Food and Chemical Toxicology 42 (2004) 1349–1358
www.elsevier.com/locate/foodchemtox

Probabilistic exposure assessment to food chemicals based on extreme value theory. Application to heavy metals from ...sh and sea products

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Received 23 October 2003; accepted 27 March 2004

Abstract

This paper presents new statistical methods in the ...eld of exposure assessment. We focus on the estimation of the probability for the exposure to exceed a ...xed safe level such as the provisional tolerable weekly intake (PTWI), when both consumption data and contamination data are independently available. Various calculations of exposure are proposed and compared. For many con− taminants, PTWI belongs to the exposure tail distribution, which suggests the use of extreme value theory (EVT) to evaluate the risk. Our approach consists in modelling the exposure tail by a Pareto type distribution characterized by a Pareto index which may be seen as a measure of the risk of exceeding the PTWI. Using propositions by EVT specialists, we correct the bias of the usual Hill estimator to accurately estimate this risk index. We compare the results with an empirical plug−in method and show that the Pareto adjustment is relevant and e...cient when exposure is low compared to the PTWI while the plug−in method should be used when exposure is higher. To illustrate our approach, we present some exposure assessment for heavy metals (lead, cadmium, mercury) via sea product consumption.

Keywords: Food risk assessment; Extreme value theory; Pareto index; Heavy metals; Sea product consumption

1. Introduction

Quantitative assessment of consumer exposure to contaminants via food consists in a stepwise procedure as recommended by FAO/WHO (1997). Exposure can be de...ned as the cross product of contamination and consumption data for given food items and contami− nants. Total exposure is a summation over all these exposure values. First, the assessment is realized for maximum levels of contamination in order to be con− servative and then if the estimated exposure exceeds its safety limit, a more accurate method of dietary exposure is applied to get a more realistic estimator. One simple way to do so is to consider mean levels of contamina− tion. However, to precisely assess the individual expo− sure of a given population, one should take into account both the individual variability and the global structure of the food basket of each consumer but also the vari− ability and the speci...city (left censorship) of the con− tamination data. Several attempts have been done to account for the individual variability when repeated measures are available (Nusser et al., 1996). In the present paper, most attention is paid to the quantitative assessment of the exposure to contaminants when both individual consumption data and contamination data are available.

In this study, the parameter of interest is the proba− bility that the individual exposure, due to several food items, exceeds a given level. This level may be ...xed a priori, for instance it can be the provisional tolera− ble weekly intake (PTWI) or any other toxicological

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0278−6915/$ − see front matter
doi:10.1016/j.fct.2004.03.016

reference level or safety limit. From a statistical point of view, the estimation of this probability highly depends on the tail behavior of the exposure distribution, more precisely on the extreme exposures. The main statistical tool for this is extreme value theory (EVT). EVT has encountered a great success in many application ...elds, such as ...ood or stock exchange prediction, see Embrechts et al. (1999). In these ...elds, extreme values are more interesting than averages because ‘‘extraordinary’’ events are more interesting than ‘‘ordinary’’. Contamination and consumption data present the same properties i.e. risk mainly concerns high consumers or highly polluted food items, which are extreme values. EVT is also of interest for nutrients in order to compare intakes with the tolerable upper level of intake. At the opposite, lowest nutrient values are the most relevant when dealing with nutrient de...ciencies. However we will focus only on exposure to contaminants in this paper. The originality of EVT is to fully take into account the very high (or very low) observed values. The principle is to model the tail of the exposure distribution by a Pareto type distribution, characterized by a Pareto index which can be interpreted as a risk index. The well-known instability of the classical Hill estimator of the Pareto index may be greatly improved by using bias correction techniques introduced by Feuerverger and Hall (1999) and Beirlant et al. (1999). This study will give some empirical evidence of the interest and the feasibility of EVT for the estimation of the probability that the individual exposure exceeds a given level. Results will be compared to a more empirical approach based on Monte-Carlo estimators of the distribution.

As an application, the exposures to lead, cadmium and methylmercury contained in sea products—wild ...sh, farmed ...sh, mollusk and shell...sh—will be evaluated using French data. The purpose here is not to evaluate the global food exposure risk but rather to study the risks related to the exposure to heavy metals from sea products. These contaminants were chosen for both methodological and practical reasons. Human beings can be exposed to heavy metals through out di...erent pathways: air inhalation, drinking water, contaminated soils and contaminated foods. Heavy metals like lead (Pb), mercury (Hg) and cadmium (Cd) are dangerous for human health because of their accumulation properties. Heavy metals are particularly toxic to children because they may ingest relatively higher amounts of metals from food than adults, in terms of consumption per body weight (WHO-IPCS-EHCs, website). Food sources, such as ...sh and shell...sh, can be contaminated by any heavy metal through trophic bioaccumulation, but mercury and methylmercury (MeHg), the toxic form of mercury, are almost exclusively present in sea products (WHO-IPCS-EHCs, website). These remarks indicate that in order to describe the risk exposure to these heavy metals via sea products, it is necessary to separately consider lead and cadmium which are present in many other products and methylmercury. The exposure to lead and cadmium due to sea product consumption is expected to be low in comparison to the overall exposure. In particular, empirical methods even tends to predict a null probability to exceed the PTWI; the proposed EVT techniques allows to obtain a better extrapolation. Methylmercury is a toxic naturally occurring in ...sh after ingesting mercury polluted feed. The associated risk is thus completely speci...c to sea product consumption: a precise exposure assessment is thus of great interest. Furthermore, for the exposure to methylmercury, it will be interesting to separately assess children exposure to adult since long term health e...ects are more important for this sensitive population (Grandjean et al., 1997).

