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

W.L. Claeys[a]*, S. De Voghel[b], J.-F. Schmit[a], V. Vromman[a] and L. Pussemier[b]

[a]*FASFC, Policy Control, Brussels, Belgium;* [b]*CODA-CERVA-VAR, Tervuren, Belgium*

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Exposure of the Belgian consumer to pesticide residues from the consumption of fruit and vegetables was determined based on data collected in the Belgian food consumption survey performed by the Scientific Institute for Public Health and data from the Belgian Federal Agency for the Safety of the Food Chain 2005 monitoring programme. A first screening of pesticide residue exposure was performed by a deterministic approach. For most pesticide residues studied, the exposure was 100 times lower than the acceptable daily intake (ADI). However, for a high consumer (97.5th percentile of consumption) the intake could reach 23% of the ADI for imazalil, 15% for chlorpropham, 14% for the dithiocarbamates, 10% for dimethoate and lambda-cyhalothrin, and 9% for chlorpyriphos. Nevertheless, probabilistic exposure assessment performed on these pesticides in a second phase of the study indicated that, except for chlorpropham, the probability to exceed the ADI is much lower than 0.1%.

Keywords: exposure assessment; probabilistic modelling; pesticides; fruit; vegetables

Introduction

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

To assess the exposure or intake, two main approaches can be distinguished: the deterministic and probabilistic approach. The deterministic approach is based on single-point estimates that are used for each variable within the model (such as an average value or the 97.5th percentile), whereas in the probabilistic approach, the variables are described in terms of distributions. In this way, all possible values for each variable are taken into account and each possible model outcome is weighted by the probability of its occurrence. Different techniques are available to calculate the outcome distribution, such as the Monte Carlo simulation (Vose 2006).

Advantages of deterministic modelling are that there is less need for extensive databases to support the input variables, that default standard assumptions can be used, that it is relatively easy to carry out, and that the single-risk estimate output is easy to understand and interpret. Advantages of probabilistic modelling are that all available data and knowledge are used, that the exposure estimate is presented as a distribution, with each value having a probability attached to it, and that variability and uncertainty can be quantified. The point estimate approach, due to its simplicity and its worldwide use and acceptance, may be used as a first screening tool to identify possible pesticides that may pose a problem. If so, the probabilistic approach can then be applied to study if the point estimate outcome really gives reason for concern.

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*Corresponding author. Email: wendie.claeys@favv.be

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An overview of current knowledge on intake or exposure assessment in general and probabilistic modelling in particular (input models, methodologies, stakeholders involved, etc.) is given, amongst others, by Vose (2006), Leclerq et al. (2003), Ferrier et al. (2002), and Kroes et al. (2002).

The aim of the present study was to evaluate whether the estimated daily intake of pesticides through fruit and vegetables consumption by the Belgian adult population is a cause for health concern, based on the results of the monitoring programme 2005 performed by the Belgian Federal Agency for the Safety of the Food Chain (FASFC 2006).

Materials and methods

Pesticide residue data

Contents of pesticides in the different commodities were taken from the pesticide monitoring programme 2005 of the Belgian Federal Agency for the Safety of the Food Chain (FASFC 2006), including 1322 samples of fruit and vegetables (tea and cacao included). During 2005, no less than 134,940 residue/food combinations were analysed. From this vast amount of data, only those residue/food combinations were selected that could be considered as being authorized in June 2005 according to Fytoweb, a Belgian website supported by the Federal Public Service Health, Food Chain Safety and Environment containing information on authorized pesticides (http://www.fytoweb.fgov.be/indexEn.asp).

The monitoring programme does not provide a full random analysis, but is based on risk assessment. Several factors were taken into account including the importance of the food commodity in the diets, exceedings observed in previous years, European Rapid Alert System for Food and Feed (RASFF) messages, pesticides authorized in Belgium, analytical and budgetary possibilities, etc. Sampling was performed by trained officials according to Directive 2002/63/EG at auctions, importers, wholesalers, processors and, exceptionally, in retail outlets. Samples were analysed in three officially recognized laboratories accredited following ISO 17025 (FASFC 2006). Multi- and single-residue methods were used for analysing pesticide residues.

With respect to samples exceeding the MRL, note that MRL values are not yet completely harmonized in the European Union. As such, imported food can exceed the Belgian MRL but comply with the MRL of the country of origin.

Food consumption data

Intake estimates were based on consumption data collected in the Belgian nationwide food consumption survey performed in 2004 by the Belgian Institute for Public Health (IPH 2006). The representative sample of participants included 3214 people over 15 years of age, who were questioned twice about their last 24-h consumption. The selection of interviewed people and the moment of the interview were chosen in order to obtain a representative consumption profile of the Belgian population over 1 year.

Concerning the consumption data recorded by the IPH, a selection had to be made and certain commodities had to be combined as a group (e.g. consumption data on Chinese, Savoy, red and white cabbage, and sauerkraut were aggregated into the group 'cabbage') in order to match the matrices analysed in the residue monitoring campaign of the FASFC.

The total data set, including zero intakes ('zero consumption days'), was used as part of an 'average' diet for long-term consumer exposure assessment since the main interest of this study is to evaluate the probability that the ADI is exceeded. When one is, however, interested in assessing the safety of actually eating a commodity, the 'consumers only' approach is preferable (Hamilton et al. 2004, Pieters et al. 2005).

Estimation of exposure

The dietary exposure to pesticides (mg kg body weight^{-1} day^{-1}) was calculated based on consumption data and individual body weights of the IPH diet study, and residue monitoring data of the Belgian FASFC monitoring programme 2005.

In a first phase, exposure was calculated deterministically. For a given residue/food combination, the average residue concentration was multiplied by the average consumption as well as with the 97.5th percentile of consumption of the whole population in order to have an idea of the chronic exposure of the population to pesticide residues.

