[U(0, c_{i_{\min(\pi_0),j}})]$ is the continuous uniform distribution defined on the interval $(0, c_{i_{\min(\pi_0),j}})$ with $c_{i_{\min(\pi_0),j}}$ as the minimal consumption of the foodstuff j, in the sample π_0 (this part corresponds to the non-consumer class). $[\Gamma(r, \lambda, \theta)]_{\pi_{0j}}$ is the gamma or lognormal fitted consumption distribution for the foodstuff j (this part corresponds to the consumer class). $U_{\pi_0,j}^{[D]}$ means a sampling from a discrete uniform distribution: a random number u is drawn from a continuous uniform distribution defined on [0; 1]. If u is less than or equal to h (proportion of the number of consumers), then a new random number u' is drawn from $[U(0, c_{i_{\min(\pi_0),j}})]_j$, otherwise a new random number is drawn from $[\Gamma(r, \lambda, \theta)]_{\pi_{0j}}$.

Food contamination treated censored data (as stated above) were fitted in most cases to the gamma distribution and in some cases to the lognormal distribution for each population group. As in the deterministic estimation, two exposure scenarios were obtained (ND replaced by zero and ND replaced by the LOD).

The MP-P method used to estimate the normalised exposure k of the S simulation set was built as follows:

$$\hat{E}_k^{[MP-P]} = \sum_{j=1}^{p} \tilde{c}_{k(\hat{F}_{Uj}),j} \tilde{t}_{k(\hat{F}_{Tj}),j} \quad (2)$$

where $\tilde{c}_{k(\hat{F}_{Uj}),j}$ is a random normalised consumption for the foodstuff j; the random deviate k is drawn from $\hat{F}_{U,j}$, with the corresponding cumulative distribution function (cdf) adjusted from the distribution consumption defined above. $\tilde{t}_{k(\hat{F}_{Tj}),j}$ is a random contamination for the foodstuff j; the random deviate k is drawn from $\hat{F}_{T_j}$, the fitted gamma cdf of contamination. In the case of wheat flakes, the mean contamination was used in the simulations because not enough data were available for computations.

The mean of normalised exposures over the simulation set S was then estimated using the following equation:

$$\hat{E}_S^{[MP-P]} = \frac{1}{n} \sum_{k=1}^{n} \hat{E}_k^{[MP-P]} \quad (3)$$

where n is the number of random deviates drawn (10 000 in the present study).

Other descriptive statistics were directly computed on the histogram built with the simulations of the S set.

Pseudo-parametric bootstrap confidence intervals were built as reported by Gauchi and Leblanc (2002), by randomly drawing B samples of size n_{π_0} in the exposure simulation set S (with $B = 10{,}000$). The boundaries of the 95% confidence interval were

Table 2. OTA levels ($ng\,g^{-1}$ and $ng\,ml^{-1}$ for beer and wines) in food composites and samples of wines.

Foodstuffs	*n*	Positive samples	Percentage of positive samples	Mean of positives	SD	Median	Minimum	Maximum
Baby foods	69	6	8.7	0.233	0.036	0.225	0.195	0.293
Beer	71	63	88.7	0.022	0.023	0.015	0.004	0.126
Breakfast cereals								
Corn-based	71	2	2.8	0.728	0.764	0.728	0.188	1.268
Wheat/rice-based	28	7	25.0	0.293	0.141	0.270	0.180	0.570
Coffee[a]	72	35	48.6	2.171	0.790	1.960	1.210	4.210
Loaf bread	70	9	12.9	0.283	0.181	0.196	0.162	0.658
Peanuts	72	30	41.7	0.214	0.138	0.173	0.084	0.774
Pistachios	70	2	2.9	0.228	0.133	0.228	0.134	0.321
Red wine[b]	120	18	15.0	0.513	0.807	0.165	0.070	3.190
Dessert wine[c]	141	70	49.6	3.288	6.890	0.797	0.057	48.680

Notes: [a]Data taken from Coronel et al. (2011).
[b]Data taken from Bellí et al. (2004).
[c]Data taken from Bellí et al. (2004) and Valero et al. (2008).

calculated taking the 0.025th and 0.975th empirical quantiles of the final bootstrap distribution.

Statistical program SAS 9.0 (Cary, NC, USA) was used for the probabilistic analysis.

Other statistical analysis

Differences between population groups (sorted by gender and age) were evaluated by means of the non-parametric Wilcoxon and Kruskal–Wallis tests.

Results and discussion

Presence of OTA in foodstuffs sampled in Catalonia

Table 2 shows the occurrence of OTA in the foodstuffs considered in this work; Figures 1 and 2 show the distributions of the contamination by OTA. Data for contamination in loaf bread were corrected for water content, as the measured dry weight was 77% of the fresh samples. The correction factor was then 0.77. Data for contamination in pistachios were corrected for the proportion of shells. It was observed that the edible part comprised 56% of the total weight. Thus, assuming the worst case in which all the detected toxin was in the edible portion, the correction factor was 0.56^{-1}, which equals to 1.79.

With the exception of beer, the frequency of contamination of the analysed foodstuffs was lower than 50%. The mean of positives, as well as maximum values of baby foods, breakfast cereals and loaf bread were below the maximum levels established by the European Commission (2006): 0.50 and $3\,ng\,g^{-1}$, respectively, considering breakfast cereals and loaf bread as products derived from cereals. Still no legislation has been set regarding beer and nuts, but the mean of positives of these foodstuffs were lower than the levels found in the rest of foodstuffs.

