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	the P-P simulation method, the Catalonian population should be expected to be exposed at moderated levels of deoxynivalenol, the infants and individuals with ethnic dietary patterns being the most exposed population groups

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Quantitative dietary exposure assessment of the Catalonian population (Spain) to the mycotoxin deoxynivalenol

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Abstract: The mycotoxin deoxynivalenol (DON) is one of the most common contaminants of cereals worldwide, and its occurrence has been widely reported in raw foods and foodstuffs, around the European region, including Catalonia. In the present work, a stochastic methodology has been applied to accurately assess the exposure of the Catalonian population (Spain) to DON through food consumption. Raw contamination data was provided by a large survey conducted in this region, in addition to the raw consumption data from a nutritional study specifically designed to assess the dietary intake of the main foodstuffs related to DON contamination for all population age groups. Contamination and consumption data were combined by simulation using an essentially parametric (P-P) method. The P-P method draws sampling values from distribution functions fitted to consumption and contamination datasets. Moreover, to quantify the accuracy and reliability of the statistics estimates, we built the related confidence intervals using a Pseudo-Parametric bootstrap method. Considering the

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3 results drawn from the P-P simulation method, the Catalonian population should be expected
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5 to be exposed at moderated levels of deoxynivalenol, the infants and individuals with ethnic
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7 dietary patterns being the most exposed population groups
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12 **Keywords:** Risk assessment; exposure; deoxynivalenol; cereal food
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15 16 17 **Introduction**

18
19 Trichothecenes are a family of related cyclic sesquiterpenoids, which are divided into four
20
21 groups (types A–D) according to their characteristic functional groups, the type A and B
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23 being the most common. Type A is represented by HT-2 toxin (HT2) and T-2 toxin (T2) and
24
25 type B is most frequently represented by deoxynivalenol (DON). Trichothecenes are produced
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27 in several cereals by species of *Fusarium*, thus a wide range of cereal-based foods have been
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29 reported to be contaminated by these toxins (JECFA 2001), DON being one of the most
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31 common contaminants of cereals worldwide (Jelinek et al. 1989; Scott 1989).
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39 Acute effects of food poisoning by DON in humans are abdominal pains, dizziness, headache,
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41 throat irritation, nausea, vomiting, diarrhoea, and blood in stool (Rotter et al. 1996). A
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43 tolerable daily intake (TDI) of 1 µg/kg body weight based on a reduction of body weight gain
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45 was established by the EC SCF (SCF 2002).
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51 Occurrence of DON has been widely reported in raw foods and foodstuffs in European
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53 countries (JECFA 2001), confirming that food processing methods do not completely remove
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55 mycotoxins (Hazel and Patel 2004). The presence of trichothecenes in Catalonia (Spain) has
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57 been recently studied by means of a thorough methodology involving a large sampling and
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59 accurate chemical analysis. That study concluded that the occurrence of DON was high in
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3 cereal-based foodstuffs from Catalonian market, especially in wheat flakes, corn flakes, corn
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5 snacks, pasta and bread (Cano-Sancho et al. 2011). Cereal-based foods are the base of the
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7 energy intake in Catalonia, as well as in the other Mediterranean countries (Serra-Majem et al.
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9 2003). Therefore, in the food safety framework, to assess the exposure of Catalonian
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11 population to DON is a priority.
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17 To our knowledge, few studies have been published to assess the exposure to DON, and the
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19 ones that have been published have generally used deterministic approaches (SCOOP 2003).
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21 Experts have recommended the use of a stochastic approach to provide a more realistic
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23 exposure assessment, taking model uncertainties and variability into consideration.
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25 Nevertheless, there is no consensus on which specific methodology should be applied in each
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27 case (Kroes 2002; WHO 2005; EFSA 2006; Verger and Fabiansson 2008). A stochastic
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29 methodology was developed by Gauchi and Leblanc (2002) to assess the exposure of human
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31 populations to food contaminants, as in the case of the exposure of the French population to
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33 ochratoxin A (OTA). The authors proposed two simulation approaches to estimate the
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35 exposure, based on Monte Carlo simulations (using NonParametric-NonParametric and
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37 Mixed Parametric-Parametric methods), leading to the assessment of four types of bootstrap
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39 confidence intervals. The aim of this paper is to quantitatively assess the exposure of the
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41 Catalonian population (Spain) to DON.
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50 **Materials and Methods**

51 *Raw contamination data*

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53 Raw contamination data was mainly taken from the study of Cano-Sancho et al. (2011), and
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55 completed with some data from the Project to Assess the Exposure of the Catalonian
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57 Population to Mycotoxins (UdL-ACSA 2010).
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3 In the first work, during the months of June and July 2008, corn flakes (n=168), wheat flakes
4 (n=27), sweet corn (n=185), corn snacks (n=213), pasta (n=201), beer (n=213), sliced bread
5 (n=147), bread (n=31), gluten-free foods (n=12), ethnic foods (n=35) and baby foods (n=90)
6 were obtained in six hypermarkets and supermarkets from twelve main cities (Tortosa,
7 Tarragona, Reus, Vilanova i la Geltrú, l'Hospitalet de Llobregat, Barcelona, Terrassa,
8 Sabadell, Mataró, Girona, Manresa and Lleida) of Catalonia, Spain, representative of 72 % of
9 the population. From each supermarket or hypermarket, 3 items (if present) of each product
10 were randomly taken. The level of trichothecenes was determined in a total of 72 composite
11 samples obtained by pooling the 3 items taken from each store if available (12 cities x 6 stores
12 / city = 72 samples / category). However, in some cases, no items were available in the store,
13 thus, less than 72 composites could be obtained. Finally, a wide range of brands was obtained,
14 which can be considered the majority of market share in Catalonia of these products, as well
15 as in the rest of the Spanish market. Moreover, some commodities were selected because they
16 are highly consumed by some population groups with different dietary patterns: baby foods,
17 ethnic foods and gluten-free foods. DON was determined in breakfast cereals, snacks and
18 pasta samples following extraction, clean-up, derivatization and final analysis by GC-ECD.
19 Moreover, this mycotoxin was determined in sliced bread, sweet corn, beer, baby food, ethnic
20 food and gluten-free food by LC-DAD. In this study, non-detected samples were assumed to
21 be equal to the limit of detection (LOD) divided by 2, an assumption widely recognised to
22 reduce the uncertainty of values between 0 and the LOD (GEMs/Food-WHO, 1995). Some
23 statistics from contamination data and the related histograms are shown in Table I and Fig. 1,
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Message to the editor: Place Table I and Fig.1 approximately here.