In a second phase, exposure was evaluated in more detail by a probabilistic approach. Hereto, the Monte Carlo technique was applied. This technique involves the random sampling of each probability distribution within the model to produce hundreds or even thousands of scenarios (also called iterations or trials). Each probability is sampled in a manner that reproduces the distribution's shape. The distribution of the values calculated for the model outcome, therefore, reflects the probability of the values that could occur (Vose 2006).

In the probabilistic exposure assessment, Monte Carlo simulations were performed with 10,000 iterations. The model input distributions were randomly sampled by Latin Hypercube sampling. Calculations were performed by means of the commercially available software package @Risk® (Version 4.5.5; Palisade Corporation, NY, USA).

In order to have a good representation of consumption and pesticide residue data, both parametric and non-parametric approaches were evaluated. The fitting of probability distributions to the data in the parametric approach was performed using BestFit (Palisade). However, due to the large number of zero consumption days and of 'non-detects' in the residue concentration data, most of the consumption and contamination data could not be fitted by one of the distributions, and preference was given to the non-parametric approach. As such, model inputs (consumption and pesticide residue concentration data) were described by a discrete, uniform distribution. In this non-parametric approach the collected data points themselves are considered to form the distribution function and all possible values have the same probability of occurrence (Vose 2006).

Processing factors (peeling, cooking, boiling, etc.) and variability factors were not included in the calculations and no distinction was made between imported commodities and commodities of Belgian origin.

The exposure levels were compared with the ADI. The dithiocarbamates were considered as one group because testing methods do not differentiate between the different dithiocarbamates. Pesticide sales of the dithiocarbamates in Belgium concerns mainly mancozeb and maneb. Therefore, the ADI of mancozeb and maneb was chosen as the reference. With respect to the benomyl group, residues included are benomyl, thiofanate-methyl and carbendazim. Since benomyl is no longer authorized in Belgium and carbendazim is the common metabolite of thiofanate-methyl, the reference ADI chosen for the benomyl group was the one of carbendazim.

Levels below the limit of quantification (LOQ)

In pesticide exposure assessment, the estimated distribution typically consists of many measurements below the minimum pesticide residue concentration reported (limit of quantification, or LOQ). Such samples do not necessarily indicate that pesticide residues are not present in the sample, but only that the analytical method could not detect the possible amount of pesticide present. Therefore, samples with residue levels below the LOQ ('non-detects') were assigned either zero, LOQ/2 or LOQ, corresponding to a lower, middle and upper bound (worst-case) scenario.

Uncertainty of exposure estimates

The probabilistic method in itself generates estimates of variability in the form of an exposure distribution (one-dimensional Monte Carlo model). To estimate the uncertainty of the exposure estimate, it is necessary to apply other methods, such as bootstrap sampling (two-dimensional Monte Carlo model). Bootstrap sampling is a method to assess the reliability of percentiles of exposure and is a means to study the stability of the tail of the exposure distribution. With this method a sample of *n* observations (food consumption levels, residue levels) is resampled from the original database to obtain a bootstrap sample of *n* observations. Sampling is performed with replacement, so that every observation can occur more than once in the bootstrap sample. By repeating this process, e.g. 500 times, 500 bootstrap samples are obtained, which may be considered as alternative data sets that might have been obtained during sampling from the population of interest. Each statistic that can be calculated from the original data set (e.g. 97.5th, 99.9th percentile) can also be calculated from each bootstrap sample. This will generate a bootstrap distribution of five hundred 97.5th, 99.9th percentiles, etc. The bootstrap distribution now characterizes the uncertainty due to sampling uncertainty of the original data set (Boon et al. 2004).

Results

Deterministic exposure assessment

As a first screening, the exposure to pesticide residues was calculated by a deterministic approach on a total of 25 residues. These residues were selected out of the 200 residues analysed by the FASFC in 2005 based on their frequency of detection ($>2\%$ of analysed samples exceeded the LOQ). The residue concentration in the food commodities was assumed to be equal to the average concentration in order to take the variation of the concentrations into account. A rough estimate of the total exposure to a given pesticide residue X was obtained by summing exposures from all residue X/food combinations considered. The total exposure was compared with the ADI and expressed in terms of percentage ADI. Results for the lower-, middle- and upper-bound scenario for samples with a residue concentration below the LOQ are given in Table 1, together with their detection frequency and the ADI. Figure 1 presents an overview of the exposure for the average and the 97.5th percentile (P97.5) of consumption according to a scenario where the residue concentration of non-detected samples is equal to half the LOQ of the analytical method used (middle-bound scenario).

Probabilistic exposure assessment

Table 2 presents the exposure values based on a probabilistic assessment approach for chlorpropham, imazalil, dithiocarbamates, dimethoate, lambda-cyhalothrin and chlorpyriphos. Figure 2 gives more

Table 1. Total exposure in terms of percentage ADI of selected pesticide residues based on a deterministic exposure assessment approach (based on average residue concentration and on average and 97.5th percentile of consumption).

			Average consumption			P97.5 consumption		
	ADI (mg kg^{-1} body weight day^{-1})	Detection frequency (%)[a]	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
Chlormequat	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
Chlorpyriphos	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

Notes:[a] The total number of samples is indicated by parentheses. 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).

