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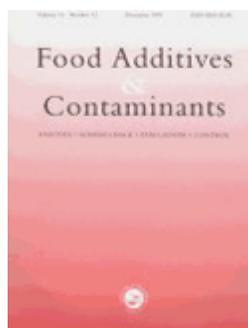
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Assessment of seasonality in exposure to dioxins, furans and dioxin-like PCBs by using long term food consumption data

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Food Types:	

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4 **1 Assessment of seasonality in exposure to dioxins, furans and dioxin-like**
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6 **2 PCBs by using long-term food consumption data**
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20 **Key words:** dioxins, dioxin-like PCBs, dietary exposure, seasonality, long-term dietary
21 survey

22

23 **Abstract**

24 According to the European Food Safety Authority (EFSA) guidance related to uncertainties in
25 dietary exposure assessment [The EFSA Journal (2006) 438:1-54], exposure assessment based
26 on short-term food consumption surveys, such as 24-hour recalls or 2 days records, tend to
27 overestimate the long term exposure because of the assumption that the dietary pattern will be
28 similar day-after-day over a lifetime. The aim of this study was to make an assessment of
29 dietary exposure to polychlorinated dibenzodioxins (PCDDs) and polychlorinated
30 dibenzofurans (PCDFs) also called 'dioxins' and 'dioxin-like PCBs' using, long-term
31 household purchase and consumption survey data collected by TNS-Secodip. Weekly
32 purchases of the major dioxins and dl-PCB vector products of these contaminants were
33 collected for 328 single person households, who participated at TNS-Secodip consumption
34 surveys from 2003 to 2005 and who were single person households in order to better estimate
35 their consumption. These data were combined with average contamination levels of food
36 products. Weekly gross average exposure was estimated at 10.2 pg Toxic Equivalent
37 (WHOTEQ)/kg body wt/week (95% confidence interval [9.6, 10.9]). According to the typical
38 shape of the distribution of individual weekly exposures, it is sensible to fit an exponential
39 law to these data. The mean value was therefore 12.1 pg WHOTEQ /kg bw/week. This value
40 is higher than the arithmetic mean because it better takes into account the inter-individual
41 variability. It was estimated that about 20% persons of this sample were exceeding the
42 current Health Based Guidance Value mainly due to high consumption of seafood and/or
43 dairy products. Thanks to long survey duration (3 years) and the weekly recording of food
44 consumption, it was possible to demonstrate the actual seasonality of dietary exposure to
45 dioxins and dl-PCBs with a maximum between March and September; similar seasonality is
46 observable for fish consumption. Autoregressive integrated moving average or ARIMA
47 models were adjusted to the time series and it could be demonstrated that the number of times
48 the upper limit of confidence intervals exceeds Provisional tolerable weekly intake (PTWI) is
49 about 15 weeks per year in average. Finally, compared to the results obtained from data
50 collected in the short term surveys (one week), this study does not suggest that short term
51 consumption surveys tend to overestimate the long-term exposure.

52

53 **Introduction**

54 The term “dioxins” encompasses a group of 75 polychlorinated dibenzo-*p*-dioxin (PCDD)
55 (among which 2,3,7,8-tetrachlorodibenzo-*p*-dioxin or TCDD plays a key-role), and 135
56 polychlorinated dibenzofuran (PCDF) congeners. Polychlorinated biphenyls (PCBs) are
57 chlorinated aromatic hydrocarbons which have been classified according to their biochemical
58 and toxicological properties. Non-*ortho* and mono-*ortho* substituted PCBs show toxicological
59 properties that are similar to dioxins. They are therefore often termed “dioxin-like PCBs” (dl-
60 PCBs).

61 Dioxins and dl-PCBs are lipophilic compounds. They are extremely resistant towards
62 chemical and biological degradation processes and therefore persist in the environment and
63 accumulate in the food chain. It is considered that 95% of the human exposure to dioxins and
64 related compounds is from the diet (De Mul, et al. 2008; Parzefall W, 2002). When ingested
65 the compounds accumulate in the body and any adverse effects are likely to appear after years
66 and to be related to the total body burden. This property underlines the importance of long-
67 term dietary exposure surveys.

68 Many toxic effects are related to dioxin exposure like reproductive effects (Mocarelli, et al.
69 2008), neurodevelopmental effects and endocrine effects via thyroid toxicity (Giacomini, et
70 al. 2006) even if various studies show controversial results (Wilhelm, et al. 2008). Recently a
71 metabolic syndrome including increase blood pressure, high triglyceride levels as well as
72 glucose intolerance was described and related to exposure to dioxins (Uemura, et al. 2009).
73 Finally, 2,3,7,8-TCDD is considered to be carcinogen for human (Schwarz and Appel, 2005;
74 IARC, 1997) but for relatively high exposure compared with the one of the general
75 population (Kociba, et al. 1978). The toxicity of all dioxins and dioxin-like PCBs is calculated
76 relatively to 2,3,7,8-TCDD, one of the most potent of the chemicals on which most
77 toxicological and epidemiological information is available. The analytical results are
78 converted into toxic equivalents (TEQ). This conversion is based on the assumption that all
79 2,3,7,8-substituted PCDDs and PCDFs, as well as the dioxin-like PCBs, bind to the same Ah
80 receptor, and show comparable qualitative effects, but with different potencies (Schwarz,
81 2005). The differences in toxicity are expressed in the toxic equivalency factors (TEFs).

82 Within all the congeners, 17 dioxins and 12 dioxin-like PCBs are of toxicological concern.
83 These 29 substances are considered all together for risk assessment.

84 For the current study, the TEFs of dioxins and dioxin-like PCBs established in 1998 and
85 revised in 2006 by the World Health Organization (WHO) were considered and noted
86 WHOTEQ (Van den Berg, et al. 1998; Van den Berg, et al. 2006). WHO also established a
87 Provisional Tolerable Monthly Intake (PTMI) of 70 pg WHOTEQ/kg body weight (BW) for
88 dioxins and dl-PCBs (WHO, 2002). Similarly the EU established a Provisional Tolerable
89 Weekly Intake (PTWI) of 14 pg WHOTEQ/kg body weight (BW) for dioxins and dl-PCBs
90 (SCF, 2001).

91 The results of the most recent dietary exposure assessments show that this health based
92 guidance value is often exceeded by a significant part of the population (SCF, 2001; WHO,
93 2001; AFSSA, 2005). However these assessments rely on food consumption data collected
94 over short periods of time between 1 and 7 days which are expected to overestimate the
95 exposure due to the under-estimation of the intra-individual variability i.e. food consumption
96 observed on short period of time are likely to vary on longer term and therefore the resulting
97 exposure is likely to be lower (WHO, 2005).

98 The objective of the present study was to estimate the long-term dietary exposure to dioxins
99 and related compounds based on French household food consumption data collected over
100 multiple years of consumption recording. This approach should allow highlighting the inter-
101 individual variability as well as the specific contribution to dietary exposure of rarely
102 consumed food like fish.

