



HAL
open science

Exposure assessment of the Belgian population to pesticide residues through fruit and vegetable consumption

Wendie Liliane Claeys, Simon de Voghel, Jean-François Schmit, Valérie Vromman, Luc Pussemier

► **To cite this version:**

Wendie Liliane Claeys, Simon de Voghel, Jean-François Schmit, Valérie Vromman, Luc Pussemier. Exposure assessment of the Belgian population to pesticide residues through fruit and vegetable consumption. *Food Additives and Contaminants*, 2008, 25 (07), pp.851-863. 10.1080/02652030701854741 . hal-00577445

HAL Id: hal-00577445

<https://hal.science/hal-00577445>

Submitted on 17 Mar 2011

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

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



Exposure assessment of the Belgian population to pesticide residues through fruit and vegetable consumption

Journal:	<i>Food Additives and Contaminants</i>
Manuscript ID:	TFAC-2007-346.R1
Manuscript Type:	Original Research Paper
Date Submitted by the Author:	27-Nov-2007
Complete List of Authors:	Claeys, Wendie; FASFC, Policy Control de Voghel, Simon; CODA-CERVA-VAR Schmit, Jean-François; FASFC Vromman, Valérie; FASFC Pussemier, Luc; CODA-CERVA-VAR
Methods/Techniques:	Exposure assessment, Probabalistic modelling
Additives/Contaminants:	Pesticides
Food Types:	Fruit, Vegetables

SCHOLARONE™
Manuscripts

1
2
3 1 **EXPOSURE ASSESSMENT OF THE BELGIAN POPULATION TO**
4
5 2 **PESTICIDE RESIDUES THROUGH FRUIT AND VEGETABLE**
6
7 3 **CONSUMPTION**
8
9
10 4
11
12 5
13
14 6
15
16 7
17
18
19 8 **Abstract**
20

21 9 Exposure of the Belgian consumer to pesticide residues from consumption of fruit and
22
23 10 vegetables was determined based on data collected in the Belgian food consumption
24
25 11 survey performed by the Scientific Institute for Public Health and data from the 'Belgian
26
27 12 Federal Agency for the Safety of the Food Chain' 2005 monitoring programme. A first
28
29 13 screening of pesticide residue exposure was performed by a deterministic approach.
30
31 14 For most pesticide residues studied, the exposure was hundred times lower than the
32
33 15 ADI ('acceptable daily intake'). However, for a high consumer (97.5th percentile of
34
35 16 consumption) the intake could reach 23% of the ADI for imazalil, 15% for
36
37 17 chlorpropham, 14% for the dithiocarbamates, 10% for dimethoate and lambda-
38
39 18 cyhalothrin, and 9% for chlorpyrifos. Nevertheless, probabilistic exposure
40
41 19 assessment performed on these pesticides in a second phase of the study, indicated
42
43 20 that except for chlorpropham, the probability to exceed the ADI is much lower than
44
45 21 0.1%.
46
47
48
49
50
51
52
53
54
55
56
57

58 26 **Keywords:** *Exposure assessment, probabilistic modelling, pesticides, fruit, vegetables*
59
60

27 Introduction

28
29 Monitoring programmes for pesticide residues in food are carried out by regulatory
30 authorities to control if authorized pesticides are correctly applied to food crops in terms
31 of granted authorizations and registrations (application rates and pre-harvested
32 intervals), and to check if pesticide-treated products comply with fixed MRLs (maximum
33 residue limit). Although monitoring programmes provide a useful tool to verify that
34 consumers are not exposed to unacceptable pesticide residue levels, their outcome is
35 not representative as to the actual exposure to pesticides. The MRL is a product limit
36 and is based on the application of pesticides on crops according to Good Agricultural
37 Practices (GAP) in controlled field experiments, whereas health safety limits or
38 toxicological endpoint values such as the ADI (acceptable daily intake) and the ARfD
39 (acute reference dose) are based on toxicological data and insights. Exposure or intake
40 of a compound below its health safety limit, is considered to be “safe”. As such, the
41 residue concentration may be above the MRL without representing a risk to the
42 consumer. The outcome of a monitoring programme such as detection frequency and
43 number of samples exceeding the MRL, give a good indication on residues and food
44 commodities to prospect, but lack information in terms of food safety. Residue levels
45 need to be combined with consumption data in order to determine if the intake by the
46 consumer exceeds the health-based limits.

47 To assess the exposure or intake, two main approaches can be distinguished: the
48 deterministic and the probabilistic approach. The deterministic approach is based on
49 single point estimates that are used for each variable within the model (such as an
50 average value or the 97.5th percentile), whereas in the probabilistic approach the
51 variables are described in terms of distributions. In this way, all possible values for
52 each variable are taken into account and each possible model outcome is weighted by

1
2
3 53 the probability of its occurrence. Different techniques are available to calculate the
4
5 54 outcome distribution, such as the Monte Carlo simulation (Vose 2006).
6

7 55 Advantages of deterministic modelling are that there is less need for extensive
8
9 56 databases to support the input variables, that default standard assumptions can be
10
11 57 used, that it is relatively easy to carry out, and that the single-risk estimate output is
12
13 58 easy to understand and interpret. Advantages of probabilistic modelling are that all
14
15 59 available data and knowledge are used, that the exposure estimate is presented as a
16
17 60 distribution, with each value having a probability attached to it, and that variability and
18
19 61 uncertainty can be quantified. The point estimate approach, due to its simplicity and its
20
21 62 worldwide use and acceptance, may be used as a first screening tool to identify
22
23 63 possible pesticides that may pose a problem. If so, the probabilistic approach can then
24
25 64 be applied to study if the point estimate outcome really gives reason for concern.
26
27
28

29 65 An overview of current knowledge on intake or exposure assessment in general
30
31 66 and probabilistic modelling in particular (input models, methodologies, stakeholders
32
33 67 involved, etc.) is given amongst others by Vose (2006), Leclercq et al. (2003), Ferrier et
34
35 68 al. (2002), Kroes et al. (2002).
36
37

38 69 The aim of the present study was to evaluate whether the estimated daily intake of
39
40 70 pesticides through fruit and vegetables consumption by the Belgian adult population, is
41
42 71 a cause for health concern, based on the results of the monitoring programme 2005 of
43
44 72 the Belgian Federal Agency for the Safety of the Food Chain (FASFC 2006).
45
46
47
48
49
50

51 **Materials and methods**

52 **Pesticide residue data**

53 77 Contents of pesticides in the different commodities were taken from the pesticide
54
55 78 monitoring programme 2005 of the Belgian Federal Agency for the Safety of the Food
56
57 79 Chain (FASFC 2006), including 1 322 samples of fruit and vegetables (tea and cacao
58
59 80 included). During the year 2005, no less than 134 940 residue/food combinations were
60

1
2
3 81 analysed. From this vast amount of data, only those residue/food combinations were
4
5 82 selected that could be considered as being authorized in June 2005 according to
6
7 83 Fytoweb, a Belgian website supported by the Federal Public Service Health, Food
8
9
10 84 Chain Safety and Environment containing information on authorized pesticides
11
12 85 (<http://www.fytoweb.fgov.be/indexEn.asp>).

13
14 86 The monitoring programme does not provide a full random analysis, but is based on
15
16 87 risk assessment. Several factors were taken into account including the importance of
17
18 88 the food commodity in the diets, exceedings observed in previous years, RASFF
19
20 89 messages (European Rapid Alert System for Food and Feed), pesticides authorized in
21
22 90 Belgium, analytical and budgetary possibilities, etc. Sampling was done by trained
23
24 91 officials according to directive 2002/63/EG at auctions, importers, wholesalers,
25
26 92 processors and exceptionally in retail. Samples were analysed in three officially
27
28 93 recognised laboratories accredited following ISO 17025 (FASFC 2006). Multi-residue
29
30 94 and single-residue methods were used for analysing pesticide residues.

31
32
33 95 With respect to samples exceeding the MRL, it has to be noted that MRL values
34
35 96 are not yet completely harmonised in the EU. As such, imported food can exceed the
36
37 97 Belgian MRL but comply to the MRL of the country of origin.