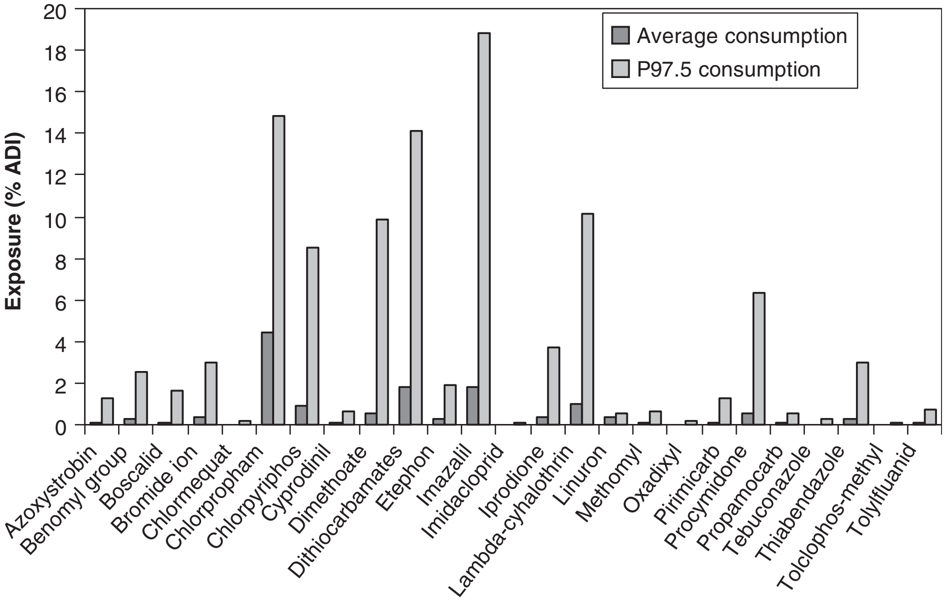

Figure 1. Exposure assessment by the deterministic approach for the middle-bound scenario (LOQ/2 for residue concentrations < LOQ) in terms of percentage ADI (based on average residue concentration and average and P97.5 consumption).

Table 2. Total exposure in terms of percentage ADI of selected pesticide residues based on a probabilistic exposure assessment approach.

				Average			P97.5			P99.9			P99.99		
	ADI (mg kg^{-1} body weight day^{-1})	ARfD (mg kg^{-1} body weight day^{-1})	Detection frequency (%)[a]	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[b]	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
Chlorpyriphos	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

Notes: [a] The total number of samples is indicated in parentheses.
[b] ARfD of Maneb and Mancozeb, respectively.

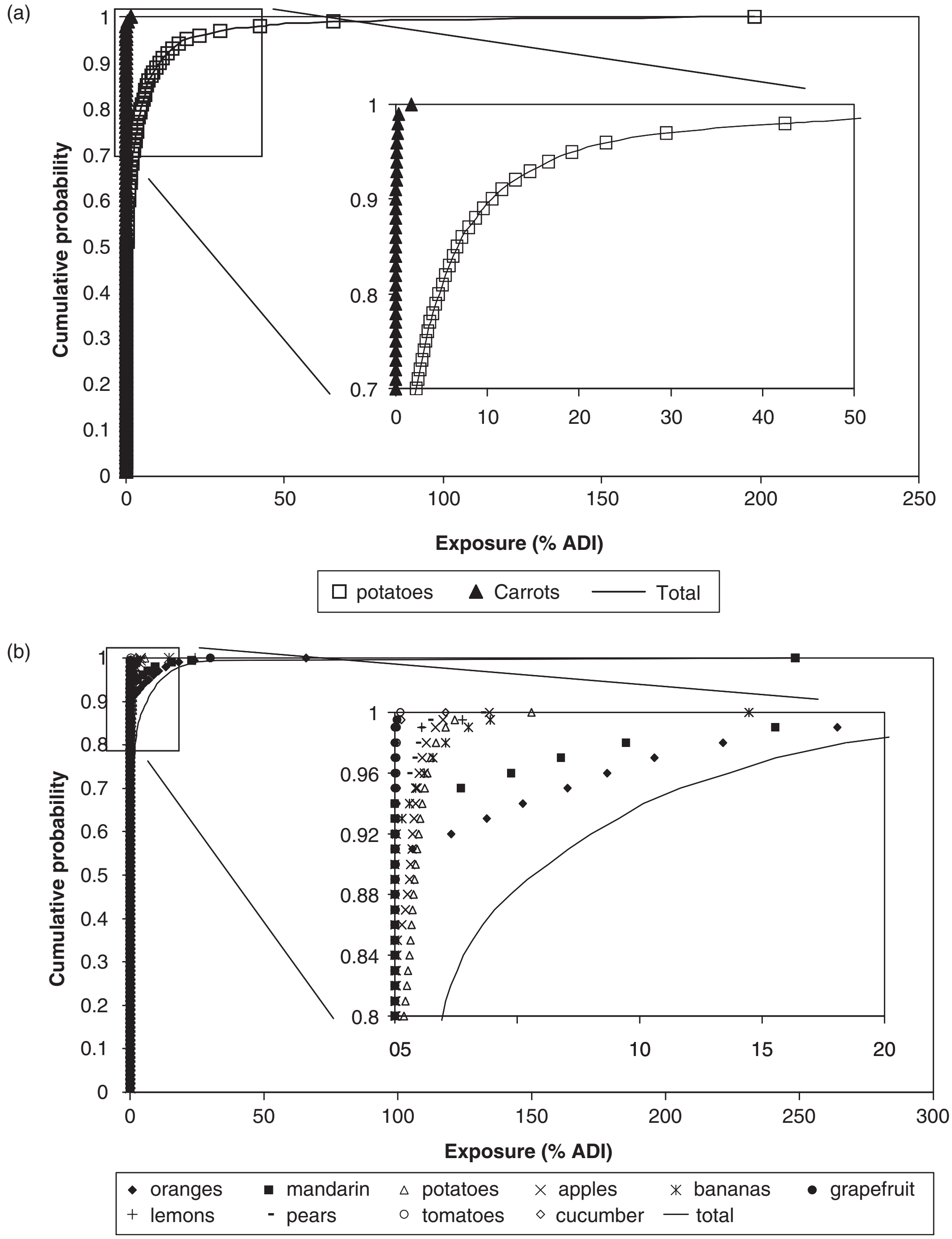

Figure 2. Contribution of different food items to the total exposure (in terms of percentage ADI, middle-bound scenario) of chlorpropham (a), imazalil (b), dimethoate (c), dithiocarbamates (d), chlorpyriphos (e) and lambda-cyhalothrin (f).

specific information on the contribution of several food items to the exposure of a given pesticide residue, and this for the middle-bound scenario.