103 **Material and methods**

104 *Food consumption survey*

105 Various types of surveys can be used to assess dietary exposure (Kroes et al., 2002) but most
106 of them are based on survey durations between 1 and 7 days. The current study used the
107 household budget survey performed by TNS-Secodip (Boizot, 2005) and hereafter called
108 TNS-Secodip in the text. In household consumption surveys, the amounts of foods and drinks
109 purchased into the household are recorded. The TNS-Secodip panel corresponds to French
110 households included by random sampling combined with quotas method in order to obtain a
111 correct representativeness together with a relatively high response rate (EFSA, 2006;

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4 112 Feinberg, et al. 2006). Household surveys do collect data every week during multiple years.
5 113 Unfortunately, they do not provide information on actual consumption by household's
6 114 individual members. Therefore, following rules were applied for selecting the sample of
7 115 households adapted to this study:

- 11 116 – Select households made of only one individual, i.e. single person households;
- 12 117 – Select same single person households who regularly reported over the three years
- 13 118 between 2003 and 2005;
- 14 119 – Discard households with less than 40 weeks/year;
- 15 120 – Discard households with partial recording of total consumption controlled from total
- 16 121 energy intake;
- 17 122 – Discard households with missing data, such as absent body weight.

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26 123 In the rest of the text we used the term “household” for “single person household”. In addition
27 124 to the food consumption, the TNS-Secodip survey provides information on subjects like their
28 125 socio-professional categories, their age, sex, body weight and place of residence.

29
30
31
32 126 Within the food items recorded by the survey participants, only those which can significantly
33 127 contribute to the exposure to dioxins i.e. product of animal origin containing fat (De Mul, et
34 128 al. 2008; Parzefall W, 2002, WHO, 2002), were further analyzed. In practice, all food
35 129 containing less than 30% of ingredients potentially contaminated by dioxins and related
36 130 compounds were excluded. For example, a pizza containing 10% of ham was not taken into
37 131 account. Finally 31 food categories were considered and listed in Table 1.

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43 132 (Table 1)

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46 133 Within these food categories, 19 are foods containing fat from animal origin and which are
47 134 therefore the main vectors for dioxins and related compounds. The other 12 food groups are
48 135 composite foodstuffs containing at least 30 % of the main vectors as ingredient. For these
49 136 composite foods, an adjustment factor of 30 % was systematically applied on the level of
50 137 occurrence in the absence of accurate recipes.

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55 138 The consumption was calculated per week, per person and per food category over 3 years.
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4 139 ***Contamination data***

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6 140 Analytical results were collected by the French Ministry of Agriculture and Fisheries between
7
8 141 2002 and 2006 on foods on the French market. Seven congeners were analyzed for PCDDs,
9
10 142 10 congeners for PCDFs and 12 congeners for dl-PCBs (Commission Regulation, 2006).
11
12 143 Results are expressed in pg WHOTEQ/g of fresh weight.

13
14 144 The weekly exposure was assessed for each household in a deterministic way by multiplying
15
16 145 the individual weekly consumption of the food categories by the corresponding mean level of
17
18 146 contamination. It should be mentioned that results below the Limit of Reporting (left-censored
19
20 147 data) were replaced by the corresponding Limit of Reporting (Limit of Detection or Limit of
21
22 148 Quantification). Due to the relatively low number of non-quantified results, the impact of
23
24 149 substitution by the limit of reporting was limited when compared with the substitution by 0.
25
26 150 This approach has also the advantage of not underestimating the exposure (WHO, 1995).

27 151 For dairy products, dioxin concentrations were extrapolated from the value analyzed in fat
28
29 152 milk. For animal fat, dioxin concentrations were extrapolated from the values analyzed in
30
31 153 meat and standardized accordingly to fat content. The percentage of fat in each food group
32
33 154 was extracted from the French table of composition (REGAL) (Feinberg, et al. 1991).

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35 155 ***Dietary exposure assessment***

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38 156 In order to be able to use household consumption survey data in the purpose of assessing
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40 157 dioxin dietary exposure several assumptions must be made:

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42 158 — Food quantities that were purchased and recorded correspond to food quantities that
43
44 159 were consumed and edible part was assumed to be 100% for all foods; this hypothesis
45
46 160 may generate an overestimate of exposure;
47
48 161 — Only foods consumed at home are taken into account and selected foods when
49
50 162 consumed out of home (restoration) are neglected; this hypothesis may generate an
51
52 163 underestimate of exposure.

53
54 164 Individual household weekly exposure to PCDD/F and dl-PCB is calculated using a
55
56 165 deterministic approach, given by equation (Eq. 1):

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58
59 166
$$E_{i(k)} = \frac{\sum_{j=1}^J C_{ij(k)} \times T_j}{P_i} \quad \text{Eq.1}$$

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4 167 Where:

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6 168 — $E_{i(k)}$ household exposure i ($1 < i < I$) for week k ($1 < k < K$), expressed in pg

7
8 169 WHOTEQ/kg BW/week;

9
10 170 — $C_{ij(k)}$ food quantity consumed for food category j ($1 < j < J$), for week k , for individual

11
12 171 i , expressed in g/week;

13
14 172 — T_j estimated contaminant concentration for food category j , in pg kg^{-1} fresh weight

15
16 173 (FW);

17
18 174 — P_i body weight (BW) of individual i , in kg.

19
20 175 Statistical data processing was performed using JMP software release 7.0.1 (SAS Institute,

21
22 176 Cary, NC, USA).

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26 177 **Results**

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29 178 ***Sample description***

30
31 179 Three consecutive household consumption surveys were carried out by TNS-Secodip during 3
32 180 years, from 2003 up to 2005, i.e. over 157 weeks. A preliminary step consisted in
33 181 harmonizing the different coding systems used over this period of time in order to merge the
34 182 three surveys. Several single person households participated over all survey period and were
35 183 selected. A preliminary sub-sample of 1161 single person households was then sorted out. But
36 184 after applying the selection criteria described above, a working sample of 328 single person
37 185 households was selected and used for the study.

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44 186 Major descriptive characteristics of the working sample are given in Table 2. When referring
45 187 to classical Body Mass Index (BMI) criterion, 50% of individuals are normal weight, 35% are
46 188 overweight ($25 \text{ kg/m}^2 < \text{BMI} < 30 \text{ kg/m}^2$), 12% are obese ($\text{BMI} > 30 \text{ kg/m}^2$), and 3% are
47 189 underweight ($\text{BMI} < 18.5 \text{ kg/m}^2$). Most represented geographical regions are Paris and its
48 190 surroundings (25%), East-central region (10%), and South-east region (8%). Moreover, 14
49 191 French departments were not represented in the working sample, and TNS-Secodip purposely
50 192 discards several departments, such as overseas departments, from the household purchase
51 193 surveys because local consumption behavior is known to be different from the rest of the
52 194 country. Main occupational categories of selected single person households were: 50%

195 occupied government and private positions and 44% were retired. Only 6% of the panel
196 members were unemployed (not including retired persons), and no student participated.

