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Exposure assessment for dioxin-like PCBs intake from organic and conventional meat integrating cooking and digestion effects

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ABSTRACT

In this paper, exposure to Polychlorinated biphenyls (PCBs) related to bovine meat consumption is assessed based on multiples sources of data, namely data collected within the national research project "SoMeat" that objectively assesses the potential risks and benefits of organic and conventional food production systems in terms of contaminants respective contents. The work focuses on dioxin like PCBs in bovine meat in France. A modular Bayesian approach is proposed including measures after production, effect of cooking, levels and frequency of consumption and effect of digestion. In each module, a model is built and prior information can be integrated through previously acquired data commonly used in food risk assessment or vague priors. The output of the global model is the exposure including both production modes (organic and conventional) for three different cooking intensities (rare, medium, and well-done), before digestion and after digestion. The main results show that organic meat is more contaminated than conventional meat in mean after production stage and after cooking although cooking reduces the contamination level. This work is a first step of refined risk assessment integrating different steps such as cooking and digestion in the context of chemical risk assessment similarly to current microbiological risk assessments.

1. Introduction

Food represents one of the main sources of human exposure to harmful pollutants, of which meat in particular represents a non-negligible source (Engel et al., 2015). Even if there is no clear evidence that organic food better protect consumers from chemical contamination than conventional products (Van Loo et al., 2012), this remains one of the major forces driving consumers to buy organic (Ghidini et al., 2012). The awareness on the harmful effects of some chemicals present in food is increasing among the consumers according to an EFSA report (EFSA, 2010), and consumers concern over the safety of conventional food has intensified, becoming thus the main force for increasing demand for organic produced food. In order to assess the merits of these allegations, a first study (Dervilly-Pinel et al., 2017) enabled to compare French organic and conventional food production systems in terms of their content of more than 250 contaminants including environmental micropollutants, mycotoxins and residues from veterinary or phytosanitary treatments. Organic meat, while complying with regulatory limits, was shown to be significantly more contaminated by environmental micropollutants than conventional products, which could be explained by longer rearing periods and systematic outdoor access. Interestingly, median contamination levels in DL-PCBs were observed more than twice higher in organic meat compared to conventional meat while about 90–98% of the average exposure of humans to PCBs results from dietary intake, with food of animal origin being the predominant sources (Malisch and Kotz, 2014).

PCBs are synthetic chemical contaminants that are released from several sources including electrical appliances, heat exchange fluids, etc. They accumulate and persist in the environment contaminating both animals and humans. Although their production was forbidden from the mid-1970s, they are still found in the environment given their important persistence.

Several experiments on the toxicity of these chemicals have been conducted in the last decades, and experiments on animals and health studies have demonstrated that PCBs may be associated to several adverse health effects. In particular, they were classified as a probable

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carcinogen factor to humans. This raised an increasing public concern about the health hazards of these components and several risk studies were conducted to evaluate the risk of exposure (Lauby-Secretan et al., 2013).

PCBs are a group of organochlorine compounds that are synthesized by catalyzed chlorination of biphenyl. Depending on the number of chlorine atoms and their position, there are 209 theoretically possible congeners. They were massively produced for over four decades, from 1929 until they were banned in the 1980s. Based on structural characteristics and toxicological effects, PCBs can be divided into two groups. One group consists of 12 congeners (77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189) that easily can adopt a coplanar structure and show toxicological properties similar to dioxins (PCDD/F). This group is therefore often termed 'dioxin-like PCBs' (DL-PCBs). Each of these DL-PCBs contributes to the global toxicity with a different degree. The overall contamination of DL-PCB (TEQ) is therefore expressed as weighted sum of the concentration of the different molecules, the weight being the toxicity equivalence factor (TEF) that accounts for the relative toxicity power of every molecule with respect to that of the most toxic molecules among the dioxins. The PCB 126 contributes to nearly 95% of the overall toxicity. Most other PCBs do not show dioxin-like toxicity and are therefore termed 'non dioxin-like PCBs' (NDL-PCBs). Among the NDL-PCBs, six are considered as appropriate indicators for different PCB patterns in various sample matrices: PCB-28, 52, 101, 138, 153 and 180.

The main routes of PCB human exposure are air, human milk, food and drinking water. Since PCBs are lipophilic and accumulate in fatty tissues, dietary exposure from animal products is the predominant source of exposure and contributes to nearly 90% of total dioxin and PCB exposure (JECFA, 2002).

In chemical food risk assessment, the exposure is commonly evaluated through the combination of consumption data and contamination data either coming from total diet studies –that is analyses of food as consumed- or from surveillance plans – that is analyses of raw food. However, for a more realistic estimation of the toxicity of the contaminants, it is necessary to account for any changes in the concentrations that may occur during processing such as cooking (Domingo, 2017), or gastrointestinal digestion.

Different models exist for calculating exposure. Deterministic, probabilistic and semi-probabilistic approaches have been used (Tressou et al., 2004). The most common approach in assessing the dietary exposure uses point estimates (average values, maximum values) of all the variables involved in the chain (the consumption, the contamination, etc.). Such an approach, because it does not account for the variability and does not provide measure of uncertainty, may under or overestimate the estimated risk. An alternative approach would be to assign probabilities to all the uncertain quantities. Such a probabilistic approach better describes the available evidence.

In this work, we propose to estimate the exposure to DL-PCBs due to meat consumption using a Bayesian approach. This approach is similar to that which was proposed by Albert et al. (2008) in the context of microbiological risk assessment. A core model is constructed to gather the data available in each module of the process to exposure. Therefore, all the uncertainties carried by the data propagate throughout the chain. Moreover, when available, historical data are introduced as priors thanks to the Bayesian learning paradigm.

The paper is organized as follows. First, we describe the exposure assessment. In particular, we describe the available data in each module from production to digestion, the model likelihood which will link the data distribution to the core model and the included prior information from historical data or from vague priors if historical data are not available. At last, we describe the construction of the stochastic core model via a Bayesian network based on the likelihoods and the prior information. The last part is dedicated to the analysis and discussion of the results.

2. Materials and methods

2.1. Production

2.1.1. The data

Within a recent nationally funded project (SoMeat, http://www.so-meat.fr/), a study was undertaken to determine the concentrations of PCBs in meat products. 85 samples of beef muscle meat were collected in different French slaughterhouses, right after production. The representativeness of the sample to the national production level sampling procedure was conducted in such a way to ensure the representativeness (region, seasons, etc.), and it targeted equally two production modes: organic ($n = 43$) and non-organic or conventional ($n = 42$). All samples were analyzed in the same French accredited national reference laboratory (LABERCA lab) for their PCB concentrations (Dervilly-Pinel et al., 2017). In this work, we focus on DL-PCBs, the ones with the potentially worse effects on human health and which shows the greatest difference of concentration according to the production mode, with a greater contamination for organic production.