Discussion

Deterministic analysis

Based on the P97.5 of consumption, relatively high exposure values were observed for chlorpropham, imazalil, dithiocarbamates, dimethoate, lambda-cyhalothrin and chlorpyriphos in the deterministic analysis (Table 1 and Figure 1). Chlorpropham is a selective systemic herbicide and plant growth regulator belonging to the *N*-phenylcarbamate group of pesticides. It is mainly applied as a sprouting inhibitor in potato storage. Imazalil is a systemic fungicide used to control a wide range of fungi on fruit, vegetables, and ornamentals. It is also used as a seed dressing and for post-harvest treatment of citrus, banana, and other fruit to control storage decay. Dithiocarbamates are widely used antisporulant (contact) fungicides that are very often associated with other fungicides. Lambda-cyhalothrin is a pyrethroid insecticide. Dimethoate and chlorpyriphos are organophosphate insecticides.

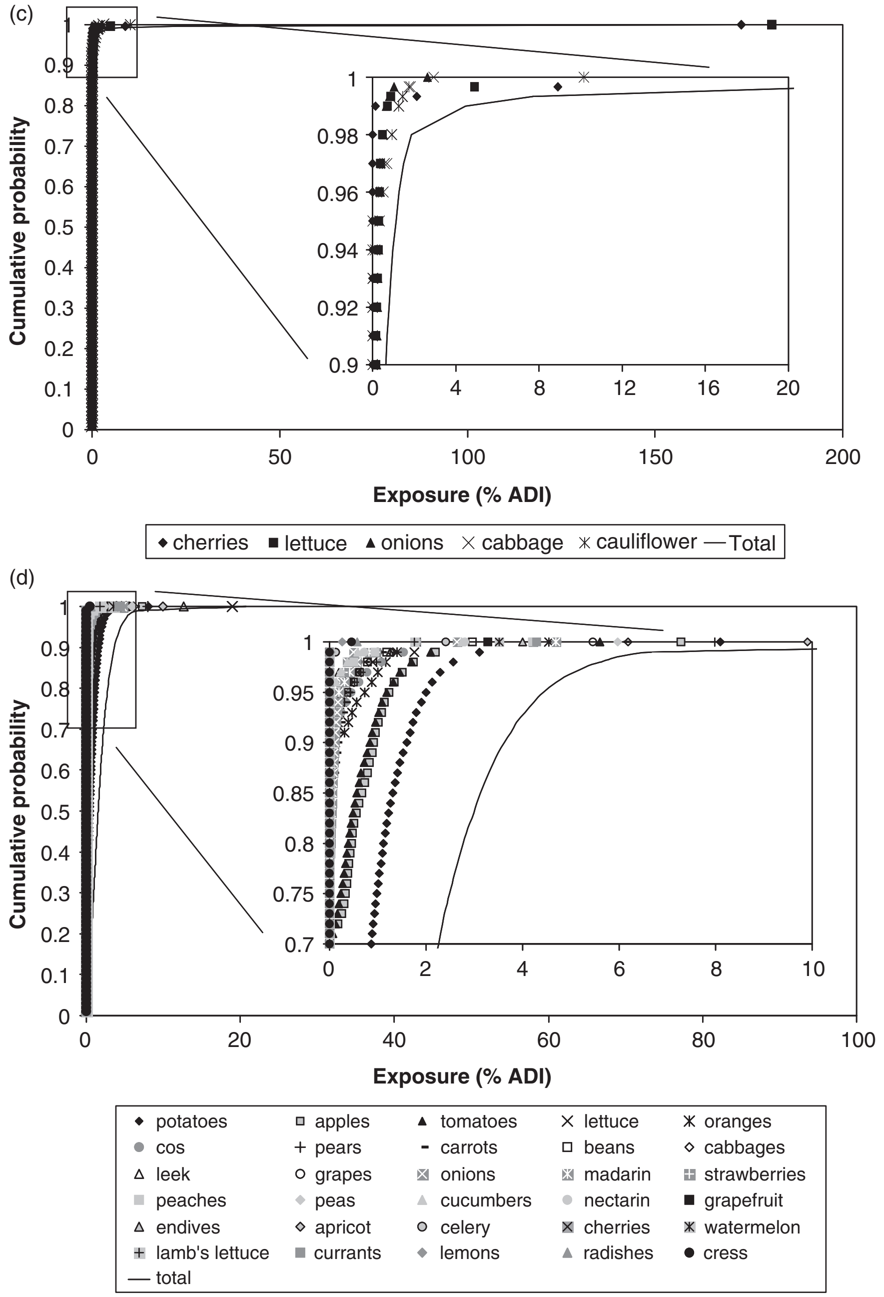

Figure 2. Continued.

These pesticides were selected for further probabilistic analysis.

Probabilistic analysis

As is illustrated in Figure 2, the exposure distributions are extremely right-skewed. Except for chlorpyriphos and lambda-cyhalothrin, the exposure estimates at the P99.99 are approximately two to three times higher than the exposure at the P99.9, which is three (to ten) times higher than the exposure at the P97.5. Because both food consumption and residue monitoring distributions are right-skewed, the resulting product is also a right-skewed distribution. It can be questioned if the consumption data set and the monitoring data set are robust enough to support exposure values calculated at the P99.9 and higher. These values can be more sensitive to uncertainties in data collection (sample size, reporting mistakes such as over reporting, analytical uncertainties) making these estimations of exposure less reliable. This is illustrated in Table 3, where the 95% confidence intervals of the higher percentiles of exposure for some residue/food combinations are given, including chlorpropham on potatoes, imazalil on oranges and mandarin, and dimethoate on cherries and lettuce. These 95% confidence intervals were calculated by the bootstrap sampling method.