Section 2 gives the description of the data, the methods retained for exposure assessment and a precise presentation of the methodology based on EVT and tail estimation. Contents of Section 3 is the exposure assessment for lead, cadmium and mercury via sea product consumption and a discussion about the different methods of quanti...cation.

2. Material and methods

2.1. Data description

2.1.1. Food consumption data

Consumption data come from the French survey INCA detailed in CREDOC-AFFSA-DGAL (1999) which concerns the food consumption of 3003 individuals aged 3 years old and more. This food record survey concerns all consumptions at home or outside, during one week: it was realized in four ways through a period of 11 months in order to integrate the seasonal e...ects. The portion sizes were estimated by duplicate weighing for food consumed at home and by photographs for food consumed outside. This is currently in France the only survey which provides individual consumptions (at home and outside). Besides of a detailed food nomenclature of about 900 food items clustered in 45 groups, individual sociodemographic data are available, including the individual body weight and age.

Among this food list, 92 food items containing ...sh or sea products were found in the groups ‘‘Fish’’, ‘‘Shell...sh and Mollusk’’, ‘‘Mixed dishes’’, ‘‘Soups’’ and miscellaneous (Fish in ‘‘Meat products’’). For some of these items, such as breaded ...sh, consumption data were weighed by a recipe factor. The operational study ...le contained the properly weighed consumption values for 92 products and n ¼ 2513 sea product consumers, including sociodemographic informations.

2.1.2. Contamination data

Sea product contamination data were collected through di...erent analytical surveys performed by several French institutions (MAAPAR, 1998–2002; IFREMER, 1994–1998). For each of the three studied contaminants (Pb, Cd and Hg), there were respectively 3089, 3017 and 2643 contamination values expressed in mg per kg of fresh weight. These values were clustered into three categories (‘‘Wild Fish’’, ‘‘Farmed ...sh’’ and ‘‘Mollusks and shell...sh’’) according to their contamination level.

According to Cossa et al. (1989) and Claisse et al. (2001), methylmercury in sea products can be extrapolated from mercury contents. Therefore, conversion factors were applied to analytical data in order to get the corresponding methylmercury (MeHg) concentration in food: 0.84 for ...shes, 0.43 for mollusks and 0.36 for shell...sh.

2.2. Scenarios for exposure calculation

Various strategies for exposure calculation can be achieved depending on the nature of the available data: this is extensively described in Kroes et al. (2002). A quick review will help in understanding the various assumptions and the di...erent methods compared in this work.

First, since PTWI is expressed as contaminant unit per kilogram of body weight it is of great interest to know the consumer body weights from consumption surveys. In this study, food consumption data are collected at the individual level and body weight is available so that no body weight approximation is needed.

Due to the detection or quanti...cation limits of analytical methods, contamination data are very often left-censored. This rounding e...ect is related to the physical chemical phenomena involved in any analytical measurement. According to their proportion, these censored data are usually replaced either by the limit of detection (LOD) or limit of quanti...cation (LOQ) or by half of these limits or by zero (GEMS/Food-EURO, 1995). Because there are very few censored data (<10%) in our application, the ...rst assumption, which is conservative, will be used: censored data are replaced by LOD or LOQ in this study. The ‘‘choice’’ between LOD and LOQ is made according to the declaration of the analysts.

When coupling contamination and food consumption data, di...erent levels of aggregation are possible depending on the calculus mode and the size of the data set. For small contamination data sets, it is useless to consider a large number of food items in consumption data. On the contrary, the calculation will be more accurate if each food consumption may be weighed by the correct contamination data. In order to evaluate the impact of aggregation or disaggregation, two levels noted AL and DL ranking from the most to the less aggregated are considered. More precisely, as contamination data were clustered into three categories (‘‘Wild Fish’’, ‘‘Farmed ...sh’’ and ‘‘Mollusks and shell...sh’’), each of the 92 food items was linked to one of these categories (see also Crepet and Leblanc, 2003). This leads to two levels of aggregation which are noted as:

- DL: disaggregated level, C^i_j is the consumption of product j for sea product consumer i, with j varying from 1 to 92.
- AL: aggregated level, $C^i_{(j)}$ is the consumption of product from category (j) for consumer i, with (j) being ‘‘Wild Fish’’, ‘‘Farmed ...sh’’ or ‘‘Mollusks and shell...sh’’.

So that a consumer is more generally de...ned by C^i, a 92-dimensional (DL) or a three-dimensional vector (AL) and his body weight w_i for i varying from 1 to n.