197 (Table 2)

198 When considering these raw figures, it is obvious that the working sample was not
199 representative of whole French population, as it is usually defined when using quota sampling
200 for sampling plan. This bias was considered as negligible because the purpose of this study
201 was to assess a possible seasonal effect on dioxin food exposure and the number of weeks
202 with consistent food records was considered as the most important objective.

203 ***Household consumption estimate***

204 According to notations used in equation 1, the working sample consisted in $I = 328$
205 households, $J = 31$ food categories, and $K = 157$ weeks. If all households should have
206 consumed, at least, one time each food category each week, the theoretical number of records
207 $C_{ij(k)}$ should be equal to $328 \times 31 \times 157 \approx 566\,000$ food consumption, expressed in g/kg
208 BW/week. However, despite the selection criteria used to sample households, many
209 individuals never consumed certain foods over this long period of time (3 years). The
210 importance of non consumers is indicated in the column of Table 1 where percents of
211 consumers are reported for each food category. Altogether, about 425 000 weekly food
212 consumption were calculated for the 328 households, each value being, by itself, the weekly
213 summation of several possible daily consumption.

214 Food categories consumed in larger quantities, i.e. purchased, by at least 95% of households
215 over the survey period were fishes, milk and dairy products, beef meat, pork meat and poultry.
216 On the contrary, foods consumed in smaller quantities were animal fats excluding butter and
217 creams (less than 10%), butter and composite dishes containing cephalopods and mutton
218 offal.

219 When looking at the gross average food consumption, results are consistent with those
220 observed in other French food consumption surveys based on individual surveys. By range of
221 magnitude, most frequent food categories were milk and yoghurts (14 g/kg BW/week), pork
222 meat (5.33 g/kg BW/week), beef meat (3g/kg BW/week), and cheese (2.37 g/kg BW/week).
223 When grouping food categories by principal types – dairy products, animal foods, seafood,
224 eggs, dishes excluding seafood, and fats – it appears in the bottom of Table 1, that dairy
225 products and animal foods embody the major part of the total consumption of dioxin food

226 sources with almost 91% of total diet, complemented by seafood and composite dishes based
227 on seafood (8%) and eggs (3%); fats and others composite dishes are negligible.

228 Because several foods, such as vegetable and fruits or beverages, were removed from the
229 selection, it must be reminded that these data are not representative of some “average diet”.
230 Only food categories that were recognized as potential vectors for dioxins were selected and
231 used in the calculation of the global food consumption. Considering the aim of the study, it
232 was a realistic choice, as a poorly consumed highly contaminated foods category may
233 represent an important source for exposure.

234 *Average dioxin concentration in foods*

235 The average concentrations in the 31 food categories were estimated from analytical results,
236 when available. The major source of data consisted in analytical results collected during
237 different official control surveys performed. These values are put together in the last columns
238 of Table 1.

239 For several foods, the average sum of pollutants (PCDD/F + dl-PCB) is not equal to the sum
240 of average concentrations of individual pollutants (PCDD/F and dl-PCB). This is due to the
241 fact that the number of analytical data used to calculate each mean is not the same for both
242 pollutants. In column “Number”, first number indicates the total number of measurements, the
243 second the number of measurements below LOD/LOQ. When data were left-censored, i.e.
244 below LOD/LOQ, censored data were substituted by LOD or LOQ reported by the
245 laboratories. For several food categories i.e. dairy products, animal fats and composite foods,
246 no analytical measurements were available and “average” values were imputed as described
247 in section Material and methods; they are marked by letter “E”.

248 According to these data, fishes are most contaminated foods with an average content close to
249 2.5 pg WHOTEQ/g fresh weight (FW). Other seafood are also significantly contaminated, but
250 weakly consumed. All average PCDD/F + dl-PCB concentrations of animal foods are below 1
251 pg WHOTEQ/g FW. Because of their high fat contents, offal is always more polluted than
252 meat, except for poultry. By decreasing order, the concentrations in animal foods are mutton,
253 beef, pork and poultry. Average concentration in eggs is 0.14 pg WHOTEQ/g FW and is
254 comparable to mutton meat while milk, with 0.06 pg WHOTEQ/g FW, is slightly polluted.

255 Relative proportions of PCDD/F compared to dl-PCB were estimated from available data (but
256 are not given here) and are highly variable depending on the food category. This can be

257 explained by the fact that the origins of both these types of chemicals are different as they are
258 differently accumulated in the various environmental compartments. As a rule of thumb, dl-
259 PCB represents about 80% total contaminants burden for fishes, and falls to 50 to 60% for
260 fats, animal foods, and eggs.

261 *Estimation of dietary exposure*

262 Average population exposure

263 A rough point estimate of dietary exposure can be directly calculated from data in Table 1 by
264 multiplying food consumption by food concentration. The average exposures to PCDD/F +
265 dl-PCB for each food categories are reported in the last column of the table. Thus, total
266 average exposure is about 10.42 pg WHOTEQ /kg BW/week; this value can be compared to
267 the Provisional tolerable weekly intake (PTWI) 14 pg WHOTEQ/kg BW/week, and the global
268 situation seems to be satisfactory. The mean exposure from dairy products, fish and other sea
269 foods represents 88% of the total (9.35 vs. 10.42 pg WHOTEQ/kg body weight/week). The
270 first category is a major contributor because of its high and widely distributed consumption
271 across the French population. The second is a major contributor because of a high mean level
272 of contamination and despite of lower levels of consumption as well as percentage of
273 consumers. In the bottom of Table 1, the contributions of the six principal food types were
274 estimated in the same way. The major role of fishes and seafood can then be exhibited
275 whereas they only represent 8% of total food consumption but 62% of dietary exposure. Next,
276 dairy products contribute to 27% of dietary exposure but represent 65% of total food
277 consumption.

278 Individual weekly exposures

279 However, for purpose of identifying any group at risk, average population exposure may be
280 misleading as it does not give any indication on individual exposure distribution nor seasonal
281 influence. Therefore, individual weekly exposure, denoted $E_{i(k)}$, were calculated from
282 consumption data and average concentrations of PCDD/F + dl-PCB reported in Table 1 by
283 applying equation 1. Because the number of weeks was 157 and the number of households
284 328 the total number of values should be $328 \times 157 = 51\,496$. But several records were
285 missing due to holiday periods. The total number of individual weekly exposures was 41\,789,
286 i.e. 81% of expected number. This percent is large enough to be considered as representative
287 of the seasonal/individual variations of dietary exposure to dioxin. Figure 1 illustrates the

288 histogram of $E_{i(k)}$ whatever the week or the household. Thus, it can be shown that there is a
289 very large dispersion of data as they are ranging from 0.11 pg to 327 pg WHOTEQ/kg
290 BW/week, while about 26% of estimated weekly dietary exposures are above PTWI and 0.5%
291 above 100 pg WHOTEQ/kg BW/week.