2.1.2. The model likelihood

To account for the correlation among isomers, we consider a multivariate model with a Gaussian mixture distribution that classifies the data into two classes (organic and non-organic):

$$\ln C^{P}_{1,\ldots,I} \sim p_{org} \times N_I\left(\boldsymbol{\mu}^{P}_{org}, \Sigma^{P}\right) + p_{conv} \times N_I(\boldsymbol{\mu}^{P}_{conv}, \Sigma^{P}), \quad (1)$$

where $\ln C^{P}_{1,\ldots,I}$, is the vector of the natural logarithms of the concentrations expressed in pictograms per gram of fat in meat products after production (P) for isomer $i = 1, \ldots, I$; p_{org} and p_{conv} are the proportions of organic and conventional meat in the sample respectively ($p_{org} + p_{conv} = 1$), $\boldsymbol{\mu}^{P}_{org}$ and $\boldsymbol{\mu}^{P}_{conv}$ are the vectors of the concentration means for organic and conventional production of the multivariate normal distribution $N_I(\,,)$, Σ^{P} is the $I \times I$ variance-covariance matrix of the error measurements of the production data, and I is the number of isomers ($I = 12$ for DL-PCBs).

2.1.3. Prior information

Additional contamination data are available from a separate study conducted by the Direction Générale De l'ALimentation (DGAL), France, in the frame of its annual monitoring and survey plans. Annual datasets record the concentration of different chemicals in biological matrices sampled following monitoring and control schemes. Note that, contrary to the SoMeat analysis, the sampling was not concerned with the production mode. From the most recent DGAL data (collected in 2013), we extracted $n = 54$ concentration data comparable to monitoring scheme of the SoMeat project (PCBs in beef). But as we did not know the mode of production of each sample (organic or conventional), we let the class of the prior data to be learned during the inference as suggested by Bowmaker et al. (1985). Each observation is assumed drawn from one of the two groups and an unknown fraction p_{org} of observations are in group 1 and $1 - p_{org}$ in group 2. To link the group 1 and 2 to the mode of production, we assigned the SoMeat concentration data from organic production to group 1 and the SoMeat concentration data from conventional production to group 2, in the same spirit of solving identifiability problem where the lower observation is associated to one group and the higher to the other (see the OpenBUGS code available upon request to the authors). Therefore, each data will be assigned to a group (with an uncertainty measure on the assignment) regarding its similarity with the SoMeat data where the class of the data is known. This approach allows us to take advantage of the previously acquired data despite the absence of information regarding the mode of production. It could be viewed as information synthesis or data merging (of complete data and incomplete data with missing labels), separated from

original pre-sample (uninformative) priors. (Albert et al., 2011).

For both the DGAL data and the SoMeat data, the contamination values used for estimating the dietary intake of DL-PCBs corresponded to the upper bound approach (WHO, 2005), meaning that values below the LOD/LOQs are replaced with the corresponding values (conservatory approach). On our data about PCB in bovine meat, there are very few nondetects (less than 1% for SoMeat data and up to 10% in DGAL data).

2.2. Cooking

Apart from total diet studies, the dietary risk associated with the consumption of animal products is most often assessed from the levels of contaminants in raw products. However, the technological processes the food undergoes before consumption, in particular cooking, have to be considered because levels of contaminants in food products can be affected by cooking (Hori et al., 2005). For PCBs, even if these lipophilic compounds are known to be thermostable, so they are not sensitive to thermal degradation, their levels in food products tend to be decreased during cooking via their release with fat into juice (Bayen et al., 2005). More recently however, Perelló et al. (2010) concluded that cooking processes are only of limited value as a means of reducing concentrations of PCDD/PCDFs, PCBs and PCDEs in food (meat & fish), highlighting the lack of consensus on the impact of cooking on POPs concentrations.

2.2.1. The data

Within the SoMeat project, experimental studies were conducted to quantify the effect of cooking on beef meat. Experiments were carried out on spiked samples ($n = 9$ in total, 3 repetitions in each cooking intensity) of beef steaks. The detailed experimental standards are described in Planche et al. (2017). Briefly, each sample (26 g, 10.6% fat) was spiked with a mixture of 12 DL-PCB congeners and cooked in a stainless steel frying pan on a controlled-temperature induction hob. Three different cooking conditions were used to simulate rare (core 50 °C), medium (core 70 °C, according to WHO recommendations for ground meats) and well-done (core 85 °C) meat and PCB losses induced by these cooking conditions were determined. The validity of the results obtained with spiked meat was discussed in Planche et al. (2017) in the light of contaminant levels measured after cooking in naturally contaminated meat samples ($n = 9$).

The SoMeat experiments show that the changes in weight and fat percentage lead to a decrease but also some increases in the concentrations of some congeners. According to Planche et al. (2017), these results are independent of the PCB concentration in meat.

2.2.2. The model likelihood

We consider a multivariate Gaussian model for each cooking intensity (rare, medium or well-done):

$$D^{rare}_{1,\dots,I} \sim N_I(\boldsymbol{\mu}^{rare}_D, \Sigma^D),$$

$$D^{medium}_{1,\dots,I} \sim N_I(\boldsymbol{\mu}^{medium}_D, \Sigma^D),$$

$$D^{well-done}_{1,\dots,I} \sim N_I(\boldsymbol{\mu}^{well-done}_D, \Sigma^D),$$

where $D^{rare}_{1,\dots,I}$, $D^{medium}_{1,\dots,I}$ and $D^{well-done}_{1,\dots,I}$ are the recovery rates in meat products for the I isomers, $\boldsymbol{\mu}^{rare}_D$, $\boldsymbol{\mu}^{medium}_D$ and $\boldsymbol{\mu}^{well-done}_D$ are the vectors of the recovery rate means for rare, medium and well-done cooking intensity respectively, and finally Σ^D is the $I \times I$ variance-covariance matrix of the error measurements of the recovery rates.

2.2.3. Prior information

Vague multivariate normal priors were chosen for the means $\boldsymbol{\mu}^{cook}_D$ (cook = rare, medium or well-done) and a vague inverse Wishart distribution was chosen for Σ^D in lack of prior information (see OpenBUGS code for more details).

2.3. Meat consumption

2.3.1. The data

Within the SoMeat project, a survey was conducted on-line to gather data on the consumption of organic and non-organic meat on the adult population. Consumers were recruited in organic specialized outlets and among short channel customers (markets, farm sales) mainly located at the South-West of France. 392 consumers provided estimates of their meat consumption (in grams per week) with detail of the proportion of organic meat and non-organic meat. 297 out of 392 consumed bovine meat over the last 12 months, with details of their weekly consumed quantity (ranging from 10 to 1650 g per week, with mean 210 g per week). 187 out of 297 consumed organic meat, with detail of the proportion of organic meat (ranging from 12.5% to 100%, with mean 59.2%).

2.3.2. The model likelihood

For the meat consumption model, there are several steps: first, individuals can consume meat or not, then when they do consume, the weekly quantity is determined, and within this quantity a certain proportion is organic if the individual is a consumer of organic meat. From this analysis, the model comprises 4 variables:

- Variable X_i indicates whether the individual i consumed meat ($X_i = 1$) or not ($X_i = 0$) over the year. We assume

$$X_i \sim Bernoulli(p_Q),$$

where p_Q is the probability of consuming meat over the year.

- Variable lQ_i stands for the natural logarithm of the weekly meat consumed quantity (in grams per week). The following model likelihood is assumed:

$$lQ_i \sim N(\mu_{lQ}, \sigma^2_{lQ}), \quad (2)$$

where μ_{lQ} is the mean and σ^2_{lQ} the variance of the natural logarithm of the weekly meat consumed quantity.

- Variable Y_i indicates whether the individual i is a consumer of organic meat ($Y_i = 1$) or not ($Y_i = 0$). We assume that Y_i has a Bernoulli distribution:

$$Y_i \sim Bernoulli(q_{org}),$$

where q_{org} is the probability of consuming organic meat over the year.