As concerns baby foods, the mean contamination of the positive samples was similar to the mean contamination levels found in Turkey ($0.221\,ng\,g^{-1}$; Kabak 2009), but higher than those found in a previous study in Spain in which the mean level was $0.187\,ng\,g^{-1}$ (Araguás et al. 2005). However, two samples in that survey exceeded the European limit of $0.5\,ng\,g^{-1}$ (0.706 and $0.740\,ng\,g^{-1}$), while the maximum level found in the current survey was $0.293\,ng\,g^{-1}$. The observed mean contamination was lower than the found in a study in Canada, where means of positive samples ranged from 0.28 to $2.40\,ng\,g^{-1}$, according to the type of cereal used in the formula (Lombaert et al. 2003). Considering ranges, levels detected in an Italian study were in the range of <0.06–0.74 (Beretta et al. 2002), with four samples above the European limit of $0.5\,ng\,g^{-1}$. In Portugal, levels of OTA in baby foods ranged between 0.034 and $0.212\,ng\,g^{-1}$ (Alvito et al. 2010). The percentage of positive samples in the present study (8.7%) was lower than in the rest of the named studies: Canada (26.1%), Italy (16.8%), Portugal (37%), Spain (70%), and Turkey (17%).

Many studies on the occurrence of OTA in beer have been done. A summary of the sample collections carried out from 1992 to 2007 in several countries of beers from diverse origins was presented by Kabak (2009). Apart from those, there are studies done in Belgium (Anselme et al. 2006), Brazil (Kawashima et al. 2007), and Japan (Kumagai et al. 2008; Aoyama et al. 2010). In most cases, mean OTA levels were below $0.070\,ng\,ml^{-1}$, with the exception of those found in Korea ($0.25\,ng\,ml^{-1}$; Park et al. 2005), a sample of Scottish origin ($0.201\,ng\,ml^{-1}$; Medina et al. 2005), Belgian beers ($0.103\,ng\,ml^{-1}$; Anselme et al. 2006), and non-alcoholic beers purchased in Iranian supermarkets ($0.108\,ng\,ml^{-1}$; Mahdavi et al. 2007). Incidence ranged