(Figure 1)

293 The right-skew of data distribution is typical of many risk assessment studies on food
294 contaminants when individual exposures are estimated instead of average population exposure
295 (Tressou, et al. 2004; Crépet, et al. 2005). Obviously, data are not distributed following
296 normal distribution. Because, the mean of normal distribution is estimated by the arithmetic
297 mean, this means that this statistic is not well adapted to estimating population exposure.
298 According to the typical shape of the histogram, it is sensible to fit an exponential law to these
299 data. The probability density function of the exponential distribution is described by the
300 following function (when $x \geq 0$) and fully defined by a single parameter λ :

$$f(x) = \lambda \exp(-\lambda x)$$

302 Then, the mean of an exponential law is equal to $1/\lambda$. Exponential probability density function
303 was adjusted to individual weekly exposures and confirmed by a Kolmogorow-Smirnov
304 adjustment test. The mean was estimated thanks to this new parameterization and observed
305 value was 12.1 pg WHOTEQ /kg BW/week. This value is higher than the former arithmetic
306 mean because it better takes into account the inter-individual variability.

307 It is also possible to use this statistical modeling to estimate the percent of weekly exposure
308 data which are above the PTWI; about 27.1% weekly exposures exceed this Health Based
309 Guidance Value. Separate exponential distribution laws were also fitted for each year and the
310 percent of exceeding exposure values were estimated; they are ranging from 21 to 22%; this is
311 still very high but also demonstrates the great stability of this percent over time.

312 At this stage it can be concluded from these data that there is a real risk of sporadic
313 overexposure to dioxin for a non negligible portion of French population: about 1/5 weekly
314 dietary exposures may be too high. It means that in average about 10 weeks per year,
315 consumers are exposed to too high dioxin intake.

316 Contribution of fish to the overall dietary exposure

317 More in detail, the analysis of contributions in the case of dietary exposure exceeding the
318 Health Based Guidance Value shows that it is mainly related to an increase in the
319 consumption of fish and other seafood.

320 A complementary approach consists in selecting within all the individual weekly exposures,
321 the ones in which a consumption of fish and other seafood was reported. This happens for
322 about half of the cases, i.e. 19,070 persons \times weeks over 41,789. It is therefore possible to
323 estimate the contribution from fish and seafood noted $E_{i(k)}^*$, relative to the overall exposure
324 noted $E_{i(k)}$ by calculating the ratio $B_{i(k)}$ for a considered individual during a considered week:

$$325 \quad B_{i(k)} = \frac{E_{i(k)}^*}{E_{i(k)}} \quad \text{Eq. 2}$$

326 Figure 2 illustrates the frequency distribution of this ratio. It shows that in about 3% of cases
327 fish and other seafood are the only contributors to the dietary exposure. In 25% of cases they
328 represent 88% and in half of cases they represent 75% of the total dietary exposure.

329 These results globally emphasize the predominant role of fish consumption regarding dietary
330 exposure to dioxins and related compounds. However, they may hide many specific situations
331 where fish consumption poorly contributes to overall exposure as illustrated by the minimum
332 value of the distribution, at 0.6%.

333 (Figure 2)

335 Average individual weekly exposures

336 When considering the distributions of $E_{i(k)}$ for each individual 157 weeks, the same typical
337 exponential distribution can be observed. Average individual weekly exposures were then
338 calculated for each of the 328 households, all years and weeks confounded, and reported in
339 Figure 3. A log-normal distribution can be fitted to the data of this new histogram and
340 confirmed by a Kolmogorow-Smirnov adjustment test. The mean of a log-normal distribution
341 can be estimated by the geometric mean. The estimated value of the geometric mean is 10.2
342 pg WHOTEQ/kg BW/week and the 95% confidence interval is [9.6, 10.9]. Most interesting
343 percentiles values are: 99.5 % is 77.8 pg; 97.5 % is 38.9 pg, and 50.0 % (median) is 10.5 pg.

344 (Figure 3)

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4 345 Modeling weekly exposure as a function of time

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6 346 Also starting from $E_{i(k)}$ values, it is possible to estimate the weekly average by summing over
7 347 all households. The 157 obtained data can be represented as a function of the week when they
8 348 were calculated. They form a time series that can be used to show evidence of seasonal
9 349 variation. In order to ascertain this dependence with time, time series statistical techniques
10 350 were used as described in (Pandit and Wu, 1983). It can be assumed that there is no trend
11 351 because it was demonstrated that global average exposure was stable over year as illustrated
12 352 in Table 3. Thus, it is not necessary to use detrending techniques and a simple exponential
13 353 smoothing is sufficient to show the seasonality as illustrated by Figure 4. Observed averages
14 354 and averages predicted by using the smoothing model with their 95% confidence intervals are
15 355 simultaneously presented on the graphics.

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24 356 (Table 3)

25
26 357 Average exposure to dioxin is actually constant over the three-year period of the study
27 358 (around 12 WHOTEQ/kg BW/week), but increase up to a maximum of 13 pg WHOTEQ/kg
28 359 BW/week between March and September and decrease down to a minimum of 11 pg
29 360 WHOTEQ/kg BW/week between December and February. The magnitude of these average
30 361 variations is small if confidence intervals are not taken into consideration. But if they are
31 362 considered, the risk to exceed PTWI is rather frequent. For instance, the upper limits of the
32 363 confidence intervals are exceeding PTWI about 35 weeks per year.

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39 364 (Figure 4)

40
41 365 Complete and partial autocorrelation functions were also calculated in order to extract
42 366 parameters for what is called the seasonal Autoregressive integrated moving average
43 367 (ARIMA) model in time series theory. This kind of model can be used for predicting no
44 368 observed situations, and the results observed during 2003 to 2005 were used to build the
45 369 model and predict exposures for 2006. The seasonal coefficient used for the ARIMA model
46 370 was 13 and the order coefficient 7; according to classical nomenclature, this model is defined
47 371 as ARIMA (7,0,7)13 and illustrated on Figure 5. Confidence intervals are narrower because
48 372 this model is more explicative than simple smoothing as it takes into account the role of
49 373 seasons. Therefore, the number of times the upper limit of confidence intervals are exceeding
50 374 PTWI is smaller and only 15 weeks per year in average. However, the variations from one
51 375 week to another may still be very large and can be explained by the uncertainty of exposure
52 376 estimates.

377 (Figure 5)

378 Obviously, the observed stability over time is emphasized by the fact point estimate and
379 unique dioxin concentrations were used for exposure assessment. The application of
380 probabilistic risk assessment technique and several concentration levels should increase the
381 variability and slightly modify these conclusions, although no actual decrease of dioxin
382 concentration in fish was observed during the considered years.

383 **Discussion and conclusion**

384 Long-term single person household purchase survey was used to estimate long-term food
385 consumption for 328 single person households. These data were combined to average
386 concentrations of PCDD/F + dl-PCB in 31 selected foods categories. It was then possible to
387 derive different dietary exposure values. It was mainly possible to demonstrate that dietary
388 exposure to dioxin may vary with the season and that, if the global point estimates may lead
389 to conclude that they are below PTWI, individual variations and seasonal effect are important
390 and may induce frequent exposure above the Health Based Guidance Value. Several
391 important methodological conclusions may also be drawn from this study.