38
39
40 98

41
42 99 *Food consumption data*

43
44 100

45
46 101 Intake estimates were based on consumption data collected in the Belgian nation-wide
47
48 102 food consumption survey performed in 2004 by the Belgian Institute for Public Health
49
50 103 (IPH 2006). The representative sample of participants included 3 214 people over 15
51
52 104 years old, which were questioned two times about their last-24-hours-consumption.
53
54 105 The selection of interviewed people and the moment of the interview were chosen in
55
56 106 order to obtain a representative consumption profile of the Belgian population over one
57
58 107 year.
59
60

1
2
3 108 Concerning the consumption data recorded by the IPH, a selection had to be made
4
5 109 and certain commodities had to be combined as a group (e.g. consumption data on
6
7 110 Chinese, Savoy, red and white cabbage, and sauerkraut were aggregated into the
8
9
10 111 group “cabbage”) in order to match the matrices analysed in the residue monitoring
11
12 112 campaign of the FASFC.

13
14 113 The total dataset, including zero intakes (“zero consumption days”), was used as
15
16 114 part of producing an “average” diet for long-term consumer exposure assessment since
17
18 115 the main interest of this study is to evaluate the probability that the ADI is exceeded.
19
20 116 When one is however interested in assessing the safety of actually eating a
21
22 117 commodity, it is preferred to use the “consumers only” approach (Pieters et al. 2005;
23
24
25 118 Hamilton et al. 2004).

26
27 119

28
29 120 *Estimation of exposure*

30
31 121 The dietary exposure to pesticides ($\text{mg kg body weight}^{-1} \text{ day}^{-1}$) was calculated based
32
33 122 on consumption data and individual body weights of the IPH diet study, and residue
34
35 123 monitoring data of the Belgian FASFC monitoring programme 2005.

36
37 124 In a first phase, the exposure was calculated deterministically. For a given
38
39 125 residue/food combination, the average residue concentration was multiplied with the
40
41 126 average consumption as well as with the 97.5th percentile of consumption of the whole
42
43 127 population in order to have an idea of the chronic exposure of the population to
44
45 128 pesticide residues.

46
47 129 In a second phase, the exposure was evaluated in more detail by a probabilistic
48
49 130 approach. Hereto, the Monte Carlo technique was applied. This technique involves the
50
51 131 random sampling of each probability distribution within the model to produce hundreds
52
53 132 or even thousands of scenarios (also called iterations or trials). Each probability is
54
55 133 sampled in a manner that reproduces the distribution’s shape. The distribution of the
56
57 134 values calculated for the model outcome therefore reflects the probability of the values
58
59 135 that could occur (Vose 2006).

1
2
3 136 In the probabilistic exposure assessment, Monte Carlo simulations were performed
4
5 137 with 10000 iterations. The model input distributions were randomly sampled by Latin
6
7 138 Hypercube sampling. Calculations were performed by means of the commercially
8
9 139 available software package @Risk[®] (Palisade Corporation, Version 4.5.5, NY, US).

10
11 140 In order to have a good representation of consumption and pesticide residue data,
12
13 141 both parametric and non-parametric approaches were evaluated. Fitting of probability
14
15 142 distributions to the data in the parametric approach was performed using BestFit
16
17 143 (Palisade, NY, US). However, due to the large number of zero consumption days and
18
19 144 of 'non-detects' in the residue concentration data, most of the consumption and
20
21 145 contamination data could not be fitted by one of the distributions, and preference was
22
23 146 given to the non-parametric approach. As such, model inputs (consumption and
24
25 147 pesticide residue concentration data) were described by a discrete, uniform distribution.
26
27 148 In this non-parametric approach the collected data points themselves are considered to
28
29 149 form the distribution function and all possible values have the same probability of
30
31 150 occurrence (Vose 2006).

32
33 151 Processing factors (peeling, cooking, boiling, ...) and variability factors were not
34
35 152 included in the calculations and no distinction was made between imported
36
37 153 commodities and commodities of Belgian origin.

38
39 154 The exposure levels were compared with the ADI. The dithiocarbamates were
40
41 155 considered as one group because testing methods do not differentiate between the
42
43 156 different dithiocarbamates. Pesticide sales of the dithiocarbamates in Belgium
44
45 157 concerns mainly mancozeb and maneb. Therefore, the ADI of mancozeb and maneb
46
47 158 was chosen as the reference. With respect to the benomyl group, residues included are
48
49 159 benomyl, thiofanate-methyl and carbendazim. Since benomyl is no longer authorized in
50
51 160 Belgium and carbendazim is the common metabolite of thiofanate-methyl, the
52
53 161 reference ADI chosen for the benomyl group was the one of carbendazim.

54
55
56
57
58
59
60 162

163 *Levels below the limit of quantification (LOQ)*

1
2
3 164 In pesticide exposure assessment, the estimated distribution typically consists of many
4
5 165 measurements below the minimum pesticide residue concentration reported (limit of
6
7 166 quantification or LOQ). Such samples do not necessarily indicate that pesticide
8
9
10 167 residues are not present in the sample, but only that the analytical method could not
11
12 168 detect the possible amount of pesticide present. Therefore, samples with residue levels
13
14 169 below the LOQ (“non-detects”) were assigned either 0, $\frac{1}{2}$ LOQ or LOQ, corresponding
15
16 170 to a lower, middle and upper bound (worst case) scenario.
17

18 171

19 172 *Uncertainty of exposure estimates*

20
21
22 173 The probabilistic method in itself generates estimates of variability in the form of an
23
24 174 exposure distribution (1-dimensional Monte Carlo model). To estimate the uncertainty
25
26 175 of the exposure estimate, it is necessary to apply other methods, such as bootstrap
27
28 176 sampling (2-dimensional Monte Carlo model). Bootstrap sampling is a method to
29
30 177 assess the reliability of percentiles of exposure and is a means to study the stability of
31
32 178 the tail of the exposure distribution. With this method a sample of n observations (food
33
34 179 consumption levels, residue levels) is resampled from the original database to obtain a
35
36 180 bootstrap sample of n observations. Sampling is performed with replacement, so that
37
38 181 every observation can occur more than once in the bootstrap sample. By repeating this
39
40 182 process e.g. 500 times, 500 bootstrap samples are obtained, which may be considered
41
42 183 as alternative data sets that might have been obtained during sampling from the
43
44 184 population of interest. Each statistic that can be calculated from the original data set
45
46 185 (e.g. 97.5th, 99.9th percentile), can also be calculated from each bootstrap sample. This
47
48 186 will generate a bootstrap distribution of 500 97.5th, 99.9th percentiles, etc. The bootstrap
49
50 187 distribution now characterises the uncertainty due to sampling uncertainty of the
51
52 188 original data set (Boon et al. 2004).
53
54
55
56
57
58
59
60

1
2
3 190 **Results**
4

5 191 *Deterministic exposure assessment*
6

7 192 As a first screening, the exposure to pesticide residues was calculated by a
8
9 193 deterministic approach on a total of 25 residues. These residues were selected out of
10
11 194 the 200 residues analysed by the FASFC in 2005 based on their frequency of detection
12
13 195 (> 2% of analysed samples exceeded the limit of quantification, LOQ). The residue
14
15 196 concentration in the food commodities was assumed to be equal to the average
16
17 197 concentration in order to take the variation of the concentrations into account. A rough
18
19 198 estimate of the total exposure to a given pesticide residue X was obtained by summing
20
21 199 exposures from all residue X / food combinations considered. The total exposure was
22
23 200 compared with the ADI and expressed in terms of % ADI. Results for the lower, middle
24
25 201 and upper bound scenario for samples with a residue concentration below the LOQ are
26
27 202 given in Table I, together with their detection frequency and the ADI. Figure 1 presents
28
29 203 an overview of the exposure for the average and the 97.5th percentile (P97.5) of
30
31 204 consumption according to a scenario where the residue concentration of non-detected
32
33 205 samples is equal to half the LOQ of the analytical method used (middle bound
34
35 206 scenario).
36
37
38
39
40
41

42

43 [Insert Table I and Figure 1 about here]

44
45

46 210 *Probabilistic exposure assessment*
47

48 211 Table II presents the exposure values based on a probabilistic assessment approach
49
50 212 for chlorpropham, imazalil, dithiocarbamates, dimethoate, lambda-cyhalothrin and
51
52 213 chlorpyrifos. Figure 2 gives more specific information on the contribution of several
53
54 214 food items to the exposure of a given pesticide residue, and this for the middle bound
55
56 215 scenario.
57
58

59
60

216
217 [Insert Table II and Figure 2 about here]