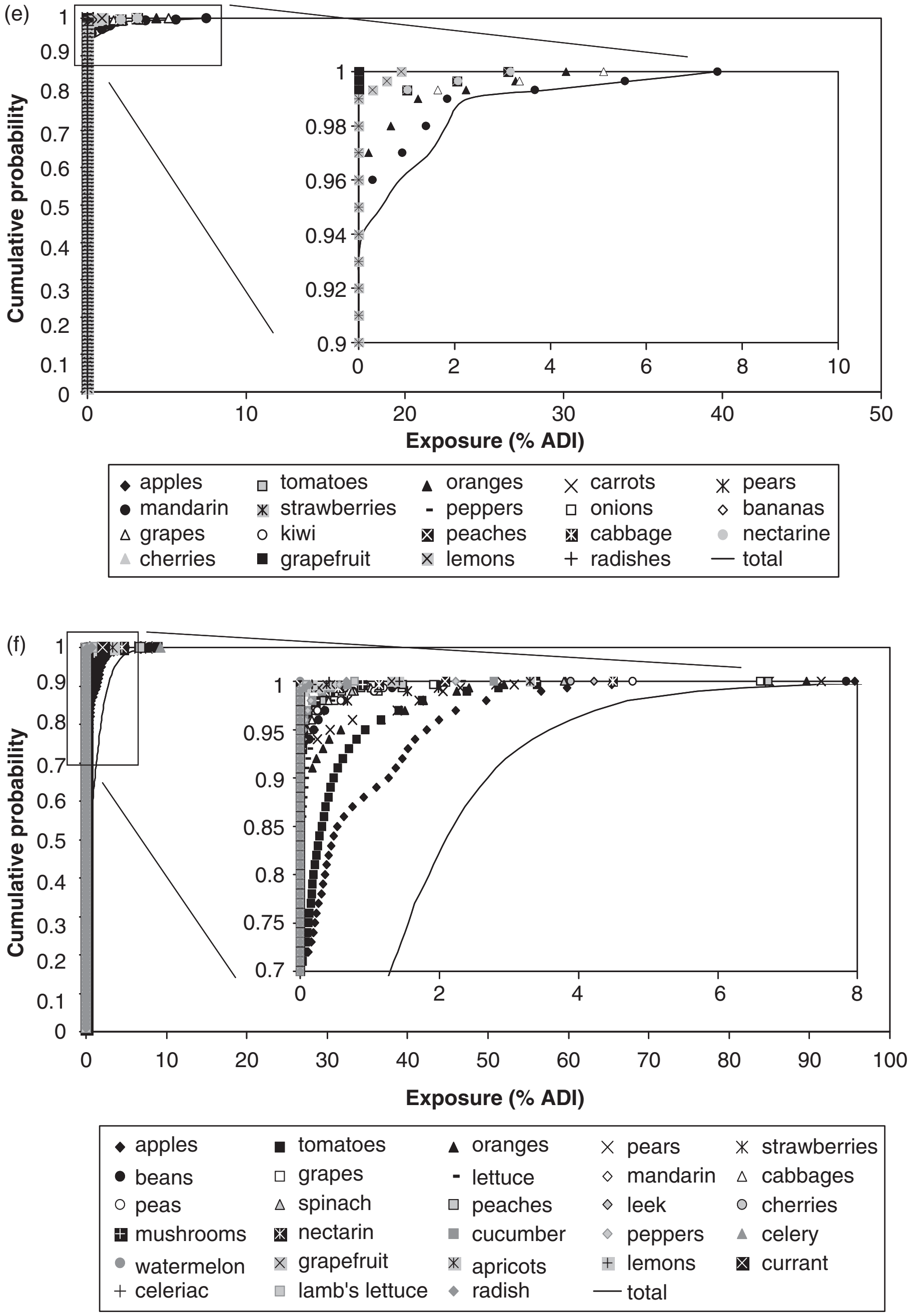

Figure 2. Continued.

'Non-detects'

The detection frequency or the number of samples with a residue concentration below the LOQ is rather high. As can be observed from Table 1, treatment of so-called left-censored data can have a large effect on the deterministically obtained outcome. For example, considering the results based on the P97.5 consumption, the intake of lambda-cyhalothrin or of the dithiocarbamates exceeds the intake of chlorpropham for the upper-bound scenario. For the lower- and middle-bound scenarios, on the other hand, the intake of lambda-cyhalothrin and of the dithiocarbamates was (much) lower than of chlorpropham. Generally, using the middle-bound scenario (LOQ/2) seems to be a good compromise. Nevertheless, it is clear that a proper treatment of censored data cannot be underestimated. Guidance on how to deal with so-called left censored data is given by the US Environmental Protection Agency (US EPA 2000).

With respect to the probabilistically obtained results, replacing non-detects by zero, the LOQ or the LOQ/2 does not affect the outcome presented in

Table 3. Consumer's exposure as percentage ADI (95% confidence interval) for different residue-food commodities (probabilistic approach).

	Detection frequency (%)[a]	% 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)

Note: [a]The total number of samples is indicated in parentheses.

Table 3 significantly. The large difference observed in Table 2 between the highest percentiles of exposure of the lower-, middle- and upper-bound scenario for some pesticides is due to a broad confidence interval. The higher the percentile under consideration, the broader the confidence interval. In Table 2, only one value out of the broad band of values was considered. (The exercise of bootstrap sampling was not performed for the complete data set of pesticide residues and commodities due to the huge amount of computational capacity required.)

Ferrier et al. (2006) evaluated the effect of uncertainty on the exposure estimate using scenarios reflecting different assumptions related to sources of uncertainty. The most influential uncertainty issue was the distribution type used to present input variables. Other sources that most affected the model output were non-detects, unit-to-unit variability and processing.