- Variable $f_{org,i}$ denotes the proportions of organic meat consumption over the week for individual i. The following model likelihood is assumed:

$$f_{org,i} \sim Beta(a_{forg}, b_{forg}), \quad (3)$$

where a_{forg} and b_{forg} are the standard positive parameters of the Beta distribution with mean $\frac{a_{forg}}{a_{forg} + b_{forg}}$.

2.3.3. Prior information

Additional data on meat consumption are available.
The main French datasets are:

- INCA 2: a national individual survey of dietary intake conducted in 2006–2007. It records the daily food consumption over 7 days of 2624 adults (18–79) and 1455 children (3–17) (INCA 2, 2006). The data on beef consumption of the adult population was extracted. From this extraction, we get prior information on meat consumption indicators (1910 out of 2624 adults consume beef meat) as well as information on weekly consumed quantities (ranging from 30 g per week to 1710 g per week, with mean 251 g per week).
- KANTAR: a panel consumption database that records food

purchases. The latest available version within the SoMeat project is 2013 (Kantar Worldpanel, 2013). A sample of 5547 active households with beef meat purchases in 2013 was extracted with information on the production mode (organic/conventional) of the beef. From this extraction, we get prior information on organic meat consumption indicators (327 of the 5547 households buy organic beef, that is nearly 6%) as well as information on the percentage of organic beef meat within the total beef meat consumption (ranging from 0.6% to 100%, with mean 23.2%).

2.4. Bioaccessibility

When quantifying exposure related to ingestion of toxic compounds in food, it is currently assumed that 100% of the contaminant is available for absorption into systemic circulation. However, only the bioaccessible fraction of this contaminant is mobilized from the contaminated food matrix into the digestive tract during digestion and induces toxic effects. The bioaccessibility of micropollutants must therefore be taken into account to make an accurate assessment of their impact on human (Guerra et al., 2012). Exposure after digestion will be referred to as "uptake" in the sequel.

2.4.1. The data

Within the SoMeat project, experimental studies were conducted to assess the bioaccessibility of PCBs in meat based on a standardized *in vitro* static digestion protocol (Minekus et al., 2014). The detailed experimental standards are described in Planche et al. (2016). Briefly, *in vitro* digestions were carried out on spiked ground beef samples ($n = 3$ for each condition) and the influence of fat level, cooking intensity and the age of consumers on the bioaccessibility of PCBs in meat was studied.

For this work, we only used data related to cooking intensities (rare, medium, or well done) with the bioaccessibility of the 12 DL-PCBs (3 repetitions for each cooking intensity) for the adult population and a percentage of fat of 15%.

2.4.2. The model likelihood

We consider a multivariate Gaussian model for each cooking intensity (rare, medium or well-done):

$$B^{rare}_{1,\ldots,I} \sim N_I(\boldsymbol{\mu}^{rare}_B, \Sigma^B),$$

$$B^{medium}_{1,\ldots,I} \sim N_I(\boldsymbol{\mu}^{medium}_B, \Sigma^B),$$

$$B^{well-done}_{1,\ldots,I} \sim N_I(\boldsymbol{\mu}^{well-done}_B, \Sigma^B),$$

where $B^{rare}_{1,\ldots,I}$, $B^{medium}_{1,\ldots,I}$ and $B^{well-done}_{1,\ldots,I}$ are the recovery rates in meat products for the I isomers, $\boldsymbol{\mu}^{rare}_B$, $\boldsymbol{\mu}^{medium}_B$ and $\boldsymbol{\mu}^{well-done}_B$ are the vectors of the recovery rate means for rare, medium and well-done cooking intensity respectively, and finally Σ^B is the $I \times I$ variance-covariance matrix of the error measurements of the recovery rates.

2.4.3. Prior information

Vague multivariate normal priors were chosen for the means $\boldsymbol{\mu}^{cook}_B$ ($cook = rare, medium, well-done$) and a vague inverse Wishart distribution was chosen for Σ^B in lack of prior information (see the OpenBUGS code available upon request to the authors for more details).

2.5. Core model

The aim of the core model is to obtain an estimation of the exposure/intake before digestion and exposure/uptake after digestion for a person taken at random in the French population. Because risks related to PCBs are chronic, we focus on the long run average and use the mean intake and uptake.

At the production step, the variables of interest are the estimated mean concentration for each isomer. From the likelihood model (1), we derive their absolute values based on the means of the log normal distribution:

$$m^P_{mode,i} = \exp(\mu^P_{mode,i} + \sigma_i^2/2)$$

where $m^P_{mode,i}$ is the estimated mean absolute concentration of isomer i ($i = 1, \ldots,12$) after production (P), $\mu^P_{mode,i}$ is the ith element of the vector of the concentration mean $\boldsymbol{\mu}^P_{mode}$ where $mode = org$ for organic production or $conv$ for conventional production, and $\sigma_i^2 = diag(\Sigma^P)_i$. Then, we derive the concentration in isomer i after cooking for each production mode ($mode = org$ or $conv$) and each cooking intensity (where $cook = rare$, $medium$ or $well-done$) from:

$$m^{cook}_{mode,i} = \mu^{cook}_{D,i} \times m^P_{mode,i},$$

where $\mu^{cook}_{D,i}$ is the ith element of the vector of the mean recovery rate $\boldsymbol{\mu}^{cook}_D$.

From the consumption module and the log normal model likelihood given in (2), we get the estimated beef meat weekly absolute estimated consumption of an individual taken at random in the adult population by:

$$\overline{Q} = p_Q \times \exp(\mu_{lQ} + \sigma^2_{lQ}/2).$$

From the model likelihood given in (3), we get the estimated proportion of organic meat within the total of a beef meat consumer taken at random by:

$$\overline{f}_{org} = q_{org} \times \frac{a_{forg}}{a_{forg} + b_{forg}}$$

and the estimated consumption of organic and conventional beef an individual taken at random in the adult population as:

$$\overline{Q}_{org} = \overline{f}_{org} \times \overline{Q},$$

$$\overline{Q}_{conv} = (1 - \overline{f}_{org}) \times \overline{Q}.$$

Mean exposure to each isomer i before digestion for each cooking intensity is obtained by:

$$E^{cook}_i = \frac{\overline{Q}_{org} \times m^{cook}_{org,i} + \overline{Q}_{conv} \times m^{cook}_{conv,i}}{w},$$

where w is the body weight of the individual fixed at 70 kg as commonly done.

They are then combined using toxicological equivalence factors: the toxic equivalency (TEQ) of a mixture is defined by the sum of the concentrations of individual compounds multiplied by their relative toxicity (TEF). For DL-PCBs, TEF were taken from The International Programme of Safety, WHO, 2005. This results into the exposure to DL-PCBs for a given cooking intensity:

$$E^{cook}_{DL} = \frac{\overline{Q}_{org} \times TEQ^{cook}_{org} + \overline{Q}_{conv} \times TEQ^{cook}_{conv}}{w}.$$

Similarly exposure after digestion, noted $E^{cook,B}_{DL}$, is obtained by replacing the concentrations after cooking with the concentrations after digestion, that are bioaccessible estimated concentrations (with an exponent "B" for bioaccessibility), for each production mode and cooking intensity:

$$m^{cook,B}_{mode,i} = \mu^{cook}_{B,i} \times m^{cook}_{mode,i}.$$

This core model can be summarized in the directed acyclic graph (DAG) of the model presented in Fig. 1. In the DAG, triangles are used for the parameters of the models, rectangles are used for data and circles are used for quantities of interest.