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[Table I. Occurrence of deoxynivalenol in food matrices from food available on the
Catalonian market]

[Fig 1. Contamination histograms (relative frequencies), in $\mu\text{g g}^{-1}$ for each food]

Raw consumption data

Consumption data were taken from the Project to Assess the Exposure of the Catalonian Population to Mycotoxins (UdL-ACSA 2010). This study involved a wide nutritional survey designed to specifically identify the dietary patterns of the Catalonian population in relation to the main foodstuffs susceptible to mycotoxins contamination.

Food dietary intake was assessed through a specific Food Frequency Questionnaire (FFQ), developed for the Catalonian population including those foods typically consumed in the region that may be potentially contaminated with these mycotoxins. According to World Health Organization (WHO) recommendations, studies to assess the dietary intake of chemical contaminants should show the significant intake within the standard population, as well as within all population groups that could have different dietary patterns. Therefore, five different age population groups were considered: infants (0-3 years), children (4-9 years), adolescents (10-19 years), adults (20-65 years) and seniors (> 65 years). Moreover, adult celiac sufferers and adults with ethnic dietary pattern were included in the nutritional study, and studied separately. The FFQ included 38 specific food items known to be the major foods contaminated by mycotoxins worldwide, excluding those foods not consumed in the region. Concerning consumption frequency, five response options, ranging from 'never' to 'annually', were considered. Quantities were assessed by portion size with the aid of a series of colour photograph models. Finally, 76 seniors, 720 adults, 235 adolescents, 69 children, 164 parents of infants, 70 adult celiac sufferers and 56 adults with ethnic dietary pattern were interviewed in 2008-2009 by trained interviewers. In the present study, the adults were treated separately

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3 according to their sex, with a major population group of 336 adult males and another of 384
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5 adult females. Individuals were from 89 cities and towns from Catalonia. An example of some
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7 statistics from the consumption data of adult females and the related histograms is shown in
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9 Table II and Fig. 2, respectively. They are in the line of those obtained for males.
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15 *Message to the editor: Place Table II and Fig.2 approximately here.*

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17 [Table II. Normalised consumption of the main foodstuff related to DON contamination by the
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19 384 adult females (g kg^{-1} body weight day^{-1}).]
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22 [Fig 2. Consumption histograms (relative frequencies) for adult female consumers, in $\mu\text{g kg}^{-1}$
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24 bw day^{-1}]
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29 Dependencies on consumption patterns can be quantified by Spearman correlation
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31 coefficients and can be taken into account by the Iman and Conover method (Iman and
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33 Conover 1982). However, Gauchi and Leblanc (2002) did not report significant differences in
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35 the results, regardless of whether or not dependencies were taken into account. We therefore
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37 did not consider dependencies in the present work.
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42 43 *Methodology used to calculate the exposure*

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45 In this section we recall some general aspects concerning the exposure calculations, already
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47 given in Gauchi and Leblanc (2002).
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51 The main limitation to estimate the normalised exposure distribution of consumer populations
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53 is derived from the equation:
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$$55 \quad E_{\pi} = \sum_{j=1}^p C_{\pi,j} T_j \quad (1)$$

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57 where the random variable “normalised global exposure in a consumer population π ” (E_{π}) is
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59 a function of the random variables: “normalised consumption of the foodstuff j in a consumer
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3 population π ” ($C_{\pi, j}$) and “DON concentration level of the foodstuff j ” (T_j). The difficulty is
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5 due to the probability density functions (*pdf*) since these variables are generally unknown and
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7 even if they are known, they are generally different and not independent. Moreover, the $C_{\pi, j}$
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9 $\times T_j$ *pdf* products are unknown, too.

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11 If we assume independency between consumption ($C_{\pi, j}$) and contamination (T_j), as well as
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13 between their products, we can estimate the mean exposure of the population π with the
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15 population sample π_0 as follows:

$$\hat{E}_{\pi_0} = \sum_{j=1}^p \bar{C}_{\pi_0, j} \bar{T}_j \quad (2)$$

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17 Where $\bar{C}_{\pi_0, j}$ is the arithmetical mean of the normalised consumption of the foodstuff j in the
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19 population group π_0 , and \bar{T}_j is the arithmetical mean of the available contamination data of
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21 the related food.

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23 Under these assumptions we can also calculate the variance estimate $\hat{V}(E_{\pi_0})$ using Eq. (5)
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25 and Eq. (6) in Gauchi and Leblanc (2002). We could consider this approach to be a theoretical
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27 approach, but because of the postulated assumptions, it would have to be a (very) simplified
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29 theoretical approach. For example, this simplified approach does not enable us to calculate
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31 complicated statistics such as high quantiles. Simulation methods would therefore be required
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33 to obtain estimates of these statistics. Finally, we will consider this approach to be a direct
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35 approach.

50 51 *Stochastic procedure to calculate the exposure: Parametric-Parametric (P-P) method*

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53 In this section we recall some elements of the methodology used to calculate the exposure by
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55 means of the stochastic procedure, already given in Gauchi and Leblanc (2002).