1
2
3 218 **Discussion**

4
5 219 *Deterministic analysis*

6
7 220 Based on the P97.5 of consumption, relatively high exposure values were observed for
8
9 221 chlorpropham, imazalil, dithiocarbamates, dimethoate, lambda-cyhalothrin and
10
11 222 chlorpyrifos in the deterministic analysis (Table I, Figure 1). Chlorpropham is a
12
13 223 selective systemic herbicide and plant growth regulator belonging to the N-
14
15 224 phenylcarbamate group of pesticides. It is mainly applied as sprouting inhibitor in
16
17 225 potato storage. Imazalil is a systemic fungicide used to control a wide range of fungi on
18
19 226 fruit, vegetables, and ornamentals. It is also used as a seed dressing and for
20
21 227 postharvest treatment of citrus, banana, and other fruit to control storage decay.
22
23 228 Dithiocarbamates are widely used antisporeulant (contact) fungicides that are very often
24
25 229 associated with other fungicides. Lambda-cyhalothrin is a pyrethroid insecticide.
26
27 230 Dimethoate and chlorpyrifos are organophosphate insecticides. These pesticides
28
29 231 were selected for further probabilistic analysis.
30
31
32

33 232

34
35 233 *Probabilistic analysis*

36
37 234 As is illustrated in Figure 2, the exposure distributions are extremely right-skewed.
38
39 235 Except for chlorpyrifos and lambda-cyhalothrin, the exposure estimates at the P99.99
40
41 236 are approximately 2 to 3 times higher than the exposure at the P99.9, which is 3 (to 10)
42
43 237 times higher than the exposure at P97.5. Because both food consumption and residue
44
45 238 monitoring distributions are right-skewed, the resulting product is also a right-skewed
46
47 239 distribution. It can be questioned if the consumption data set and the monitoring data
48
49 240 set are robust enough to support exposure values calculated at the P99.9 and higher.
50
51 241 These values can be more sensitive to uncertainties in data collection (sample size,
52
53 242 reporting mistakes such as over reporting, analytical uncertainties) making these
54
55 243 estimations of exposure less reliable. This is illustrated in Table III, where the 95 %
56
57 244 confidence intervals of the higher percentiles of exposure for some residue/food
58
59 245 combinations are given, including chlorpropham on potatoes, imazalil on oranges and
60

1
2
3 246 mandarin, and dimethoate on cherries and lettuce. These 95% confidence intervals
4
5 247 were calculated by the bootstrap sampling method.
6

7 248

8
9
10 249 [Insert Table III about here]
11

12 250

13
14 251 *“Non-detects”*
15

16 252 The detection frequency or the number of samples with a residue concentration below
17
18 253 the LOQ is rather high. As can be observed from Table I, treatment of so-called left
19
20 254 censored data can have a large effect on the deterministically obtained outcome. For
21
22 255 example, considering the results based on the P97.5 consumption, the intake of
23
24 256 lambda-cyhalothrin or of the dithiocarbamates exceeds the intake of chlorpropham for
25
26 257 the upper bound scenario. For the lower and middle bound scenarios on the other
27
28 258 hand, the intake of lambda-cyhalothrin and of the dithiocarbamates was (much) lower
29
30 259 than of chlorpropham. Generally, using the middle bound scenario (LOQ/2) seems to
31
32 260 be a good compromise. Nevertheless, it is clear that a proper treatment of censored
33
34 261 data cannot be underestimated. Guidance on how to deal with so-called left censored
35
36 262 data, is given by the U.S. Environmental Protection Agency (US EPA 2000).
37
38
39

40 263 With respect to the probabilistically obtained results, replacing non-detects by 0, the
41
42 264 LOQ or the LOQ/2 does not affect the outcome presented in Table III significantly. The
43
44 265 large difference observed in Table II between the highest percentiles of exposure of the
45
46 266 lower, middle and upper bound scenario for some pesticide, is due to a broad
47
48 267 confidence interval. The higher the percentile under consideration, the broader the
49
50 268 confidence interval. In Table II, only one value out of the broad band of values was
51
52 269 considered. (The exercise of bootstrap sampling was not performed for the complete
53
54 270 data set of pesticide residues and commodities due to the huge amount of
55
56 271 computational capacity required.)
57
58
59
60

1
2
3 272 Ferrier et al. (2006) evaluated the effect of uncertainty on the exposure estimate
4
5 273 using scenarios reflecting different assumptions related to sources of uncertainty. The
6
7 274 most influential uncertainty issue was the distribution type used to present input
8
9
10 275 variables. Other sources that most affected the model output were non-detects, unit-to-
11
12 276 unit variability and processing.

13
14 277

15
16 278 *Comparison of the deterministic and probabilistic analysis*

17
18 279 Comparison between the deterministic and probabilistic approach has to be done with
19
20 280 care. Historically, the P97.5 has been chosen as the upper percentile for deterministic
21
22 281 acute intake assessments, i.e. the P97.5 of consumers for the specific food and the
23
24
25 282 P97.5 residue level, which in combination amounts to the P99.94 for residue intake
26
27 283 $(100 \times (1 - ((1 - 0.975) \times (1 - 0.975))))$ (Hamilton et al. 2004).

28
29 284 In the present study, the deterministic and probabilistic P97.5 exposure values are
30
31 285 not comparable since they are essentially based on different notions. The
32
33 286 deterministically obtained exposure values are based on percentiles of consumption,
34
35 287 whereas the probabilistically determined results are actual percentiles of exposure. In
36
37 288 contrast, the average exposure values are very similar.

38
39
40 289 Additionally, it has to be remarked that in the deterministic approach, total exposure
41
42 290 for a given residue was obtained by summing exposures from all residue/food
43
44 291 combinations, whereas deterministic intake assessments should deal with only one
45
46 292 food at a time. Generally, summing point estimates across foods leads to an estimate
47
48 293 of food consumption that is extremely high and unrealistic. Because of the inherent
49
50 294 conservatism in the point estimate, summing these estimates over commodities would
51
52 295 be equivalent to assuming that large portions of all commodities (P97.5 of a
53
54 296 consumption distribution) are consumed on 1 day or frequently by one individual and
55
56 297 that all these commodities are contaminated. For a realistic estimate of short-term
57
58 298 dietary exposure through multiple foods, a probabilistic approach is recommended
59
60 299 (Hamilton et al. 2004).

1
2
3 300 Nevertheless, when calculating the total exposure by the probabilistic approach,
4
5 301 potential correlations between daily consumption rates of different food items need to
6
7 302 be taken into account. As a pragmatic approach, recorded days were considered as
8
9 303 such, without discriminating between variation among individuals and variation among
10
11 304 days (within individuals), resulting in a distribution of “person-days”. The drawback of
12
13 305 this approach is that it cannot be determined if few persons are at risk but each of them
14
15 306 at many days, or that many persons are at risk but each of them only rarely (Pieters et
16
17 307 al. 2005). Since the focus of the present study concerns mainly the intake of frequently
18
19 308 exposed individuals, the intake was compared to the ADI rather than to the ARfD.
20
21
22
23

309

310 *Upper percentiles of exposure*

24
25
26
27 311 Since in exposure assessment of pesticides, the regulatory threshold risk is at the
28
29 312 upper tail of the exposure distribution (P99 and higher), also the P99.9 and P99.99
30
31 313 values of exposure are presented in Table II. From this table, it can be concluded that
32
33 314 except for chlorpropham, the probability that a person exceeds the ADI by eating one
34
35 315 or more servings of contaminated a food item is less than 0.1%. The relatively high
36
37 316 exposure to chlorpopham is solely due to 1 crop, namely potatoes (Figure 2a).

38
39
40 317 Next to chlorpropham, a relatively high intake in terms of % ADI is also observed for
41
42 318 dimethoate and to a lesser extent, for imazalil. This high exposure value for dimethoate
43
44 319 is mainly due to a high or frequent consumption of cherries and lettuce (Figure 2c).
45
46 320 With respect to imazalil, citrus fruit, orange and mandarin in particular, contributed most
47
48 321 to the intake (Figure 2b).