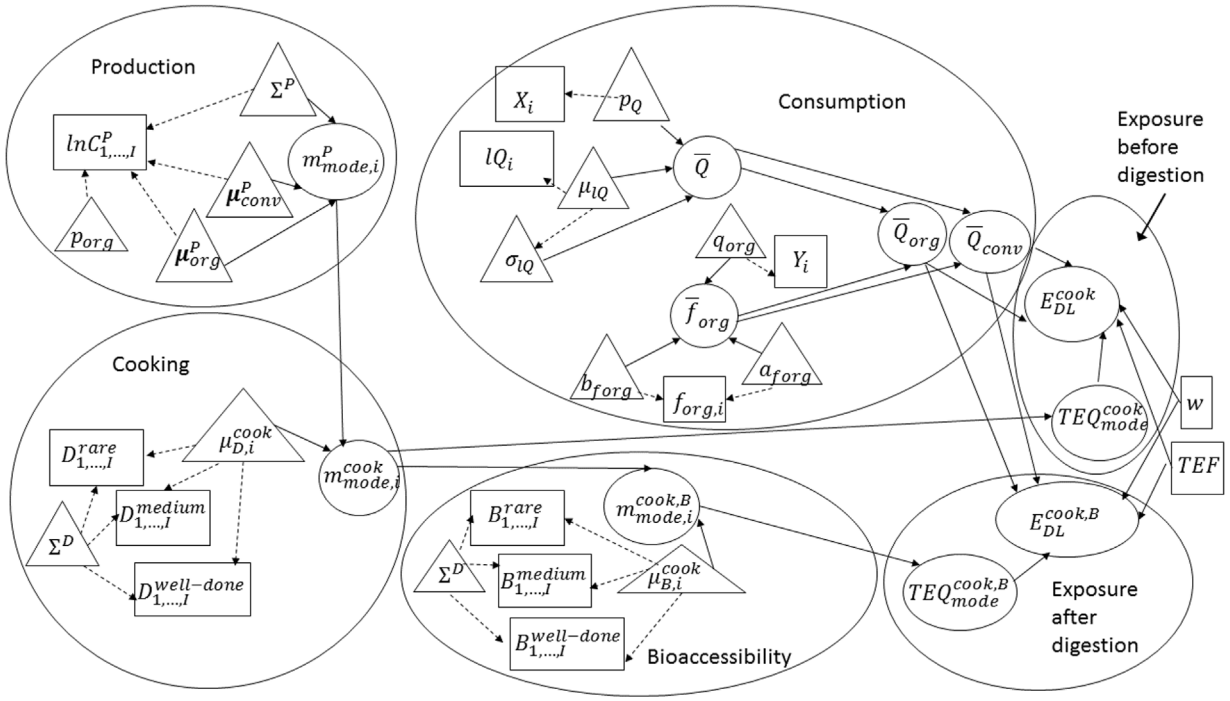

Fig. 1. DAG of the model.

3. Results and discussion

3.1. Results

3.1.1. Production module

Looking at the concentrations of the 12 isomers of DL-PCBs for conventional and organic production mode using the data collected within the SoMeat project and the historical data from DGAL (surveillance plans from the French ministry of Agriculture), we observe that concentrations in organic production are generally higher, see Fig. 2.

A side result of this module is the distribution of the probability for a given data in the pooled sample of being organic: p_{org} has mean 0.512 and 95% credibility interval (0.413–0.613), which means that half of the data was classified as organic and the other half as conventional as it is the case in the SoMeat data.

3.1.2. Cooking module

We observe here two kinds of pattern in the reduction of contamination according to the different cooking intensities: a first set of DL-PCBs only show a significant reduction of concentration for well-done cooking intensity (PCBs 118, 123, 156, 189 similar to PCB-105 & 126 in Fig. 3) and another set of DL-PCBs shows an increase of the reduction when the cooking intensity gets more intense (PCBs 81, 157, 167, 169 similar to PCB-114 & 77 in Fig. 3). This decrease for well-done cooking can be explained by the expulsion of lipophilic PCBs into cooking juices along with lipids contained in the food matrix. The different physicochemical properties of PCB congeners partly explain that there are different kinds of pattern during cooking, see Planche et al. (2017) for more details. Note that when cooking intensity is “rare”, the quantity of contaminant may increase due to the change of mass and percentage of fat, see Planche et al. (2017) for more details.

Fig. 4 illustrates how concentration in PCB-126 changes with cooking according to production mode being organic or conventional. The reduction is more important for well-done cooking still with higher concentrations in organic meat with mean concentration in organic meat changing from 0.23 pg/g fw for raw meat to 0.20 pg/g fw for rare cooked meat, 0.21 pg/g fw for medium cooked meat, and 0.18 pg/g fw for well-done meat and mean concentrations in conventional meat going from 0.18 pg/g fw for raw meat to 0.15 pg/g fat for rare cooked meat, 0.16 pg/g fat for medium cooked meat, and 0.14 pg/g fw for well-done meat.

3.1.3. Consumption module

Important results from the consumption module are the estimated quantity of beef meat consumed weekly in France, with distribution given in Fig. 5 (left graph) and the estimated percentage of organic meat, with distribution given in Fig. 5 (right graph). The mean meat consumption is 269 g per week, varying from 263 to 275 g/w. The mean percentage of organic meat consumption is 3.8%, varying from 3.4% to 4.2%, for an individual taken at random in the French population.

3.1.4. Bioaccessibility module

Bioaccessibility of DL-PCBs in meat (for adult population and meat with 15% fat) is not so much sensitive to cooking intensity as shown in Fig. 6. Indeed, the mean bioaccessibility of DL-PCBs is not significantly different between rare- and WHO-cooked meat (both around 25%). Only a well-done cooking intensity induces a decrease in the DL-PCBs bioaccessibility (around 22%).

3.1.5. Intake and uptake

Fig. 7 shows the intake distribution of exposure obtained according to different cooking intensities before (intake) and after digestion (uptake). Before digestion, mean intake is 0.066 pg WHO-TEQ_{2005}/kgbw/w for rare cooking, 0.069 pg WHO- TEQ_{2005}/kg bw/w for medium cooking and 0.058 pg WHO- TEQ_{2005}/kg bw/w for well-done cooking (95% credibility intervals are respectively (0.052–0.083) for rare, (0.053–0.086) for medium, (0.045–0.074) for well-done). After digestion, mean uptake is reduced to 0.017 pg WHO- TEQ_{2005}/kg bw/w with CI (0.007–0.027) for rare cooking, 0.018 pg WHO- TEQ_{2005}/kg bw/w with CI (0.007–0.030) for medium cooking and 0.012 pg WHO-TEQ_{2005}/kg bw/w with CI (0.003–0.021). Fig. 8 illustrates what would be the mean intake and uptake for specific consumption behaviors, with the example of medium cooked meat either 100% conventional or 100% organic.