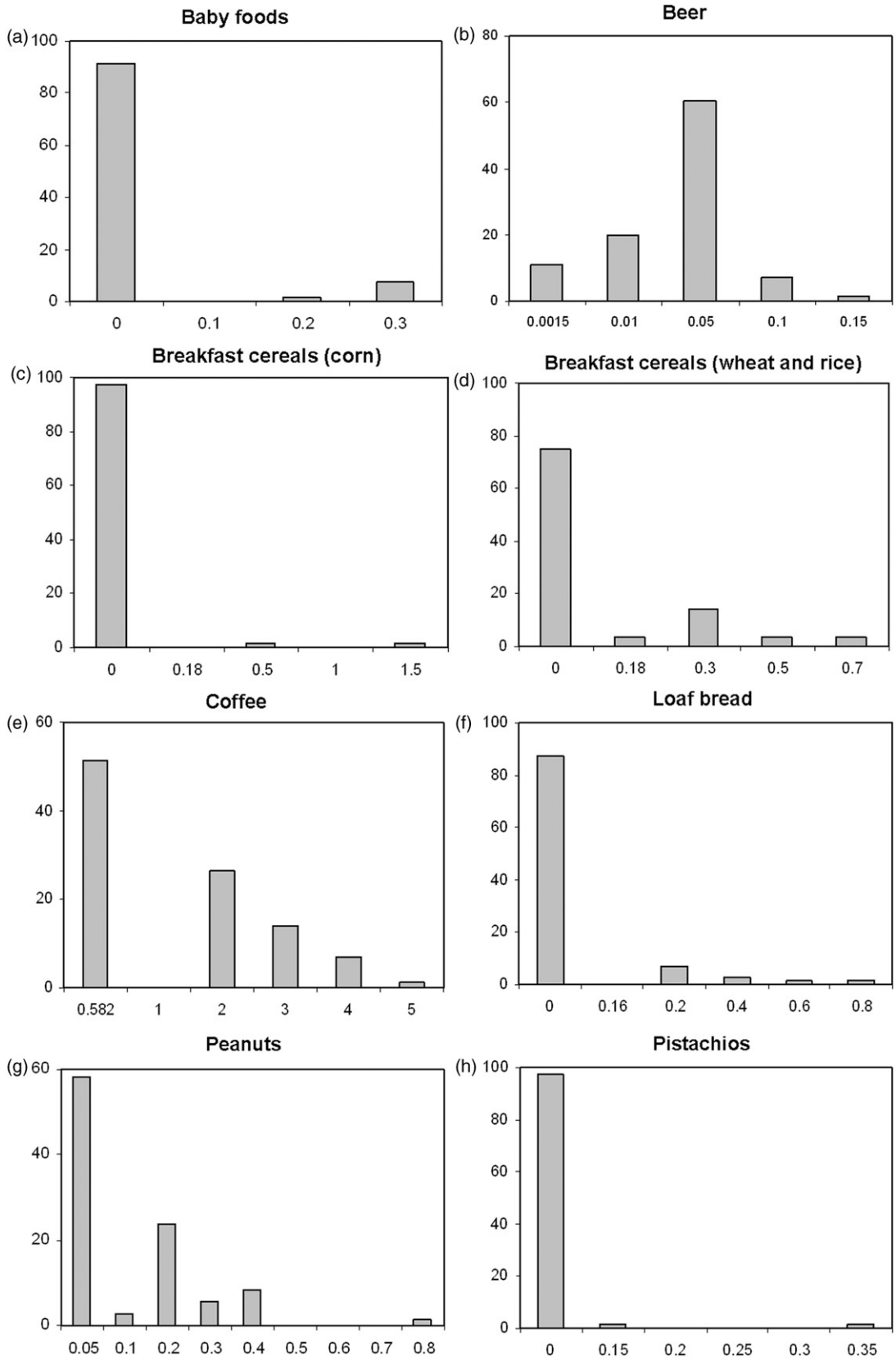

Figure 1. Contamination histograms (relative frequencies versus OTA contamination, ng g^{-1}, and ng ml^{-1} in the case of beer) in food composites. In order to build these plots, non-detected values were replaced by zero (baby foods, breakfast cereals, loaf bread, pistachios) or by the LOD/2 (beer, coffee, peanuts).

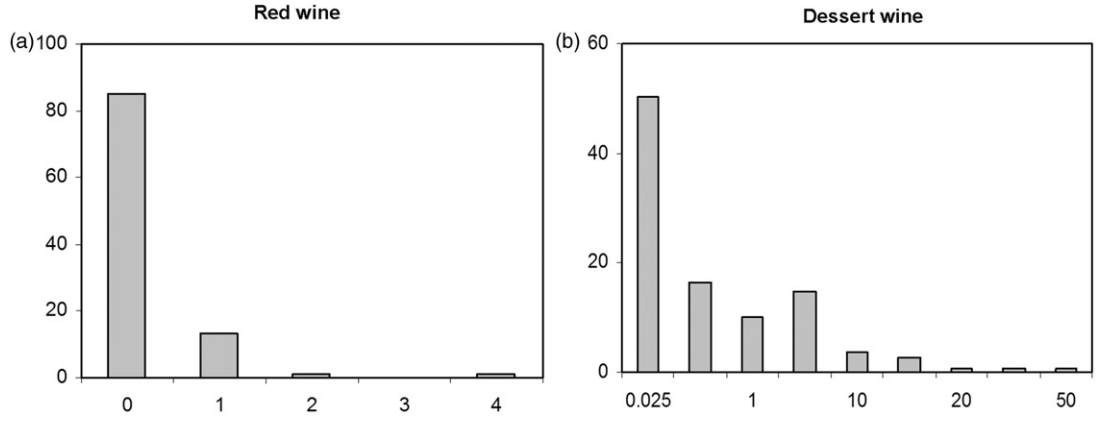

Figure 2. Contamination histograms in wine samples (relative frequencies versus OTA contamination, $ng\,ml^{-1}$). In order to build these plots, non-detected values were replaced by zero (red wine) or by the LOD/2 (dessert wine).

from zero to 100%, but was mostly above 50%. Bertuzzi et al. (2011) analysed 106 beer samples collected in 25 European countries. The incidence was 67.9% and levels ranged between <0.002 and $0.189\,ng\,ml^{-1}$, with a mean of the total samples of $0.019\,ng\,ml^{-1}$. Particularly, we observed that the mean value of this study ($0.022\,ng\,ml^{-1}$) was lower than other means found in Spain of $0.044\,ng\,ml^{-1}$ (Araguás et al. 2005) and $0.0358\,ng\,ml^{-1}$ (Medina et al. 2005). Thus, the mean level of this study was in the range of those found in the literature, and incidence was also similar to the observed in previous studies.

Mean contamination levels of positive samples in corn-based breakfast cereals were around two-fold higher than in the case of wheat and rice-based breakfast cereals, and the same was observed in the median and maximum values of both distributions, although the occurrence was higher in the second case. Roscoe et al. (2008) and Ibáñez-Vea et al. (2011) determined the contamination of OTA and other mycotoxins in breakfast cereals of different compositions sampled in Canada and Spain, respectively. In both cases a higher incidence was also observed in the wheat and rice-based ones, but mean contamination was lower in the corn-based (0.12 and $0.10\,ng\,g^{-1}$, respectively) than in the wheat and rice-based (0.30 and $0.16\,ng\,g^{-1}$, respectively). Both studies showed lower mean contaminations than in this study, which also occurred in samples collected in Greece ($0.18\,ng\,g^{-1}$; Villa and Markaki 2009). Mean contamination of positive samples in the two types of cereals of this study matched with that observed by Araguás et al. (2005) ($0.265\,ng\,g^{-1}$) and Kabak (2009) ($0.752\,ng\,g^{-1}$). In this study, the incidence in both types of breakfast cereals was low, especially in the case of the corn-based. Such incidences were lower than the found in other studies: up to 100% (Araguás et al. 2005), 60% (Villa and Markaki 2009), 18% (corn-based) to 38% (wheat-based) (Roscoe et al. 2008), 5% (corn-based) to 88% (wheat and rice-based) (Ibáñez-Vea et al. 2011).

Regarding bread contamination, Duarte et al. (2010) compiled the occurrence of OTA in different types of bread worldwide. Mean values of wheat bread positive samples ranged from $0.07\,ng\,g^{-1}$ in Switzerland (Legarda and Burdaspal 2001) to $13\,ng\,g^{-1}$ in Morocco (Zinedine, Juan, et al. 2007), although most were below $0.50\,ng\,g^{-1}$. Incidence was between 65% and 100% for most of the listed studies, and in some exceptions it was below 20%. Therefore, the present results were similar to most of the data of previous studies. In addition, when comparing our results with other samples collected in Spain, we could observe that mean values were lower than those found by Legarda and Burdaspal (2001) and Osnaya et al. (2006), with 0.45 and $2.19\,ng\,g^{-1}$, respectively.