49
50
51 322 Different profiles can be distinguished between the high exposure levels. Exposure
52
53 323 to imazalil is relatively high since the amount of commodities on which it was detected
54
55 324 and its use is authorized, is high. For chlorpropham on the other hand, the high
56
57 325 exposure was due to some high levels of pesticide residue detected on a single food
58
59 326 item, potatoes, and the relatively high consumption of that food item.
60

1
2
3 327 The effect of consumption data on the outcome can be illustrated by Table III. For
4
5 328 example, the P97.5 exposure to dimethoate by the consumption of cherries is zero and
6
7 329 the P99.99 exposure values range from zero to values higher than the ADI, although a
8
9 330 detection frequency of 84.6% was observed. Cherries are food items that are eaten
10
11 331 occasionally. Their consumption is seasonal resulting in extreme right-censored
12
13 332 consumption data (1.2% non-zero consumption days). Due to the high detection
14
15 333 frequency of dimethoate on cherries, exceeding of the ADI may occur for a minority of
16
17 334 consumers (P99.99), although rarely. From this example, it is also clear that caution
18
19 335 should be taken when interpreting high exposure percentiles without knowledge of the
20
21 336 confidence interval. Performing a 1-dimensional Monte Carlo simulation could result in
22
23 337 extreme values of a confidence interval leading to unnecessary concern.
24
25
26
27
28

29 339 *Comparison with other studies*

30
31 340 Different probabilistic exposure assessment studies of pesticides from food are
32
33 341 reported in literature, but are still scarce (Brüschweiler 2004; Boon et al. 2003; López et
34
35 342 al. 2003; Wright et al. 2002; Hamey 2000). When comparing exposure assessments in
36
37 343 literature, some caution has to be taken. One difficulty is that studies are performed
38
39 344 using different (national) food consumption databases. The way in which food
40
41 345 consumption data of different countries has been and is collected, is not harmonised
42
43 346 and can vary considerably. This diversity is related to the population addressed (e.g.
44
45 347 children included or not), the method of data collection (24-h recall, dietary method),
46
47 348 duration of the survey, number of respondents involved, coding of food consumption
48
49 349 data and method of quantifying amount consumed (actual weighing vs. estimations on
50
51 350 the basis of portion sizes) (Kroes et al. 2002). These variables should be kept in mind
52
53 351 when comparing exposure assessments using different databases.
54
55
56

57 352 Another critical aspect is the sampling of commodities for pesticide residue
58
59 353 analysis. This is not always according to an ad random procedure, but often based on
60

1
2
3 354 a risk approach (directed or targeted sampling). The use of these data to estimate
4
5 355 dietary exposure may thus lead to an overestimation of exposure.
6

7
8 356 In order to be able to compare exposure assessment results internationally,
9
10 357 standardized ways of collecting and reporting data is needed.
11

12 358

13
14 359 The exposure estimates presented here for chlorpyrifos, compare favourably to
15
16 360 estimates published in literature. The US FDA's total Diet studies from 1989 and 1990
17
18 361 estimated chlorpyrifos exposure at 0.041×10^{-4} mg/kg bw/day for children aged 14 to
19
20 362 16 years (FDA 1991; FDA 1990). This estimate is similar to the average value given for
21
22 363 the lower bound scenario in Table II (0.045×10^{-4} mg/kg bw/day). Based on monitoring
23
24 364 programmes, Wright et al. (2002) estimated a P99.9 exposure of 4.8×10^{-4} mg/kg
25
26 365 bw/day for the US population, and 9.4×10^{-4} mg/kg bw/day for children between 1 and
27
28 366 6 years old, which is of the same order of magnitude as calculated for the higher
29
30 367 percentiles of exposure in Table II.
31
32

33
34 368 In a Dutch study where a probabilistic model of dietary exposure of infants to
35
36 369 pesticides was compared with a duplicate diet study, the 95 % confidence interval for
37
38 370 the P99 of exposure to chlorpyrifos was $0.2-4.3 \times 10^{-4}$ mg/kg bw/day for the duplicate
39
40 371 diet study assuming a log-normal exposure distribution and $24.5-43.3 \times 10^{-4}$ mg/kg
41
42 372 bw/day for the probabilistic model in which variability and processing was taken into
43
44 373 account an non-detects were replaced with the limit of reporting or LOR (Boon et al.
45
46 374 2003). The latter value exceeds even the worst case (non-detects equal to LOQ)
47
48 375 P99.99 value estimated in this study (14.2×10^{-4} mg/kg bw/day). This can be amongst
49
50 376 others due to the fact that in the Dutch study toddlers aged 8 to 12 months were
51
52 377 considered and the sampling strategy was designed to include as many as children as
53
54 378 possible that were fed home-made meals of fruits and vegetables.
55
56

57
58 379 In a Swiss study, the probabilistic intake to chlorpropham was assessed for potato
59
60 380 dishes and raw potatoes (Brüschweiler 2004). Applying processing factors, the mean
381
381 381 daily intake was 0.0056 mg per person and the P99 0.15 mg per person corresponding

1
2
3 382 to 0.93×10^{-4} and 0.0025 mg/kg bw/day respectively for a person weighing 60 kg. Both
4
5 383 results are lower than observed in this study, where the average intake of
6
7 384 chlorpropham by uncooked and unpeeled potatoes was calculated as 0.0021 mg/kg
8
9
10 385 bw/day. As remarked by the author, calculated exposures depend strongly on the
11
12 386 processing factors applied, which differ markedly in literature.
13

14 387

15
16 388 *Risk assessment*

17
18 389 As already indicated in the introduction, detection frequency and the number of
19
20 390 samples for which the MRL is exceeded, give an indication on residues and food
21
22 391 commodities to prospect, but lack information in terms of food safety. This can be
23
24 392 illustrated by comparing e.g. chlorpropham with imazalil and the dithiocarbamates. The
25
26 393 detection frequency of chlorpropham in potatoes was lower than the one for imazalil in
27
28 394 mandarin or oranges (69% compared to 87 and 100% respectively) (Table III).
29
30 395 Nevertheless, exposure values for imazalil were lower. In 2005 and with respect to the
31
32 396 considered food items, two MRL exceedings were reported for chlorpropham on
33
34 397 potatoes (non-harmonised, national MRL of 5 mg/kg), whereas 6 MRL exceedings
35
36 398 were reported for dithiocarbamates including 1 for cress, 3 for lettuce, 1 for peas and 1
37
38 399 for radishes (EC MRLs of 0.3, 3-5, 0.1 and 0.05 mg/kg respectively) (FASFC 2006). In
39
40 400 terms of exposure, chlorpropham is ranked higher than the dithiocarbamates.
41
42

43
44 401 Based on results, it can be concluded that chronic intakes of 25 selected pesticide
45
46 402 residues are rather low compared to the ADI (mostly < 1 % of ADI) and that the public
47
48 403 safety in Belgium seems to be generally under control in terms of (single) pesticide
49
50 404 intakes through consumption of fruit and vegetables. However, some residues such as
51
52 405 chlorpropham, imazalil, dithiocarbamates, dimethoate, lambda-cyhalothrin and
53
54 406 chlorpyrifos, needed to be considered more closely since for a high consumer
55
56 407 (P97.5) the intake can reach 23% of the ADI for imazalil, 15% for chlorpropham, 14%
57
58 408 for the dithiocarbamates and 10% for lambda-cyhalothrin and dimethoate, and 9% for
59
60 409 chlorpyrifos for the middle bound scenario. Nevertheless, probabilistic analysis

1
2
3 410 indicated that except for chlorpropham, the probability to exceed the ADI will be much
4
5 411 lower than 0.1% when exposed to one of the selected pesticides.
6

7
8 412 Considering acute exposure to pesticide residues, interpretation of exposure levels
9
10 413 determined in this study in terms of % of the ARfD will indicate the absence of acute
11
12 414 risks, even for high consumers, given that ADI values for pesticides are lower than
13
14 415 ARfD values (Table II).
15

16 416
17
18 417 It seems that the exposure to the pesticide residues considered is mainly due to the
19
20 418 consumption of only one or two food items (e.g. potatoes in case of chlorpropham,
21
22 419 citrus fruit in case of imazalil). With regard to fruit and vegetables, attention should also
23
24
25 420 be given to chemical contaminants other than pesticides such as natural (e.g.
26
27 421 mycotoxins), environmental (e.g. cadmium, PCBs) or process contaminants (e.g. 3-
28
29 422 MCPD, acrylamide). As such, e.g. exposure to chlorpropham (and other sprouting
30
31 423 inhibitors) on potatoes could be linked to the acrylamide issue. It has been shown that
32
33 424 more acrylamide is formed during cooking of potatoes that have been stored at cold
34
35 425 temperature (Claeys et al. 2005). If less potatoes are stored in cold-storage depots, it is
36
37 426 not inconceivable that the application of sprouting inhibitors like chlorpropham
38
39 427 increases.
40
41

42 428 It has to be noted that the present study focused on fruits and vegetables only.
43
44 429 Therefore, it is an underestimation of the total exposure of pesticides studied. On the
45
46 430 other hand, processing factors were ignored, whereas fruit and vegetables are often
47
48 431 peeled, cooked or boiled before consumption, resulting in an overestimation of the
49
50 432 actual exposure to pesticide residues. Other variables affecting the pesticide residue
51
52 433 concentration include storage, transport, shelf life, use patterns, lab-to-lab variation and
53
54 434 analytical methods used to measure chemicals (Kroes et al. 2002). Additionally, it has
55
56 435 to be noted that the dietary consumption data provided by the IPH did not include data
57
58 436 for children under 15 years old. A special attention to this sensitive group could be
59
60 437 given in further research.