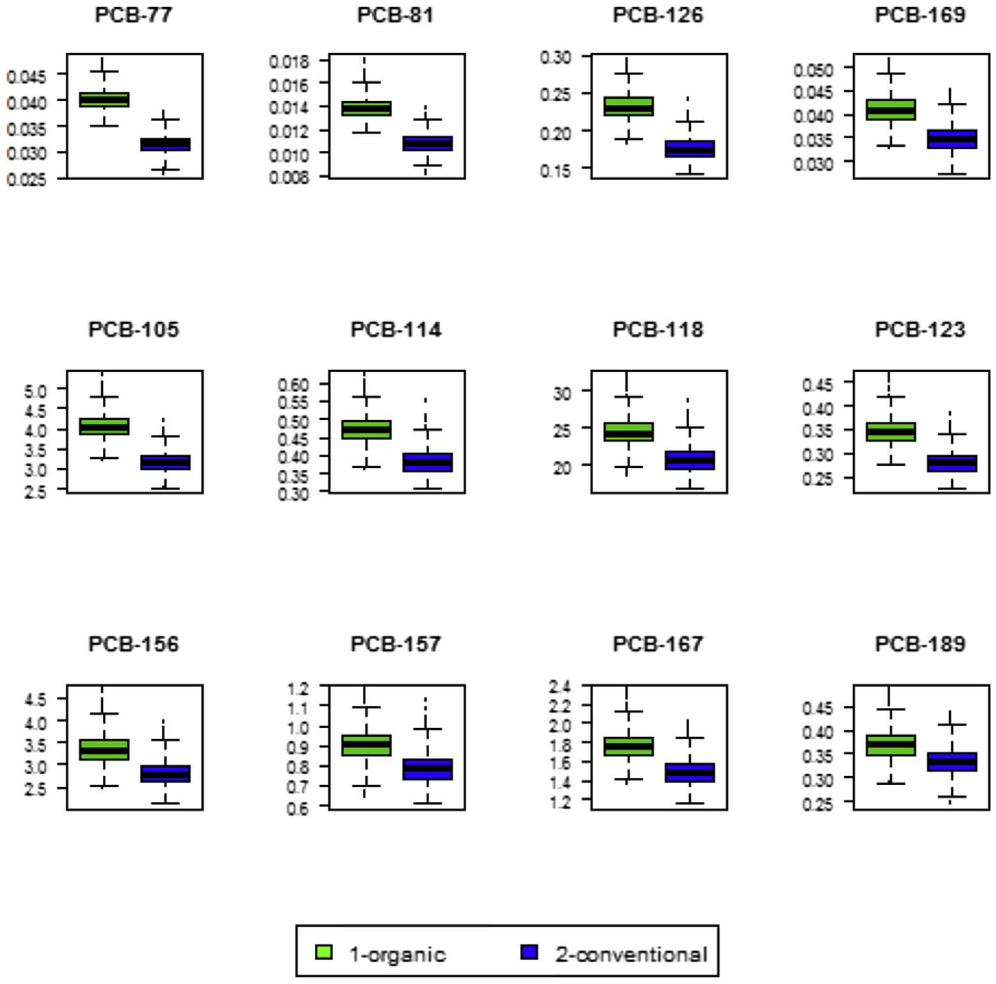

Fig. 2. Boxplots for mean contamination at Production ($m^{P}_{mode,i}$ with mode = organic or conventional, unit: pg/g fresh weight).

3.2. Discussion

3.2.1. Differences between data given as prior and data collected within the project

DL-PCBs contamination data used in the present study originated from two distinct sources. Although both sources corresponded to contamination levels after the production stage, they exhibited some particularities as follows: (i) the data collected within the SoMeat project were measured in the French National Reference Laboratory, in meat and using a single analytical method. Further, this project specifically recorded the mode of production (organic or conventional) and compared the contamination levels in both cases (Dervilly-Pinel et al., 2017), (ii) the set of data corresponding to national monitoring and control plans corresponded to fat samples characterization; it was obtained in different laboratories, whose analytical methods although complying with regulatory requirements in terms of sensitivity of the detection, exhibited different performances in terms of LOD/LOQ with subsequent consequence on the upper-bound reported levels (with limited effect on our data because of a low percentage of censored data). Furthermore, no information regarding the production mode of the considered animals (e.g. organic, etc …) was available. Despite the differences in their design, both data sets were used in the present study, which consequently induced a bias in the estimation of the probability for a sample within the official control data set to belong to organic production. Indeed, as associated sampling is designed to be representative of the national production, and since no more than 1% of the French production is organic, it is expected that the data set mostly correspond to conventional production. In the present study, our calculations led to an estimation of 51.2% organic in the data set (§ 3.1.1) which can therefore nor reflect neither be interpreted as the chances that samples from the surveillance plans originated from organic productions but rather as a lack of knowledge on the production type. Such bias, which has been clearly identified during the project, did not interfere with the exposure results presented since the model does not rely on the proportion of organic production but directly on the proportion of organic consumption.

At consumption step, the data collected within the project targeted the organic meat consumer: it is therefore biased toward a greater percentage of organic meat consumption. On the other hand, purchase data used to estimate this percentage of organic meat as a prior dated from 2013 and it is clear that the offer of organic meat in supermarkets has been expanded in the recent years, even if it remains still limited (Sans and Boizot-Szantaï, 2017). Also, it is well known in food risk assessment that estimating parameters from household data induces biases compared to individual data (only some of the household members may consume organic meat) but estimating individual intakes from household purchases is a difficult problem (Allais and Tressou, 2009). About the consumed quantity, the estimation in the prior data relies on a greater number of observations but again the INCA2 was collected in 2006–2007 and since then messages on reducing meat consumption have been more numerous.

3.2.2. Comparisons of the obtained average estimates with those obtained in other studies

Although estimated dietary intakes (EDIs) can hardly be compared as the studies refer to different countries and different reference years, the results reported in this work are consistent with other international publications related to meat and presenting either DL-PCBs intake