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57 The main advantage of the use of a parametric method is derived from the fact that
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59 NonParametric-NonParametric method could lead us to less reliable estimations, especially of
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3 the high quantiles (Gauchi and Leblanc, 2002). Taking the asymmetrical appearance of
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5 consumption and contamination histograms and our previous experience into account, we
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7 fitted the probability density functions, choosing the gamma distribution to be the best
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9 candidate. Therefore, for each consumption and contamination dataset, the gamma *pdf* was
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11 fitted with the method of maximum likelihood using the CAPABILITY procedure of SAS
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13 software (SAS 2010). The Chi-Square statistic was used in the goodness-of-fit test,
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15 considering a significance level greater than 95%, to accept the distribution as a suitable
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17 candidate. Moreover, several graphical methods such as quantile-quantile plot exist, making it
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19 possible to quickly check if the fitted distribution is adapted to the selected hypothesis (Smut
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21 et al. 2000). Shape and scale parameters were estimated for each contamination set, in
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23 accordance with the methodology described above.
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29 Concerning consumption datasets, the foodstuffs selected are commonly consumed within the
30
31 Catalonian population, despite several exceptions related to age-group dietary patterns. The
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33 appearance of the histograms is therefore continuous and only one distribution was considered
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35 pooling all age groups data, fitting all data to gamma *pdf*. In case of gluten-free foods, the
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37 mean contamination was used in the simulations because few data were available.
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39

40 The *P-P* method used to estimate the normalised exposure k of the S simulation set was built
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42 as follows:
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$$45 \hat{E}_k^{[P-P]} = \sum_{j=1}^P \tilde{c}_{i(j)} \tilde{t}_j \quad (4)$$

46 where $\tilde{c}_{i(j)}$ is a random normalised consumption i for the foodstuff j , drawn from \hat{C}_j , the
47
48 corresponding adjusted gamma *pdf*, and \tilde{t}_j is a random contamination for the foodstuff j ,
49
50 drawn from \hat{T}_j , the corresponding adjusted gamma *pdf*. The density parameter estimates for
51
52 \hat{C}_j and \hat{T}_j are shown in Table V.
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3 The mean of normalised exposures over the simulation set S was then estimated using the
4 following equation, where n was the number of random deviates drawn (10,000 in the present
5 study):
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$$\hat{E}_S^{[P-P]} = \frac{1}{n} \sum_{k=1}^n \hat{E}_k^{[P-P]} \quad (5)$$

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16 Other statistics were directly computed on the histogram built with the simulations of the S
17 set, and statistics were also estimated from the lognormal and gamma fitted *pdfs*.
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23 *Method to build the bootstrap confidence intervals*

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25 To know the validity and accuracy of the high quantiles estimated for the simulation method,
26 the confidence intervals are required. Therefore, this section focuses on building bootstrap
27 confidence intervals in order to determine the reliability of the simulation methods. Several
28 methods to build bootstrap (Efron 1993) confidence intervals (CI_b) were assessed by Gauchi
29 and Leblanc (2002), revealing the difficulties involved in applying the nonparametric CI_b and
30 highlighting the “*pseudo-parametric CI_b* ” as the best choice from among the other parametric
31 procedures and analytical methods proposed. We therefore built CI_b according to this method.
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“*Pseudo-parametric CI_b* ”, referred to as Type 1, was built by randomly drawing B samples of
size n_{π_0} in the exposure simulation set S . Typically, B is equal to 10,000

The boundaries of the 95% confidence interval are calculated taking the 0.025th and 0.975th
empirical quantiles of the final bootstrap distribution.

54 **Results**

57 **Results of the Direct Approach**

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3 The parameter estimates of this direct approach, \hat{E}_{π_0} and $\hat{V}(E_{\pi_0})$, for each population
4 group are shown in Table III. They should be compared with the results of the subsequent
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The parameter estimates of this direct approach, \hat{E}_{π_0} and $\hat{V}(E_{\pi_0})$, for each population group are shown in Table III. They should be compared with the results of the subsequent tables. We observed that they were quite different; the infants and ethnics being the most exposed groups.

Message to the editor: Place Table III approximately here (if possible).

[Table III. Results of exposure using the direct approach.]

Results of the P-P Method

The scale and shape parameters estimated from the gamma *pdf* fitted to consumption and contamination datasets are shown in Table IV.

Message to the editor: Place Table IV approximately here (if possible).

[Table IV. Parameters of the gamma pdf fitted to the subclasses of normalized consumption.]

Table V provides statistics calculated from the simulation outputs. The estimated statistics presented are the mean, standard deviation, skewness and kurtosis, as well as the median and the main high quantiles (90th, 95th and 99th). The highest values of skewness and kurtosis were found in the celiac sufferers group, while it was assumed that the most exposed group would be the infants and ethnics, with the highest mean and quantiles, especially the related 99th quantile. An example of exposure output histograms of relative frequencies, obtained for adult females is given in Fig. 3. When the estimated means from the simulation method are compared to those obtained through the direct approach, it is observed that they are quite higher.

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3 *Message to the editor: Place Figure 3 and Table V approximately here (if possible).*

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6 *[Fig. 3. Exposure output histograms (relative frequencies) obtained with the P-P*
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8 *method, for adult females. Exposure in $\mu\text{g kg}^{-1} \text{bw day}^{-1}$]*

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11 *[Table V. Results of exposure assessment of the population groups from P-P*
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13 *simulation method. These results were obtained directly from the N simulation outputs*
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15 *without pdf fittings; they must be compared to the results of Table VI (in $\mu\text{g kg}^{-1} \text{bw day}^{-1}$).]*

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23 Table VI shows the estimated statistics of exposure to DON by lognormal and gamma
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25 *pdf* fitted to the outputs of the simulation method. An example of the exposure histograms is
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27 given in Fig. 4 where the fitted gamma and lognormal *pdfs* for adolescents are given. The
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29 Anderson-Darling and Chi-Square statistics showed that lognormal and gamma distributions
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31 can be acceptable, with the exception of fitted lognormal *pdf* for adult females and ethnics.
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33 When comparing the means from lognormal and gamma *pdf* fittings to the simulation outputs,
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35 the values were close for all age groups with the exception of infants, the highest value of
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37 which was estimated for lognormal *pdf* fittings. On the other hand, large differences were
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39 found for the high quantiles (95th and 99th quantiles), with the biggest values observed again
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41 for infants.
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48 *Message to the editor: Table VI approximately here (if possible).*

