



Using decomposed household food acquisitions as inputs of a kinetic dietary exposure model

Olivier Allais, Jessica Tressou

► To cite this version:

Olivier Allais, Jessica Tressou. Using decomposed household food acquisitions as inputs of a kinetic dietary exposure model. , 30 p., 2007. hal-01189236

HAL Id: hal-01189236

<https://hal.science/hal-01189236>

Submitted on 6 Jun 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Using decomposed household food acquisitions as inputs of a Kinetic Dietary Exposure Model.

Olivier Allais*and Jessica Tressou[†]

October 2, 2007

Abstract

Foods naturally contain a number of contaminants that may have different and long term toxic effects. This paper introduces a novel approach for the assessment of such chronic food risk that integrates the pharmacokinetic properties of a given contaminant. The estimation of such a Kinetic Dietary Exposure Model (KDEM) should be based on long term consumption data which, for the moment, can only be provided by Household Budget Surveys such as the SECODIP panel in France. A semi parametric model is proposed to decompose a series of household quantities into individual quantities which are then used as inputs of the KDEM. As an illustration, the risk assessment related to the presence of methylmercury in seafoods is revisited using this novel approach.

Keywords: household surveys, individualization, linear mixed model, risk assessment, spline-estimation.

*INRA-CORELA, Laboratoire de recherche sur la consommation, 94205 Ivry sur Seine, France.(*olivier.allais@ivry.inra.fr*)

[†]INRA-Mét@risk, Méthodologies d'analyse des risques alimentaires, INA P-G, 16 rue Claude Bernard, 75231 Paris Cedex 5, France (*jessica.tressou@agroparistech.fr*)

Introduction

The quantitative assessment of dietary exposure to certain contaminants is of high priority to the Food and Agricultural Organization and the World Health Organization (FAO/WHO). For example, excessive exposure to methylmercury, a contaminant mainly found in fish and other seafood (mollusks and shellfish) may have neurotoxic effects such as neuronal loss, ataxia, visual disturbance, impaired hearing, and paralysis (WHO, 1990). Quantitative risk assessments for such chronic risk require the comparison between a tolerable dose of the contaminant called Provisional Tolerable Weekly Intake (PTWI) and the population’s usual intake. The usual intake distribution is generally estimated from independent individual food consumption surveys (generally not exceeding 7 days) and food contamination data. Several models have been developed to estimate the distribution of usual dietary intake from short-term measurements (see for example, Nusser *et al.*, 1996; Hoffmann *et al.*, 2002). The proportion of consumers whose usual weekly intake exceeds the PTWI can then be viewed as a risk indicator (see for example, Tressou *et al.*, 2004). This kind of risk assessment does not account for the underlying dynamic process, *i.e.* for the fact that the contaminant is ingested over time and naturally eliminated at a certain rate by the human body. Moreover, longer term measurements of consumption are available through household budget surveys (HBS).

In this paper, we propose to use HBS data to quantify individual long term exposure to a contaminant. This data provides long time series of household food acquisitions which are first used in a decomposition model, similar to the one proposed by Chesher (1997, 1998) in the nutrition field, in order to obtain time series of individual intakes. Then, the pharmacokinetic properties of the contaminant are integrated into an autoregressive model in which the current body burden is defined as a fraction of the previous one plus the current intake.

From a toxicological point of view, this approach is, to our knowledge, novel and hence requires the definition of an ad-hoc long term safe dose as proposed in the next section. We refer to this autoregressive model as Kinetic Dietary Exposure Model (KDEM).

From a statistical point of view, such autoregressive models are well known in general time series analysis (see for example, Hamilton, 1994) and most of the paper is devoted to the description of the decomposition model. This statistical model aims at estimating individual quantities from total household quantities and structures. This problem is similar to that studied by Engle *et al.* (1986),

Chesher (1997, 1998), and Vasdekis and Trichopoulou (2000), and is addressed in a slightly different way. In the present article, the individual contaminant intake is firstly viewed as a nonlinear function of age within each gender, with time and socioeconomic characteristics being secondly introduced in a linear way. The nonlinear function is represented by a truncated polynomial spline of order 1 that admits a mixed model spline representation (section 4.9 in Ruppert *et al.*, 2003). These choices yield a simple linear mixed model which is estimated by REstricted Maximum Likelihood (REML, Patterson and Thompson, 1971). One major extension of the proposed model compared to Chesher (1997) is the introduction of dependence between the individual intakes of a given household.

In the next section, focusing on the methylmercury example even though the method is much more general and could be applied to any chronic food risk, SECODIP data are described along with the construction of a household intake series and the individual cumulative and long term exposure concepts yielding the KDEM. Section 2 is devoted to the statistical methodology used to decompose the household intake series into individual intake series, namely the presentation of the model and its estimation and tests. Section 3 displays the application of the methodology to the case of methylmercury exposure in the French population using the 2001 SECODIP panel. It includes an empirical validation of the proposed methodology on individual data as well as the results of our estimation procedure and some tests on the structure of the decomposition model. Finally, a discussion on the use of household acquisition data, with the focus on the French SECODIP panel, is conducted in section 4 with respect to the proposed long term risk analysis.

1 Motivating example: risk related to methylmercury in seafoods in the French population

In this section, the Kinetic Dietary Exposure Model (KDEM) and the concept of long term risk are defined (see also Verger *et al.*, 2007, for a different presentation of the same model). Then a brief panorama of consumption data in France is given and the way the SECODIP HBS data will be used as an input of the KDEM is described.

1.1 Cumulative exposure and long term risk: the Kinetic Dietary Exposure Model (KDEM)

The main objective of the analysis is to assess individuals' long term exposure to a contaminant to deduce whether these individuals are at risk or not. As mentioned in the introduction the only "safe dose" reference is the PTWI expressed in terms of body weight (*relative* intake). Unfortunately, TNS SECODIP did not record the body weight of the individuals until 2001. The body weights are thus estimated from independent data sets; namely the French national survey on individual consumption (INCA, CREDOC-AFSSA-DGAL, 1999) for people older than 18, and the weekly body weight distribution available from French health records (Sempé *et al.* (1979)) for individuals under 18. In both cases, gender differentiation is introduced.

Assume that estimations of the individual weekly intakes are available, that is $y_{i,h,t}$ denotes the intake of individual i belonging to household h for the t^{th} week (with $i = 1, \dots, n_{h,t}$; $h = 1, \dots, H$ and $t = 1, \dots, T$), and $D_{i,h,t}$ denotes the same quantity expressed on a body weight basis. The cumulative exposure up to the t^{th} week of this individual is then given by

$$S_{i,h,t} = \exp(-\eta) \cdot S_{i,h,t-1} + D_{i,h,t}, \quad (1)$$

where $\eta > 0$ is the natural dissipation rate of the contaminant in the organism. This dissipation parameter is defined from the so called *half life* of the contaminant, which is the time required for the body burden to decrease by half in the absence of any new intake. For methylmercury, the half life, denoted by $l_{1/2}$, is estimated to 6 weeks, so that $\eta = \ln(2)/l_{1/2} := \ln(2)/6$ (Smith and Farris, 1996).

The autoregressive model defined by (1) and a given initial state $S_{i,h,0} = D_{i,h,0}$ has a stationary solution since $\exp(-\eta) < 1$. As a convention, $S_{i,h,0}$ is set to the mean of all positive exposures $(D_{i,h,t})_{t=1, \dots, T}$. However, this convention has little impact on the level of an individual's long term exposure since the contribution of the initial state $S_{i,h,0}$ tends to zero as t increases. We call this autoregressive model "KDEM" for Kinetic Dietary Exposure Model.

The individual cumulative exposure $S_{i,h,t}$ can be considered to be the long term exposure of an individual for sufficiently large values of t . For methylmercury, the long term steady state of the

individual exposure to a contaminant is reached after 5 or 6 half lives according to Dr P. Granjean, a methylmercury expert. Thus, the long term individual's exposure to methylmercury is defined as the cumulative exposure reached after say $6l_{1/2} = 36$ weeks.

The risk assessment usually consists of comparing the exposure with the so called Provisional Tolerable Weekly Intake (PTWI). This tolerable dose, determined from animal experiments and extrapolated to humans, refers to the dose an individual can ingest throughout his entire life without appreciable risk. For methylmercury, the PTWI is set to 1.6 microgram per kilogram of body weight per week (1.6 $\mu\text{g}/\text{kg}$ bw, see FAO/WHO, 2003).