Few data are available on OTA contamination of nuts. From these, we could observe that OTA levels in this study were below those found in Tunisia (0.1–3.0, Ghali et al. 2009; and 11–$203\,ng\,g^{-1}$, Zaied et al. 2010). The low incidence of OTA in pistachios observed here (2.9%) was even lower than the observed in the mentioned studies (16% and 25%, respectively), whereas in Morocco, Zinedine, Soriano, et al. (2007) found no contamination above the limit of quantification of their detection method ($0.027\,ng\,g^{-1}$). A higher incidence than in the pistachio samples was observed for peanuts (41.7%), and it was similar to the observed in Tunisia (44%; Ghali et al. 2009) and higher than in the samples analysed in Morocco and in other study in Tunisia (25% and 24%, Zinedine, Soriano, et al. 2007; and Zaied et al. 2010, respectively). Mean of positives were lower than the observed in Côte d'Ivoire ($0.373\,ng\,g^{-1}$; Sangare-Tigori et al. 2006), Morocco ($0.68\,ng\,g^{-1}$, Zinedine, Soriano, et al. 2007), and Tunisia (2.4 and $60\,ng\,g^{-1}$; Ghali et al. 2009; and Zaied et al. 2010, respectively).

Contamination and occurrence data of red and dessert wines and coffee were previously discussed in the articles from which data were taken (Bellí et al. 2004; Valero et al. 2008; Coronel et al. 2011).

Table 3. Percentage of the consumer population for each foodstuff.

	Infants ($n=164$)		Children ($n=68$)		Adolescents ($n=211$)		Adults ($n=905$)	
	Number of consumers	Consumer population (%)	Number of consumers	Consumer population (%)	Number of consumers	Consumer population (%)	Number of consumers	Consumer population (%)
Baby foods	133	81.1						
Beer					44	20.9	586	64.8
Breakfast cereals (corn)			49	72.1	143	67.8	382	42.2
Breakfast cereals (wheat and rice)			49	72.1	143	67.8	383	42.3
Coffee			0	0	93	44.1	767	84.8
Loaf bread			38	55.9	140	66.4	440	48.6
Peanuts			0	0	84	39.8	407	45.0
Pistachios			0	0	71	33.6	460	50.8
Red wine					26	12.3	597	66.0
Dessert wine					25	11.8	184	20.3
Total	133	81.1	55	80.9	201	95.3	900	99.4

The comparison of the present results with other works indicates that most of the values found were similar or lower than previously published data. Figures 1 and 2 show the histograms for contamination levels of the foodstuffs listed in Table 2. In all cases it can be observed that the shape of the distributions does not show any pattern in common, and that for most of the samples the levels of contamination were non-detectable.

Consumption of foodstuffs

Table 3 shows the proportion of the total population, classified by age groups, of the people who consumed the studied foods, sorted by each kind of foodstuff. In the case of infants, even though there was only one surveyed food product, a high percentage of consumers could be observed. FFQ for children included the same foodstuffs as for adolescents and adults (except alcoholic beverages), however, this age group showed no consumption of nuts or coffee. Clear differences were observed in the percentages of consumers of each foodstuff when comparing the age groups. For example, in the case of breakfast cereals and loaf bread, the consumer percentage decreased as the age increased. These foodstuffs presented the highest percentages of consumers in the groups of children and adolescents. Regarding adults, the highest consumer percentages were observed for beer, coffee and red wine.

Further information about the normalised quantities of food consumption is listed in Tables 4 and 5 for total population and the consumer population.

As an example, histograms of consumption for each foodstuff by the adults are shown in Figure 3. The shapes of these histograms were irregular, and showed the proportion of consumers and non-consumers.

Estimation of the daily intake of OTA

Deterministic estimation

The results of the mean contamination values derived from the treatment of the ND data are listed in Table 6. These alternatives were used in the calculation of the OTA daily intake of each foodstuff, and results of the estimations for the total population are shown in Table 7.

The total population (consumers and non-consumers) of each age group was considered in this analysis, as the percentage of consumers in all cases was high (above 80%), and in the case of adolescents and adults above 95%. Another reason was to obtain results to make possible the comparison with the probabilistic estimation, in which the percentages of consumers and number of consumers were taken into account. An example of the shapes of the estimated daily intake distribution obtained by the deterministic method is shown in Figure 4, where adult exposure in the two exposure scenarios (ND = 0 and ND = LOD) is described.

In the best-case exposure scenario (ND = 0), adults presented the highest mean daily intake, followed by infants, adolescents and children. In the worst-case scenario (ND = LOD), the highest mean was observed for infants, followed by adults, children and adolescents. The observed increase in the descriptive values was not proportional for each age group; instead they depended on the values of the LOD of the different methods of analysis of the considered foodstuffs (consumption of each age group was the same for both estimations). However, results agreed with the fact that the most exposed groups were infants and adults.

In all groups, mean and median estimated OTA daily intakes due to the consumption of the studied

Table 4. Descriptives of the normalised consumption ($g\,kg^{-1}\,bw\,day^{-1}$) of foodstuffs by population groups, considering all the surveyed population.