For example, if data are available for trout, salmon and bass, the aggregated level (AL) will consist in using the same value of contamination for the three species since they all belong to the ‘‘Farmed ...sh’’ category, for example the average of contamination; on the contrary, for the disaggregated level (DL) each species is separately considered. Only two aggregation levels are used but it is possible to de...ned a whole continuum of aggregation levels.

Two kinds of calculus will be considered:

- Deterministic calculus. The contaminant concentration for each food will be expressed according to three way: (i) D-AVE the average of all available contamination data for this food; (ii) D-97.5 for the 97.5th percentile and (iii) D-MAX for the maximum. In this notation, D stands for deterministic because no randomization is assumed concerning contamination data. Each consumer faces the same contamination levels. The D-AVE calculation corresponds the usual realistic methods mentioned in Section 1.
- Double random sampling. This exposure assessment method is a non-parametric Monte-Carlo method, also described in Gauchi and Leblanc (2002). It consists in randomly selecting, on one hand a consumer that is a basket of food consumption values and his associated body weight, and, on the other hand as many contamination values as food items in the basket. The random sampling size is denoted by B. This method is denoted 2R since both consumption and contamination distributions are Randomly used.

More precisely, such random selection among the available data is a selection according to the empirical cumulative distribution function (c.d.f.) of the data. For instance, for consumption data, each consumer may be selected with probability $1/n$

The deterministic calculus (at least D-AVE and D-MAX) can be achieved for both AL and DL aggregation levels but the 2R calculus (and the D-97.5) need much more data and cannot be achieved at the DL level. Indeed, AL is necessary for random sampling so that contamination data set is large enough. Concerning the DL level, it was necessary to associate to each 92 food items the corresponding analytical data by scanning all the available analyses. For instance, for "Fried sole" or "Steam-cooked sole", all the contamination data concerning "sole" were used to calculate average or maximum, while for vaguer named items, such as "Fish soup" or "Fried fish", all analytical data from the clusters "Wild fish" and "Farmed fish" were taken.

For the 2R calculus mode, according to U-statistic arguments presented in another paper by Bertail and Tressou (2003), it is necessary that $B \gg N$, where N is the sum of all the sample sizes (consumption, contamination in each category "Wild fish", "Farmed" and "Mollusks and shellfish"). For example, for lead, there are 592 analyses concerning "Fish", 532 for "Farmed fish" and 1965 for "Mollusks and shellfish", and $n = 2513$ sea product consumers, so that $B \gg 5602$, which is fulfilled with $B = 10{,}000$. Although this value of B allows for a certain stability of the exposure distribution, the results, presented in next section, correspond to the mean over 100 repetitions of the same calculus.

To summarize, for each exposure computation, the calculation is performed according to the following points:

- the aggregation level (AL or DL),
- the calculus mode (D-AVE, D-97.5, D-MAX or 2R).

Left censorship and body weight treatments are fixed here but could also generate other calculation scenarios. Such decomposition could serve as guidelines for further exposure assessment to test the sensitivity of the results.

Furthermore, individual consumptions are assumed to be independent and identically distributed as well as contamination data.

2.3. Risk characterization: definition of the parameter of interest

Chemical food risks to human health are assessed by comparing the dietary exposure with an adequate safe exposure level, such as provisional tolerable weekly intake (PTWI) proposed by the Joint FAO/WHO Expert Committee on Food Additives (JECFA). This step of the risk assessment is well described in Renwick et al. (2003). Our goal is to estimate the probability that the exposure of an individual from a given population exceeds the PTWI. In a large population, a precise estimation of this quantity is of great importance since even a difference of 1‰ involves a large number of individuals.

2.3.1. The plug-in (PI) estimator

If X_i is defined as the exposure value to a given contaminant for an individual i $(i = 1, \ldots, n)$ and assuming that exposure values are available for all individuals and expressed in the same unit as the PTWI, a simple way to estimate the risk is to use the plug-in (PI) or empirical estimator of the probability to exceed the PTWI, defined as:

$$\frac{\#(X_i > \mathrm{PTWI})}{n}$$

where $\#(X_i > \mathrm{PTWI})$ denotes the number of exposure values that exceed the PTWI. For example, if this quantity is equal to 0.05 for a given population, it means that an unknown individual belonging to that population may exceed the PTWI with a probability of 5%.

The results obtained with the PI estimator will be compared to those issued with the tail estimation (TE) method extensively described in next section.

2.3.2. The tail estimation (TE) based estimator

2.3.2.1. Extreme value theory (EVT) for risk assessment. A few basic facts about EVT are now recalled (Embrechts et al., 1999). We give here the results concerning the high extreme values (right tail of the distribution) but they can be transposed to lowest values (left tail of the distribution) if one is interested by nutrient deficiencies.

Let $X_1, \ldots, X_n$ be a n-sample, that is n independent and identically distributed (i.i.d.) random variables (r.v.). F denotes its cumulative distribution function (c.d.f.), i.e. $F(x) = \Pr(X_i \leq x)$ for any X_i, $i = 1, \ldots, n$. $X_{1;n} \leq \cdots \leq X_{n;n}$ denotes the associated ordered sample so that $X_{i;n}$ is the ith smallest variable among the $(X_i)_i$.