392 1) Household purchase surveys are interesting to show evidence of seasonality for long-
393 term dietary exposure and consequently food risk assessment. Because they are
394 performed over a period of time much longer than few days records or 24-hour recalls,
395 it is possible assess more accurately the consumption habits of a large panel. For this
396 study, it appears that fish consumption is not constant over months and influences the
397 exposure to dioxin. Recent report from the French fish marketing board (Office
398 FranceAgrimer) which is in charge of the regulation of fish markets in France,
399 partially confirms the seasonality of fish consumption in relation to the seasonal
400 variations of stocks and fishing campaigns (FranceAgrimer, 2008).

401 2) Because seasonality of dioxin exposure covers one year collecting data over a short
402 period of time, likewise few days record or 24-hour recalls may conceal the season
403 effect and neglect the modifications of consumption behavior as a function of time On
404 the contrary, it may be sufficient to collect data over one year or to correct the
405 sampling for seasonal variations over a period of 1 year. A recent study realized by
406 Tard *et al.* 2007 has been used to define the French exposure to total dioxins expressed
407 in toxic equivalents (TEQs). The study combines concentrations data from French

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4 408 monitoring programs (2001-2004) with the first French Individual Consumption
5 409 Survey (INCA 1999). This survey is based on seven 24-hour diet records corrected for
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7 410 seasonal variations by a sampling of subjects distributed over 1 year. The daily
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9 411 exposure to total dioxins was estimated to be on average 1.8 pg WHOTEQ/kg
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11 412 BW/day, i.e. 12.6 pg WHOTEQ/kg BW/week, and very similar to our own average
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13 413 estimates. Therefore, it seems that there is a good convergence of results for average
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15 414 point estimates whatever the food survey methodology. But only long-term surveys
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17 415 may help identifying a seasonal effect.

- 18 416 3) This study also confirms the results of other risk assessment studies and demonstrates
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20 417 that the possibility for an individual to exceed PTWI is a rather frequent event as it is
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22 418 observable in about 20% of weeks; it is not a rare event.

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24 419 From a general point of view, the estimation of the uncertainty of dietary exposure
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26 420 simultaneously depends on uncertainty of consumption data and uncertainty of contamination
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28 421 data. Obviously, this study may present some major sources of uncertainty that may explain
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30 422 the large variability of observed data.

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32 423 Firstly, when considering consumption data:

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34 424 Consumption outside home, auto-consumption and non-consumption were not
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36 425 accounted and total food consumption is certainly systematically biased. Food
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38 426 categories were selected for their assumed usual high concentration in dioxin. But,
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40 427 other food categories may also contribute to global exposure. For instance, in its study
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42 428 of 2005, AFSSA stated that cereals and fruits and vegetables may represent 1% and
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44 429 6% respectively of total PCDD-PCDF + dl-PCB dietary exposure for adults
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46 430 (Moccarelli, et al. 2008). As consumption of vegetal foods was neglected and
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48 431 consumption of butter seemed very low in our study, some underestimation of total
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50 432 dioxin dietary exposure is likely. Although vegetal foods are little contaminated, they
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52 433 are highly consumed in France and they may represent a non negligible contribution to
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54 434 total exposure.
- 55 435 The fact that we were able to use three year-survey must be put into balance. Such a
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57 436 long-term survey cannot be easily achieved by other recording methods. The working
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59 437 sample was sampled by only selecting single person household. Consequently, it is a
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438 small sample which presents its own characteristics and extrapolation to the rest of

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4 439 French population must be cautiously made, as it is not evident that single person
5 440 household consumption behavior is comparable to other households.

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7 441 However, no food survey methodology can be claimed as unbiased and each may
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9 442 underestimate or overestimate some aspects of food consumption. Therefore, we were
10 443 always very cautious in contrasting our results with those obtained by other means.
11 444 This was only possible for global average exposure estimates and presented results can
12 445 be considered as very consistent. But the great advantage of used data was their ability
13 446 to demonstrate the seasonality of dioxin exposure.

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19 447 Secondly, when considering contamination data:

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21 448 Only “average” values were used and exposure was calculated in a deterministic way
22 449 while it is more interesting to apply a probabilistic approach (Feinberg, et al. 2006).
23 450 But this requires more contamination data than we were actually able to gather
24 451 (mainly for these food categories where average values were imputed) and a more
25 452 important computational effort.

26 453 Because left-censored data were substituted by detection limit, exposure was not
27 454 underestimated. But, for some foods, the small number of analytical data for
28 455 calculating the average, or the absence of any analytical data, greatly contributes to
29 456 total uncertainty.

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38 457 Despite these precautions, if one extrapolates the results to entire French population, it can
39 458 concluded that, if the estimate is restricted to global average, the average weekly exposure is
40 459 12 pg WHO_{TEQ}/kg BW/week and below the health based guidance value of 14 pg
41 460 WHO_{TEQ}/kg BW/week. This value is comparable to the estimates published by the Scientific
42 461 Committee on Food (SCF) of the European Commission in 2001 (SCF, 2001). These
43 462 estimates were derived from various short-term studies, and it seems it unnecessary to use
44 463 long-term studies to assess as average exposures and reduce the uncertainty. However, our
45 464 study shows that in average, people who consume foods contaminated by dioxin are above
46 465 PTWI 10 weeks/year, because about 20% of individual estimated weekly exposures are above
47 466 PTWI each year.

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56 467 Finally, because dioxins and related compounds are accumulating in the body, this study
57 468 could be completed by Kinetic Dietary Exposure Model (Verger et al. 2007; Bertail et al.
58 469 2010) which take into account the period of time between two food consumption and the half-
59 470 life of PCDD/F and dl-PCB in the body, in order to evaluate the total body burden of

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4 471 consumers and estimate more realistically the probability to exceed the Health Based
5 472 Guidance Value. Dynamic models were developed for methyl mercury from fish but they
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7 473 require more sophisticated modeling to calculate the dietary exposure from various inter-
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9 474 dependent food groups.

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Tables

Table 1: Food categories selected for the study: average weekly consumption expressed as g/kg Body Weight/week, % of consumers and average dioxin contents. Remarkable values are underlined

Table 2: Description of the survey sample

Table 3: Average individual weekly exposure per year to PCDD/PCDF/dl-PCB (expressed in pg WHOTEQ /kg BW/week

Figures

Figure 1: Distribution of individual weekly dietary exposures for the 238 households during 3 years (i.e. 41,789 weeks). The grey area represents the weekly exposure higher than the Health Based Guidance Value of 14 pg WHOTEQ/ kg BW/week, (27.1% of values).