All population	Mean	SD	Median	99th quantile	Mean	SD	Median	99th quantile
	Infants ($n=164$)				Children ($n=68$)			
Baby foods	12.65	12.20	10.00	49.13				
Breakfast cereals (corn)					0.71	0.81	0.26	3.26
Breakfast cereals (wheat and rice)					0.71	0.81	0.26	3.26
Loaf bread					0.53	0.83	0.08	3.00
Total consumption	12.65	12.20	10.00	49.13	1.94	1.80	1.86	7.21
	Adolescents ($n=211$)				Adults ($n=905$)			
Beer	0.19	0.71	0.00	3.34	1.30	2.20	0.38	9.63
Breakfast cereals (corn)	0.26	0.40	0.08	2.05	0.10	0.19	0.00	0.60
Breakfast cereals (wheat and rice)	0.26	0.40	0.08	2.05	0.10	0.19	0.00	0.60
Coffee	0.03	0.07	0.00	0.30	0.14	0.14	0.11	0.56
Loaf bread	0.36	0.62	0.07	2.78	0.22	2.18	0.00	1.74
Peanuts	0.05	0.19	0.00	0.82	0.03	0.15	0.00	0.57
Pistachios	0.03	0.12	0.00	0.78	0.03	0.19	0.00	0.48
Red wine	0.05	0.34	0.00	0.98	0.93	1.94	0.22	7.66
Dessert wine	0.02	0.17	0.00	0.46	0.03	0.20	0.00	0.41
Total consumption	1.25	1.54	1.07	6.02	2.89	4.07	1.83	13.88

Table 5. Descriptives of the normalised consumption ($g\,kg^{-1}\,bw\,day^{-1}$) of foodstuffs by population groups, considering the consumer population.

Only consumers	Mean	SD	Median	99th quantile	Mean	SD	Median	99th quantile
	Infants ($n=133$)				Children ($n=55$)			
Baby foods	133.00	15.60	15.00	50.08				
Breakfast cereals (corn)					0.98	0.81	1.00	3.37
Breakfast cereals (wheat and rice)					0.98	0.81	1.00	3.37
Loaf bread					0.94	0.93	0.54	3.00
Total consumption	133.00	15.60	15.00	50.08	2.40	1.70	2.22	7.32
	Adolescents ($n=201$)				Adults ($n=900$)			
Beer	0.91	1.34	0.48	5.89	2.01	2.47	1.14	11.39
Breakfast cereals (corn)	0.38	0.44	0.24	2.43	0.24	0.22	0.18	0.85
Breakfast cereals (wheat and rice)	0.38	0.44	0.24	2.43	0.24	0.22	0.18	0.85
Coffee	0.07	0.09	0.02	0.35	0.16	0.14	0.13	0.59
Loaf bread	0.54	0.70	0.20	2.85	0.46	3.12	0.12	2.43
Peanuts	0.12	0.29	0.03	1.16	0.08	0.22	0.02	1.16
Pistachios	0.10	0.19	0.05	0.94	0.06	0.26	0.02	0.57
Red wine	0.43	0.90	0.12	3.65	1.42	2.25	0.75	7.88
Dessert wine	0.21	0.48	0.03	1.88	0.14	0.43	0.04	2.44
Total consumption	1.32	1.56	1.12	6.13	2.91	4.07	1.86	13.88

foodstuffs were below the latest PTDIs for both exposure scenarios, and the same was observed for the high quantiles: values reached 14% of the PTDI of $14\,ng\,kg^{-1}\,bw\,day^{-1}$ and 11% of the PTDI of $17\,ng\,kg^{-1}\,bw\,day^{-1}$, respectively, excluding infants. The highest values were observed for the high quantiles in the infants group, especially in the ND = LOD scenario and these were closer to the PTDI (65% and 53% of the mentioned PTDIs). Even though the contamination of the baby foods was similar to the foods consumed by the rest of the population (Table 6), it is worth noting that infants have a less varied diet than the rest of the population, and thus the contribution of a specific foodstuff to the total diet (in this case baby foods) may be higher when comparing it with the dietary habits of the other age groups.

Differences among age groups were tested in total and by pairs by taking into account the data of the