EVT main theorem (Fisher Tippett theorem) gives the asymptotic behavior of the sample maxima $X_{n;n}$ when n goes to infinity. There are only three possibilities for the asymptotic distribution G of $X_{n;n}$: Gumbel, Fréchet or Weibull distributions. The Jenkinson representation allows to write the c.d.f. of G as a function G_c depending on an index c. The limit case $c \to 0$ corresponds to the Gumbel distribution, the case $c > 0$ to the Fréchet distribution and the case $c < 0$ to the Weibull distribution.

These laws are called extreme value distributions and each one corresponds to a special tail behavior: Gumbel law is related to light-tailed distribution such as normal, log-normal or exponential distributions; Fréchet law to heavy-tailed distributions such as Pareto, Cauchy or Student distributions and Weibull law to finite support distributions that is for instance uniform distribution.

This limit distribution G_c of $X_{n;n}$ is highly related to the tail behavior of the $(X_i)_i$ so that one way to use EVT is to adjust a distribution to the tail of the $(X_i)_i$, i.e. the largest $(X_i)_i$.

The application of EVT to risk assessment consists in adjusting a distribution to the distribution tail of the exposure. Here, the realizations of the $(X_i)_i$ are the exposure levels obtained from the calculation procedure described in the previous section.

An example of distribution of exposure is given in Fig. 1. The zoom on the tail of the distribution shows that very high values are reached. The first assumption is thus that exposure has a heavy-tailed distribution. A standard way to model such heavy tail phenomenon is to use a Pareto law. For any x belonging to the tail of distribution, i.e. for sufficiently large x, it is assumed that $1 - F(x) = Cx^{-1/c}$. In that case, the maximum is of Fréchet type with index $c > 0$ which may be interpreted as a risk index.

In our model, the parameter of interest is the probability that individual exposure exceeds the PTWI: $\Pr(X_i > \mathrm{PTWI}) = C[\mathrm{PTWI}]^{-1/c}$ which is an increasing function of c. Fig. 2 clearly illustrates the influence of c on the thickness of the distribution tail and consequently on the risk as defined earlier. Indeed, the probability to exceed a fixed level d of the x-axis, represented by the surface delimited by the x-axis, a vertical line at the d level and the left part of the curve, increases when c increases.

2.3.2.2. Estimation of parameters.

Fitting the distribution tail to a Pareto law consists in estimating the parameters C and c for x large enough. This notion of

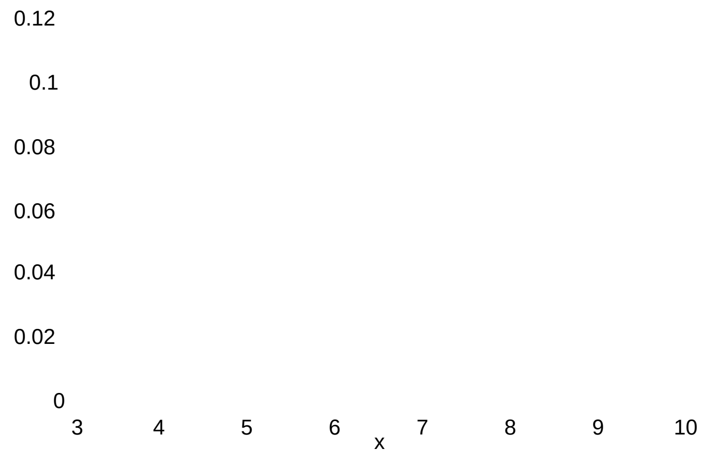

Fig. 2. Pareto distribution tail for different values of c: $c = 1$ (solid line), $c = 0.5$ (dots) and $c = 0.3$ (dashed line).

"sufficiently large" is quantified by selecting a fraction of the sample, i.e. the k largest observed values.

If $(X_i)_{i=1,\dots,n}$ are independent and identically distributed (i.i.d.), conditionally to k, maximum likelihood technique allows to estimate c and C by

$$\begin{cases} \hat{c}_{MV}(k) = H_{k,n} = \frac{1}{k}\sum_{i=1}^{k} \log \frac{X_{n-i+1,n}}{X_{n-k,n}} \\ \hat{C}_{MV}(k) = \frac{k}{n}(X_{n-k,n})^{1/H_{k,n}} \end{cases}$$

where $X_{i;n}$ as before denotes the ith order statistic and $H_{k;n}$ is the Hill estimator (Embrechts et al., 1999).

The Hill estimator is very sensitive to the choice of k as shown in Fig. 3 (the Hill estimator is the dashed line).