Figure 2. Distribution of ratios between exposure from fish and other sea food and from total food for weeks during when fishes were consumed

Figure 3: Distribution of average weekly exposures of each of the 328 households. Log-normal distribution law can be fitted to the data.

Figure 4: Application of simple exponential smoothing to observed average weekly exposures (all households confounded) over 3-year period. Legend: full squares represent observed values; continuous line predicted values, dot lines the limits of 95% confidence interval of predicted values, and Provisional Tolerable Weekly Intake (PTWI).

Figure 5: Adjustment of average weekly exposures by seasonal ARIMA (7,0,7)13 model. Data from 2003 to 2005 were used to estimate the parameters of the model that was used to predict exposures in 2006. Legend is the same than in Figure 3.

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Tables

Table 1: Food categories selected for the study: average weekly consumption expressed as g/kg Body Weight/week, % of consumers and average dioxin contents. Remarkable values are underlined

Food categories	Food consumption		Average contamination pg WHOTEQ/g FW ⁽¹⁾				Exposure pg/kg BW/week
	Average g/kg BW/week	% consumers	Number ⁽²⁾	PCDD/F	dl-PCB	PCDD/F + dl- PCB	
Fishes	2.8	100%	578 – 583	0.340	<u>2.130</u>	<u>2.470</u>	<u>5.9360</u>
Mollusks	0.27	46%	42 – 43	0.290	0.570	0.860	0.2295
Shellfishes	0.53	79%	70 – 71	0.240	0.220	0.460	0.2385
Cephalopods	0.02	15%	55	0.180	0.370	0.550	0.0170
Milk	<u>14.1</u>	98%	35	0.020	0.040	0.060	0.8442
Butter	0.001	3%	24	0.200	0.350	0.550	0.0006
Cheese and creams	2.4	98%	E ⁽³⁾	0.150	0.330	0.480	<u>1.1376</u>
White cheese	0.49	58%	E	0.080	0.170	0.250	0.1225
Yoghurts, dairy desserts and ice-creams	<u>15.0</u>	100%	E	0.020	0.040	0.060	0.7485
Pork meat and delicatessen	5.3	99%	13	0.020	0.020	0.040	0.2665
Pork offal	0.16	52%	7	0.060	0.030	0.090	0.0144
Mutton offal	0.03	18%	2	0.420	0.460	0.880	0.0264
Mutton meat	0.58	75%	10	0.040	0.070	0.110	0.0638
Poultry offal	0.15	70%	6	0.020	0.130	0.150	0.0225
Poultry meat	1.97	99%	46	0.020	0.020	0.040	0.0788
Beef meat	3.0	97%	23	0.030	0.060	0.090	0.3000
Beef offal	0.27	64%	8	0.040	0.070	0.110	0.0297
Animal fats excluding butter and creams	0.01	8%	E	0.450	0.540	0.990	0.0099
Eggs	1.7	94%	155	0.070	0.070	0.140	0.2366
Dishes with >30% Fishes	0.11	51%	E	0.100	0.640	0.740	0.0704
Dishes with >30% Mollusks	0.004	9%	E	0.090	0.170	0.260	0.0010
Dishes with >30% Shellfishes	0.01	15%	E	0.070	0.070	0.140	0.0014
Dishes with >30% Cheese	0.09	35%	E	0.040	0.100	0.140	0.0135
Dishes with >30% Pork meat	0.09	47%	E	0.010	0.010	0.020	0.0009
Dishes with >30% Pork offal	0.01	16%	E	0.020	0.010	0.030	0.0003
Dishes with >30% Mutton offal	0.02	13%	E	0.120	0.140	0.260	0.0052
Dishes with >30% Poultry offal	0.0001	1%	E	0.005	0.040	0.045	0.0000
Dishes with >30% Poultry meat	0.14	54%	E	0.005	0.010	0.015	0.0014
Dishes with >30% Beef meat	0.17	54%	E	0.010	0.020	0.030	0.0051
Dishes with >30% Beef offal	0.002	4%	E	0.010	0.020	0.030	0.0001
Dishes with >30% Eggs	0.01	16%	E	0.020	0.020	0.040	0.0004
Grouping							
Dairy products	31.9	65%					<u>2.85</u>
Animal foods	11.5	23%					0.80
Seafood	3.7	8%					<u>6.49</u>
Eggs	1.7	3%					0.24
Dishes (excluding seafood)	0.5	1%					0.03
Fats	0.01	0%					0.01
Total	49.4						10.42

Footnotes

- (1) Data from official control plans of Direction Générale de l'Alimentation (DGAL) a division of French Ministry of Agriculture. FW stands for fresh weight.
- (2) Number: first number indicates the number of quantified data and the the second number includes left-censored data below determination limits (LOD or LOQ).
- (3) E extrapolated concentration

Table 2: Description of the survey sample

	Women	Men	Total
Number	225	103	328
% total	69%	31%	
Average age (years)	62.1 (± 14.3)	54.3 (± 14.4)	59.7 (± 14.8)
- More than 50 years	80%	58%	
Average BMI	24.90 (± 4.52)	25.40 (± 3.77)	25.06 (± 4.30)

Footnotes

BMI : Body mass Index expressed as kg/m² (SD between parentheses)

**Table 3: Average individual weekly exposure per year to PCDD/PCDF/dl-PCB
(expressed in pg WHOTEQ /kg BW/week**

	Average	Standard deviation	Minimum	Maximum	(1)	%	(2)	%
2003	10.7	7.8	1.4	54.5	71	22%		
2004	10.5	7.2	0.8	49.2	64	20%		
2005	10.3	7.3	1.0	56.6	67	20%	99	30%
2003 et 2004	10.6	7.3	1.4	48.7	65	20%		
2004 et 2005	10.4	7.0	0.9	52.6	56	17%	59	18%
2003 à 2005	10.5	7.1	1.6	50.2	61	19%	44	13%

Footnotes

- (1) Number of persons exceeding PTWI
(2) Number of persons with at least one yearly average exposure that exceeds PTWI