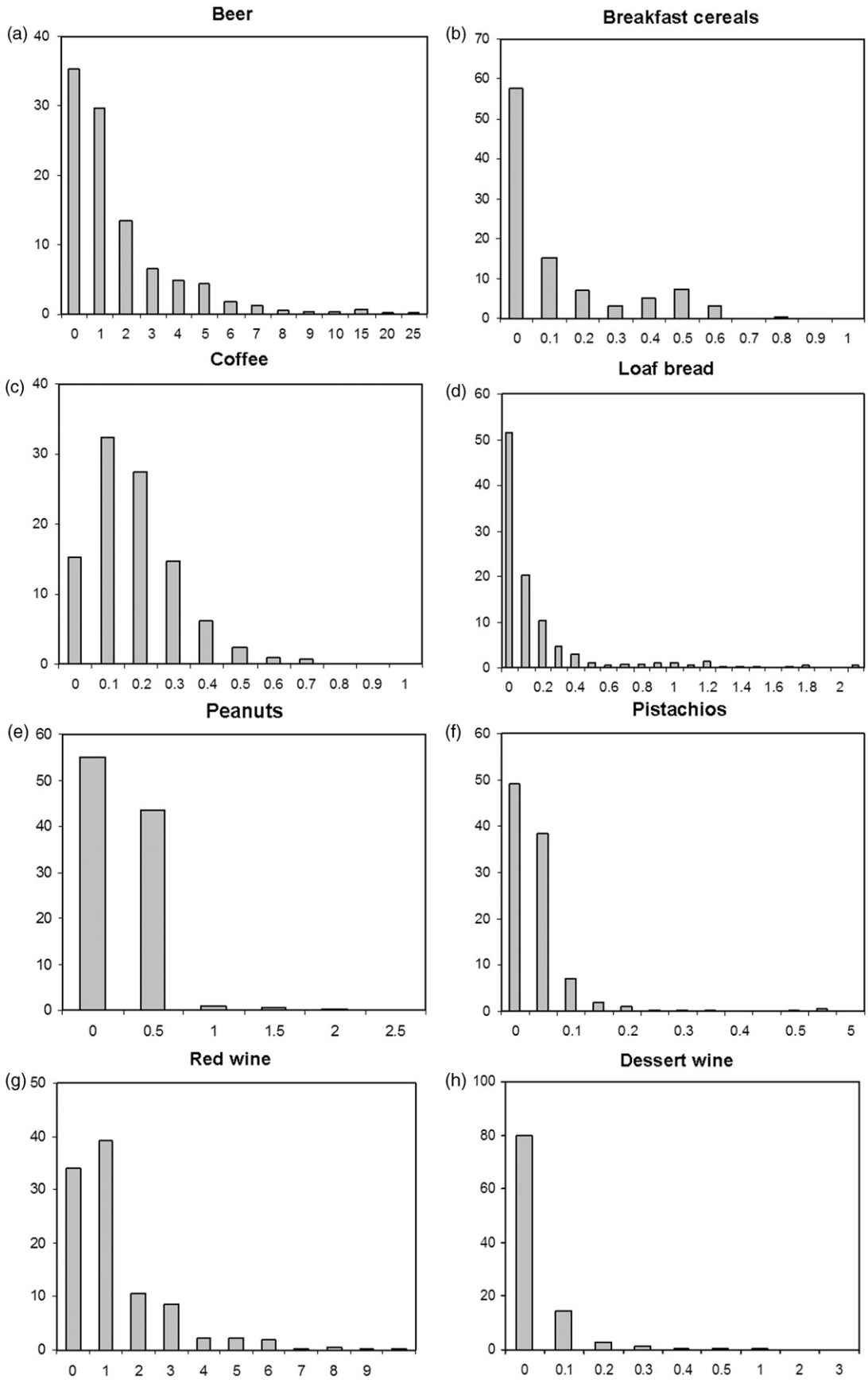

Figure 3. Consumption histograms (relative frequencies) of the adult population for each foodstuff ($g\,kg^{-1}\,bw\,day^{-1}$).

Table 6. Mean values of OTA contamination in different foodstuffs derived from treated ND data used to perform the deterministic estimation.

			Censored data	Mean contamination, ND replaced by:		
	n	% ND	Treatment[a]	LOD/2	ZERO	LOD
Baby foods	69	91.3	C	–	0.020	0.185
Beer	71	11.3	A	0.020	–	–
Breakfast cereals						
Corn-based	71	97.2	C	–	0.020	0.195
Wheat/rice-based	28	75.0	C	–	0.073	0.208
Coffee	72	51.4	A	1.354	–	–
Loaf bread	70	87.1	C	–	0.036	0.158
Peanuts	72	58.3	A	0.110	–	–
Pistachios	70	97.1	C	–	0.007	0.132
Red wine	120	85.0	C	–	0.077	0.119
Dessert wine	141	50.4	A	1.639	–	–

Note: [a]According to criteria described in the Materials and methods.

Table 7. Deterministic estimation of the daily intake of OTA (ng kg^{-1} bw day^{-1}) for the total surveyed population.

	ND = 0	ND = LOD	ND = 0	ND = LOD
	Infants		Children	
n	164	164	68	68
Mean	0.25	2.34	0.08	0.37
SD	0.24	2.26	0.08	0.35
Median	0.20	1.85	0.07	0.32
0.90th quantile	0.51	4.69	0.19	0.81
0.95th quantile	0.73	6.79	0.19	0.84
0.99th quantile	0.98	9.09	0.33	1.42
Skewness	1.07	1.07	1.05	1.05
Kurtosis	1.17	1.17	1.26	1.25
	Adolescents		Adults	
n	211	211	905	905
Mean	0.16	0.29	0.47	0.57
SD	0.40	0.45	0.53	0.68
Median	0.06	0.21	0.39	0.47
0.90th quantile	0.33	0.63	0.87	1.01
0.95th quantile	0.58	0.81	1.03	1.22
0.99th quantile	1.24	1.42	1.76	1.91
Skewness	8.08	6.73	7.30	9.36
Kurtosis	80.82	63.08	84.49	125.55

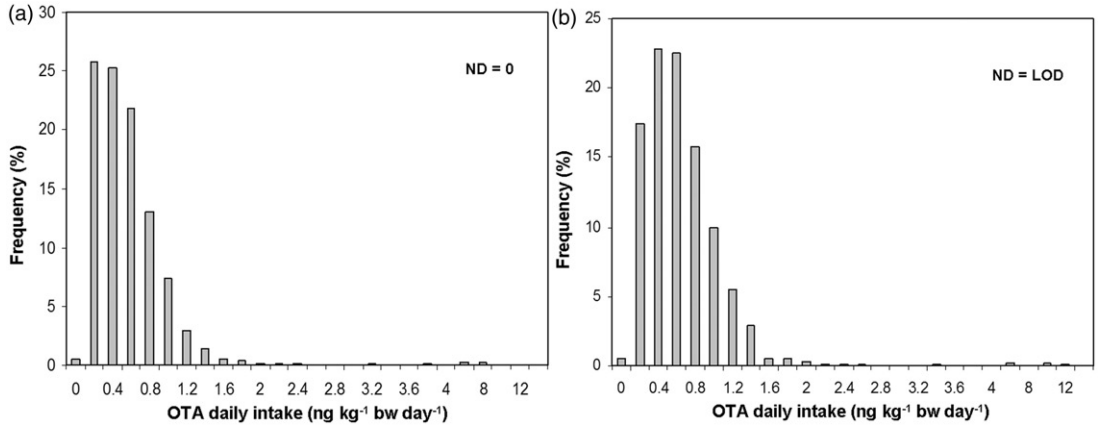

Figure 4. Histograms of the OTA daily intake by the adult population estimated by the deterministic method, in the ND = 0 and ND = LOD scenarios.