In our dynamic approach, the long term exposure is compared to a reference long term exposure denoted by S^{ref} , and defined as the cumulative exposure of an individual whose weekly intake is equal to the PTWI, d , such as

$$S^{ref} = \lim_{t \rightarrow \infty} S_t^{ref} = \frac{d}{1 - \exp(-\eta)}, \quad (2)$$

where

$$S_t^{ref} = \sum_{s=0}^t d \exp(-\eta(t-s)) = d \frac{\exp(-\eta(t+1)) - 1}{\exp(-\eta) - 1}. \quad (3)$$

For methylmercury, the reference for long term exposure S^{ref} is 14.6 $\mu\text{g}/\text{kg}$ bw. An individual is then assumed to be at risk if his cumulative exposure $S_{i,h,t}$ exceeds the reference S_t^{ref} for any $t > 6l_{1/2}$.

This KDEM model requires some long surveys of individual intakes which are not monitored and can only be approximated from available consumption data and contamination data.

1.2 From household acquisition data to household intake series

Two current major consumption data sources in France are the national survey on individual consumption (INCA, CREDOC-AFSSA-DGAL, 1999) and the SECODIP panel managed by the company TNS SECODIP. Most quantitative risk assessments conducted by the French agency for food safety (AFSSA) use the 7 day individual consumption data of the INCA survey jointly with contamination data collected by several French institutions. Regarding methylmercury, seafood contamination data have been collected through different analytical surveys (MAAPAR, 1998-2002;

IFREMER, 1994-1998) and were used in Tressou *et al.* (2004) and Crépet *et al.* (2005) combined with the INCA survey. In this paper, a methodology using the SECODIP data is developed (see Boizot, 2005, for a full description of this database). Furthermore, as it is commonly admitted in chronic risk assessment (see Kroes *et al.*, 2002, for a description of the common practices in food risk assessment), mean contamination levels are used rather than the distributions of contamination of the different foods.

The company TNS SECODIP has been collecting the weekly food acquisition data of about five thousand households since 1989. All participating households register grocery purchases through the use of EAN bar codes but other grocery purchases are registered differently: the fresh fruit and vegetable purchases are recorded by the FL sub-panel while fresh meat, fresh fish and wine purchases are recorded by the VP sub-panel. The households are selected by stratification according to several socioeconomic variables and stay in the survey for about 4 years. TNS SECODIP provides weights for each sub-panel and each period of 4 weeks to make sure of the representativeness of the results in terms of several socioeconomic variables. TNS SECODIP also defines the notion of household activity which refers to the correct and regular reporting of household purchases over a year. For each household, the age and gender of each member of the household are retained in our decomposition model with some socioeconomic variables: the region, the social class (from modest to well-to-do), the occupation category and level of education of the principal household earner.

For methylmercury risk assessment, the households of the VP panel are considered; in the 2001 data set, there are $H = 3229$ active households (corresponding to 9288 individuals) and $T = 53$ weeks during which the households may or may not acquire seafood. The weekly purchases of seafood are clustered into two categories ("Fish" and "Mollusks and Shellfish") for which the mean contamination levels are calculated from the MAAPAR-IFREMER data and are given in table 1.

Table 1 around here, see page 24

Household intake series $((y_{h,t})_{h=1,\dots,H;t=1,\dots,T})$ are computed as the cross product between weekly purchases of seafoods which are assimilated to weekly consumptions, and mean contamination levels. They are expressed in micrograms per week ($\mu g/w$).

Before turning to the model description, a few remarks are in order.

1. The use of mean contamination levels in the computation of exposures is a simplification of the model. However it is a current practice in food risk assessment related to chemicals. A comparison between this deterministic approach and a distributional one in which contamination levels are sampled according to their empirical distribution or a chosen parametric distribution was conducted in Tressou *et al.* (2004). It mainly shows that the distribution of exposure shows less variability when using mean contamination but that the quality/quantity of observed measures of contamination should be carefully checked as well as the parametric distribution.
2. One underlying assumption of most quantitative food risk assessments based on consumption and contamination data is the independence of the two sets of data. This implies that consumption levels are not correlated to the contamination levels, which in turn can not be considered as determinants of the consumers' choices. Concerning the MeHg health risk, some information campaigns were conducted in France but their impact seems limited as shown in Roosen *et al.* (2007). From an economic point of view, we can therefore assume that the contamination levels of fish and seafood do not enter as inputs of the consumers' utility function (mostly because they are not aware of the risks and also because of the persistence of the consumption habits). This provides some justification to the hypothesis commonly made in food risk assessment.
3. The food "purchase-consumption" assimilation is of course arguable and will be the main topic of the final discussion (see section 4).
4. An additional assumption concerns the household size, denoted by $n_{h,t}$ for the household h and the week t . This can indeed vary over time in the case of a birth or death of a household member. Since a new born baby will not consume fish in his first few months, we assume that food diversification (and hence consumption of seafoods) starts at one year of age, yielding a total sample of 8913 individuals for the 2001 panel. These household intake series are then decomposed into individual intake series using the model described in the next section. These individual intake series are then used as inputs of the KDEM.

2 Statistical methodology

In this section, the decomposition model is described and compared to similar models described in the literature, namely Chesher (1997, 1998); Vasdekis and Trichopoulou (2000). Its estimation and some structure tests are then presented.

2.1 The decomposition model

2.1.1 General principle

Consider a household composed of $n_{h,t}$ members, each member having unobserved weekly intakes $y_{i,h,t}$, with $i = 1, \dots, n_{h,t}$, $h = 1, \dots, H$, and $t = 1, \dots, T$. The week t intake of a household h is simply the sum across household members of the individual weekly intakes, such as

$$y_{h,t} = \sum_{i=1}^{n_{h,t}} y_{i,h,t}. \quad (4)$$

As detailed below, the individual weekly intake $y_{i,h,t}$ is assumed to depend on

- the age and gender of the individual via a function f ,
- some socioeconomic characteristics of the household,
- time (seasonal variations).

There are obviously several ways to model the individual intake under these assumptions and this choice leads to more or less simple estimation procedures. In Chesher (1997, 1998); Vasdekis and Trichopoulou (2000), a discretization argument on age is used leading to a penalized least square estimation of a great number of parameters, that is one parameter for each year of age and gender. We propose to use a truncated polynomial spline of order 1 for each gender, which admits a mixed model spline representation for f . As far as socioeconomic characteristics are concerned, Chesher (1997) retained a multiplicative specification whereas Vasdekis and Trichopoulou (2000) chose the additive one. In the multiplicative model, a change in income for example would proportionally affect all the individual intakes whereas in the additive setting, they would be affected by the same value. Following Vasdekis and Trichopoulou (2000), we retained the additive specification

since the difference between the two specifications may not be notable, and the additive setting yields to a much simpler estimation procedure (linear model). Finally, time dependency is only introduced in Chesher (1998) to track changes with age within cohorts: this time dependency is directly introduced into the function f that is bivariate smoothed according to age and time (cf. Green and Silverman, 1994). Again, we adopt a simpler specification in which time is introduced as a dummy variable. All these assumptions yield an individual model of the form

$$y_{i,h,t} = x_{i,h,t}\beta + z_{i,h,t}u + w_{h,t}\gamma + \delta_t\alpha + \varepsilon_{i,h,t}, \quad (5)$$

where the terms $x_{i,h,t}\beta + z_{i,h,t}u$ stand for the mixed model spline representation of the function f , the term $w_{h,t}\gamma$ denotes the socioeconomic effects, the term $\delta_t\alpha$ the time effect, and $\varepsilon_{i,h,t}$ is the individual error term.

Combining (4) and (5), we obtain the final rescaled household model given by

$$Y_{h,t} = X_{h,t}\beta + Z_{h,t}u + \sqrt{n_{h,t}}w_{h,t}\gamma + \sqrt{n_{h,t}}\delta_t\alpha + \varepsilon_{h,t}, \quad (6)$$

where $Y_{h,t} \equiv \sum_{i=1}^{n_{h,t}} y_{i,h,t}/\sqrt{n_{h,t}}$, $X_{h,t} \equiv \sum_{i=1}^{n_{h,t}} x_{i,h,t}/\sqrt{n_{h,t}}$, $Z_{h,t} \equiv \sum_{i=1}^{n_{h,t}} z_{i,h,t}/\sqrt{n_{h,t}}$, and $\varepsilon_{h,t} \equiv \sum_{i=1}^{n_{h,t}} \varepsilon_{i,h,t}/\sqrt{n_{h,t}}$.