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51 *[Table VI. Estimated statistics of exposure to DON by lognormal and gamma pdf fitted to the*
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53 *outputs of the P-P method. See the rigorous definitions of the shape and scale parameters in*
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55 *the appendix. (in $\mu\text{g kg}^{-1} \text{bw day}^{-1}$).]*

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58 *[Fig. 4. Fitted gamma (solid line) and lognormal (broken line) pdfs for adult females.*
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60 *Relative parameters are given in Table VI. Exposure in $\mu\text{g kg}^{-1} \text{bw day}^{-1}$]*

Results of the Bootstrap Confidence Intervals

Confidence intervals built using the bootstrap pseudo-parametric method for the outputs of P - P method are given in Table VII. The estimates given in Tables IV and VI were always contained in the confidence intervals. It was therefore considered that the P - P method provided reliable estimates, and that this hypothesis was valid for both those statistics estimated from lognormal and gamma pdf fitted to the outputs of this simulation method.

Another point to highlight in this section is the progressive decrease in accuracy when we estimate the CI_b of high quantiles, particularly excessive in the case of the 95th and 99th quantiles. It is an accepted fact that it is very difficult to obtain good accuracy for the 99th-quantile, in particular (Breiman et al. 1990; Beirlant et al. 1996, 1999).

Message to the editor: Table VII approximately here (if possible).

[Table VII. Bootstrap confidence intervals obtained from the results of the P - P method. (in $\mu\text{g kg}^{-1} \text{ bw day}^{-1}$).]

Discussion

In the present work the stochastic methodology developed by Gauchi and Leblanc (2002) was applied to quantify the exposure of Catalonian population to deoxynivalenol. A raw contamination data set obtained by means of thorough chemical analysis of many foodstuff samples was used. Moreover, consumption data was taken from a specific nutritional study to assess the consumption of those commodities susceptible to mycotoxin contamination, by Catalonian population. The exposure was estimated, in one hand, through a direct method, the

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3 most commonly method used in the previous studies, and in the other hand, by means of a
4 simulation method (*P-P*). Finally, the pseudo-parametric bootstrap confidence intervals were
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6 calculated for the parameters obtained through the simulation method.
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10 In both cases, direct method and simulation method, the most exposed groups to DON were
11 infants and ethnics. The means estimated through the simulation method were 0.15 and 0.96
12 $\mu\text{g kg}^{-1} \text{ bw day}^{-1}$, for celiac sufferers and ethnics, respectively. The median was quite low in
13 all cases (0.04-0.36 $\mu\text{g kg}^{-1} \text{ bw day}^{-1}$), but the high percentiles increased these estimated
14 intakes until worrying values, for example 3.82 $\mu\text{g kg}^{-1} \text{ bw day}^{-1}$ (percentile 95 for ethnic
15 group). When the estimates were compared to the TDI of 1 $\mu\text{g kg}^{-1}$ body weight (SCF 2002),
16 it was observed that the mean values were between 15% and 96% of TDI, and the high
17 percentiles commonly exceeded it.
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20
21 Other studies have already estimated a high DON consumption worldwide, for example,
22 JECFA estimated human dietary intake of DON in five regional diets, the highest one
23 occurring in the Middle Eastern region (2.4 $\mu\text{g kg}^{-1} \text{ bw day}^{-1}$), followed by Far Eastern and
24 European (1.6 and 1.4 $\mu\text{g kg}^{-1} \text{ bw day}^{-1}$, respectively) (Canady et al. 2001). In the First French
25 Total Diet Study, Leblanc et al. (2005) estimated DON daily intakes of 0.28 and 0.57 $\mu\text{g kg}^{-1}$
26 bw day^{-1} , for mean and percentile 95 for adults, respectively; while 0.45 and 0.93 $\mu\text{g kg}^{-1} \text{ bw}$
27 day^{-1} , were estimated for children. A probabilistic methodology to assess the exposure to
28 DON was conducted in Netherlands, concluding that 1-year-old children was the most
29 exposed group, with estimated intakes of 0.46 and 1.00 $\mu\text{g kg}^{-1} \text{ bw day}^{-1}$, for the median and
30 percentile 95, respectively (Pieters et al. 2004).
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34 To our knowledge, this is the first study to assess the exposure of special population groups to
35 deoxynivalenol. The special groups selected in the present study had dietary habits markedly
36 different from the general population. In one hand, celiac sufferers substitute the wheat-based
37 food for other gluten free cereals, and in the other hand, some ethnic groups partially maintain
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3 the dietary patterns from their country of origin. In Catalonia during 2009, the immigrant
4 population represented 15.9 % of the total population (Migracat 2010), therefore, their
5 specificities should be taken into account to assess the exposure of this collective in other
6 close regions.
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12 To sum up, considering our results from the *P-P* simulation method, the Catalanian
13 population should be expected to be exposed at moderated levels of deoxynivalenol, being the
14 infants and individuals with ethnic dietary patterns, the most exposed population groups.
15
16 Although the majority of the population do not exceed the TDI of $1 \mu\text{g kg}^{-1} \text{ bw day}^{-1}$, there is
17 still a large population exceeding this safety value. A reduction of DON levels in foodstuffs
18 would entail to reach safety levels on global DON intake.
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61 **Appendix: Definitions of the lognormal and gamma probability density functions**

62 The lognormal *pdf* for the continuous random variable Y defined in $[0, +\infty[$ is:

$$g(y) = \frac{1}{\sqrt{2\pi} y \sigma_x} \exp\left(-\frac{1}{2} \left(\frac{\text{Log}(y) - m_x}{\sigma_x}\right)^2\right)$$

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3 where m_X and σ_X are λ and r , the scale and the shape parameters, respectively. X then follows
4
5 a normal distribution with mean m_X and variance σ_X^2 . The mean and the variance of Y are
6
7 defined by $E(Y) = \exp(m_X + \sigma_X^2/2)$ and $V(Y) = [\exp(2m_X + \sigma_X^2)] [\exp(\sigma_X^2) - 1]$, respectively.
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11 The gamma *pdf* for a continuous random variable X defined in $[0, +\infty[$ is:
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$$f(x) = \frac{1}{\lambda \Gamma(r)} \left(\frac{x - \theta}{\lambda} \right)^{r-1} \exp\left(-\frac{x - \theta}{\lambda} \right)$$

13
14 where r , λ and θ are the shape, scale, and the threshold parameters, respectively, and $\Gamma(r)$ is
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16 the usual Euler's integral. The mean and variance of the gamma distribution are related to r
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18 and λ in the following way: $E(X) = r\lambda$ and $V(X) = r\lambda^2$.
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Table I. Occurrence of deoxynivalenol in food products available in the Catalanian market.