1
2
3 438 Further research could also deal with exposure assessment related to the presence
4
5 439 of multiple residues on a single food or serving, i.e. cumulative dietary intake
6
7 440 assessment. Cumulative exposure to various residues of pesticides in food is a
8
9 441 potential area of concern and gaining more and more interest. This issue is especially
10
11 442 relevant for pesticides with a common mechanism of toxicity (e.g. organophosphates,
12
13 443 carbamates) (EFSA 2007; van Raaij et al. 2005; Jensen et al. 2003).
14
15
16 444

17
18 445 Finally, it has to be noted that compared to point estimates, making use of
19
20 446 probabilities and revealing the extreme upper tail of the exposure distribution may
21
22 447 create difficulties for both consumers and regulators in terms of their understanding of
23
24 448 the assessment. However, a full probability distribution also enables more informed
25
26 449 decisions to be made for policy and regulation. Regulation based on unrealistic
27
28 450 exposure estimates could lead to unnecessary restrictions on products, or potentially
29
30 451 put consumers at risk. It is therefore important to understand the influence that sources
31
32 452 of uncertainty exert on predictions of exposure and to include this knowledge in the
33
34 453 decision making process (Ferrier et al. 2006).
35
36
37 454

38 39 40 455 **Conclusion**

41
42 456 In 2005, the FASFC controlled 1,322 samples of fruit and vegetables. Pesticide
43
44 457 residues were found in 56% of the samples and standards were exceeded for 7.9% of
45
46 458 the samples (FASFC 2006). These numbers could give unnecessary rise to
47
48 459 consumers' concern. A more nuanced and different picture is obtained when the actual
49
50 460 exposure of consumers to pesticide residues is considered. Based on results of the
51
52 461 present study, chronic exposure to pesticide residues due to the consumption of fruit
53
54 462 and vegetables seems to be under control, even for frequent consumers.
55
56
57
58
59
60 464

1
2
3 465 **Acknowledgement**
4

5 466 Part of this work has been financed in the framework of the Belgian Federal
6
7 467 Programme on Reduction of Pesticides and Biocides. Prof. W. Steurbaut (UGent), Prof.
8
9
10 468 B. Schiffers (FUSAGX, Gembloux) and dr. P. Delahaut (CER, Marloie) are
11
12 469 acknowledged for their guidance of this study.
13

14 470

15
16 471 **References**
17

- 18 472 Boon P, Lignell S, van Klaveren J, Tjoe Nij E. 2004. Estimation of the acute dietary exposure to
19
20 473 pesticides using the probabilistic approach and the point estimate methodology. Rikilt
21
22 474 Report 2004.008. Available: [http://www.rikilt.wur.nl/NR/rdonlyres/BDEEDD31-F58C-](http://www.rikilt.wur.nl/NR/rdonlyres/BDEEDD31-F58C-47EB-A0AA-23CB9956CE18/10727/R2004008.pdf)
23
24 475 [47EB-A0AA-23CB9956CE18/10727/R2004008.pdf](http://www.rikilt.wur.nl/NR/rdonlyres/BDEEDD31-F58C-47EB-A0AA-23CB9956CE18/10727/R2004008.pdf) via the INTERNET. Accessed 2007
25
26 476 Sep 10.
27
28 477 Boon P, van der Voet H, van Klaveren J. 2003. Validation of a probabilistic model of dietary
29
30 478 exposure to selected pesticides in Dutch infants. Food Additives and Contaminants 20
31
32 479 (Suppl. 1): S36-S49.
33
34 480 Brüscheweiler B. 2004. Probabilistic exposure assessment for chlorpropham through potato
35
36 481 consumption. Mitteilungen aus Lebensmitteluntersuchung und Hygiene 95(5): 421-436.
37
38 482 Claeys W, De Vleeschouwer K, Hendrickx M. 2005. Quantifying the formation of carcinogens
39
40 483 during food processing: acrylamide. Trends in Food Science and Technology 16(5): 181-
41
42 484 193.
43
44 485 EFSA. 2007. Cumulative risk assessment of pesticides to human health: the way forward. EFSA
45
46 486 Scientific Colloquium 7. Summary Report. Available:
47
48 487 [http://www.efsa.europa.eu/EFSA/ScientificOpinionPublicationReport/EFSA_Scientific_Collo](http://www.efsa.europa.eu/EFSA/ScientificOpinionPublicationReport/EFSA_Scientific_Colloquium_Reports/efsa_locale-1178620753812_RiskAssessmentPesticidesHumanHealth.htm)
49
50 488 [quiumReports/efsa_locale-1178620753812_RiskAssessmentPesticidesHumanHealth.htm](http://www.efsa.europa.eu/EFSA/ScientificOpinionPublicationReport/EFSA_Scientific_Colloquium_Reports/efsa_locale-1178620753812_RiskAssessmentPesticidesHumanHealth.htm)
51
52 489 via the INTERNET. Accessed 2007 Nov 3.
53
54 490 FASFC. 2006. Report of Monitoring results concerning Directives 90/642/EEC, 76/895/EEC and
55
56 491 Commission Recommendation 2005/178/EC.
57
58 492 FDA. 1991. Residues in food – 1990. Journal - Association of Official Analytical Chemists 63:
59
60 493 121a-141a.

- 1
2
3 494 FDA. 1990. Residues in food – 1989. Journal - Association of Official Analytical Chemists 62:
4
5 495 127a-145a.
6
7 496 Ferrier H, Shaw G, Nieuwenhuijsen M, Boobis A, Elliott P. 2006. Assessment of uncertainty in a
8
9 497 probabilistic model of consumer exposure to pesticide residues in food. Food Additives
10
11 498 and Contaminants 23(6): 601-615.
12
13 499 Ferrier H, Nieuwenhuijsen M, Boobis A, Elliott P. 2002 Current knowledge and recent
14
15 500 developments in consumer exposure assessment of pesticides: a UK perspective. Food
16
17 501 Additives and Contaminants 19(9): 837-852.
18
19 502 Hamey P. 2000. A practical application of probabilistic modelling in assessment of dietary
20
21 503 exposure of fruit consumers to pesticide residues. Food Additives and Contaminants
22
23 504 17(7): 601-610.
24
25 505 Hamilton D, Ambrus A, Dieterle R, Felsot A, Harris C, Petersen B, Racke K, Wong S-S,
26
27 506 Gonzalez R, Tanaka K, Earl M, Roberts G, Bhula R. 2004. Pesticide residues in food –
28
29 507 acute dietary exposure. Pest Management Science 60(4): 311-339.
30
31 508 IPH. 2006. De Belgische Voedselconsumptiepeiling 1 – 2004. Devriese S, Huybrechts I, Moreau
32
33 509 M, Van Oyen H. Afdeling Epidemiologie, 2006; Brussel Wetenschappelijk Instituut
34
35 510 Volksgezondheid, Depotnummer : D/2006/2505/17, IPH/EPI REPORTS N° 2006 – 016.
36
37 511 Available: <http://www.iph.fgov.be/epidemio/epien/index5.htm> via the INTERNET.
38
39 512 Accessed 2007 Sep 10.
40
41 513 Jensen A, Petersen A, Granby K. 2003. Cumulative risk assessment of the intake of
42
43 514 organophosphorus and carbamate pesticides in the Danish diet. Food Additives and
44
45 515 Contaminants 20(8): 776-785.
46
47 516 Kroes R, Müller D, Lambe J, Löwik M, van Klaveren J, Kleiner J, Massey R, Mayer S, Urieta I,
48
49 517 Verger P, Visconti A. 2002. Assessment of intake from the diet. Food and Chemical
50
51 518 Toxicology 40: 327-385.
52
53 519 Leclercq C, Arcella D, Armentia A, Boon P, Kruizinga A, Gilsenan M, Thompson R. 2003.
54
55 520 Development of databases for use in validation studies of probabilistic models of dietary
56
57 521 exposure to food chemicals and nutrients. Food Additives and Contaminants 20(Suppl. 1):
58
59 522 S27-S35.
60