Comparison of the deterministic and probabilistic analysis

Comparison between the deterministic and probabilistic approach has to be done with care. Historically, the P97.5 has been chosen as the upper percentile for deterministic acute intake assessments, i.e. the P97.5 of consumers for the specific food and the P97.5 residue level, which in combination amounts to the P99.94 for residue intake (Hamilton et al. 2004):

$$(100 \times (1 - ((1 - 0.975) \times (1 - 0.975)))).$$

In the present study, the deterministic and probabilistic P97.5 exposure values are not comparable since they are essentially based on different notions. The deterministically obtained exposure values are based on percentiles of consumption, whereas the probabilistically determined results are actual percentiles of exposure. In contrast, the average exposure values are very similar.

Additionally, it has to be remarked that in the deterministic approach, total exposure for a given residue was obtained by summing exposures from all residue/food combinations, whereas deterministic intake assessments should deal with only one food at a time. Generally, summing point estimates across foods leads to an estimate of food consumption that is extremely high and unrealistic. Because of the inherent conservatism in the point estimate, summing these estimates over commodities would be equivalent to assuming that large portions of all commodities (P97.5 of a consumption distribution) are consumed on 1 day or frequently by one individual and that all these commodities are contaminated. For a realistic estimate of short-term dietary exposure through multiple foods, a probabilistic approach is recommended (Hamilton et al. 2004).

Nevertheless, when calculating the total exposure by the probabilistic approach, potential correlations between daily consumption rates of different food items need to be taken into account. As a pragmatic approach, recorded days were considered as such, without discriminating between variation among individuals and variation among days (within individuals), resulting in a distribution of 'person-days'. The drawback of this approach is that it cannot be determined if few persons are at risk, but each of them at many days, or that many persons are at risk but each of them only rarely (Pieters et al. 2005). Since the focus of the present study concerns mainly the intake of frequently exposed individuals, the intake was compared with the ADI rather than to the ARfD.

Upper percentiles of exposure

Since in the exposure assessment of pesticides the regulatory threshold risk is at the upper tail of the exposure distribution (P99 and higher), also the P99.9 and P99.99 values of exposure are presented in Table 2. From this table, it can be concluded that, except for chlorpropham, the probability that a person exceeds the ADI by eating one or more servings of a contaminated food item is less than 0.1%. The relatively high exposure to chlorpropham is solely due to one crop, namely potatoes (Figure 2a).

Next to chlorpropham, a relatively high intake in terms of percentage ADI is also observed for dimethoate and, to a lesser extent, for imazalil. This high exposure value for dimethoate is mainly due to a high or frequent consumption of cherries and lettuce (Figure 2c). With respect to imazalil, citrus fruit, orange and mandarin in particular contributed most to the intake (Figure 2b).

Different profiles can be distinguished between the high exposure levels. Exposure to imazalil is relatively high since the amount of commodities on which it was detected and its use is authorized is high. For chlorpropham, on the other hand, the high exposure was due to some high levels of pesticide residue detected on a single food item (potatoes) and the relatively high consumption of that food item.

The effect of consumption data on the outcome can be illustrated by Table 3. For example, the P97.5 exposure to dimethoate by the consumption of cherries is zero and the P99.99 exposure values range from zero to values higher than the ADI, although a detection frequency of 84.6% was observed. Cherries are food items that are eaten occasionally. Their consumption is seasonal resulting in extreme right-censored consumption data (1.2% non-zero consumption days). Due to the high detection frequency of dimethoate on cherries, exceeding of the ADI may occur for a minority of consumers (P99.99), although rarely. From this example, it is also clear that caution should be taken when interpreting high exposure percentiles without knowledge of the confidence interval. Performing a one-dimensional Monte Carlo simulation could result in extreme values of a confidence interval leading to unnecessary concern.

Comparison with other studies

Different probabilistic exposure assessment studies of pesticides from food are reported in the literature, but are still scarce (Hamey 2000, Wright et al. 2002, Boon et al. 2003, López et al. 2003, Brüschweiler 2004). When comparing exposure assessments in the literature, some caution has to be taken. One difficulty is that studies are performed using different (national) food consumption databases. The way in which food consumption data of different countries have been and is collected, is not harmonized and can vary considerably. This diversity is related to the population addressed (e.g. children included or not), the method of data collection (24-h recall, dietary method), duration of the survey, the number of respondents involved, coding of food consumption data, and the method of quantifying the amount consumed (actual weighing versus estimations on the basis of portion sizes) (Kroes et al. 2002). These variables should be kept in mind when comparing exposure assessments using different databases.

Another critical aspect is the sampling of commodities for pesticide residue analysis. This is not always according to a random procedure, but often based on a risk approach (directed or targeted sampling). The use of these data to estimate dietary exposure may thus lead to an overestimation of exposure.

In order to be able to compare exposure assessment results internationally, standardized ways of collecting and reporting data are needed.

The exposure estimates presented here for chlorpyriphos compare favourably with estimates published in the literature. The US FDA's Total Diet Studies from 1989 and 1990 estimated chlorpyriphos exposure at $0.041 \times 10^{-4}\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$ for children aged 14–16 years (FDA 1990, 1991). This estimate is similar to the average value given for the lower-bound scenario in Table 2 ($0.045 \times 10^{-4}\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$). Based on monitoring programmes, Wright et al. (2002) estimated a P99.9 exposure of $4.8 \times 10^{-4}\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$ for the US population, and $9.4 \times 10^{-4}\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$ for children between 1 and 6 years old, which is of the same order of magnitude as calculated for the higher percentiles of exposure in Table 2.