	N individual	N composite	Samples >LOQ	Mean±sd µg/g	Max µg/g
<i>Wheat flakes</i> ¹	27	27	20/27	0.190±0.117	0.437
<i>Corn flakes</i> ¹	168	65	49/65	0.109±0.078	0.580
<i>Beer</i> ¹	213	71	1/70	0.012	0.012
<i>Sweet corn</i> ¹	185	72	2/72	0.114±0.036	0.139
<i>Corn snacks</i> ¹	213	71	56/71	0.153±0.058	0.304
<i>Pasta</i> ¹	201	70	52/70	0.226±0.177	0.946
<i>Sliced bread</i> ¹	147	72	12/72	0.068±0.018	0.098
<i>Bread</i> ¹	31	31	31/31	0.246±0.158	0.739
<i>Ethnic foods</i> ²	35	35	20/35	0.406±0.272	1.080
<i>Gluten-free Bread</i> ²	5	5	1/5	0.270	0.270
<i>Gluten-free pasta</i> ²	7	7	1/7	0.163	0.163
<i>Baby food</i> ^x	30	30	12/30	0.131±0.054	0.286

¹ Contamination data provided by Cano-Sancho et al. (2010).² Contamination data provided by UdL-ACSA et al. (2010).

Table II. Normalised consumption of the main foodstuffs related to DON contamination by the 384 adult females (g/kg body weight/day).

	Consumers	% Consumers	Mean	<i>SD</i>	Max
Breakfast cereals	153	39.8	0.48	0.46	2.35
Sliced Bread	161	41.9	0.22	0.32	1.74
Bread	365	95.1	0.91	0.72	4.78
Pasta	378	98.4	0.34	0.29	3.51
Corn Snacks	114	29.7	0.07	0.11	0.91
Beer	204	53.1	1.43	1.97	10.14
Sweet Corn	143	37.2	0.50	1.08	8.33

Table III. Results of exposure using the direct approach.

	\hat{E}_{π_0}	$\hat{V}(E_{\pi_0})$
Seniors	0.04	$(0.04)^2$
Celiac sufferers	0.13	$(0.09)^2$
Ethnics	0.57	$(0.47)^2$
Adults females	0.09	$(0.07)^2$
Adults males	0.10	$(0.08)^2$
Adolescents	0.15	$(0.13)^2$
Children	0.36	$(0.27)^2$
Infants	0.74	$(0.71)^2$

($\mu\text{g kg}^{-1} \text{ bw day}^{-1}$)

Table IV. Parameters of the gamma *pdf* fitted to the subclasses of normalized consumption.

Foodstuff ^a	Elders ($\hat{r}, \hat{\lambda}$)	Adult females ($\hat{r}, \hat{\lambda}$)	Adult males ($\hat{r}, \hat{\lambda}$)	Teenagers ($\hat{r}, \hat{\lambda}$)
Breakfast cereals (0.457; 1.437)	not enough data	0.895 ; 0.542	1.229 ; 0.354	0.873 ; 0.891
Bread (0.458; 1.436)	1.999; 0.381	0.765; 1.089	0.705; 1.701	0.554; 2.218
Sliced bread (0.824; 52.632)	2.840 ; 0.0384	0.762 ; 0.294	0.611 ; 0.624	0.674 ; 0.785
Pasta (0.747; 4.386)	1.423 ; 0.1616	2.263 ; 0.151	2.149 ; 0.195	1.770 ; 0.292
Corn snacks (1.410; 11.364)	not enough data	0.635 ; 0.787	1.396 ; 0.044	0.790 ; 0.211
Beer (not enough data)	0.606 ; 3.039	0.735 ; 1.945	0.937 ; 2.471	0.759 ; 1.532
Sweet corn (not enough data)	not enough data	0.635 ; 0.787	0.777 ; 0.475	0.653 ; 0.543
Ethnic foods (0.892; 0.535)	-	-	-	-
Baby food (0.458; 1.436)	-	-	-	-
Foodstuff ^a	Infants ($\hat{r}, \hat{\lambda}$)	Babies ($\hat{r}, \hat{\lambda}$)	Ethnics ($\hat{r}, \hat{\lambda}$)	Celiacs ($\hat{r}, \hat{\lambda}$)
Breakfast cereals (0.457; 1.437)	1.106 ; 1.772	-	0.130; 1.739	0.193; 2.020
Bread (0.458; 1.436)	0.741; 5.707	-	0.892; 1.869	0.550; 2.096
Sliced bread (0.824; 52.632)	0.853 ; 1.102	-	not enough data	not enough data
Pasta (0.747; 4.386)	2.256 ; 0.449	-	1.231; 0.242	0.786; 0.236
Corn snacks (1.410; 11.364)	1.074 ; 0.212	-	0.152; 0.716	0.258; 0.272
Beer (not enough data)	-	-	0.157; 14.485	0.133; 2.413
Sweet corn (not enough data)	0.484 ; 1.77	-	0.140; 0.461	0.167; 0.908
Ethnic foods (0.892; 0.535)	-	-	0.295; 1.297	-
Baby food (0.458; 1.436)	-	1.474 ; 10.579	-	-

^a Values between parentheses are \hat{r} and $\hat{\lambda}$ of the Gamma *pdf* fitted to the contamination data.