2.1.2 Specification details

Age-gender function specification Let $a_{i,h,t}$ and $s_{i,h}$ denote the age and sex of individual i of household h for the t^{th} week. Individual dietary intake is generally different according to the gender of individuals, so the function f takes the following form

$$f(a_{i,h,t}, s_{i,h}) = f_M(a_{i,h,t})\mathbb{1}_{\{s_{i,h}=M\}} + f_F(a_{i,h,t})\mathbb{1}_{\{s_{i,h}=F\}},$$

where $f_M(\cdot)$ and $f_F(\cdot)$ are age-intake relationships for males (M) and females (F) respectively, and $\mathbb{1}_{\{A\}}$ is the indicator function of event A . The function $f_S(\cdot)$ is approximated by a spline of order

one with a truncated polynomial basis for either sex, such as

$$f_S(a_{i,h,t}) = \beta_0^S + \beta_1^S a_{i,h,t} + \sum_{k=1}^{K_S} u_k^S (a_{i,h,t} - \kappa_{S,k})_+, \quad (7)$$

where the $(\kappa_{S,k})_{k=1,\dots,K_S}$ are nodes chosen from an age list and

$$(a_{i,h,t} - \kappa_{S,k})_+ \equiv (a_{i,h,t} - \kappa_{S,k}) \mathbb{I}_{\{a_{i,h,t} - \kappa_{S,k} > 0\}}$$

denotes the positive part of the difference between the age of the individual $a_{i,h,t}$ and the node $\kappa_{S,k}$ and the u_k^S are random effects assumed to be i.i.d. Gaussian with distribution $\mathcal{N}(0, \sigma_{u_S}^2)$. This last assumption allows us to introduce some penalties into the model and to smooth the function f_S yielding a mixed model representation for the spline as shown in Speed (1991); Verbyla (1999); Brumback *et al.* (1999); Ruppert *et al.* (2003). As in Ruppert *et al.* (2003), page 125, the total number of nodes K_S is set to $\min\left\{\left\lceil \frac{a_{S,d}}{4} \right\rceil, 35\right\}$, where $a_{S,d}$ is the list of distinct ages for individuals of sex S , and the nodes $\kappa_{S,k}$ are defined as the $\left(\frac{k+1}{K_S+2}\right)^{th}$ percentile of vector $a_{S,d}$ for $k = 1, \dots, K_S$.

Defining $x_{i,h,t}$ as a line vector $\begin{pmatrix} \mathbb{I}_{\{s_{i,h}=M\}} & a_{i,h,t} \mathbb{I}_{\{s_{i,h}=M\}} & \mathbb{I}_{\{s_{i,h}=F\}} & a_{i,h,t} \mathbb{I}_{\{s_{i,h}=F\}} \end{pmatrix}$, and $z_{i,h,t}$ as the line vector $\left\{ (a_{i,h,t} - \kappa_{S,k})_+ \mathbb{I}_{\{s_{i,h}=S\}} \right\}_{k=1,\dots,K_S; S=M,F}$, we finally obtain the first terms of (5), that is $f(a_{i,h,t}, s_{i,h}) = x_{i,h,t} \beta + z_{i,h,t} u$.

Socioeconomic characteristics and time dependency In the application, all the socioeconomic characteristics are categorical variables. Consider the Q categorical variables $W_{h,t}^{(q)}$, $q = 1, \dots, Q$, with m_q modalities, and fix the m_q^{th} modality as the reference modality, then the socioeconomic effect term in (5) and (6) is

$$w_{h,t} \gamma = \sum_{q=1}^Q \sum_{m=1}^{m_q-1} \gamma_{q,m} \mathbb{I}_{\{W_{h,t}^{(q)}=m\}},$$

where $\gamma_{q,m}$ is the effect of the m^{th} modality of the socioeconomic variable q .

In section 3.1, tests are conducted to select the most relevant socioeconomic variables and their modalities.

Similarly, time is only measured by weekly counts throughout the year so that the time effect

in (5) and (6) is simply

$$\delta_t \alpha = \sum_{\substack{\tau=1 \\ \tau \neq \tau_R}}^T \alpha_\tau \mathbb{1}_{\{\tau=t\}},$$

where α_τ is the effect of week τ and τ_R is the reference week.

Error specification The error at the individual level $\varepsilon_{i,h,t}$ is assumed to be Gaussian with zero mean, and the variance-covariance structure is such that

- households are independent, i.e. $\forall i, i', t, t'$ and $\forall h \neq h'$

$$\text{cov}(\varepsilon_{i,h,t}, \varepsilon_{i',h',t'}) = 0,$$

- members of the same household are dependent, that is for $\forall h, t$ and $i \neq i'$,

$$\text{cov}(\varepsilon_{i,h,t}, \varepsilon_{i',h,t}) = \rho \sigma_\varepsilon^2, \quad (8)$$

where ρ measures the dependence between individuals within the same household.

- there is no time dependence, that is $\forall i, i'$ and $\forall t \neq t'$

$$\text{cov}(\varepsilon_{i,h,t}, \varepsilon_{i',h,t'}) = 0. \quad (9)$$

In the rescaled household model (6), the error $\varepsilon_{h,t} \equiv \sum_{i=1}^{n_{h,t}} \varepsilon_{i,h,t} / \sqrt{n_{h,t}}$ is i.i.d. Gaussian with a zero mean and a variance R such that $\forall t, t'$ and $\forall h \neq h'$,

$$\mathbb{V}(\varepsilon_{h,t}) = \rho \sigma_\varepsilon^2 n_{h,t} + (1 - \rho) \sigma_\varepsilon^2 \text{ and } \text{cov}(\varepsilon_{h,t}, \varepsilon_{h',t'}) = 0. \quad (10)$$

These assumptions are discussed and tested in the application section (see 3.1).

2.2 Estimation and tests

The model (6) is a linear mixed model that can be estimated using restricted maximum likelihood (REML) techniques, see Ruppert *et al.* (2003) for details. An attractive consequence of the use

of the mixed model representation of a penalized spline in (7) is that mixed model methodology and software can be used to estimate the parameters and predict the random effect in the resulting household model. The amount of smoothing of the underlying functions f_S is estimated with the REML technique via the estimation of $\sigma_{u_S}^2$. The estimation was conducted using [®]SAS MIXED procedure. To get estimators for σ_ε^2 and ρ , asymptotic least square techniques combined with the linear relationship between the variance given in (10) and the household size were used. More precisely, a residual variance σ_n^2 is first estimated for each household size $n = 1, \dots, N = \max n_{h,t}$ using an option of the MIXED procedure (see the program for the detailed syntax). Then, ordinary least square regression and the delta method give estimators for σ_ε^2 and ρ and their standard deviations.

The individual intake is then predicted by

$$\widehat{y_{i,h,t}} = x_{i,h,t}\widehat{\beta} + z_{i,h,t}\widehat{u} + w_{h,t}\widehat{\gamma} + \delta_t\widehat{\alpha}, \quad (11)$$

where $\widehat{\beta}$, $\widehat{\gamma}$, and $\widehat{\alpha}$ are the estimators of β , γ , and α respectively and \widehat{u} is the best prediction of the random effect u in the model (6).

Confidence and prediction intervals can be built for the prediction $\widehat{y_{i,h,t}}$ as proposed in Ruppert *et al.* (2003) and several tests can be conducted in this model:

1. Are the random effects different according to sex? In other words, is the assertion $\sigma_{u_M}^2 = \sigma_{u_F}^2 = \sigma_u^2$ true?
2. Another question is the necessity for such random effects. Is the assertion $\sigma_u^2 = 0$ (resp. $\sigma_{u_M}^2 = 0$ or $\sigma_{u_F}^2 = 0$) true?
3. More globally, is the function f the same for both sexes? Is the assertion $f_M = f_S$ true?