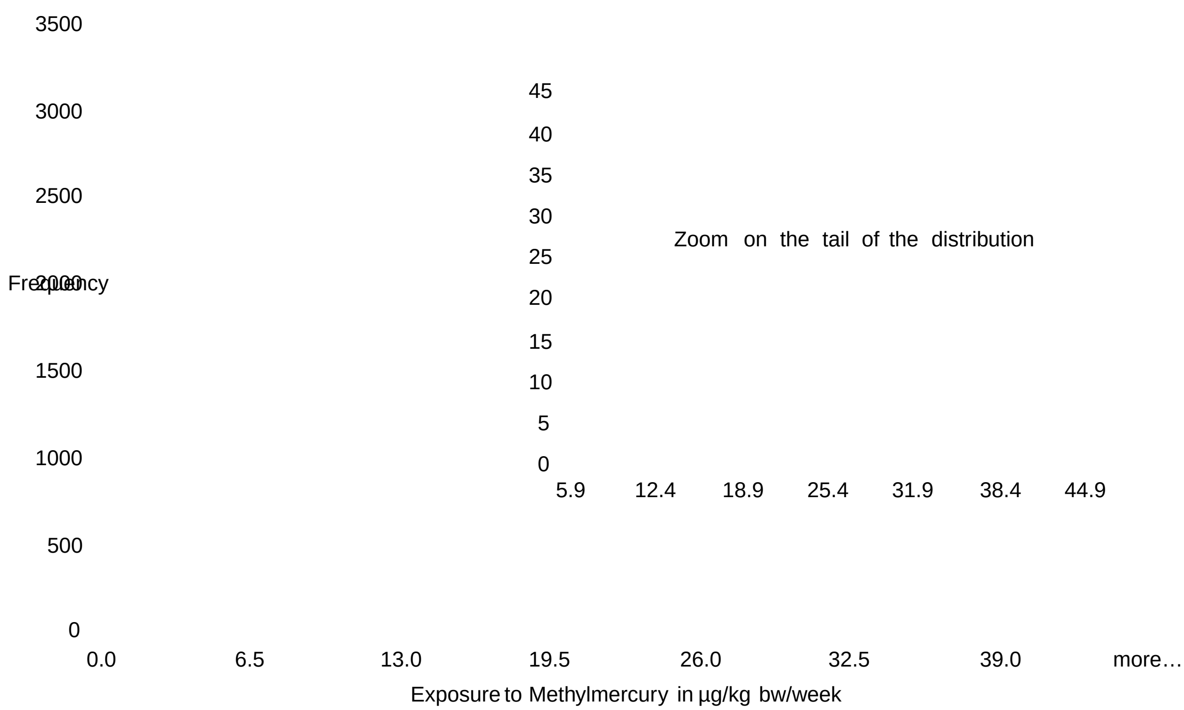

Fig. 1. Example of distribution: exposure to methylmercury, obtained by 2P procedure, INCA data.

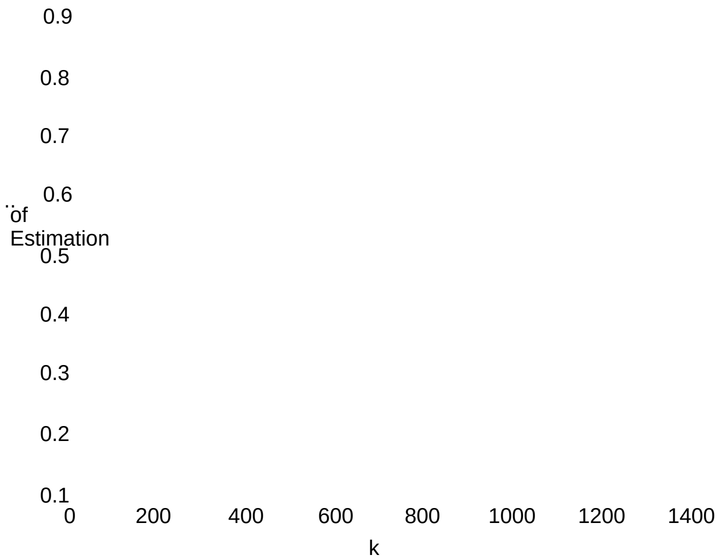

Fig. 3. Example of bias correction for the risk index c: Hill estimator (dashed line), bias corrected Hill estimator (solid line) and confidence interval for the debiased estimator (dots); the minimization of AMSE gives $k = 50$, $\hat{c} = 0.252$ and $H_{k;n} = 0.265$. Case of the exposure to lead, disaggregated level, average contamination.

Indeed its bias increases with k while its variance decreases with k.

One way to correct this is to introduce slowing varying functions, so that $1 - F(x) = Cx^{-1/c}L(x)$, where $L(x)$ denotes a satisfying for all $t > 0$, $\frac{L(tx)}{L(x)} \to 1$ as $x \to \infty$, which takes into account small deviations from the exact Pareto case (Beirlant et al., 1999; Feuerverger and Hall, 1999). All distributions of this type are of Fréchet type with index c.

One example of slowly varying function is $L(x) = 1 + Dx^{-b}$, with $b > 0$ and $D \in \mathbb{R}$. This form can for instance appear when considering a population which is a mixture of two different populations with risk exposures with two different risk indexes c_1 and c_2 ($c_1 > c_2$). In that case, the resulting distribution of exposure is not strictly Pareto but perturbed by a slowly varying function with $c = c_1$ and $b = 1/c_2 - 1/c_1 > 0$. In our case, the data are more likely to come from a mixture of several populations with different risks.