-, no consumption data

Table V. Results of exposure assessment of the population groups from *P-P* simulation method. These results were obtained directly from the N simulation outputs without *pdf* fittings; they must be compared to the results of Table VI (in $\mu\text{g kg}^{-1} \text{bw day}^{-1}$).

<i>P-P</i> Method	<i>Seniors</i>	<i>Adult females</i>	<i>Adult males</i>
N	10000	10000	10000
Mean	0.28	0.56	0.37
SD	0.48	0.77	0.48
Skewness	5.58	4.97	4.79
Kurtosis	59.47	41.65	41.08
Median	0.12	0.32	0.22
0.90 th quantile	0.71	1.26	0.82
0.95 th quantile	1.08	1.84	1.18
0.99 th quantile	2.27	3.70	2.33
	<i>Adolescents</i>	<i>Children</i>	<i>Infants</i>
N	10000	10000	10000
Mean	0.43	0.68	0.90
SD	0.55	1.83	1.51
Skewness	3.26	10.16	4.80
Kurtosis	14.39	156.74	42.15
Median	0.25	0.24	0.36
0.90 th quantile	0.99	1.38	2.33
0.95 th quantile	1.46	2.49	3.57
0.99 th quantile	2.86	8.17	7.15
	<i>Ethnics</i>	<i>Celiac sufferers</i>	
N	10000	10000	
Mean	0.96	0.15	
SD	1.98	0.55	
Skewness	6.95	19.20	
Kurtosis	79.96	724.87	
Median	0.36	0.04	
0.90 th quantile	2.24	0.29	
0.95 th quantile	3.82	0.64	
0.99 th quantile	9.38	2.18	

Table VI. Estimated statistics of exposure to DON by lognormal and gamma *pdf* fitted to the outputs of the *P-P* method. See the rigorous definitions of the shape and scale parameters in the appendix. (in $\mu\text{g kg}^{-1} \text{bw day}^{-1}$).

<i>P-P</i> Method	<i>Seniors</i>	<i>Adult Females</i>	<i>Adult Males</i>
<i>Lognormal</i>	$\hat{r}=1.614; \hat{\lambda}=-2.302$	$\hat{r}=1.061; \hat{\lambda}=-1.146$	$\hat{r}=1.036; \hat{\lambda}=-1.526$
Mean	0.37	0.56	0.37
SD	1.30	0.80	0.52
Median	0.10	0.32	0.22
0.90 th quantile	0.79	1.24	0.82
0.95 th quantile	1.42	1.82	1.19
0.99 th quantile	4.27	3.75	2.42
AD (p-value)	31.88 (<0.005)	0.25 (>0.5)	0.79 (<0.042)
<i>Gamma</i>	$\hat{r}=0.607; \hat{\lambda}=0.456$	$\hat{r}=1.027; \hat{\lambda}=0.542$	$\hat{r}=1.083; \hat{\lambda}=0.340$
Mean	0.28	0.56	0.37
SD	0.36	0.55	0.35
Median	0.15	0.39	0.26
0.90 th quantile	0.72	1.27	0.83
0.95 th quantile	0.99	1.65	1.07
0.99 th quantile	1.65	2.53	1.63
Chi-Sq (p-value)	15402.96 (<0.001)	133.80 (<0.001)	108.08 (<0.001)
	<i>Adolescents</i>	<i>Children</i>	<i>Infants</i>
<i>Lognormal</i>	$\hat{r}=1.066; \hat{\lambda}=-1.389$	$\hat{r}=1.397; \hat{\lambda}=-1.439$	$\hat{r}=1.841; \hat{\lambda}=-1.240$
Mean	0.44	0.63	1.58
SD	0.64	1.55	8.44
Median	0.25	0.24	0.29
0.90 th quantile	0.98	1.42	3.06
0.95 th quantile	1.44	2.36	5.98
0.99 th quantile	2.98	6.12	20.98
AD (p-value)	4.37 (<0.005)	3.79 (<0.005)	80.97 (<0.005)
<i>Gamma</i>	$\hat{r}=1.039; \hat{\lambda}=0.417$	$\hat{r}=0.591; \hat{\lambda}=-1.146$	$\hat{r}=0.550; \hat{\lambda}=1.644$
Mean	0.43	0.68	0.90
SD	0.43	0.88	1.22
Median	0.31	0.35	0.45
0.90 th quantile	0.99	1.77	2.40
0.95 th quantile	1.28	2.45	3.36
0.99 th quantile	0.96	4.10	5.69
Chi-Sq (p-value)	1239.06 (<0.001)	190006.68 (<0.001)	14077.63 (<0.001)

AD: Anderson Darling statistic; Chi-Sq: Chi Square statistic

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	<i>Ethnics</i>	<i>Celiacs</i>
Lognormal	$\hat{r}=1.434; \hat{\lambda}=-1.088$	$\hat{r}=1.374; \hat{\lambda}=-3.089$
Mean	1.00	0.12
SD	2.61	0.28
Median	0.36	0.05
0.90 th quantile	2.25	0.27
0.95 th quantile	3.78	0.44
0.99 th quantile	10.06	1.11
AD (p-value)	0.46 (0.267)	66.40 (<0.005)
Gamma	$\hat{r}=0.622; \hat{\lambda}=1.546$	$\hat{r}=0.517; \hat{\lambda}=0.299$
Mean	0.96	0.16
SD	1.22	0.22
Median	0.52	0.07
0.90 th quantile	2.48	0.42
0.95 th quantile	3.51	0.59
0.99 th quantile	5.67	1.01
Chi-Sq (p-value)	52310.98 (<0.001)	154786.81 (<0.001)

AD: Anderson Darling statistic; Chi-Sq : Chi Square statistic

Table VII. Bootstrap confidence intervals obtained from the results of the P-P method. (in $\mu\text{g kg}^{-1}$ bw day $^{-1}$).*P-P Method 95% Bootstrap Confidence Interval*