These tests can be conducted using classical likelihood (or restricted likelihood) ratio techniques. The likelihood ratio statistic is asymptotically distributed as a chi square with a degree of freedom being the number of tested equalities, except for point 2, where the limiting distribution is known to be a mixture of chi-square (Self and Liang, 1987; Crainiceanu *et al.*, 2003) because the test concerns the frontier of the parameter definition ($\sigma_u^2 \in [0, +\infty[$).

3 Applying our methodology to the methylmercury risk assessment

In this section, we illustrate our approach on the methylmercury risk assessment. Firstly, several tests are conducted on the decomposition model yielding a final estimation of the individual intake series based on the SECODIP data. An empirical validation on individual consumption data is then proposed. Finally, individual long term exposure obtained from our model is compared to the reference long term exposure described in section 1.

3.1 Estimation and tests on the structure of the model

Table 2 shows a preliminary REML estimation of our model, defined in (6), under the following assumptions:

- the socioeconomic variables are household income, region of residence, occupation category and level of education of the principal household earner, their modalities (inc. the reference) are given in Table 2.
- the function f differs according to the gender but the random effect does not ($f_M \neq f_F$ and $\sigma_{u_M}^2 = \sigma_{u_F}^2$),
- the maximum household size \overline{N} is set to 6 for variance-covariance estimation. Indeed, the dependence between individuals within the same household depends on the household size n_h in (10). For each household size, a variance is estimated, and estimates of ρ and σ^2 are obtained using asymptotic least square techniques as mentioned in section 2.2. Since large households are not numerous in the database, the estimations are implemented with a maximum household size, \overline{N} , set to 6; it is assumed that there is a common variance for all households with size greater than \overline{N} .

Table 2 around here, see page 24

Selection of the sociodemographic covariates/modalities In this sub-section, we show the results of several tests we carried out to simplify the interpretation of our study. These tests

have been implemented in a hierarchical way, starting with the highest-order interaction terms, combining to the reference modality the modality which does not differ significantly from the reference. All tests are performed on the 5% level of significance and each new hypothesis is tested conditionally on the results of the previous tests. Each null hypothesis and the p-value resulting from the appropriate F-test are shown in Table 3.

First of all, concerning the occupation category variable, the self-employed modality does not significantly differ from the reference modality blue collar workers ($H1$, $Pval = 0.771$). Refitting the model with the reference modality "Blue collar workers and self employed", all the socioeconomic variables are significantly different from the reference. Then, F-tests allow us to conclude that the resulting three groups are significantly different from each other ($H2$, $H3$, $H4$).

Let us now consider the region of residence variable. First, there are some very substantial differences among the 4 regions of residence ($H5$, $Pval \leq 0.001$). However, the modality "North, Brittany, and Vendee coast" and the modality "Paris and its suburbs" should be grouped ($H6$, $Pval_c = 0.881$). Then, the other tests implemented for the level of education and income variables suggest that no further simplification is possible (see p-values of null hypotheses $H7$, $H8$, $H9$ in Table 3). Finally, the overall F-test comparing our resulting final model to the original model (6) shows that no important variable has been left out of the model ($Pval = 0.59$).

Other tests on the final model Likelihood ratio tests are implemented to test the structure of the final model, that is the one resulting from the previous tests and simplifications regarding the socioeconomic variables.

First, the dependence of individual exposures to methylmercury within a household is tested. The null hypothesis $\rho = 0$ (cf. equation (10)) is rejected ($Pval < 0.001$) which confirms that individuals within the same household have correlated exposures. Moreover, a graphical comparison of the exposure curves from the two models shows that accounting for the dependence within the household reduces the mean individual exposure in the adult population and increases the children's (Tressou, 2005). Time dependence in the errors (cf. (9)) was also investigated in Tressou (2005) (see pages 139 and 147) where two models were confronted: in the first one, time independence and within household dependence were assumed, and in the second one, autoregressive correlation of order 1 and within household independence were assumed. A comparison of the Akaike criteria

favoured the first model. Unfortunately, limitations in the parametrization of the R variance structure enabled the test of joint time and within household dependences but this will be investigated in future work.

Finally, we test if the function f is the same for both genders. The null hypothesis $f_M = f_F$ is rejected ($Pval < 0.001$) but the null hypothesis $\sigma_{u_M}^2 = \sigma_{u_F}^2$ is accepted. This means that individual exposure differs with gender but both functions need the same amount of smoothing.

Regarding the error structure, basic model checks are quite disappointing. Indeed, normality is rejected by usual tests (Kolmogorov-Smirnov, Cramer-von Mises, Anderson-Darling tests have p-values lower than 1%). This is mainly due to the important disymetry exhibited by the residuals as shown in Figure 1. Moreover, since the dependent variable are positive, the residual plot is truncated by below, see Figure 2. For standard regressions, a (log) transformation is usually applied to the data but in our case this kind of (nonlinear) transformation would annihilate the possibility of prediction for individual quantity, namely $\log \Sigma_i y_i$ is not equal to $\Sigma_i \log y_i$. Other solutions could be studied to account for the disymetry: choice of some disymetric parametric distribution such as the (shifted) lognormal distribution or adopt generalized linear mixed modelling approach. Bootstrap techniques can also be adapted. However, in this premier model, we chose not to engage in such refinements, convinced that other limitations of the model would need to be addressed first, namely the failure of the proposed model to determine non-consumers (see the discussion section).

Figure 1 around here, see page 25

Figure 2 around here, see page 26

Main features of the final model Table 4 shows the parameter estimates and p-values of the Student's t-tests for all socioeconomic variables of the reduced final model. The income effects on individual exposure are those expected: the richer the households are, the higher their exposures are because seafoods are expensive. Furthermore, living in a coastal region or in Paris and its suburbs brings about larger individual exposure relatively to living in a non coastal region because of the more ready supply of seafoods in these regions. Moreover, the more educated you are, the larger the individual exposure is. The occupation category of the principal household earner has an unexpected effect on the individual exposure. Indeed a higher exposure is expected for white collar

workers and retirees when compared to blue collar workers but an opposite effect is observed. This may be explained by the fact that the reference modality for this variable is a very heterogeneous modality also comprising managers and self-employed persons (farmers and craftsmen). Another explanation could be that white collars workers have a higher propensity to eat out in restaurants whereas outside the home consumption is not included in the model.

Table 3 around here, see page 27

Table 4 around here, see page 27

Finally, the individual exposure is plotted as a function of age in Figure 3 for the female and male populations respectively. Children exposure is quite similar for both gender but the adult female subpopulation tends to be more exposed than the adult male subpopulation. This is not a body weight effect since it remains true when plotting the exposures expressed in $\mu g/w$ instead of $\mu g/kgbw/w$ (graphics not shown). Besides, when considering the 95%- confidence intervals (CI's) for the curves, also given in Figure 3 for both subpopulations, we observe that this difference is significant at least for adults between roughly 55 and 75. These CI's also illustrate that our estimation procedure yields more uncertainty and variability at the edge of the graph, that is for the younger and the older individuals. This is common to most spline estimation and also results here from an extrapolation bias in the case of children. Indeed, the model parameters are estimated from the household model and there is no household exclusively composed of children. However as we shall see in the next section, the error certainly remains within the computed confidence intervals.

Figure 3 around here, see page 28

3.2 Empirical validation of the decomposition model

In this subsection, an empirical validation of the decomposition model based on individual consumption data is proposed. Indeed, the French INCA data (mentioned in the introduction as the main database for individual food consumption in France) provides the individual consumptions of 3003 persons among which $n = 1613$ actually belong to some $H = 697$ households whose members aged over 3 were all interviewed about their individual consumptions. This dataset can therefore be

used to validate our model comparing the true exposures computed from the observed individual consumptions and the estimated exposures obtained from the decomposed household total exposures. Figures 4 and 5 display the results of this empirical validation for each gender¹. In the adult population, our model gives mean exposures very close to the true ones even though there is a slight overestimation in the adult male population. For children aged over 3, there is an underestimation of the mean exposure, namely for young girls. However the general shape of the curves is reproduced. Moreover, the body weight approximation is also investigated in this empirical validation since individual body weights are available in the INCA dataset and it is shown to have very little impact on the estimated exposures.