This slowly varying function induces a bias on the estimator and may strongly reduce the rate of convergence of the Hill estimator. The principle of the bias correction method is to interpret the Hill estimator as an estimator of the QQplot slope perturbed by a small deviation induced by the slowly varying function. Taking the weighted average of several slopes allows to reduce the bias showing that this average behaves like an exponential r.v. with mean depending on the parameters. The technical principles about the bias correction method and about the estimation of parameters are available from the authors. Simulations of the validity of these corrections are available in Feuerverger and Hall (1999). Fig. 3 gives an example of bias correction.

These estimations can be done for different values of k ($\hat{c}_k$ is the current bias-corrected estimator of c) and the optimal sample fraction k can then be chosen as the solution of the program:

$$\min_{k;k>10} \frac{\hat{c}_k^2}{k} + (H_{k;n} - \hat{c}_k)^2$$

which consists in minimizing the asymptotic mean squared error (AMSE) of the Hill estimator.

As explained above, our parameter of interest is $C[\mathrm{PTWI}]^{-1/c}$ and is estimated by $\hat{C}[\mathrm{PTWI}]^{-1/\hat{c}}$ where $\hat{c} = \hat{c}_k$ is the bias corrected estimator of c taken at the optimal sample fraction k, and $\hat{C} = \frac{k}{n}(X_{n-k;n})^{1/\hat{c}_k}$ is the resulting estimator of constant C.

This method of risk estimation is referred to as TE (tail estimation) in the application.

3. Results

3.1. Exposure to heavy metals due to sea product consumption

Results for food exposure to lead (Pb), cadmium (Cd) and methylmercury (MeHg) are given in Table 1. Each line of this table corresponds to a different calculation of exposure for a given contaminant according to the proposed assumptions, leading to six scenarios for each contaminant. For example, for scenario 1, the exposure to lead from sea products is described by its average, its 97.5th percentile and its maximum over the sea product consumers. This first scenario corresponds to a calculation with a deterministic calculus at disaggregated level (DL) using average of contamination (D-AVE). The last columns give the associated probabilities of exceeding the PTWI, calculated with our new method based on tail estimation (TE) and the plug-in method (PI).

The international toxicological references (PTWI) were established and revised by the JECFA. The most recent references were used for this study and are: 25 lg/week/kg b.w. for lead, (revision FAO/WHO, 1999), 7 lg/week/kg b.w. for cadmium (revision FAO/WHO, 2000), and 1.6 lg/week/kg b.w. for methylmercury (revision FAO/WHO, 2003).

As mentioned in Section 1, it is clear that methylmercury exposure needs a particular focus (next section) while lead and cadmium which are present in other foods, will better illustrates the proposed TE method. According to previous French reports using different calculation modes, similar to D-AVE in SCOOP 3.2.11 (2003) and to D-MAX in CREDOC (1998), the exposure due to sea products is from 3% to 11% of the total food exposure for lead and from 8% to 25% of the total food exposure for cadmium.

Table 1
Exposure assessment to lead (Pb), cadmium (Cd) and methylmercury (MeHg) for sea product consumers (for 2R, B ¼ 10; 000)

Scenario	Contaminant (PTWI in lg/kg b.w.)	Assumptions		Exposure (lg/week/kg b.w.)			Associated probability of exceeding the PTWI	
		Aggregation level	Calculus mode	Average	97.5th percentile	Maximum	TE	PI
1	Pb (25)	DL	D−AVE	0.325	1.406	5.143	3.17E)07	0
2			D−MAX	3.847	15.239	36.239	3.76E)03	4.78E)03
3		AL	D−AVE	0.387	1.774	7.735	2.90E)06	0
4			D−97.5	1.290	6.176	26.776	2.20E)04	3.98E)04
5			D−MAX	6.392	23.095	93.934	1.67%	1.87%
6			2R	0.386	2.096	21.725	1.03E)04	2.60E)05
7	Cd (7)	DL	D−AVE	0.199	1.061	3.537	7.14E)05	0
8			D−MAX	2.592	13.200	32.080	10.94%	9.15%
9		AL	D−AVE	0.235	1.211	5.434	7.54E)05	0
10			D−97.5	0.780	4.054	18.132	4.10E)03	3.18E)03
11			D−MAX	4.694	20.763	90.021	100%	20.57%
12			2R	0.234	1.422	19.391	7.92E)04	7.97E)04
13	MeHg (1.6)	DL	D−AVE	0.628	2.712	17.213	9.26%	7.40%
14			D−MAX	9.167	39.989	110.486	100%	75.05%
15		AL	D−AVE	1.113	4.202	10.796	100%	21.53%
16			D−97.5	4.807	18.270	46.760	100%	76.72%
17			D−MAX	16.039	60.573	155.832	100%	92.40%
18			2R	1.114	6.273	50.217	75.63%	18.38%

An important remark concerns the signi...cance of all these results. This assessment of exposure to heavy metals was made on e...ective sea product consumers from the INCA data. A multiplicative coe...cient of 2513/3003 ¼ 84% may be applied to risk calculated with PI in order to take into account the non−consumers and extrapolate to the whole population (adults and chil− dren) of the survey. Because of the short period of the survey, the bias due to the observed zeros is well known: individuals with null consumptions in INCA may be true non−consumers of sea products or may scarcely consume sea products, maybe in large quantity, but not during the survey week. This bias, which can be evalu− ated by comparisons with other sources on household consumptions, such as the Secodip panel survey (daily observations during a year), is not signi...cant in the case of sea products in France.