	<i>Seniors</i>	<i>Adult Females</i>	<i>Adult Males</i>
Mean	[0.19; 0.40]	[0.48; 0.64]	[0.32; 0.42]
SD	[0.24; 0.88]	[0.55; 1.05]	[0.35; 0.66]
Median	[0.07; 0.18]	[0.27; 0.36]	[0.19; 0.25]
0.90 th quantile	[0.43; 1.12]	[1.03; 1.47]	[0.68; 0.98]
0.95 th quantile	[0.66; 1.95]	[1.43; 2.26]	[0.94; 1.45]
0.99 th quantile	[1.11; 6.51]	[2.60; 5.89]	[1.61; 3.46]
	<i>Adolescents</i>	<i>Children</i>	<i>Infants</i>
Mean	[0.37; 0.51]	[0.37; 1.23]	[0.67; 1.19]
SD	[0.41; 0.70]	[0.48; 4.47]	[0.93; 2.56]
Median	[0.21; 0.29]	[0.16; 0.35]	[0.25; 0.53]
0.90 th quantile	[0.80; 1.28]	[0.80; 2.86]	[1.62; 3.21]
0.95 th quantile	[1.12; 1.98]	[1.16; 5.60]	[2.40; 5.46]
0.99 th quantile	[1.88; 3.99]	[2.41; 36.42]	[3.95; 12.29]
	<i>Ethnics</i>	<i>Celiacs</i>	
Mean	[0.56; 1.58]	[0.07; 0.33]	
SD	[0.70; 4.09]	[0.10; 1.21]	
Median	[0.22; 0.55]	[0.03; 0.06]	
0.90 th quantile	[1.23; 4.33]	[0.12; 0.81]	
0.95 th quantile	[1.89; 8.79]	[0.22; 2.07]	
0.99 th quantile	[3.21; 28.40]	[0.50; 8.23]	

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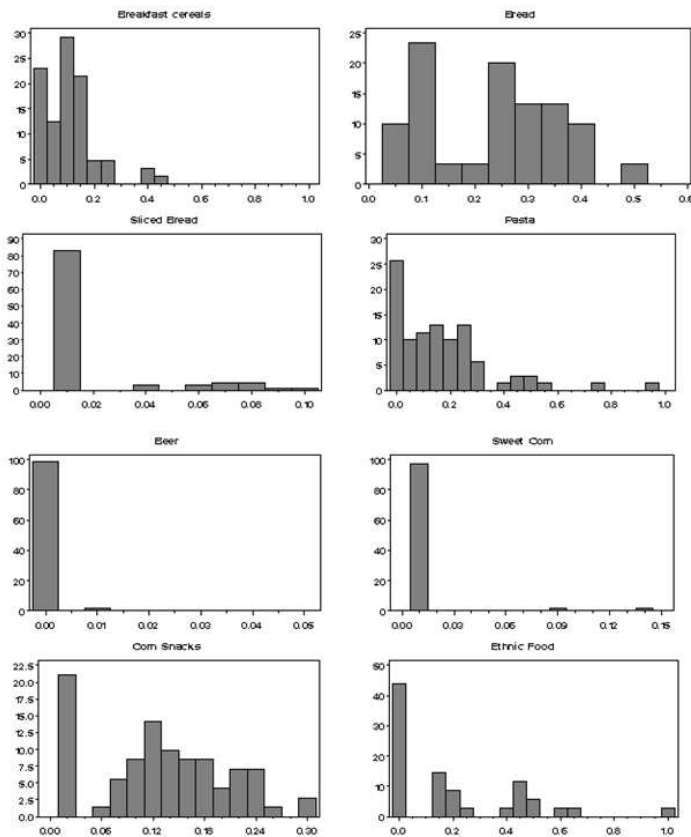


Fig 1. Contamination histograms (relative frequencies), in $\mu\text{g g}^{-1}$ for each food
190x275mm (96 x 96 DPI)

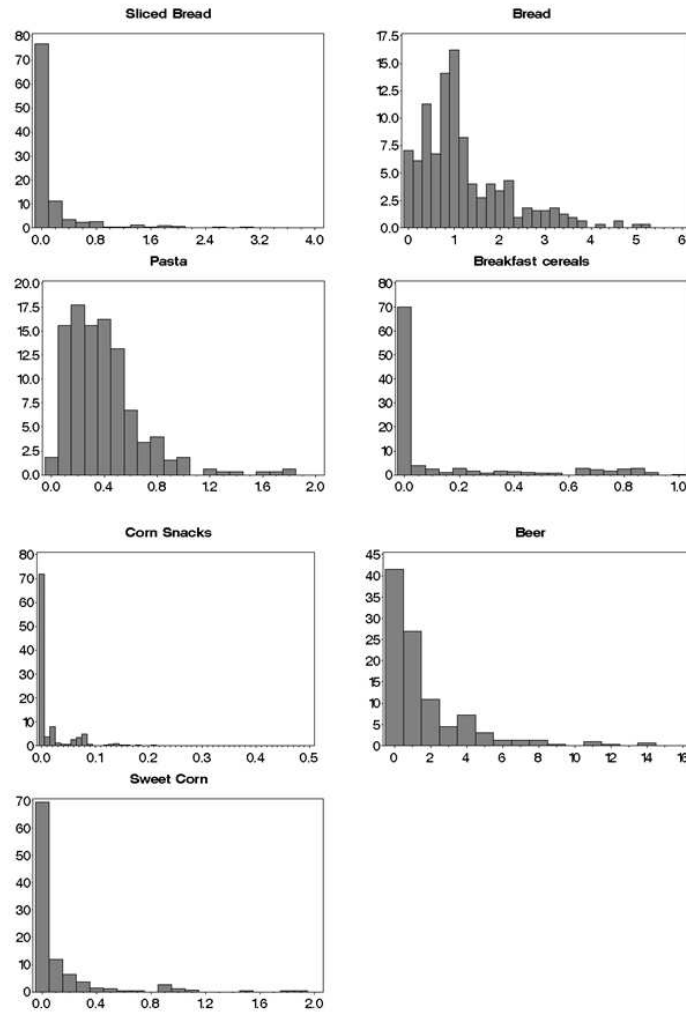


Fig 2. Consumption histograms (relative frequencies) for adult female consumers, in $\mu\text{g kg}^{-1} \text{bw day}^{-1}$.
190x275mm (96 x 96 DPI)