- 1
2
3 523 López A, Rueda C, Armentia A, Rodríguez M, Cuervo L, Ocio J. 2003. Validation and sensitivity
4
5 524 analysis of a probabilistic model for dietary exposure assessment to pesticide residues
6
7 525 with a Basque Country duplicate diet study. Food Additives & Contaminants 20(Suppl. 1):
8
9 526 S87-S101.
- 10
11 527 Pieters M, Ossendorp B, Slob W. 2005. Probabilistic modelling of dietary intake of substances.
12
13 528 The risk management question governs the method. RIVM report 320011001/2005.
14
15 529 Available: <http://rivm.openrepository.com/rivm/bitstream/10029/7301/1/320011001.pdf> via
16
17 530 the INTERNET. Accessed 2007 Sep 10.
- 18
19 531 US EPA. 2000. Assigning values to non-detected/non-quantified pesticide residues in human
20
21 532 health food exposure assessments. Available:
22
23 533 <http://www.epa.gov/pesticides/trac/science/trac3b012.pdf> via the INTERNET. Accessed
24
25 534 2007 Sep 10.
- 26
27 535 van Raaij M, Ossendorp B, Slob W, Pieters M. 2005. Cumulative exposure to cholinesterase
28
29 536 inhibiting compounds: a review of the current issues and implications for policy RIVM
30
31 537 report 320108001/2005. Available:
32
33 538 <http://www.rivm.nl/bibliotheek/rapporten/320108001.pdf> via the INTERNET. Accessed
34
35 539 2007 Sep 10.
- 36
37 540 Vose D. 2006. Risk Analysis – A quantitative guide. Second edition. John Wiley & Sons, Ltd,
38
39 541 West Sussex, England.
- 40
41 542 Wright J, Shaw M, Keeler L. 2002. Refinements in acute dietary exposure assessments for
42
43 543 chlorpyrifos. Journal of Agricultural and Food Chemistry 50: 235-241.
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

Table I. Total exposure in terms of % ADI of selected pesticide residues based on a deterministic exposure assessment approach, based on average residue concentration and an average and 97.5th percentile of consumption.

	ADI (mg kg ⁻¹ bw day ⁻¹)	Detection frequency (%) ¹	Average consumption			P 97.5 consumption		
			0	LOQ/2	LOQ	0	LOQ/2	LOQ
Azoxystrobin	0.1	2.0 (1020)	0.0	0.1	0.2	0.0	1.3	2.6
Benomyl group	0.03	18.9 (328)	0.2	0.2	0.2	1.3	1.7	2.1
Boscalid	0.04	11.7 (213)	0.1	0.1	0.1	1.4	1.6	1.7
Bromide ion	1.0	16.2 (376)	0.1	0.3	0.5	1.0	3.0	5.0
Chloromequat	0.05	5.0 (20)	0.0	0.0	0.0	0.0	0.2	0.3
Chlorpropham	0.05	41.4 (104)	4.3	4.4	4.4	14.5	14.8	15.1
Chlorpyrifos	0.01	3.3 (509)	0.1	0.9	1.7	0.9	8.5	16.2
Cyprodinil	0.03	10.3 (367)	0.0	0.0	0.1	0.3	0.6	0.9
Dimethoate	0.001	9.6 (197)	0.3	0.5	0.6	2.4	9.9	17.5
Dithiocarbamates	0.05	16.3 (857)	0.3	1.8	3.4	2.7	14.2	25.6
Ethephon	0.03	14.3 (28)	0.1	0.3	0.4	0.9	1.9	2.9
Imazalil	0.025	26.0 (323)	1.7	2.1	2.5	20.0	22.6	25.1
Imidacloprid	0.06	2.1 (47)	0.0	0.0	0.0	0.0	0.0	0.1
Iprodione	0.06	15.8 (865)	0.2	0.4	0.6	1.8	3.8	5.7
Lambda-cyhalothrin	0.005	1.6 (862)	0.0	1.0	2.1	0.1	10.1	20.2
Linuron	0.003	8.6 (70)	0.3	0.3	0.3	0.0	0.6	1.1
Methomyl	0.0025	9.0 (299)	0.2	0.6	1.0	2.1	5.4	8.7
Oxadixyl	0.125	2.5 (245)	0.0	0.0	0.0	0.0	0.2	0.3
Pirimicarb	0.035	3.7 (646)	0.0	0.1	0.1	0.1	1.3	2.5
Procymidone	0.025	8.9 (651)	0.1	0.5	0.5	2.1	6.3	10.6
Propamocarb	0.29	14.5 (379)	0.0	0.1	0.1	0.1	0.8	1.4
Tebuconazole	0.03	17.0 (47)	0.0	0.0	0.0	0.1	0.3	0.4
Thiabendazole	0.1	17.5 (331)	0.2	0.3	0.3	2.8	3.0	3.2
Tolclophos-methyl	0.064	16. (238)	0.0	0.0	0.0	0.0	0.1	0.1
Tolyfluanid	0.1	17.4 (493)	0.0	0.1	0.1	0.5	0.7	0.9

¹ total number of samples is indicated between brackets. Numbers are based on selected food items in this study and not on the total of samples reported in the monitoring programme 2005 (FASFC 2006).

Table II. Total exposure in terms of % ADI of selected pesticide residues based on a probabilistic exposure assessment approach.

	ADI (mg kg ⁻¹ bw day ⁻¹)	ARfD (mg kg ⁻¹ bw day ⁻¹)	Detection frequency (%) ¹	Average			P97.5			P99.9			P99.99		
				0	LOQ/2	LOQ	0	LOQ/2	LOQ	0	LOQ/2	LOQ	0	LOQ/2	LOQ
Chlorpropham	0.05	0.5	41.3 (104)	4.26	4.26	4.49	34.65	34.40	36.20	119.61	133.41	153.35	223.9	187.81	281.07
Imazalil	0.025	0.05	26 (323)	1.65	2.14	2.58	15.99	16.82	17.36	54.19	52.35	59.55	92.40	117.89	78.42
Dimethoate	0.001	0.01	9.6 (197)	0.25	0.44	0.63	0.00	1.63	3.25	55.92	56.61	63.70	125.52	173.04	150.20
Dithiocarbamates	0.05	0.2-0.6 ²	16.4 (861)	0.26	1.83	3.41	2.22	5.27	9.55	11.66	11.74	18.68	31.77	14.91	30.49
Chlorpyrifos	0.01	0.1	5.3 (509)	0.05	0.79	1.51	0.68	3.24	5.96	3.62	6.54	10.62	5.84	7.87	14.19
Lambda- cyhalothrin	0.005	0.0075	1.9 (855)	0.01	1.04	2.05	0.00	4.45	8.87	1.63	8.93	17.04	7.50	10.12	21.30

¹ total number of samples is indicated between brackets

² ARfD of Maneb and Mancozeb respectively

Table III. Consumer's exposure in % ADI [95% confidence interval] for different residue-food commodities (probabilistic approach).