Figures

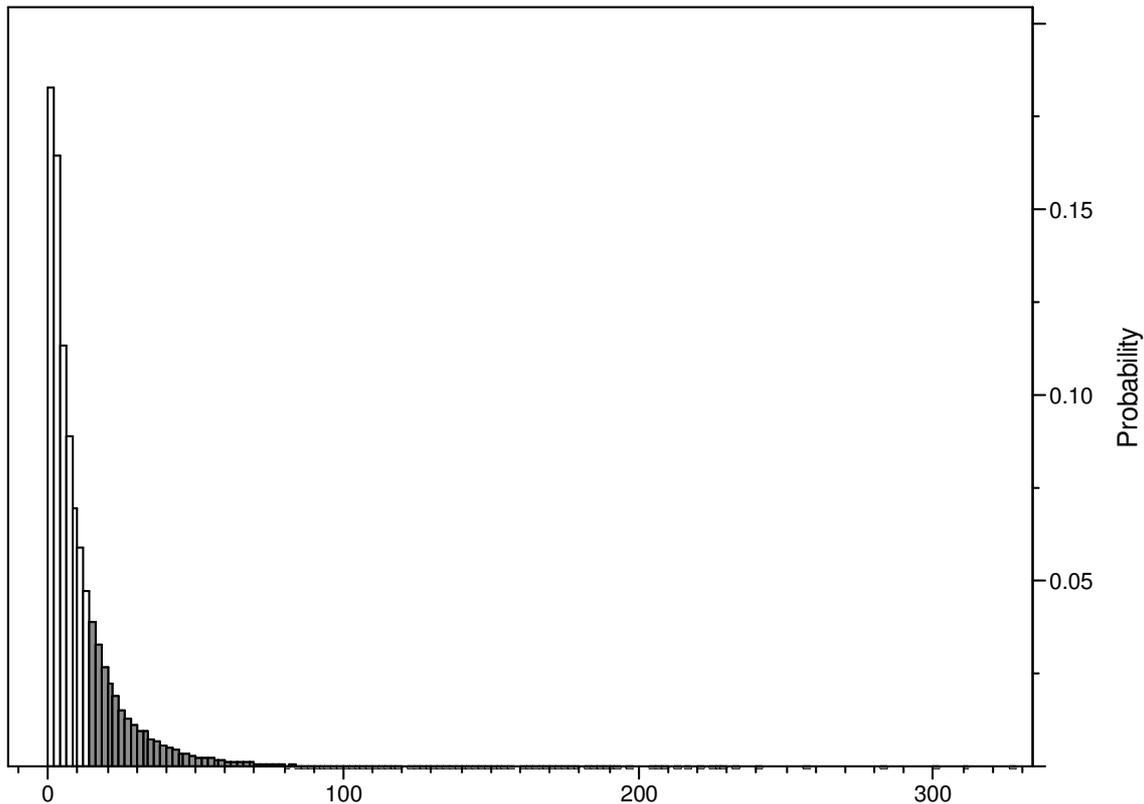


Figure 1: Distribution of individual weekly dietary exposures for the 238 households during 3 years (i.e. 41,789 weeks). The grey area represents the weekly exposure higher than the Health Based Guidance Value of 14 pg WHOTEQ/ kg BW/week, (27.1% of values).

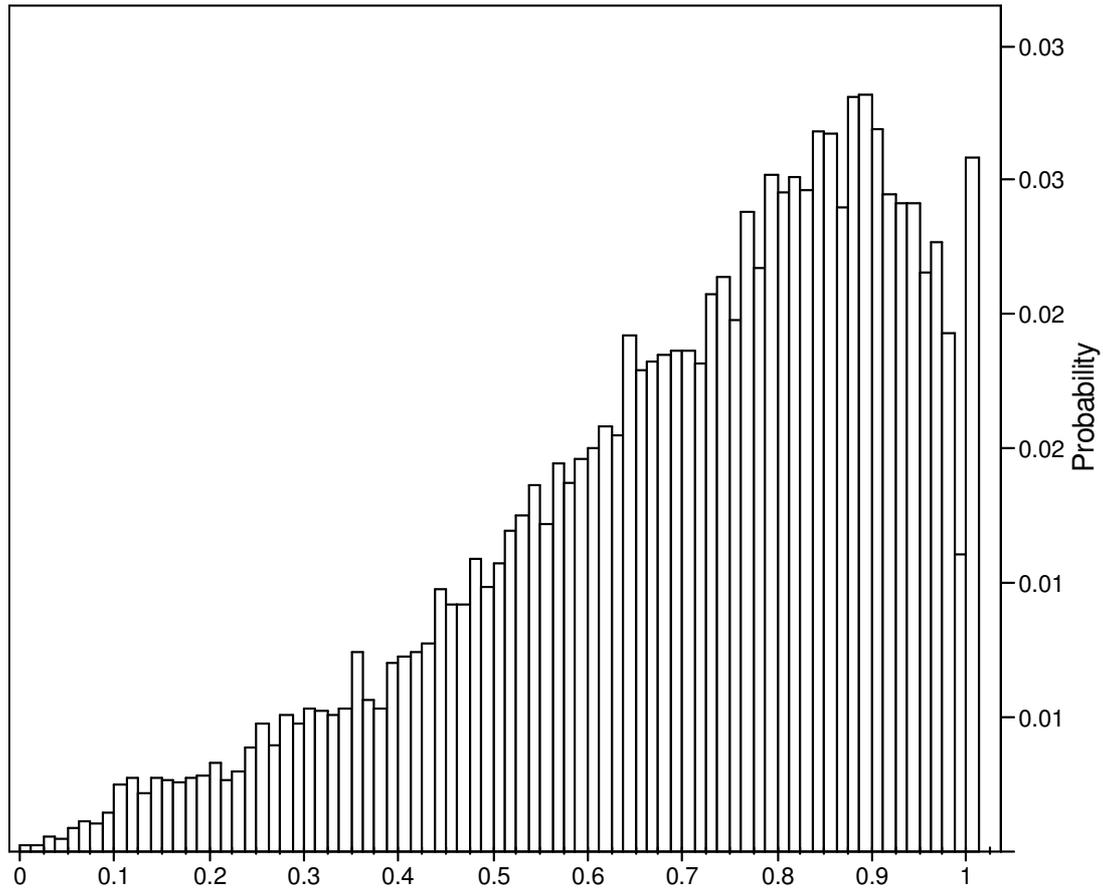


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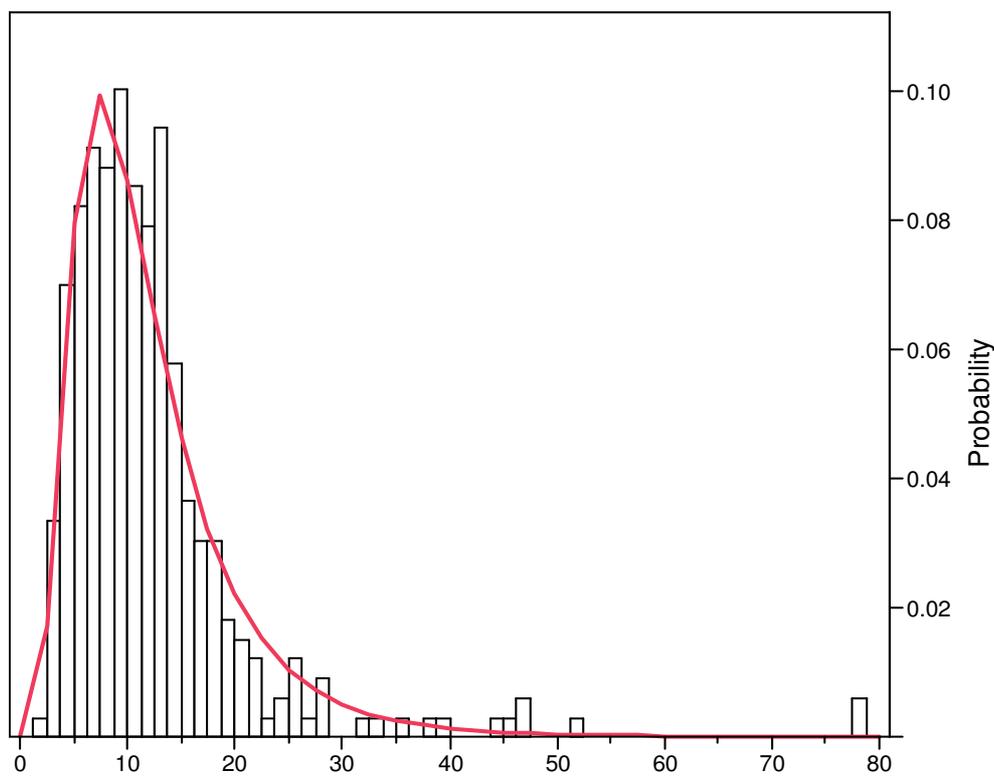