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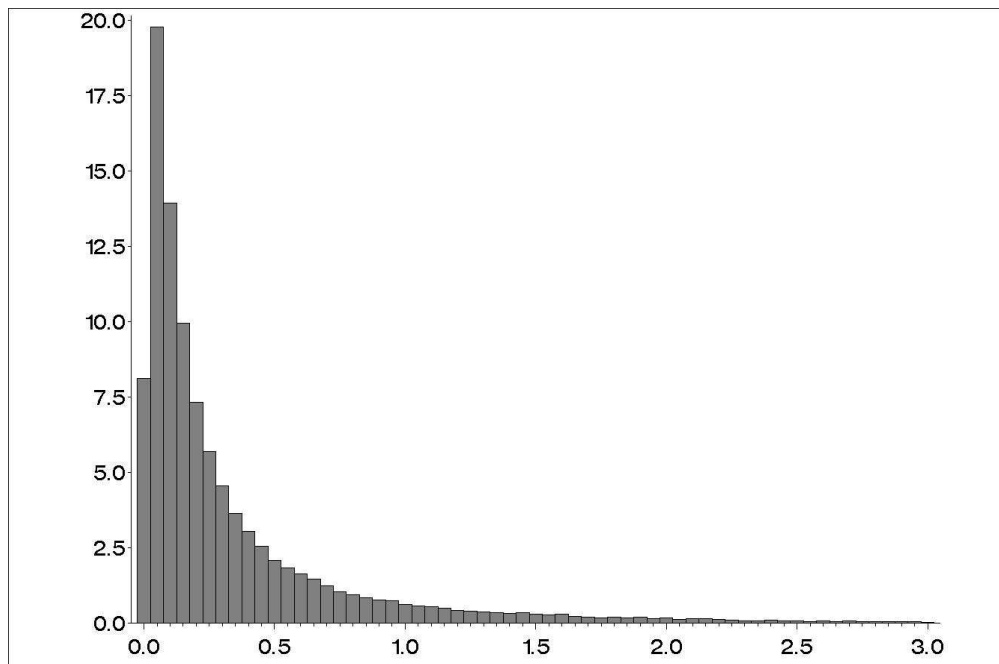


Fig. 3. Exposure output histograms (relative frequencies) obtained with the P-P method, for adult females. Exposure in $\mu\text{g kg}^{-1} \text{bw day}^{-1}$.

354x234mm (96 x 96 DPI)

View Only

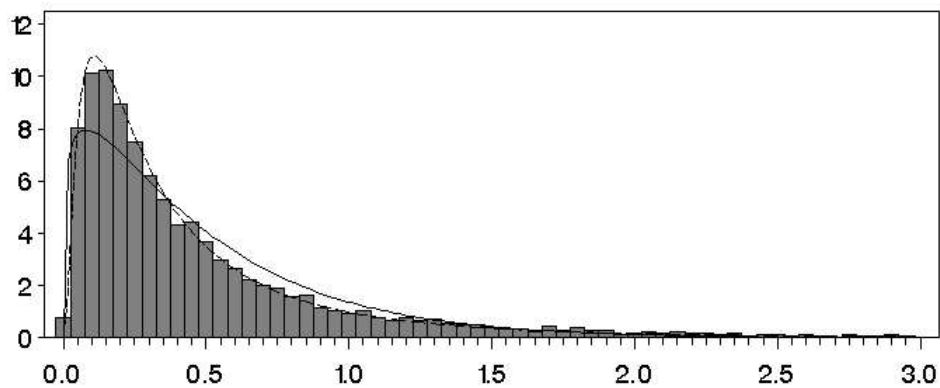


Fig. 4. Fitted gamma (solid line) and lognormal (broken line) pdfs for adult females. Relative parameters are given in Table VI. Exposure in $\mu\text{g kg}^{-1} \text{ bw day}^{-1}$.
181x83mm (100 x 100 DPI)

Tables Captions

Table I. Occurrence of deoxynivalenol in food matrices from food available on the Catalonian market.

Table II. Normalised consumption of the main foodstuff related to DON contamination by the 384 adult females (g kg^{-1} body weight day^{-1}).

Table III. Results of exposure using the direct approach.

Table IV. Parameters of the gamma pdf fitted to the subclasses of normalized consumption.

Table V. Results of exposure assessment of the population groups from P-P simulation method. These results were obtained directly from the N simulation outputs without pdf fittings; they must be compared to the results of Table VI (in $\mu\text{g kg}^{-1}$ bw day^{-1}).

Table VI. Estimated statistics of exposure to DON by lognormal and gamma pdf fitted to the outputs of the P-P method. See the rigorous definitions of the shape and scale parameters in the appendix. (in $\mu\text{g kg}^{-1}$ bw day^{-1}).

Table VII. Bootstrap confidence intervals obtained from the results of the P-P method. (in $\mu\text{g kg}^{-1}$ bw day^{-1}).

Figures Captions

Fig 1. Contamination histograms (relative frequencies), in $\mu\text{g g}^{-1}$ for each food

Fig 2. Consumption histograms (relative frequencies) for adult female consumers, in $\mu\text{g kg}^{-1}$ bw day^{-1} .

Fig. 3. Exposure output histograms (relative frequencies) obtained with the P-P method, for adult females. Exposure in $\mu\text{g kg}^{-1}$ bw day^{-1} .

Fig. 4. Fitted gamma (solid line) and lognormal (broken line) pdfs for adult females. Relative parameters are given in Table VI. Exposure in $\mu\text{g kg}^{-1}$ bw day^{-1} .