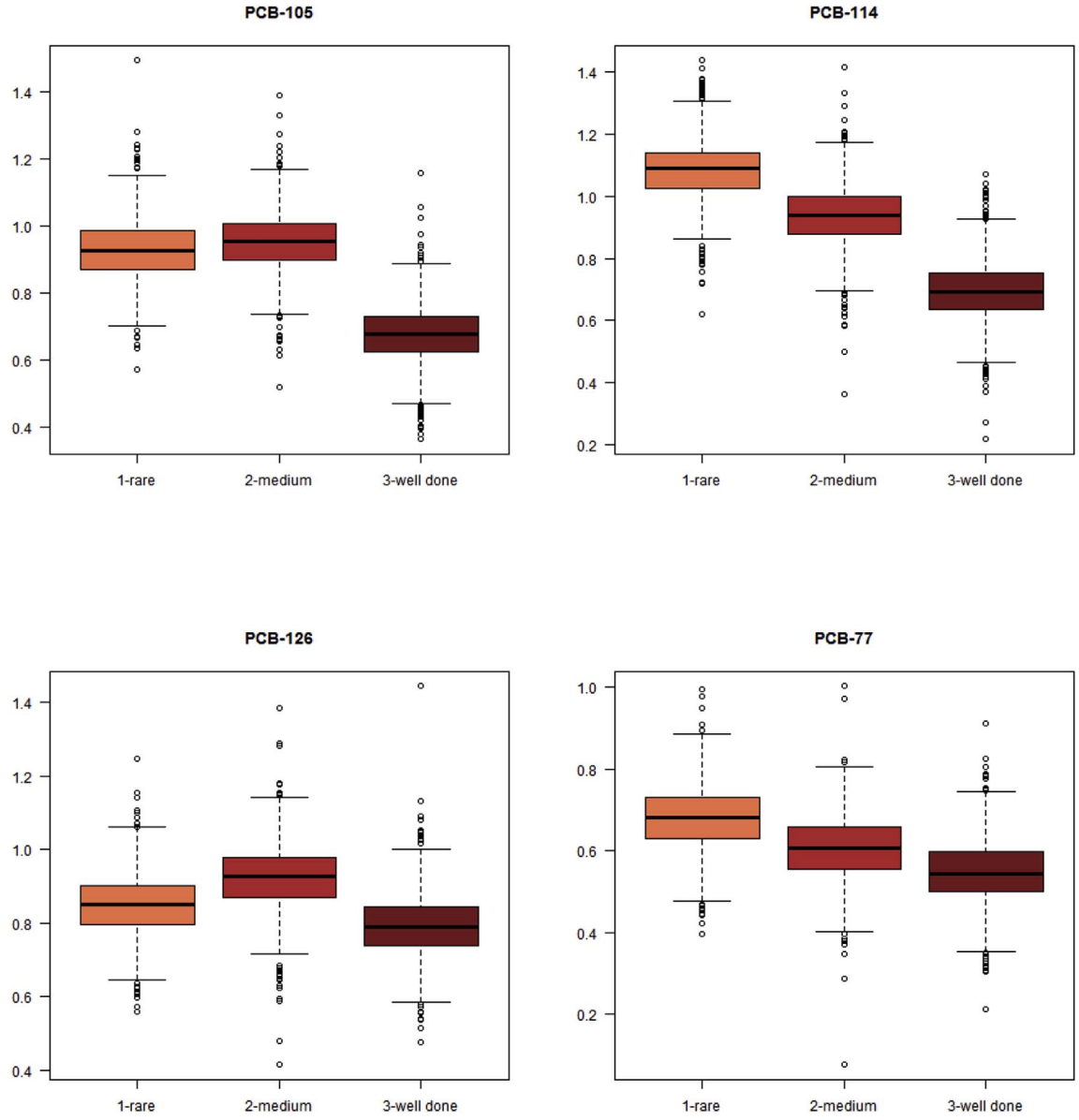

Fig. 3. Boxplots for mean cooking reduction rates (μ_D^{cook} with $cook = rare,\ medium,\ well - done$).

levels or combined with PCDD/Fs:

- Most recent exposure study at the French level is available as the result of the second Total Diet Study (TDS) (ANSES, 2011), which using a mean DL-PCBs contamination level in meat of 0.029 pg TEQ WHO_{98}/g fresh weight) (i.e 0.21 pg TEQ WHO_{98}/g lipid weight), reported for the adult population a mean intake to DL-PCBs through meat of 0.029 pg TEQ WHO_{98}/kg bw/day (i.e 2.03 pg TEQ WHO_{98}/kg bw/w). Such intake from meat corresponded to 9.8% of total intake of DL-PCBs from food. This intake level through meat consumption as reported in 2011 may be compared to those obtained in the present study after the cooking stage, i.e in the range 0.058–0.069 pg WHO-TEQ_{2005}/kg bw/w depending on cooking intensity. The 2 fold intake levels reported in the present study may be explained by the fact that only bovine meat was considered here, while in the previous TDS, meat included besides bovine also pork and sheep, which are known to present lower contamination levels (Dervilly-Pinel et al., 2017). As previously reported, beef is the most important contributor within the meat group (Winters et al., 1996; Malisch, 1998) although it is not necessarily the most consumed meat (Fürts et al., 1990).
- Also focusing on meat contribution to dietary DL-PCBs intake, a recent Chilean study (San Martin et al., 2016) specifically investigated various meat animal species contribution and reported for bovine meat dietary intake level estimations for adult of 0.007 pg WHO-TEQ_{2005}/kg bw/d (i.e 0.05 pg TEQ WHO_{98}/kg bw/w), which value is fully in accordance with results obtained in the present study.
- Related to PCDD/Fs + DL-PCBs dietary intake levels: for instance, levels from bovine and ovine consumption reported for the general population in Belgium (0.36 and 0.03 pg WHO_{98}-TEQ/kg bw/d; Focant et al. (2002) were comparable to the presently obtained levels when considering only DL-PCBs contribution. Furthermore, the present results were comparable, even if slightly higher than levels reported from Spain (Marin et al., 2011)

Finally, to complete risk assessment related to DL-PCBs in bovine meat, risk characterization may be performed when assessing the intake levels of these pollutants in relation to proposed TDI. The European Food Safety Authority (EFSA) adopted in 2001 an opinion on dioxins

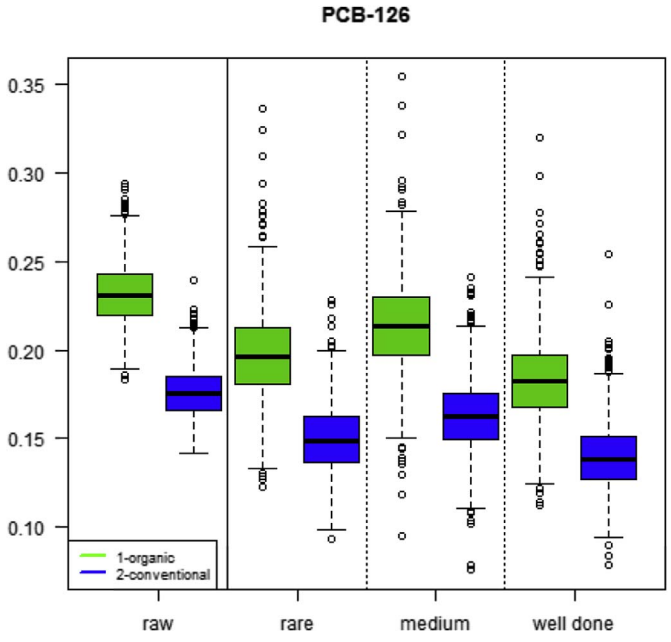

Fig. 4. Mean contamination changes with cooking for PCB-126 ($m^{cook}_{mode,i}$ *with mode = organic our conventional* and *cook = raw, rare, medium, well − done* unit pg/g fw).

and dioxin-like PCBs in food, fixing a tolerable weekly intake (TWI) of 14 pg (pg) World Health Organisation toxic equivalent (WHO-TEQ)/kg body weight (b.w.) for dioxins and dioxin-like PCBs. In parallel, the Joint Expert Committee on Food Additives (JECFA) of the WHO and from the UN Food and Agriculture Organisation (FAO) established in 2001 a provisional tolerable monthly intake (PTMI) at 70 pg/kg b.w. for dioxins and dioxin-like PCBs. Converted to a tolerable daily intake, the EFSA health-based guidance value of 2 pg/kg b.w. is in line with the JECFA value of 2.3 pg/kg bw.

Considering that foodstuffs from animal origin account for more than 90% of the human body burden, with meat, dairy, and fish products being the main contributors (Beck et al., 1989; Fürts et al., 1990), the obtained levels in the present study only represent a moderate to low health risk and are within the safety margins for human consumption.

Further market basket studies are however needed to confirm trends over time and to increase prevention efforts against the contamination of animal-origin products with these toxic compounds. A particular effort toward children exposure assessment is also required as previous studies highlighted a higher risk for this population with regard to meat consumption (San Martin et al., 2016).