Our main observations are:

- The aggregation level assumption has a high impact on the results. DL gives lower exposure levels and lower risks than AL for all contaminant for a given calculus mode. For example, for Pb, the comparison of average exposures of scenarios 1 and 3 show the importance of aggregation. This can be explained by the fact that the mean contamination for DL is lower than the mean contamination for AL. Under AL assumption, averages are taken over a larger number of observations and high values boost the average of contamination. For example, average of contamination for tuna ...sh is higher than for any other ...shes but for AL, all ...shes are assumed to be contaminated at the average level of all ...shes which is higher because of tuna. However, 2R calculus is not possible for DL assumption since there is not en− ough data to be sampled in.
- At a given aggregation level, D−AVE (deterministic− average contamination) and 2R (double random) give similar results in average but randomization of con− tamination for the 2R calculus allows to reach higher exposure levels so that 97.5th percentile and maxima are higher for 2R than for D−AVE. Likewise, risk is higher for 2R than for D−AVE (see scenarios 9 and 12 with similar averages but di...erent maximum and risks). If high consumptions are associated with high levels of contamination, some exposure may be very high and 2R allows to consider them without using an unrealistic assumption, such as D−MAX or D− 97.5. These two last methods are not realistic but present the advantage to be conservative. Indeed if D−MAX or D97.5 gives null risks or negligible risks of exceeding the PTWI, there is no need to be more accurate in the process.
- Plug−in (PI) methods gives null risk of exceeding the PTWI for D−AVE calculus for Cd and Pb (see lines 1, 3, 7 and 9). This illustrates a clear drawback of the PI estimate: risk cannot be evaluated if PTWI is too large when compared to the higher observed

values (extreme tail of the empirical distribution). Thus, when risk or sample size are small and since a null risk does not exist, precise quantification is not possible with this method. The tail estimation (TE) method allows a much more accurate quantification.

- TE mostly gives higher risks of exceeding the PTWI than PI and the difference is sometimes very important. For example, in scenario 18, the probability decreases from about 76% for TE to 18% for PI. However, TE sometimes does not allow for accurate estimation of the probability of exceeding the PTWI when this probability is too important. As shown in Fig. 4, if the PTWI is not in the distribution tail, the Pareto assumption is not sufficient to evaluate the probability of exceeding the PTWI. Indeed, Pareto c.d.f. is defined for $x \geq a$, where a is such that $F(a) = 0$, i.e. $Ca^{-1/c} = 1 \Rightarrow a = C^{c}$. Therefore, if PTWI < a, the probability to exceed the PTWI is theoretically equal to 1. In scenarios 11, 14, 15, 16 and 17, the TE method is then too conservative (a 100% probability of exceeding the PTWI) and the PI method should be used. Furthermore, if the PTWI is too close to a, the risk estimation may be too high (it may be the case for scenario 13). To summarize, we can say that: tail estimation (TE) method yields a good risk estimation if the PTWI is located in the distribution tail (PTWI$_3$ in the illustration); a conservative risk estimation is obtained for lower PTWI (PTWI$_2$ in the illustration); small PTWI relatively to the observed values leads to an overestimated value of 100% (PTWI$_1$ in the illustration).

Fig. 4. Pareto adjustment and risk index estimation.

3.2. A focus on methylmercury

Results concerning MeHg according to the age of the population are presented in Table 2. Four sub-populations are considered: the 3–8 years old sea product consumers ($n = 440$, 86% of this age class), the 9–15 years old sea product consumers ($n = 437$, 81% of this age class), the 16–60 years old sea product consumers ($n = 1280$, 83% of this age class) and the over 60 years old sea product consumers ($n = 356$, 89% of this age class). Risk of exceeding the PTWI was calculated according to PI method, since PTWI does not belong to the distribution tail, i.e. the probability of exceeding it is too high to use TE. Three calculus scenarios are presented: DL D-AVE, AL D-AVE and AL 2R.

The role of the aggregation level is even more important in this case for all population groups and especially for 3–8 year old children, where the probability of exceeding the PTWI varies from 17% (DL) to 45% (AL) for D-AVE calculus mode. However, it is clear that according to these data, the exposure of children (aged 3–8) is systematically higher than the exposure of the rest of the population. As D-AVE calculus is concerned, contamination is the same for all individuals so that the observed differences are due to the consumption behaviors. Children eat more sea products relatively to their body weight than the rest of the population. To be more accurate, confidence intervals for PI risks are currently being constructed thanks to the use of incomplete U-Statistics. The first results show that the observed differences according to the population age are significant. About the characterization of target groups, developments are needed as suggested in Bertail (2002).