The comparisons of the curves obtained from the INCA data (Figures 4 and 5) and the estimated exposure resulting from the decomposition model (Figure 3) shows quite a good adequation in the levels of exposures except for the teenagers and young adults. This difference is mainly due to the fact that consumption outside the home which is high for this age class is included in the INCA individual data and not in the SECODIP panel, see point 1 in the discussion about the use of household acquisition data.

Figures 4 and 5 around here, see page 28 and 29

3.3 The cumulative and the long term individual exposure

The cumulative individual exposure $S_{i,h,t}$ is calculated from the estimated individual weekly intakes according to equation (1) and the resulting values for $t > 35$ are compared to the reference cumulative exposure defined by (3). Figure 6 shows the cumulative individual exposure over the 53 weeks of the year 2001 for different individuals. Only certain percentiles of the distribution of the individual cumulative exposures of the last week are displayed. For example, the curve **Pmax** represents the cumulative exposure of an individual whose last week's cumulative exposure is the highest. This is the cumulative exposure of a girl who turned one year old during the 30th week of 2001, lives in Paris or its suburbs in a well to do household.

¹In this application of the decomposition model, assumptions similar to the previous section are used regarding the basic structure of the model and the introduced socioeconomic variables are the region of residence and the occupation category of the principal household earner. There is no time effect since the INCA data only records the consumption over one week.

Very few individuals have a cumulative individual exposure above the reference long term exposure. We estimate that only 0.186% of individuals are deemed at risk. This risk index should be compared to the more common one defined as the percentage of weekly intakes $D_{i,h,t}$ exceeding the PTWI, denoted $R_{1.6}$, such as $R_{1.6} = \frac{1}{nT} \sum_{t=1}^T \sum_{h=1}^H \sum_{i=1}^{n_h} \mathbb{1}(D_{i,h,t} > 1.6)$. $R_{1.6}$ is equal to 0.45%, and is slightly higher since each occasional deviation above the PTWI increases the risk index whereas only long term deviations above this PTWI should be taken into account to assess the risk.

A deeper analysis of at risk individuals shows that all these vulnerable individuals are children less than three years old. They represent 5.29% of the children aged between 1 and 3 in 2001. Further, no child of a modest households is found to be at risk.

Figure 6 around here, see page 29

4 Discussion

In our opinion, two main topics need to be discussed after this study: first the limitations of the decomposition model and then the use of household acquisition data in a food safety context.

Limitations of the decomposition model As already mentioned in section 3.1, the model does not fit very well yielding nonnormal residuals with an important disymetry so that predicted individual exposures most often sum to a quantity smaller than the observed. This is certainly due to the fact that null consumptions (and thus null exposures) are not accounted for in our model. This could be improved considering a Tobit type model at the individual level (instead of (5)). In term of risk, this is important since null consumption of one household member automatically increases the exposures of others. This should be the subject for future investigations.

Regarding the conclusion of the long term risk assessment stating that children between 1 and 3 are the more vulnerable population, the main limitation of the decomposition model is obviously the extrapolation bias concerning the children population. However, the empirical validation proposed in section 3.2 partly restores confidence in our model although the data used in the validation only concerns individuals over 3 years old. Moreover, this kind of bias is inherent to decomposition model and can also be underlined in other models of this kind (Chesher, 1997; Vasdekis and Trichopoulou,

2000). A comparison of the performances of these models was conducted in Tressou (2005) and illustrated that the mean squared error was lower in our model.

Use of household acquisition data As mentioned in section 1, the use of household acquisition data in a food safety context, and in our case the use of the SECODIP database for assessing methylmercury dietary intakes, gives rise to some approximations:

1. Consumption outside of the home is out of the scope of household acquisition data. TNS SECODIP does not provide any information on the quantities of seafoods consumed out of the home or bought for outside consumption. Nevertheless, Serra-Majem *et al.* (2003) assert that these data are good estimates for the consumption of the whole household. Vasdekis and Trichopoulou (2000) avoid this question by using the term "availability" instead of intake or consumption. However, as in Chesher (1997), auxiliary information about outdoor consumption could be introduced in the model as a correction factor accounting for the propensity to eat outside of the home according to age, sex or socioeconomic variables. The French INCA survey on individual consumptions gives details about inside / outside the home consumption for 3003 individuals people aged 3 and older. The mean outside the home consumption proportion is 20% for seafoods. Applying such a factor to all household intakes yields a long term risk of 0.226%, and $R_{1.6} = 0.791\%$. Furthermore, in this case, a small proportion of consumers older than 3 years old are vulnerable. Nevertheless, children aged between 1 and 3 in 2001 still represent the most vulnerable consumer group, at 10% of the corresponding population.
2. The amount of food bought by a household can be different from the amount actually consumed. Indeed, namely for seafoods, a non negligible part is not edible: Favier *et al.* (1995) show that on average only 61% of fresh or frozen fish is edible. Besides, Maresca and Poquet (1994) also demonstrate some part of the purchased food is thrown away, which also reduces the actual amount of food consumed by a household. However, SECODIP does not specify whether the quantity of fresh or frozen fish bought is ready to be consumed or as a whole fish that needs some preparation. Applying such a factor to all household intakes yields a long term risk of 0.00%, and $R_{1.6} = 0.043\%$. If both the 20% outside of the home consumption

correction factor and the 61% edible proportion factor are applied to our series, the long term risk is equal to 0.021%, $R_{1.6} = 0.13\%$, and 1.06% of the population of children aged between 1 and 3 are vulnerable. These results stress that applying such a correction factor to assess the actual quantity consumed is probably too strong and is certainly a crude approximation of the quantity of seafoods ingested. Thus, a more detailed database on fish and seafood is needed, to realize an accurate assessment of exposure to methylmercury, taking into account only the edible part of fish and other seafood.

3. Body weight information is crucial in a food safety context and will be included in the future SECODIP data since it has now been added to the list of required individual characteristics. The measurement error afferent to this quantity will remain however, namely for children whose body weight changes a lot throughout a year. Nevertheless, approximating the weekly body weight of young children by the median of the weekly body weight distribution available in French health records is the best approximation possible. Moreover, the empirical validation of section 3.2 clearly states that this body weight has a minor impact at least on the estimation of exposure for people older than 3.
4. The food nomenclature of the SECODIP database is not as detailed as the contamination database. Unfortunately, fish and seafood species are not well documented so it is not possible to consider more than two food categories when computing household intakes. Namely, in the case of fishes, it is clear that the contamination highly depend on the species (tuna being one the most contaminated fish) and when averaging all fish contaminations in the computation of the household intake, we reduce the actual variability of the exposure. However, this problem of nomenclature matching is ubiquitous of food risk assessments since contamination analysis are generally conducted independently from the food nomenclature of consumption data.

These arguments mainly show the disadvantages of the use of household food acquisition data such as the SECODIP database. Nevertheless, they also present many advantages compared to the individual food record survey mainly used in France in the food safety context:

- As mentioned before, households respond for a long period of time (the average is 4 years in the SECODIP panel) which allows us to observe long term behaviors and avoid some well

known biases of individual food record surveys. For example, respondents might over- (under-) declare certain foods with a good (bad) nutritional value either deliberately or just because they increased (reduced) their consumption for the short (7 days) period of the survey.

- The individual surveys are expensive and very difficult to conduct. Highly trained interviewers are required as well as an extraordinary cooperation from respondents. Household food acquisition data can serve many other applications (economics or marketing) and, at least for the SECODIP data, acquisition recording is simplified by optical scanning of food barcodes.

Conclusion

In this paper, we proposed a methodology to assess chronic risks related to food contamination using the example of methylmercury exposure through seafood consumption. This methodology includes the definition of a Kinetic Dietary Exposure Model (KDEM) that integrates the fact that contaminants are eliminated from the body at different rates, the rate being measured by the half life of the contaminant. In this paper, the estimation is based on the use of household food acquisition data which are first decomposed into individual intake data through a disaggregation model accounting for the dependence among household members. One important feature of this model is that it simplifies into a linear mixed model which can be estimated using standard routines even if the parametrization of the variance components may be difficult. Several extensions of this methodology are currently studied. First, the disaggregation model proposed here presents many limitations and would be considerably improved by considering a preliminary step in which we determine what member is an actual consumer, in the spirit of the Tobit model. The KDEM idea is also currently being developed by studying the stability and ergodic properties of the underlying continuous time piecewise deterministic Markov process (Bertail *et al.*, 2007). The parameters of this new model are the intake distribution, the inter-intake time distribution and the dissipation rate distribution. In this framework, the dissipation parameter η of the KDEM model is random and the intake and inter-intake time distributions can be estimated either from individual (INCA-type) data or household (SECODIP-type) data, see Verger *et al.* (2007) for an illustration with the INCA data.