Table 8. Probabilistic estimation of the daily intake of OTA and confidence intervals of the descriptive statistics.

	OTA daily intake ($ng\,kg^{-1}\,bw\,day^{-1}$)				Confidence intervals			
	ND = 0	ND = LOD	ND = 0	ND = LOD	ND = 0	ND = LOD	ND = 0	ND = LOD
	Infants		Children		Infants		Children	
n	10,000	10,000	10,000	10,000				
Mean	0.28	2.42	0.09	0.39	[0.14; 0.47]	[2.05; 2.81]	[0.06; 0.14]	[0.32; 0.46]
SD	1.11	2.48	0.16	0.30	[0.44; 2.07]	[2.03; 2.97]	[0.07; 0.39]	[0.24; 0.40]
Median	0.0012	1.77	0.05	0.32	[0.0003; 0.0042]	[1.34; 2.19]	[0.03; 0.07]	[0.24; 0.40]
0.90th quantile	0.58	5.74	0.21	0.80	[0.24; 1.06]	[4.72; 6.74]	[0.14; 0.33]	[0.63; 1.03]
0.95th quantile	1.46	7.23	0.30	0.98	[0.65; 2.77]	[5.85; 8.87]	[0.19; 0.55]	[0.75; 1.27]
0.99th quantile	4.94	11.00	0.68	1.39	[2.23; 12.33]	[8.18; 15.34]	[0.29; 3.20]	[0.97; 2.36]
Skewness	9.17	1.69	8.40	1.40				
Kurtosis	129.70	4.15	128.13	3.08				
	Adolescents		Adults		Adolescents		Adults	
n	10,000	10,000	10,000	10,000				
Mean	0.14	0.28	0.37	0.53	[0.10; 0.24]	[0.23; 0.36]	[0.30; 0.45]	[0.46; 0.60]
SD	0.46	0.45	0.53	0.48	[0.12; 1.12]	[0.18; 0.23]	[0.31; 0.93]	[0.34; 0.81]
Median	0.07	0.20	0.23	0.41	[0.05; 0.08]	[0.17; 0.23]	[0.19; 0.27]	[0.35; 0.47]
0.90th quantile	0.28	0.52	0.79	1.05	[0.21; 0.38]	[0.44; 0.62]	[0.62; 1.00]	[0.87; 1.21]
0.95th quantile	0.44	0.68	1.14	1.31	[0.30; 0.61]	[0.54; 0.85]	[0.83; 1.54]	[1.07; 1.58]
0.99th quantile	1.04	1.37	2.39	2.14	[0.58; 5.39]	[0.81; 5.36]	[1.46; 5.71]	[1.51; 3.87]
Skewness	25.56	15.22	6.86	4.42				
Kurtosis	1015.77	364.75	86.05	50.95				

total population (consumers and non-consumers). These were significant in all cases ($p < 0.0001$, both for ND = 0 and ND = LOD), but not when comparing children and adolescents ($p = 0.4882$ for ND = 0 and $p = 0.1083$ for ND = LOD). In a further analysis, the adult population group was divided into three subgroups of age: group A (18–29 years old, $n = 314$), group B (30–44 years old, $n = 308$), and group C (≥45 years old, $n = 283$). Significant differences were also found among these groups ($p < 0.0001$), with medians of 0.30, 0.42 and $0.45\,ng\,kg^{-1}\,bw\,day^{-1}$, respectively (for ND = 0), and 0.42, 0.47 and $0.52\,ng\,kg^{-1}\,bw\,day^{-1}$, respectively (for ND = LOD). When analysing differences between age groups by pairs, differences were significant between groups A and B ($p = 0.0006$ for ND = 0 and $p = 0.0362$ for ND = LOD) and A and C ($p < 0.0001$ for ND = 0 and $p = 0.0001$ for ND = LOD).

Gender was also considered to evaluate differences in the exposure of the adolescent and adult population. Thus, total OTA daily intake by adolescents showed no significant differences ($p = 0.4229$ for ND = 0 and $p = 0.3720$ for ND = LOD), whereas in adults differences were indeed significant ($p = 0.0175$ for ND = 0 and $p = 0.0027$ for ND = LOD), men presenting a higher median intake (0.41 and $0.50\,ng\,kg^{-1}\,bw\,day^{-1}$ for each scenario) than women (0.36 and $0.44\,ng\,kg^{-1}\,bw\,day^{-1}$).