It should be noted also that recently the US Environment Protection Agency (US EPA) confirmed the oral reference dose (RfD) of 0.7 pg/kg bw per day for dioxins (US EPA, 2012). In addition, the U.S. Agency for Toxic Substances and Disease Registry/Center for Disease Control and Prevention (ATSDR) has established a chronic-duration oral Minimal Risk Level (MRL) of 1.0 pg/kg b.w. per day for dioxins. Considering these differences in health-based guidance values established by different organizations as regards dioxins and dioxin-like PCBs, EFSA (2015) recently concluded recommended a comprehensive risk assessment on the risk for animal and public health related to the presence of dioxins and dioxin-like PCBs in feed and food to be performed. If this is the case, the more recent occurrence data of dioxins and dioxin-like PCBs in feed and food need to be taken into account.

An additional point to be considered in the discussion relates to TEFs evolution over the recent period which might impact exposure assessment and subsequent comparisons. The World Health Organisation (WHO) organized in 1997 a first re-evaluation which resulted in a revision of several mammalian TEF values and removed the di-ortho PCBs from the TEF concept (Van den Berg et al., 1998). The second re-evaluation in 2005 has been published more recently (Van den Berg et al., 2006). In comparison to WHO 1998 TEF the WHO 2005 TEF values were set at half orders of magnitude increments on a logarithmic scale of 0.03, 0.1, 0.3, etc. Values for PCB-169, PCB-81 and PCB-167 were raised to a TEF value of 0.03, 0.0003 and 0.0003, resp., while all other DL-PCBs were set to a uniform value of 0.00003. The WHO 2005 TEF scheme was primarily intended for estimating exposure and risks via oral ingestion. The impact of WHO 2005 revised TEF on the TEQs has been investigated in a range of studies which demonstrated reduced mean total TEQ after implementation of the 2005 TEFs compared to the 1998 ones, especially concerning mono-ortho PCBs (Malisch et al., 2007).

3.2.3. Possible extensions of the model

The present model has been established on the basis of a mean adult weighting 70 kg. Some refinements could be integrated accounting for

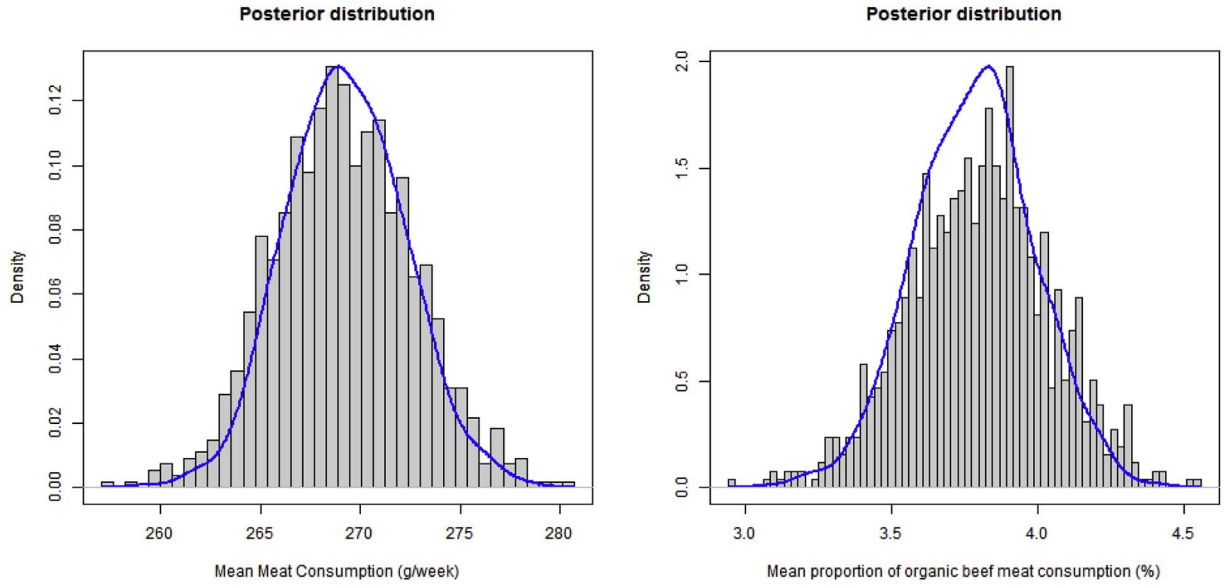

Fig. 5. Mean beef meat consumption ($\overline{Q}$, unit: g/week) and mean proportion of organic beef meat consumption ($\overline{f}_{org}$, *unit*: %).