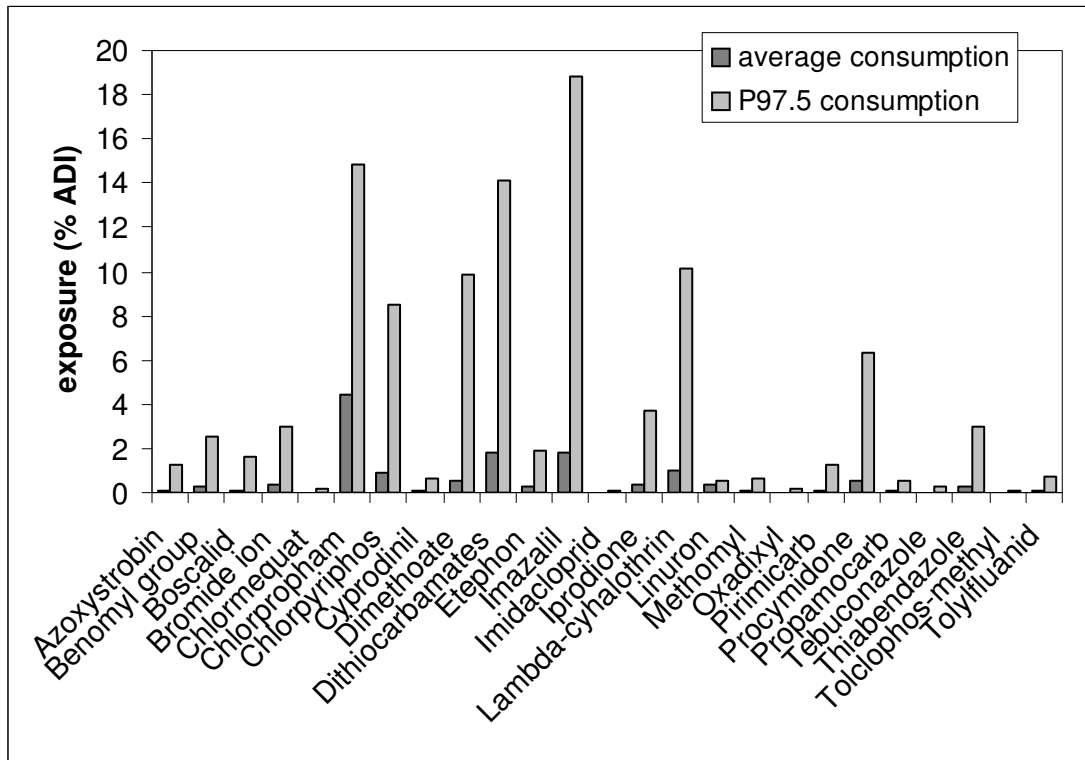
	Detection frequency (%) ¹	% of non-zero consumption days	P97.5			P99.9			P99.99		
			0	LOQ/2	LOQ	0	LOQ/2	LOQ	0	LOQ/2	LOQ
Chlorpropham											
Potatoes	69.4 (62)	67.0	31.5 [16.1-61.4]	32.9 [15.8-64.5]	32.9 [15.9-65.2]	111.5 [39.9-207.0]	108.3 [37.4-209.7]	110.8 [41.5-223.7]	121.5 [44.6-250.3]	119.9 [41.5-243.6]	120.9 [44.1-279.2]
Imazalil											
Oranges	87.1 (31)	9.6	6.3 [2.4-10.8]	6.3 [2.5-10.8]	6.2 [2.1-10.8]	24.4 [14.4-53.2]	24.6 [14.7-49.4]	24.2 [14.3-48.9]	27.1 [15.6-68.2]	27.7 [15.3-67.9]	27.0 [14.8-64.3]
mandarin	100.0 (16)	6.1	0.0 [0.0-0.5]	0.0 [0.0-0.7]	0.0 [0.0-0.6]	19.0 [6.4-58.8]	18.3 [6.8-55.4]	19.4 [6.9-58.0]	22.6 [7.2-83.0]	21.8 [7.8-83.4]	23.4 [7.2-84.4]
Dimethoate											
cherries	84.6 (13)	1.2	0.0 [0.0-0.0]	0.0 [0.0-0.0]	0.0 [0.0-0.0]	3.1 [0.0-61.1]	3.4 [0.0-65.9]	3.1 [0.0-68.2]	5.3 [0.0-107.4]	6.1 [0.0-108.7]	5.1 [0.0-122.1]
lettuce	5.6 (126)	15.0	0.0 [0.0-0.0]	0.4 [0.4-0.6]	0.8 [0.7-1.1]	11.5 [0.0-83.5]	11.2 [0.8-89.0]	10.9 [1.6-91.0]	14.6 [0.0-129.2]	14.6 [0.9-121.9]	14.0 [1.7-123.7]

¹ total number of samples is indicated between brackets

1
2
3 **Figure 1.** Exposure assessment by the deterministic approach for the middle bound
4 scenario (LOR/2 for residue concentrations < LOR) in terms of % ADI (based on
5 average residue concentration and average and P97.5 consumption).
6
7
8
9

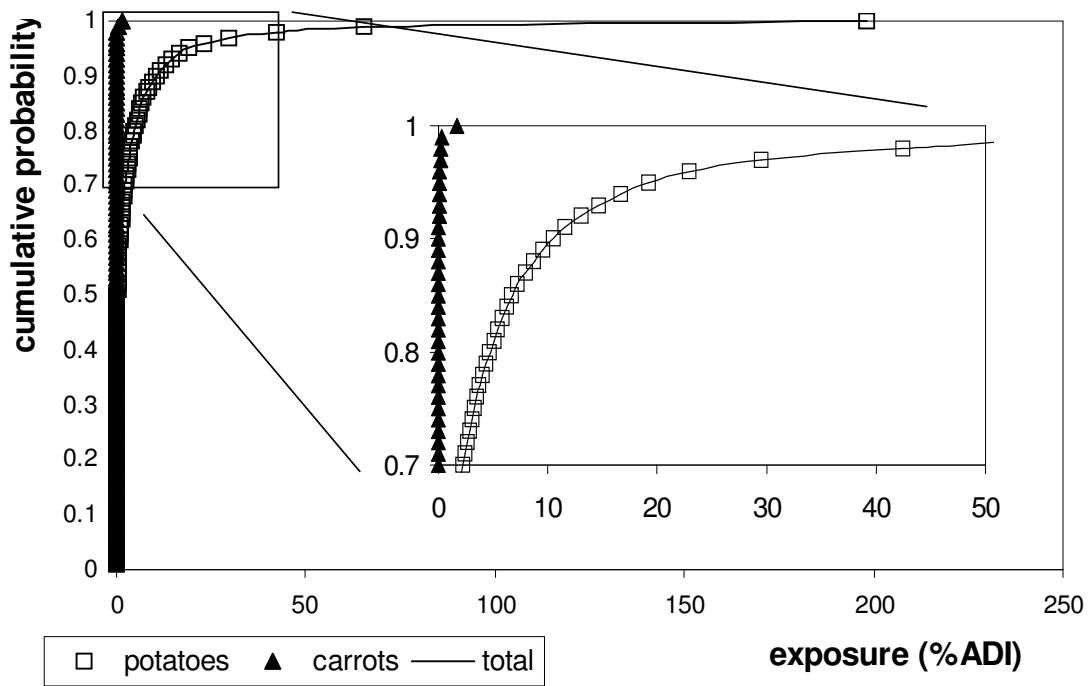
10
11 **Figure 2.** Contribution of different food items to the total exposure (in terms of % ADI,
12 middle bound scenario) of chlorpropham (a), imazalil (b), dimethoate (c),
13 dithiocarbamates (d), chlorpyrifos (e) and lambda-cyhalothrin (f).
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

Figure 1.



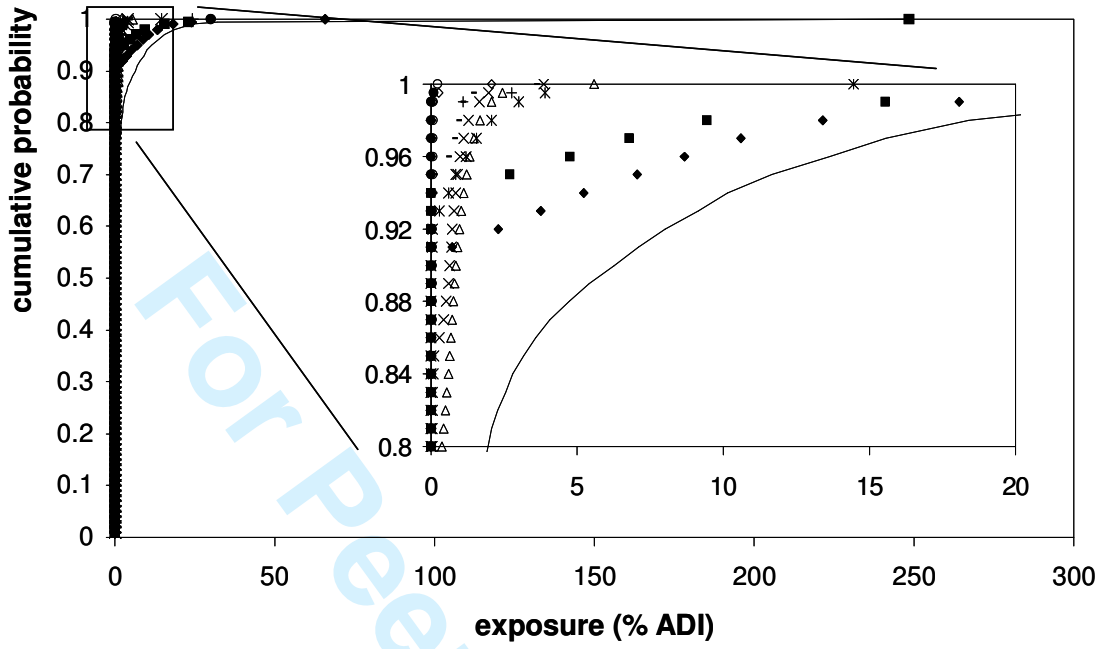
Review Only

Figure 2.a)