Taking into account the significant differences between genders in the adult population, it could be expected that such differences were found within the age groups. Therefore, differences were analysed in the same gender, for age groups and by pairs. For men, differences were significant between groups A and B ($p = 0.0182$ for ND = 0 and $p = 0.1871$ for ND = LOD) and A and C ($p = 0.0021$ for ND = 0 and $p = 0.0077$ for ND = LOD). Similarly, in the case of women, differences were significant between groups A and B ($p = 0.0206$ for ND = 0 and $p = 0.1734$ for ND = LOD) and A and C ($p < 0.0001$ for ND = 0 and $p = 0.0057$ for ND = LOD). It could be observed that when testing differences between groups A and B in men and women, differences were significant when ND = 0, but not when ND = LOD.

Probabilistic estimation and confidence intervals

Table 8 shows the results of the probabilistic estimation of the daily intake of OTA in the different population age groups for the two alternatives of replacement of the ND values.

Mean values were similar to those obtained by the deterministic estimation, slightly higher for infants and children, and slightly lower for adolescents and adults. As regards the medians, all were slightly lower, and an especial case could be observed for the infant population in which the median was almost equal to zero. Taking into account the high quantiles, in most of the cases these were higher than the deterministic estimations, but in all cases OTA daily intake was lower than the latest PTDIs for both exposure scenarios (ND = 0 and ND = LOD). Infant population was the most exposed group, too.

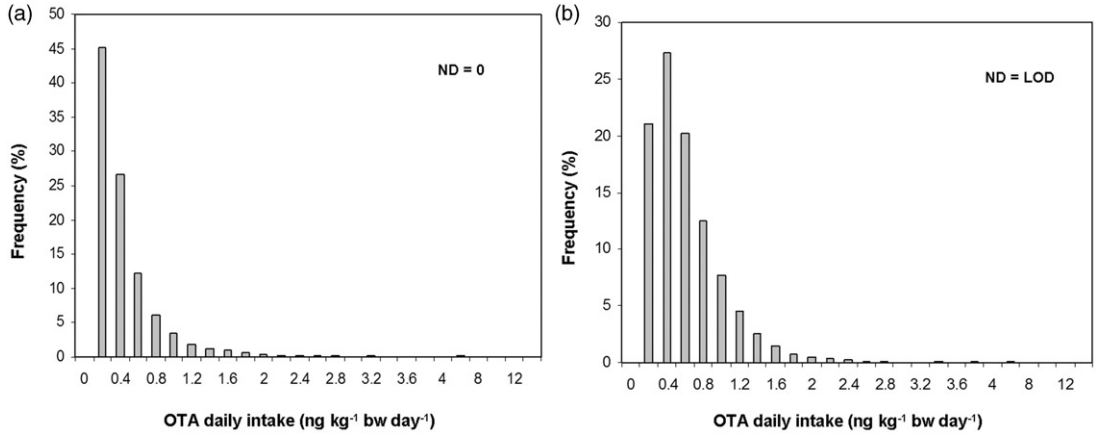

Figure 5. Histograms of the OTA daily intake by the adult population estimated by the probabilistic method, in the ND = 0 and ND = LOD scenarios.

Regarding the analysis of differences between population groups, it was performed for the same groups and pairs as in the deterministic estimation. Thus, in all cases differences were significant and in all cases $p < 0.0001$, with only one exception: differences between age groups B and C in men were not significant ($p = 0.1365$) in the ND = 0 scenario.

Figure 5 shows the distribution of the exposure obtained through the probabilistic method for the adult population. The shape of the distribution is much more defined than that obtained by the deterministic approach. This can be explained by the number of samples included in the analysis ($n = 10{,}000$) and the model used to perform the simulations. In this case the simulation process achieved exposure values that were not obtained by the deterministic estimation, as the probability to obtain a higher number of different exposures cases was higher, which is confirmed by the high values of skewness and kurtosis. The above-mentioned characteristics make possible the calculation of confidence intervals of the estimated distribution descriptives. These confidence intervals are listed in Table 6; they complete the description of the probabilistic estimation. In all cases but one the estimated daily intake values shown in Table 6 remained within the range defined by the confidence intervals. The range of the intervals increased in the highest quantiles, which was expected as the precision in the estimation of the confidence intervals of these quantiles is lower than in the case of mean or median values (Breiman et al. 1990; Beirlant and Devroye 1999).

When checking the deterministic estimations against the probabilistic confidence intervals, it could be observed that all but seven values were within these ranges. Taking into account that in addition similar descriptive statistics were obtained through the deterministic method, we can state that the probabilistic method is a useful tool for the estimation of the exposure descriptives and the obtainment of precision indicators related to those values.

Conclusions

The present work assessed the exposure of the Catalan population to OTA by determining the contamination levels of certain foodstuffs sampled in Catalonia, and by considering data of consumption for this population. The levels of contamination by OTA of the sampled foodstuffs were below the limits established by the European Commission; and the consumer population was almost the total surveyed population, which indicates that although the contamination levels were low, a major part of the population was exposed to OTA. However, a more complete exposure assessment could be reached by the inclusion of other foodstuffs previously shown to be contaminated by OTA, raisins and spices, or animal by-products, which were not included in this study due to the lack of consumption data. The analysis of pasta would have added important information to this study, but unfortunately this food product was not selected for sampling.

Exposure was quantified by the estimation of the daily intake of OTA, which was achieved by deterministic and probabilistic methods. Both estimations showed that the exposure levels were lower than the PTDIs, but differences among population groups were confirmed and still differences among adult population subgroups were also found. However, non-significant differences between population groups could be observed only when the deterministic data were analysed for differences. We observed that the probabilistic estimation gave similar results to those obtained by the deterministic methodology, but had the additional feature of the calculation of confidence intervals for the estimated descriptive values of the exposure distributions.

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