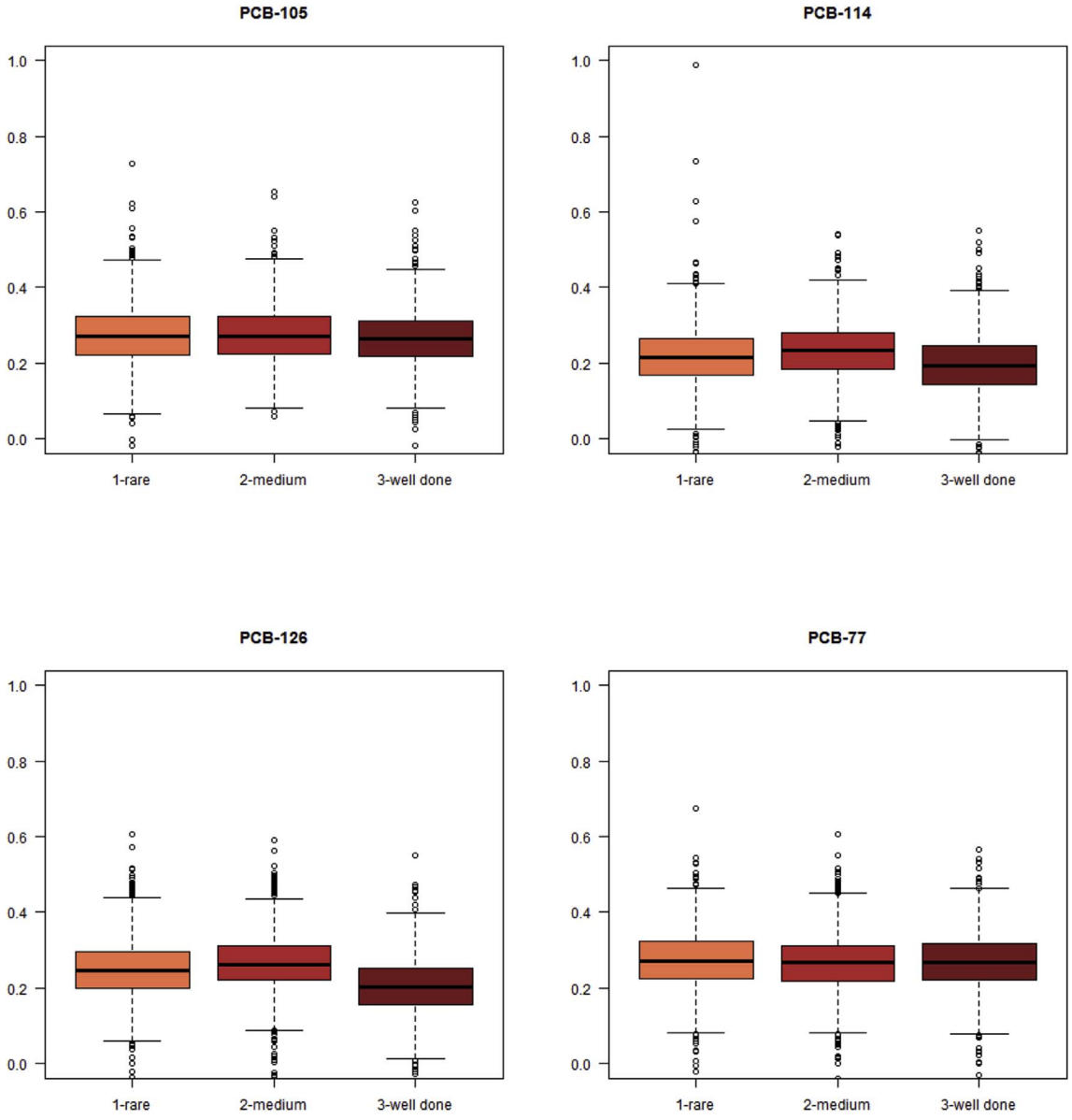

Fig. 6. Mean recovery rates after digestion ($B_{1,\ldots,I}^{cook}$, $cook = rare,\ medium,\ well-done$).

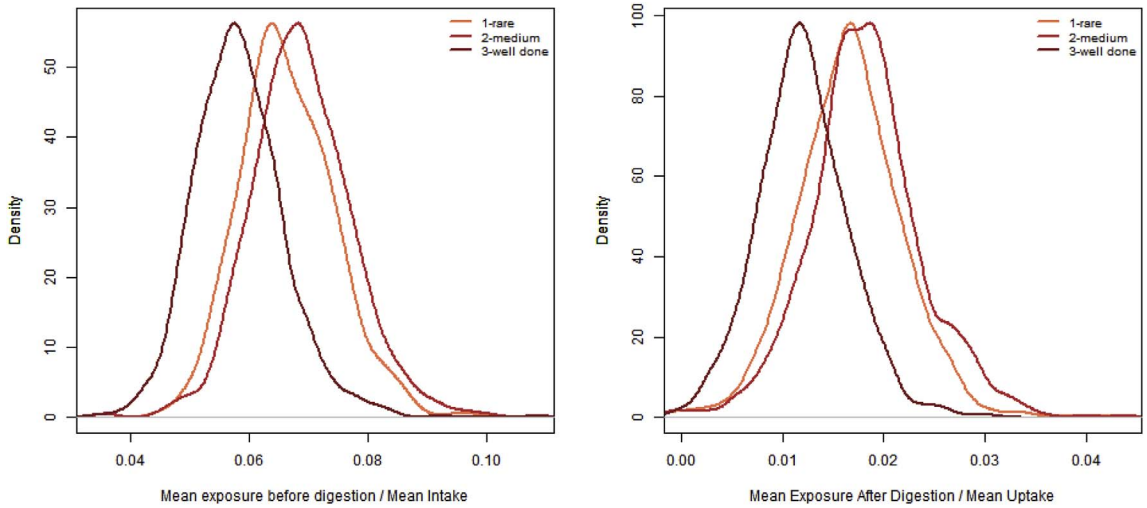

Fig. 7. Mean intakes and uptakes according to cooking intensity (intake is E_{DL}^{cook}, uptake is $E_{DL}^{cook,B}$ with cook = rare, medium, well-done) for a general behavior towards organic/conventional meat consumption.

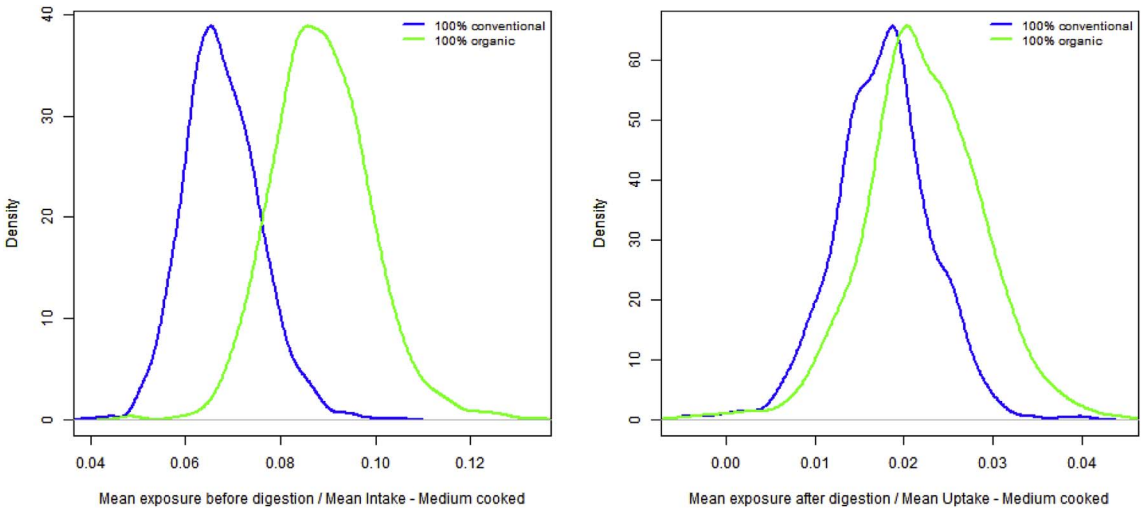

Fig. 8. Mean intakes and uptakes for medium cooked meat either 100% organic or 100%.conventional.

the weight and age of the individuals which would result into changes in the consumed quantities and in bioaccessibility. It was indeed shown in Planche et al. (2016) that bioaccessibility is around 26% for adults, 8% for children, and 17% for the elderly (when meat with 15% fat is cooked medium). The consumer choices for meat with more or less fat could also be integrated. The INCA2 survey carries that kind of precision for minced beef, but data collected within the SoMeat project did not.

4. Conclusion

In this work, we propose to estimate the exposure to DL-PCBs due to meat consumption using a Bayesian approach that allows to include in a model each step of the process from produced meat to consumption and digestion. All the uncertainties carried by the multiple available data sources propagate forward along the chain. A global mean intake to DL-PCBs varying from 0.058 pg WHO-TEQ_{2005} per kilogram of body weight per week (pg WHO-TEQ_{05}/kg bw/w) for well-done cooking intensity to 0.066 and 0.069 for rare and medium cooking intensities respectively have been calculated, as a mean for an individual randomly taken in the French adult population. After digestion, the exposures (uptakes) decrease to 0.017, 0.018, and 0.012 pg WHO-TEQ_{05}/kg bw/w respectively for rare, medium and well-done cooking intensities. Exposure before digestion (intakes) should be compared to European Food Safety Agency's provisional tolerable weekly intake of 14 pg WHO-TEQ_{98}/kg bw/w defined for the sum DL-PCBs, furans and dioxins for a full dietary intake. This work is a first step of refined risk assessment integrating different steps such as cooking and digestion in the context of chemical risk assessment and also different sources of information based on previously acquired data on one hand and data collected with a specific goal related to the comparison of organic and conventional modes of production. A similar approach could be used for other chemicals, namely pesticides.

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Transparency document

Transparency document related to this article can be found online at http://dx.doi.org/10.1016/j.fct.2017.10.032.

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