In a Dutch study where a probabilistic model of dietary exposure of infants to pesticides was compared with a duplicate diet study, the 95% confidence interval for the P99 of exposure to chlorpyriphos was $0.2\text{–}4.3 \times 10^{-4}\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$ for the duplicate diet study assuming a log-normal exposure distribution and $24.5\text{–}43.3 \times 10^{-4}\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$ for the probabilistic model in which variability and processing were taken into account and non-detects were replaced with the limit of reporting (LOR) (Boon et al. 2003). The latter value exceeds even the worst-case (non-detects equal to LOQ) P99.99 value estimated in this study ($14.2 \times 10^{-4}\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$). This can be, amongst others, due to the fact that in the Dutch study toddlers aged 8–12 months were considered and the sampling strategy was designed to include as many children as possible that were fed home-made meals of fruits and vegetables.

In a Swiss study, the probabilistic intake to chlorpropham was assessed for potato dishes and raw potatoes (Brüschweiler 2004). Applying processing factors, the mean daily intake was 0.0056 mg per person and the P99 0.15 mg per person corresponding to 0.93×10^{-4} and $0.0025\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$, respectively, for a person weighing 60 kg. Both results are lower than observed in this study, where the average intake of chlorpropham by uncooked and unpeeled potatoes was calculated as $0.0021\,\mathrm{mg\,kg^{-1}\,bw\,day^{-1}}$. As remarked by the author, calculated exposures depend strongly on the processing factors applied, which differ markedly in the literature.

Risk assessment

As already indicated in the Introduction, detection frequency and the number of samples for which the MRL is exceeded give an indication of which residues and food commodities to prospect, but lacks information in terms of food safety. This can be illustrated by comparing, for example, chlorpropham with imazalil and the dithiocarbamates. The detection frequency of chlorpropham in potatoes was lower than that for imazalil in mandarin or oranges (69% compared with 87% and 100%, respectively) (Table 3). Nevertheless, exposure values for imazalil were lower. In 2005 and with respect to the considered food items, two MRL exceedings were reported for chlorpropham in potatoes (non-harmonized, national MRL of $5\,\mathrm{mg\,kg^{-1}}$), whereas six MRL exceedings were reported for dithiocarbamates including one for cress, three for lettuce, one for peas and one for radishes (EC MRLs of 0.3, 3.0–5.0, 0.1 and $0.05\,\mathrm{mg\,kg^{-1}}$, respectively) (FASFC 2006). In terms of exposure, chlorpropham is ranked higher than the dithiocarbamates.

Based on the results, it can be concluded that chronic intakes of 25 selected pesticide residues are rather low compared with the ADI (mostly $<1\%$ of the ADI) and that public safety in Belgium seems to be generally under control in terms of (single) pesticide intakes through the consumption of fruit and vegetables. However, some residues such as chlorpropham, imazalil, dithiocarbamates, dimethoate, lambda-cyhalothrin and chlorpyriphos needed to be considered more closely since for a high consumer (P97.5) their intake can reach 23% of the ADI for imazalil, 15% for chlorpropham, 14% for the dithiocarbamates and 10% for lambda-cyhalothrin and dimethoate, and 9% for chlorpyriphos for the middle-bound scenario. Nevertheless, probabilistic analysis indicated that except for chlorpropham, the probability of exceeding the ADI will be much lower than 0.1% when exposed to one of the selected pesticides.

Considering acute exposure to pesticide residues, interpretation of exposure levels determined in this study in terms of the percentage of the ARfD will indicate the absence of acute risks, even for high consumers, given that ADI values for pesticides are lower than ARfD values (Table 2).

It seems that exposure to the pesticide residues considered is mainly due to the consumption of only one or two food items (e.g. potatoes in case of chlorpropham, citrus fruit in case of imazalil). With regard to fruit and vegetables, attention should also be given to chemical contaminants other than pesticides such as natural (e.g. mycotoxins), environmental (e.g. cadmium, polychlorinated biphenyls (PCBs)) or process contaminants (e.g. 3-monochloropropane-1, 2-diol (3-MCPD), acrylamide). As such, for example, exposure to chlorpropham (and other sprouting inhibitors) on potatoes could be linked to the acrylamide issue. It has been shown that more acrylamide is formed during the cooking of potatoes that have been stored at cold temperature (Claeys et al. 2005). If fewer potatoes are stored in cold-storage depots, it is not inconceivable that the application of sprouting inhibitors like chlorpropham increases.

It has to be noted that the present study focused on fruits and vegetables only. Therefore, it is an underestimation of the total exposure of pesticides studied. On the other hand, processing factors were ignored, whereas fruit and vegetables are often peeled, cooked or boiled before consumption, resulting in an overestimation of the actual exposure to pesticide residues. Other variables affecting pesticide residue concentration include storage, transport, shelf life, use patterns, laboratory-to-laboratory variation, and analytical methods used to measure chemicals (Kroes et al. 2002). Additionally, it has to be noted that the dietary consumption data provided by the IPH did not include data for children under 15 years old. A special attention to this sensitive group could be given in further research.

Further research could also deal with exposure assessment related to the presence of multiple residues on a single food or serving, i.e. cumulative dietary intake assessment. Cumulative exposure to various residues of pesticides in food is a potential area of concern and gaining more and more interest. This issue is especially relevant for pesticides with a common mechanism of toxicity (e.g. organophosphates, carbamates) (Jensen et al. 2003, Van Raaij et al. 2005, EFSA 2007).

Finally, it has to be noted that compared with point estimates, making use of probabilities and revealing the extreme upper tail of the exposure distribution may create difficulties for both consumers and regulators in terms of their understanding of the assessment. However, a full probability distribution also enables more informed decisions to be made for policy and regulation. Regulation based on unrealistic exposure estimates could lead to unnecessary restrictions on products, or potentially put consumers at risk. It is therefore important to understand the influence that sources of uncertainty exert on predictions of exposure and to include this knowledge in the decision-making process (Ferrier et al. 2006).