References

- Bertail, P., S. Cléménçon and J. Tressou (2007). A storage model with random release rate for modeling exposure to food contaminants. *Mathematical Biosciences and Engineering*. In press, available at <https://hal.archives-ouvertes.fr/hal-00138279>.
- Boizot, C. (2005). Présentation du panel de données SECODIP. Technical report. INRA-CORELA.
- Brumback, B., D. Ruppert and M. P. Wand (1999). Comment on "variable selection and function estimation in additive non-parametric regression using a data-based prior" by Shively, Kohn, and Wood. *Journal of the American Statistical Association* **94**, 794–797.
- Chesher, A. (1997). Diet revealed?: Semiparametric estimation of nutrient intake-age relationships. *Journal of the Royal Statistical Society A* **160**(3), 389–428.
- Chesher, A. (1998). Individual demands from household aggregates: Time and age variation in the quality of diet. *Journal of Applied Econometrics* **13**(5), 505–524.
- Crainiceanu, C. M., D. Ruppert and T. J. Vogelsang (2003). Some properties of likelihood ratio tests in linear mixed models. (Working Paper).
- CREDOC-AFSSA-DGAL (1999). *Enquête INCA (individuelle et nationale sur les consommations alimentaires)*. TEC&DOC ed.. Lavoisier, Paris. (Coordinateur : J.L. Volatier).
- Crépet, A., J. Tressou, P. Verger and J. Ch. Leblanc (2005). Management options to reduce exposure to methyl mercury through the consumption of fish and fishery products by the French population. *Regulatory Toxicology and Pharmacology* **42**(2), 179–189.
- Engle, R. F., C. W. J. Granger, J. Rice and A. Weiss (1986). Non-parametric estimation of the relationship between weather and electricity demand. *Journal of the American Statistical Association* **81**, 310–320.
- FAO/WHO (2003). Evaluation of certain food additives and contaminants for methylmercury. Sixty first report of the Joint FAO/WHO Expert Committee on Food Additives, Technical Report Series. WHO. Geneva, Switzerland.
- Favier, C., J. Ireland-Ripert, C. Toque and M. Feinberg (1995). *Répertoire Général des Aliments, Table de composition, tome 1*. TEC&DOC ed.. Lavoisier, Paris.
- Green, P.J. and B.W. Silverman (1994). *Nonparametric Regression and Generalized Linear Models*. Chapman & Hall.
- Hamilton, J. D. (1994). *Time Series Analysis*. Princeton University Press.
- Hoffmann, K., H. Boeingand, A. Dufour, J. L. Volatier, J. Telman, M. Virtanen, W. Becker and S. De Henauw (2002). Estimating the distribution of usual dietary intake by short-term measurements. *European Journal of Clinical Nutrition* **56**, 53–62.
- IFREMER (1994-1998). Résultat du réseau national d’observation de la qualité du milieu marin pour les mollusques (RNO).
- Kroes, R., D. Müller, J. Lambe, M. R. H. Löwick, J. v. Klaveren, J. Kleiner, R. Massey, S. Mayer, I. Urieta, P. Verger and A. Visconti (2002). Assessment of intake from the diet. *Food and Chemical Toxicology* **40**, 327–385.
- MAAPAR (1998-2002). Résultats des plans de surveillance pour les produits de la mer. Ministère de l’Agriculture, de l’Alimentation, de la Pêche et des Affaires Rurales.

- Maresca, B. and G. Poquet (1994). Collectes sélectives des déchets et comportements des ménages. Technical Report R146. CREDOC.
- Nusser, S.M., A.L. A.L. Carriquiry, K.W. Dodd and W.A. Fuller (1996). A semiparametric transformation approach to estimating usual intake distributions. *Journal of the American Statistical Association* **91**, 1440–1449.
- Patterson, H. D. and R. Thompson (1971). Recovery of inter-block information when block sizes are unequal. *Biometrika* **58**, 545–554.
- Roosen, J., S. Marette, S. Blanchemanche and P. Verger (2007). The effect of product health information on liking and choice. *Food Quality and Preference* **18**, 759–770.
- Ruppert, D., M .P. Wand and R. J. Carroll (2003). *Semiparametric regression*. Cambridge Series in Statistical and Probabilistic Mathematics. Cambridge University Press.
- Self, S. G. and K.Y. Liang (1987). Asymptotic properties of maximum likelihood estimators and likelihood ratio tests under nonstandard conditions. *Journal of the American Statistical Association* **82**(398), 605–610.
- Sempé, M., G. Pédrón and M. P. Roy-Pernot (1979). *Auxologie, méthode et séquences*. Théraplix. Paris.
- Serra-Majem, L., D. MacLean, L. Ribas, D. Brule, W. Sekula, R. Prattala, R. Garcia-Closas, A. Yngve and M. Lalonde and A. Petrasovits (2003). Comparative analysis of nutrition data from national, household, and individual levels: results from a WHO-CINDI collaborative project in Canada, Finland, Poland, and Spain. *Journal of Epidemiology and Community Health* **57**, 74–80.
- Smith, J. C. and F. F. Farris (1996). Methyl mercury pharmacokinetics in man: A reevaluation. *Toxicology And Applied Pharmacology* **137**, 245–252.
- Speed, T. (1991). Discussion of “that blup is a good thing: the estimation of random effects” by g. robinson. *Statistical science* **6**, 42–44.
- Tressou, J. (2005). Méthodes statistiques pour l’évaluation du risque alimentaire. PhD thesis. Université Paris X.
- Tressou, J., A. Crépet, P. Bertail, M. H. Feinberg and J. C. Leblanc (2004). Probabilistic exposure assessment to food chemicals based on extreme value theory. application to heavy metals from fish and sea products. *Food and Chemical Toxicology* **42**(8), 1349–1358.
- Vasdekis, V.G.S. and A. Trichopoulou (2000). Non parametric estimation of individual food availability along with bootstrap confidence intervals in household budget surveys. *Statistics and Probability Letters* **46**, 337–345.
- Verbyla, A. (1999). *Mixed Models for Practitioners*. Biometrics SA, Adelaide.
- Verger, P., J. Tressou and S. Cléménçon (2007). Integration of time as a description parameter in risk characterisation: application to methyl mercury. *Regulatory Toxicology and Pharmacology* **49**(1), 25–30.
- WHO (1990). Methylmercury, environmental health criteria 101. Technical report. Geneva, Switzerland.

Figures and Tables

Table 1: Description of the contamination database (Unit: microgram per kilogram)

	Mean	Min	Max	Standard Deviation	Number of analysis
Fish	0.147	0.003	3.520	0.235	1350
Mollusk and Shellfish	0.014	0.001	0.172	0.011	1293

Table 2: Restricted maximum likelihood estimates (REML) for age and all socioeconomic variables and the p-value of the Student's tests (Pval)

Effect	Parameter	REML	Pval
Income	(ref: Mean sup)		
Well to do	γ_1	6.027	<0.001
Mean inf	γ_2	2.686	<0.001
Modest	γ_3	-1.928	<0.001
Region of residence	(ref: Noncoastal regions)		
North, Brittany, Vendee coast	γ_4	0.962	0.003
South West coast	γ_5	5.232	<0.001
Mediterranean coast	γ_6	2.303	<0.001
Paris and its suburbs	γ_7	1.023	0.009
Occupation category of the principal household earner	(ref: Blue collar workers)		
self-employed persons	γ_8	-0.122	0.771
white collar workers	γ_9	-3.733	<0.001
retirees	γ_{10}	-5.261	<0.001
no activity	γ_{11}	-1.910	0.004
Level of Education of the principal household earner	(ref: BAC and higher degree)		
student	γ_{12}	5.901	<0.001
no or weak diploma	γ_{13}	-1.281	<0.001