3.3. Discussion

In this section, we discuss two points raised in the refereeing process concerning, on the one hand, the

Table 2
Estimation of the probability of exceeding the methylmercury PTWI for sea product consumers according to age class (method of risk estimation: PI, for 2R, $B = 5000$)

Assumption		3–8 years old (%)	9–15 years old (%)	16–60 years old (%)	Over 60 years old (%)	All sea product consumers (%)
Aggregation level	Calculus mode					
DL	D-AVE	17.06	5.72	5.08	5.9	7.40
AL	D-AVE	45.91	24.94	13.59	15.73	21.53
	2R	32.91	20.42	13.74	15.22	18.38

de...nition of the parameter of interest, when using transversal consumption data and, on the other hand, the absence of parametric adjustments in this paper.

When using our available data for the estimation of the probability to exceed the PTWI (de...ned over life time), one underlying hypothesis is that individuals are facing a constant distribution of exposure over time and keep the same consumption behavior over their lifetime. Indeed, if this assumption is omitted, the comparison between a one week exposure and the PTWI de...ned over lifetime is nonsense. This is a strong assumption which cannot be avoided with our available data, but might be relaxed by combining our methods with some proposals by Nusser et al. (1996) or Wallace et al. (1994) if time series of consumption (or at least repeated measures) are observed. These methods are compared and discussed in Ho...mann et al. (2002). Moreover, it is assumed that occasional short−term excursions above the PTWI would have no major health consequences, provided that the average intake over long periods is not exceeding the PTWI. Therefore, the parameter of interest may rather be interpreted as the probability of occasional short−term excursions above the PTWI than a true probability to develop a disease because of the exposure to the contaminant.

In this paper, we deliberately do not use any para−metric adjustment to well known distributions, such as log−normal or exponential, neither in the exposure assessment step, nor on the estimation of the parameter of interest step. This is one important principle when dealing with extreme values: these parametric adjust−ments are rather e...cient in measuring mean behavior but irrelevant when dealing with risks and focusing on extremes. Indeed, adjustment tests, such as Kolmogorov or v^2, give more importance to the central tendency than to extreme values and have a very little power. In addition, parametric adjustment of marginal consump−tions do not re...ect the wholesome phenomenon, be−cause they do not account for the correlation structure of the consumptions of products that may be comple−mentary or substitute. Modelling the distribution of the whole vector of consumptions is generally impossible as it lies in a space of large dimension, but also because of the problems of possible null consumptions for several items, which makes a mixture approach very di...cult to implement.

Another objection to the use of marginal parametric adjustments is that they do not allow a good control of the error level (of types I and II). For x contaminated item groups, there is a need to ...t 2x distributions to get the exposure (x for the consumption data, x for the contamination data), some of them on sample sizes smaller than 30. Even if we accept by some test, each parametric adjustments, the global (statistical) error of types I and II may be bigger than 100%, unless we have a huge amount of data.

For these reasons, we only consider here the empirical distribution of the consumption vectors, which is the best non−parametric estimator of the multidimensional distribution of the consumption vector.

4. Conclusion

This paper leads to several types of conclusions.

First, it is important to note that the scenarios of the exposure calculation (such as levels of aggregation used to couple data, calculus mode,. . .) have a strong impact on the values of the exposure so that one must not use numerical results without indicating them.

Deterministic methods for exposure assessment have many drawbacks. If the mean of contamination is used, the exposure is systematically under−evaluated because the extreme contamination are not taken into account. Using high ...xed percentiles of contamination leads to hide a part of the population at risk. Such phenom−enon is well known in other ...elds (...nance, hydrology) which currently use the methods described here. Mod−elling the tail behavior of the exposure by a Pareto distribution is empirically consistent with the available data and allows for very accurate (or at least conser−vative) estimation of the probability to exceed a given level. However, one speci...city of this application to food exposure assessment is the heterogeneity of con−sumption behaviors. From a statistical point of view, this leads to several bias problems which may be solved by using recent developments in the ...eld of extreme value theory (EVT).

Concerning the feasibility of the method based on EVT, it is important to check whether the exposure to the studied contaminant is actually close to the toxico−logical limit or not. Indeed, if the PTWI does not belong to the distribution tail, Pareto tail adjustment is useless while, on the opposite case, it allows to accurately quantify low probability of exceeding the PTWI. Devel−opments are still needed concerning con...dence interval for such probabilities to exceed a given toxicological level.

As far as exposure assessment is concerned, according to the data used and by comparison to the PTWI, methylmercury intake via the consumption of sea products seems important for a signi...cant part of the population, above all children. The case of lead and cadmium clearly illustrates the fact that EVT allows to quantify the risk to exceed the toxicological reference when it is low.

Acknowledgements

We would like to thank Ph. Verger and E. Counil for their comments and careful reading of the manuscript.

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