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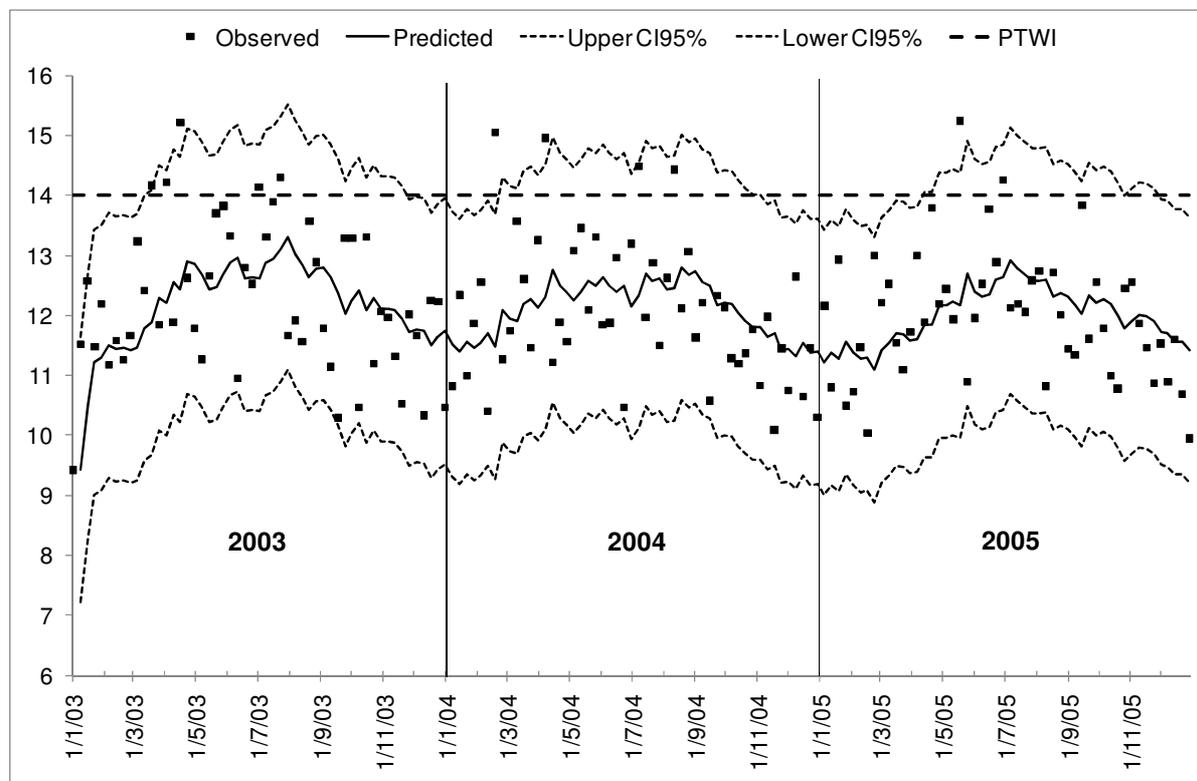


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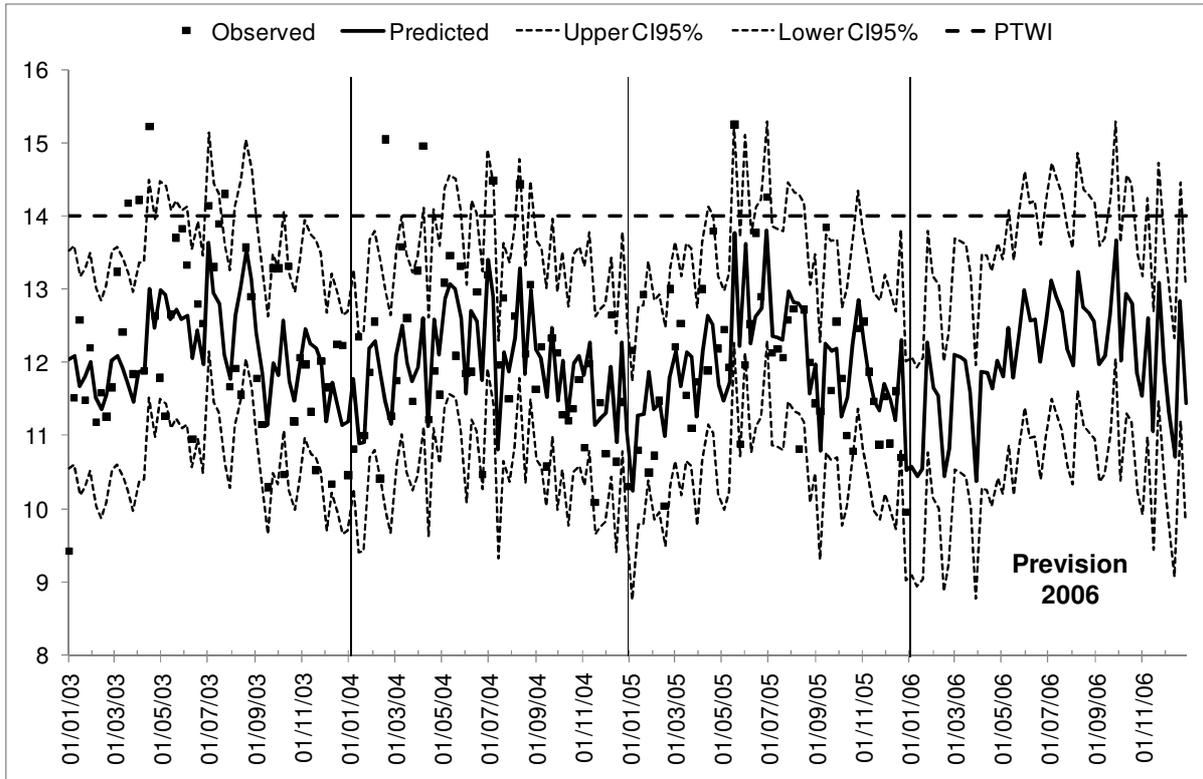


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