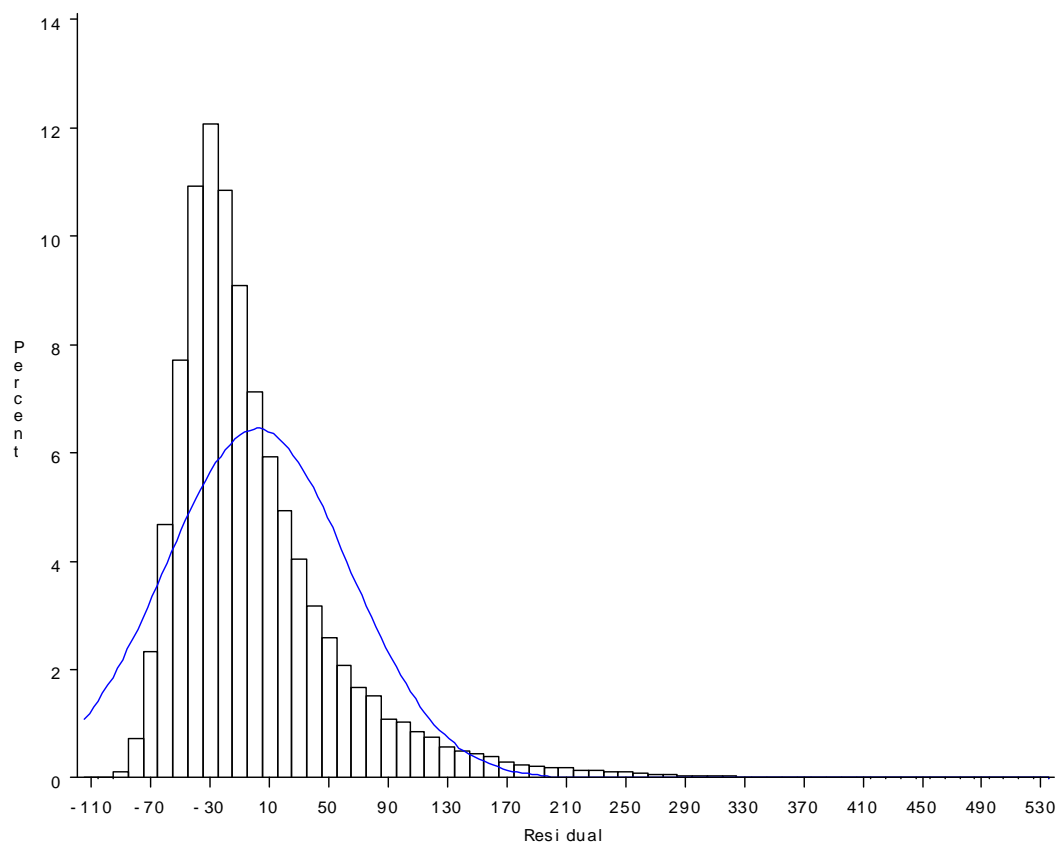


Figure 1: Model check: histogram and normal density adjustment (final model, Secodip data).

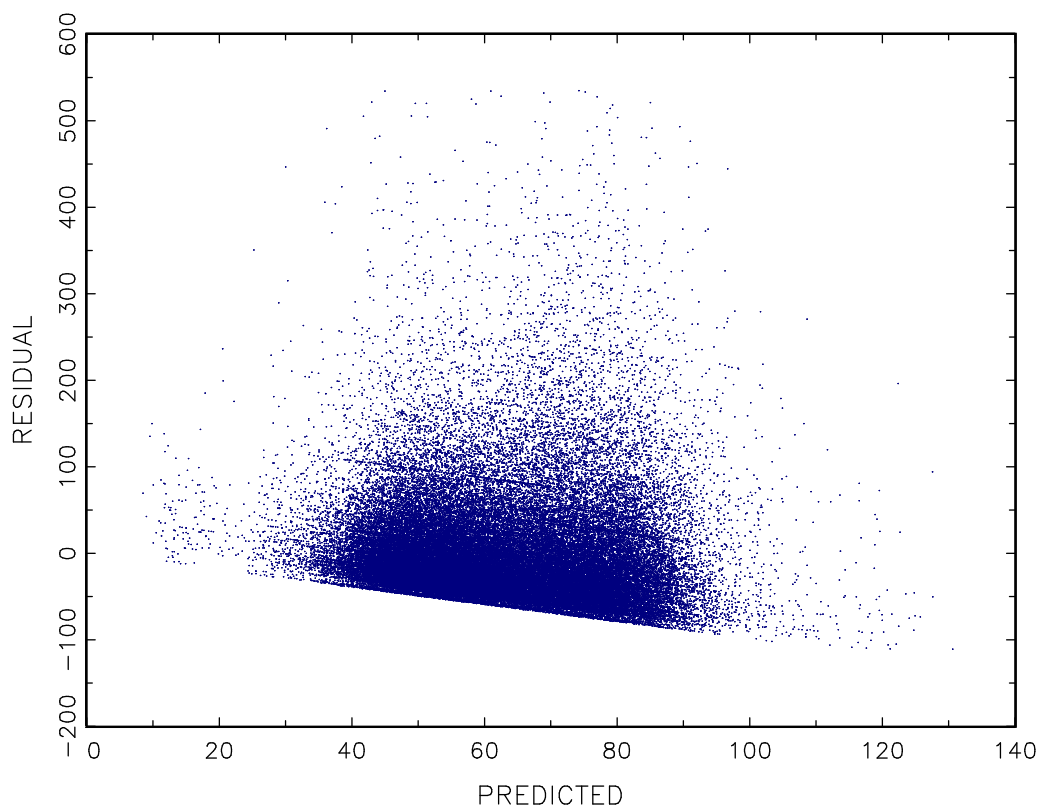


Figure 2: Model check: residual plot (final model, Secodip data).

Table 3: The different steps performed in testing the socioeconomic part of our model. For each step, the null hypothesis tested and the p-value resulting from the appropriate F-test are shown. All tests are performed conditionally on the results of the previous tests (Pval)

Null hypothesis	Pval
H1 : $\gamma_8 = 0$	0.771
H2 : $\gamma_9 = \gamma_{10}$	0.030
H3 : $\gamma_9 = \gamma_{11}$	0.018
H4 : $\gamma_{10} = \gamma_{11}$	<0.001
H5 : $\gamma_4 = \gamma_5 = \gamma_6 = \gamma_7$	<0.001
H6 : a : $\gamma_4 = \gamma_5$	<0.001
b : $\gamma_4 = \gamma_6$	<0.001
c : $\gamma_4 = \gamma_7$	0.881
d : $\gamma_5 = \gamma_6$	<0.001
e : $\gamma_5 = \gamma_7$	<0.001
f : $\gamma_6 = \gamma_7$	0.0103
H7 : $\gamma_{12} = \gamma_{13}$	<0.001
H8 : $\gamma_1 = \gamma_2 = \gamma_3$	<0.001
H9 : a : $\gamma_1 = \gamma_2$	<0.001
b : $\gamma_1 = \gamma_3$	<0.001
c : $\gamma_2 = \gamma_3$	<0.001

Table 4: Restricted maximum likelihood estimates (REML) for all age and socioeconomic variables of the reduced final model with all variance components and their standard errors (s.e)

Effect	Parameter	REML	Pval
Income	(ref: Mean inf)		
Well to do	γ_1	6.062	<0.001
Mean inf	γ_2	2.708	<0.001
Modest	γ_3	-1.931	<0.001
Region of residence	(ref: Non coastal regions)		
Paris and North, Brittany, Vendee coast	$\gamma_4 = \gamma_7$	0.984	<0.001
South west coast	γ_5	5.232	<0.001
Mediterranean coast	γ_6	2.297	<0.001
Occupation category of the principal household earner	(ref: Blue collar workers and self employed persons)		
white collar workers	γ_9	-3.704	<0.001
retirees	γ_{10}	-5.242	<0.001
no activity	γ_{11}	-1.877	0.005
Level of education of the principal household earner	(ref: BAC and higher degree)		
student	γ_{12}	5.901	<0.001
no or weak diploma	γ_{13}	-1.275	<0.001
		REML	s.e
Variance of the random effect	σ_u	24.832	6.7316
Variance-covariance structure			
variance	σ^2	1260705	282309
correlation	ρ	-0.22	0.0434