Conclusion

In 2005, the FASFC controlled 1322 samples of fruit and vegetables. Pesticide residues were found in 56% of the samples and standards were exceeded for 7.9% of the samples (FASFC 2006). These numbers could give unnecessary rise to consumer concern. A more nuanced and different picture is obtained when the actual exposure of consumers to pesticide residues is considered. Based on the results of the present study, chronic exposure to pesticide residues due to the consumption of fruit and vegetables seems to be under control, even for frequent consumers.

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References

Boon P, Lignell S, Van Klaveren J, Tjoe Nij E. 2004. Estimation of the acute dietary exposure to pesticides using the probabilistic approach and the point estimate methodology. Rikilt. Report No. 2004.008; [cited 2007 Sep 10]. Available from: http://www.rikilt.wur.nl/NR/rdonlyres/BDEEDD31-F58C-47EB-A0AA-23CB9956CE18/10727/R2004008.pdf

Boon P, Van der Voet H, Van Klaveren J. 2003. Validation of a probabilistic model of dietary exposure to selected pesticides in Dutch infants. Food Add Contam. 20(Suppl. 1):S36–S49.

Brüschweiler B. 2004. Probabilistic exposure assessment for chlorpropham through potato consumption. Mitteilungen aus Lebensmitteluntersuchung und Hygiene. 95:421–436.

Claeys W, De Vleeschouwer K, Hendrickx M. 2005. Quantifying the formation of carcinogens during food processing: acrylamide. Trends Food Sci Tech. 16:181–193.

EFSA. 2007. Cumulative risk assessment of pesticides to human health: the way forward. EFSA Scientific Colloquium 7. Summary Report; [cited 2007 Nov 3]. Available from: http://www.efsa.europa.eu/EFSA/ScientificOpinionPublicationReport/EFSAScientificColloquiumReports/efsa_locale-1178620753812_RiskAssessmentPesticidesHumanHealth.htm

European Commission. 2002. Commission Directive 2002/63/EC of 11 July 2002 establishing community methods of sampling for the official control of pesticide residues in and on products of plant and animal origin and repealing Directive 79/700/EEC.

FASFC. 2006. Report of monitoring results concerning Directives 90/642/EEC, 76/895/EEC and Commission Recommendation 2005/178/EC. Brussels (Belgium).

FDA. 1990. Residues in food – 1989. J Assoc Off Analyt Chem. 62:127a–145a.

FDA. 1991. Residues in food – 1990. J Assoc Off Analyt Chem. 63:121a–141a.

Ferrier H, Nieuwenhuijsen M, Boobis A, Elliott P. 2002. Current knowledge and recent developments in consumer exposure assessment of pesticides: a UK perspective. Food Add Contam. 19:837–852.

Ferrier H, Shaw G, Nieuwenhuijsen M, Boobis A, Elliott P. 2006. Assessment of uncertainty in a probabilistic model of consumer exposure to pesticide residues in food. Food Add Contam. 23:601–615.

Hamey P. 2000. A practical application of probabilistic modelling in assessment of dietary exposure of fruit consumers to pesticide residues. Food Add Contam. 17:601–610.

Hamilton D, Ambrus A, Dieterle R, Felsot A, Harris C, Petersen B, Racke K, Wong S-S, Gonzalez R, Tanaka K, et al. 2004. Pesticide residues in food – acute dietary exposure. Pest Manag Sci. 60:311–339.

IPH. 2006. De Belgische Voedselconsumptiepeiling 1-2004, Devriese S, Huybrechts I, Moreau M, Van Oyen H. Afdeling Epidemiologie, 2006. Brussels (Belgium): Wetenschappelijk Instituut Volksgezondheid. Depotnummer D/2006/2505/17, IPH/EPI Reports No. 2006-016; [cited 2007 Sep 10]. Available from: http://www.iph.fgov.be/epidemio/epien/index5.htm/

Jensen A, Petersen A, Granby K. 2003. Cumulative risk assessment of the intake of organophosphorus and carbamate pesticides in the Danish diet. Food Add Contam. 20:776–785.

Kroes R, Müller D, Lambe J, Löwik M, Van Klaveren J, Kleiner J, Massey R, Mayer S, Urieta I, Verger P, et al. 2002. Assessment of intake from the diet. Food Chem Toxicol. 40:327–385.

Leclerq C, Arcella D, Armentia A, Boon P, Kruizinga A, Gilsenan M, Thompson R. 2003. Development of databases for use in validation studies of probabilistic models of dietary exposure to food chemicals and nutrients. Food Add Contam. 20(Suppl. 1):S27–S35.

López A, Rueda C, Armentia A, Rodríguez M, Cuervo L, Ocio J. 2003. Validation and sensitivity analysis of a probabilistic model for dietary exposure assessment to pesticide residues with a Basque Country duplicate diet study. Food Add Contam. 20(Suppl. 1):S87–S101.

Pieters M, Ossendorp B, Slob W. 2005. Probabilistic modelling of dietary intake of substances. The risk management question governs the method. RIVM. Bilthoven (The Netherlands) Report No. 320011001/2005; [cited 2007 Sep 10]. Available from: http://rivm.openrepository.com/rivm/bitstream/10029/7301/1/320011001.pdf

US EPA. 2000. Assigning values to non-detected/non-quantified pesticide residues in human health food exposure assessments; Washington DC (US) [cited 2007 Sep 10]. Available from: http://www.epa.gov/pesticides/trac/science/trac3b012.pdf/

Van Raaij M, Ossendorp B, Slob W, Pieters M. 2005. Cumulative exposure to cholinesterase inhibiting compounds: a review of the current issues and implications for policy. RIVM. Bilthoven (The Netherlands) Report No. 320108001/2005; [cited 2007 Sep 10]. Available from: http://www.rivm.nl/bibliotheek/rapporten/320108001.pdf

Vose D. 2006. Risk analysis – a quantitative guide. 2nd ed. Chichester (UK): Wiley.

Wright J, Shaw M, Keeler L. 2002. Refinements in acute dietary exposure assessments for chlorpyrifos. J Agric Food Chem. 50:235–241.