Review Only

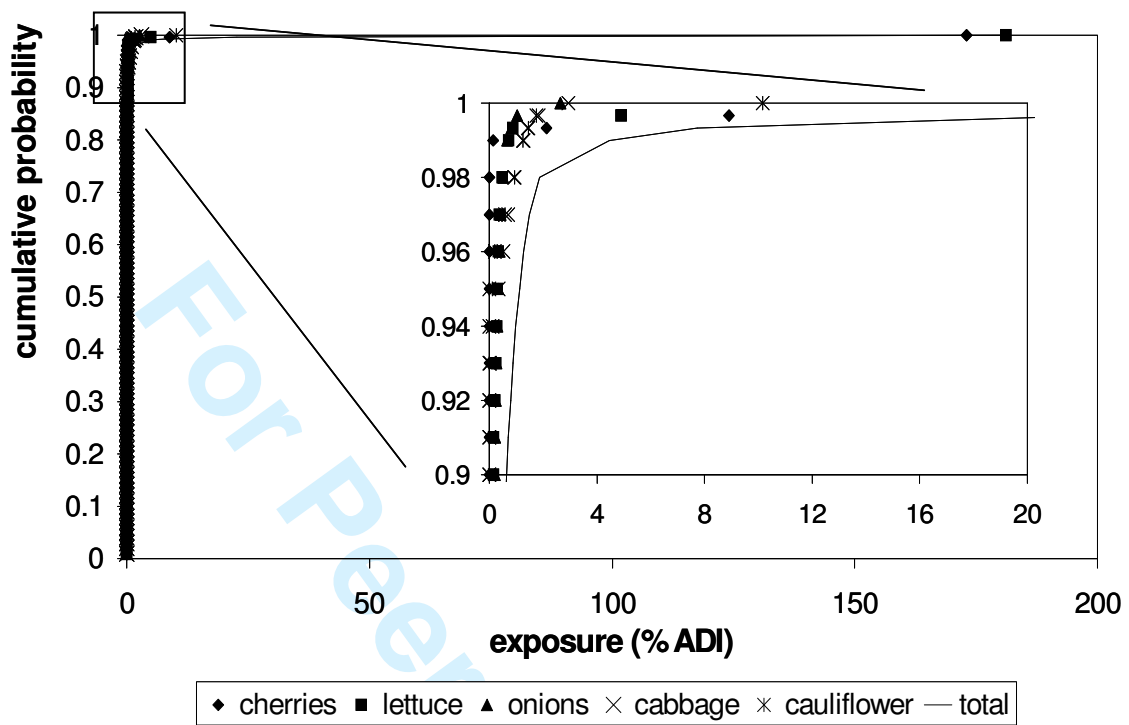
Figure 2.b)



◆ oranges	■ mandarin	△ potatoes	× apples	* bananas	● grapefruit
+ lemons	- pears	○ tomatoes	◇ cucumber	— total	

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

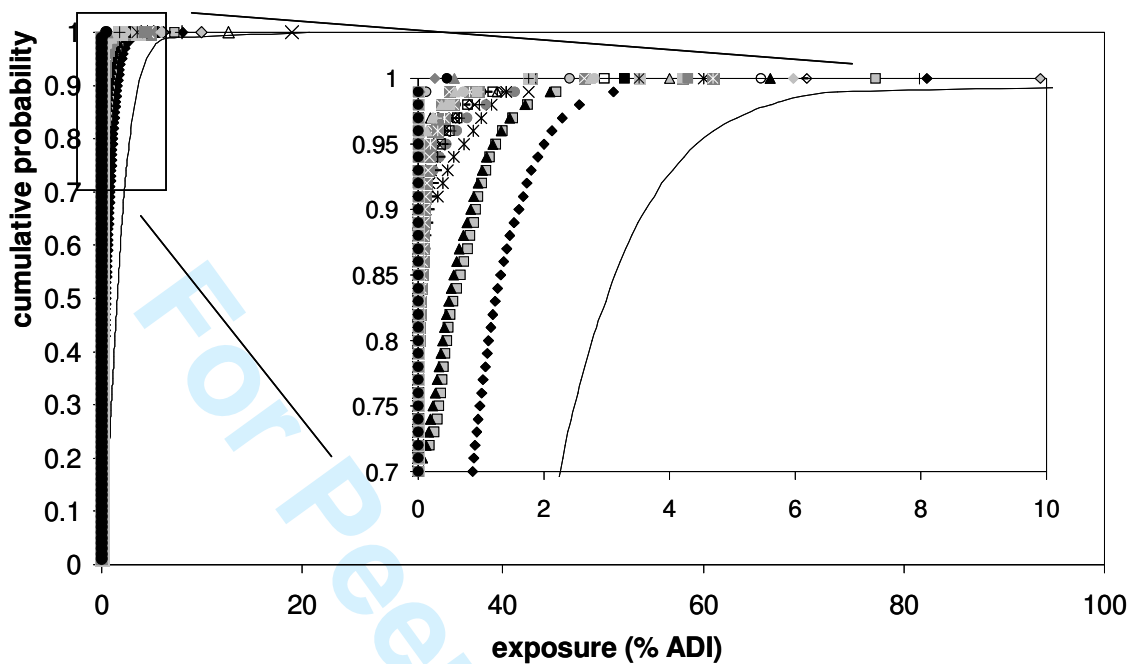
Figure 2.c)



For Peer Review Only

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

Figure 2.d)



◆ potatoes	□ apples	▲ tomatoes	× lettuce	✱ oranges
● cos	+ pears	- carrots	□ beans	◇ cabbages
△ leek	○ grapes	⊠ onions	⊠ madarin	⊠ strawberries
■ peaches	◆ peas	▲ cucumbers	● nectarin	■ grapefruit
△ endives	◇ apricot	○ celery	⊠ cherries	✱ watermelon
⊠ lamb's lettuce	■ currants	◆ lemons	▲ radishes	● cress
— total				

Figure 2.e)

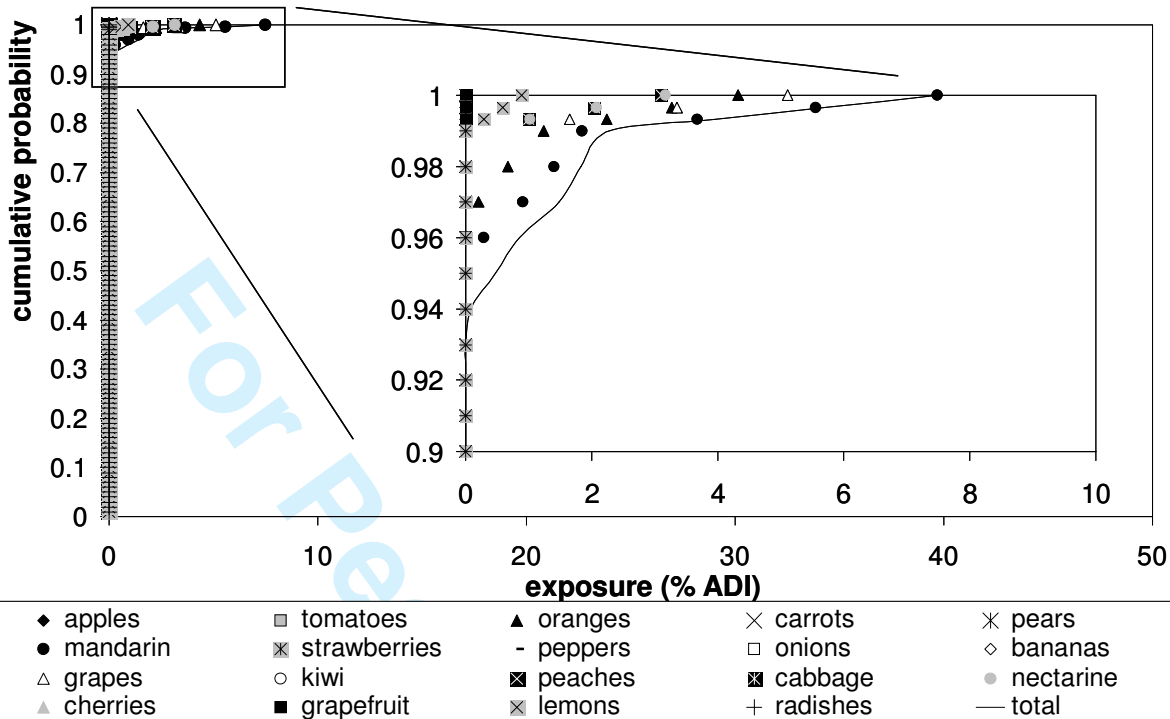


Figure 2.f)