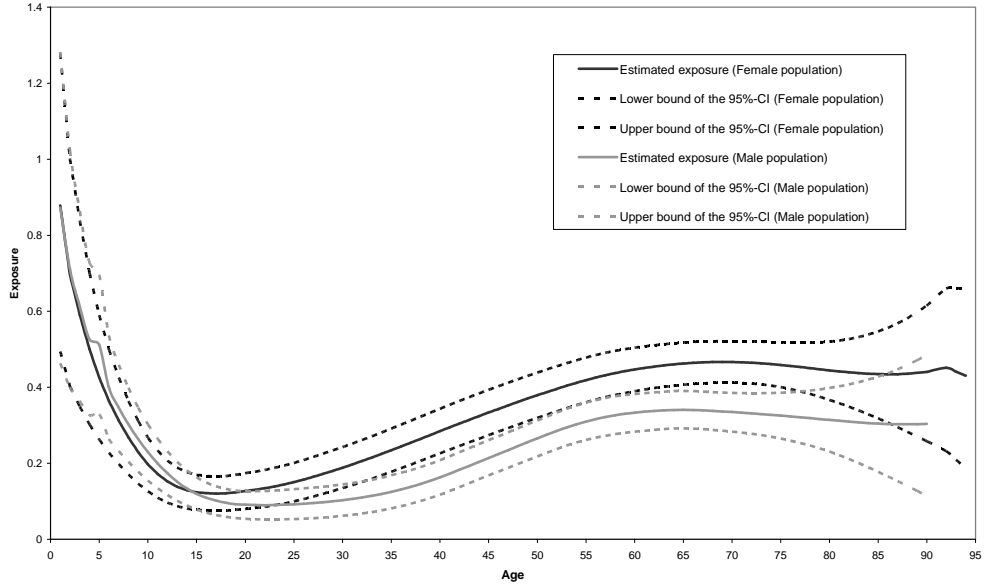


Figure 3: Individual exposure as a function of age according to gender (unit: μg per kilogram of body weight per week).

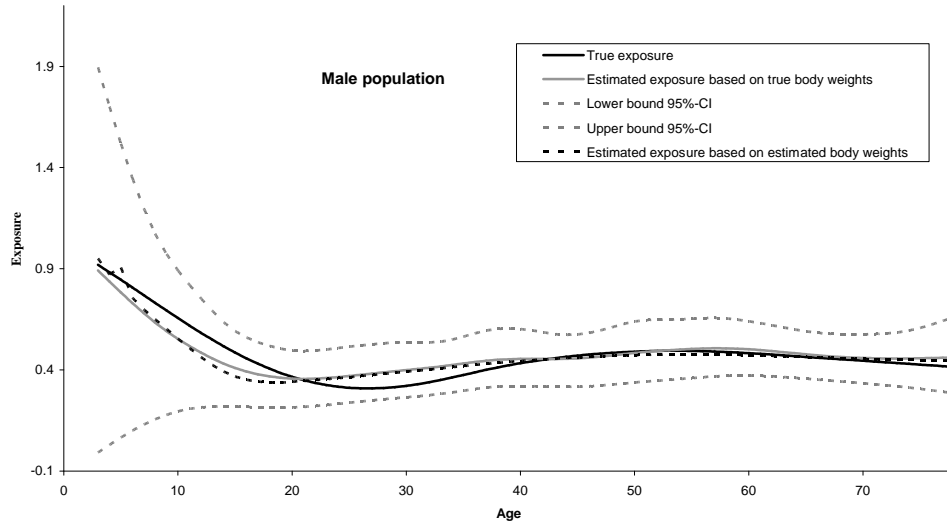


Figure 4: Empirical validation of the decomposition model using individual consumption data (Male population, unit: μg per kilogram of body weight per week).

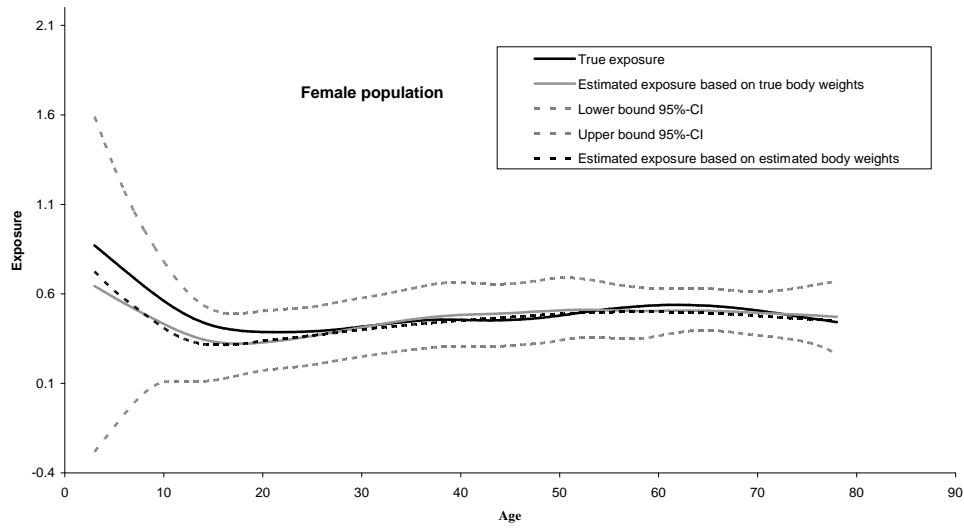


Figure 5: Empirical validation of the decomposition model using individual consumption data (Female population, unit: μg per kilogram of body weight per week).

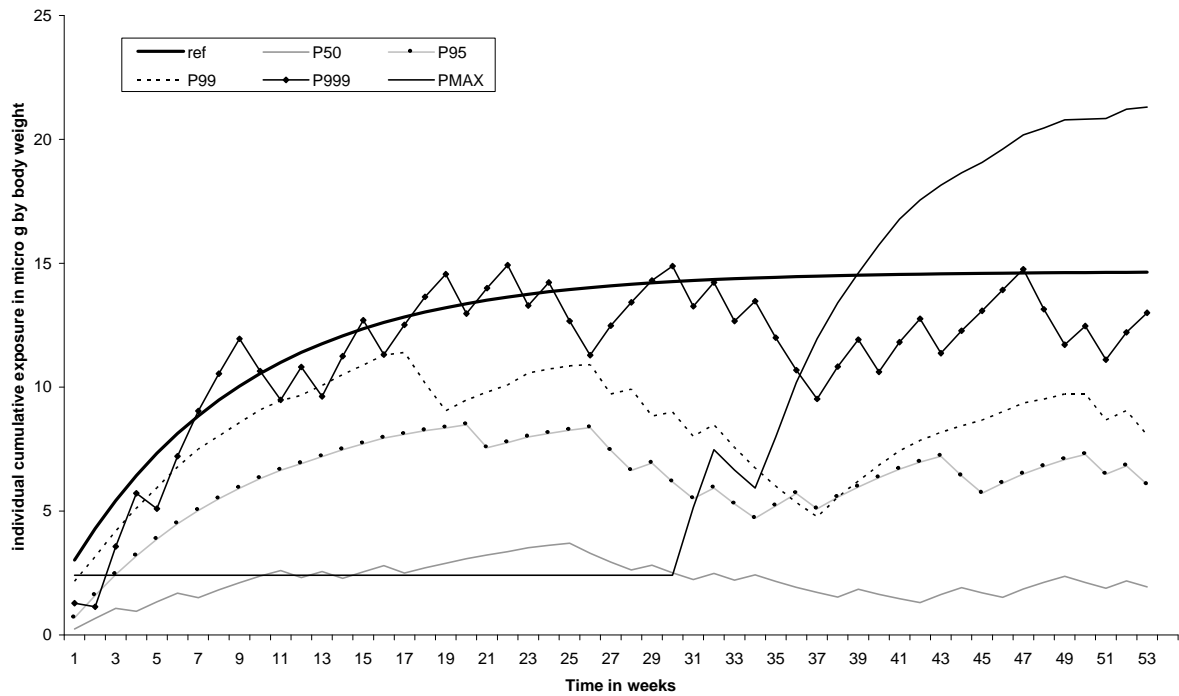


Figure 6: Cumulative exposure to MeHg (unit: μg per kilogram of body weight)