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Dietary exposure to pesticides – relevant variables and probabilistic modelling

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SUMMARY

This report focuses on the variables that influence the exposure to pesticides in the diet, and on how to model these variables in a probabilistic approach to assess the dietary exposure to these compounds. The main variables addressed are processing, unit variability, and percentage of the crop that has been treated with the compound combined with the level that should be assigned to those samples with concentrations below the reporting level. Different suggestions are made on how to include these factors in a probabilistic model. To assess the influence on dietary pesticide exposure of including these factors in the model, numeric experiments were performed using data on food consumption and pesticide levels in the Netherlands. Another issue addressed is the modelling of pesticide levels in foods when using a probabilistic approach. Pesticide levels can either enter a probabilistic model as raw data or via parametric modelling. This last approach is preferable when the number of levels available is low and/or when there are doubts that the available levels represent the true range of levels encountered in real life. To establish which type of distribution fitted pesticide levels best, 10 pesticide–commodity combinations were fitted to 21 distribution types. The results demonstrated that the lognormal distribution fitted concentrations best.

Another important issue in pesticide exposure assessment addressed in this report is what to do when pesticide data are scarce. In pesticide exposure assessment the data on pesticide residue levels on different crops will typically be limited. We addressed the question when we can speak of such a situation and a possible approach to deal with it is discussed, namely grouping of products. By grouping different products into product groups composed of related products (e.g. product groups consisting of cabbage or all kinds of berries) the number of measurements per group increases and may give sufficient data to estimate the parameters of the lognormal distribution that models the pesticide levels.

Pesticide levels in the EU are usually reported in a tabulated (histogram) form. This report describes how this type of data can be transformed into a lognormal distribution and can thus be used in a probabilistic assessment of exposure. A numerical experiment is described in which full data were classified in histograms and then transformed in a lognormal distribution. Results showed that histogram data (as reported in the EU) can be used to estimate the dietary exposure to pesticides.

This report encompasses work performed as part of Workpackage 2 of the Monte Carlo project funded by the European Commission Quality of Life and Management of Living Resources Fifth Framework Programme (contract n°QLRT-CT-1999-00155).

1 INTRODUCTION

This report focuses on the variables that influence the exposure to pesticides in the diet (chapter 2), and on how to model these variables in a probabilistic model for assessing the dietary exposure to these compounds (chapter 3).

The work reported here was carried out as part of Workpackage 2 of the Monte Carlo project funded by the European Commission Quality of Life and Management of Living Resources Fifth Framework Programme (contract nºQLRT-CT-1999-00155). This report, in a slightly different form, is available under the title 'Chemical concentrations in food – pesticide residues' on the internet (http://www.tchpc.tcd.ie/montecarlo).

2 VARIABLES THAT INFLUENCE DIETARY EXPOSURE TO PESTICIDES

Food may contain many different chemicals, such as nutrients (e.g. carbohydrates, vitamins), additives and process aids (e.g. antioxidants, emulsifiers), agricultural chemicals (pesticides and veterinary drugs) and toxins and environmental contaminants (e.g. mycotoxins, dioxins). The levels of these chemicals vary considerable. For example, agricultural chemicals may be present in high concentrations (levels above permitted levels) or be completely absent (Klaveren 1999).

In the following we will focus on the intake of pesticides through the diet. To estimate the exposure to these chemicals, different issues need to be addressed to make the estimation as accurate and realistic as possible. These include the conversion of food as eaten into raw agricultural products, pesticide residue levels and the effect of different variables on these levels. These variables are processing, variability in levels within composite samples, and percentage of the crop that has been treated with the compound combined with the level that should be assigned to those samples with concentrations below the reporting level. All these issues should be included in a model for estimating the dietary exposure to pesticides (Crossley 2000, Petersen 2000, US EPA 2000a).

Other factors are known to influence the chemical level to which a consumer is exposed or which is analysed in a certain commodity. These include storage, transport, shelf-life, use patterns, lab-to-lab variation, and analytical methods used to measure chemicals. Country of origin of the product is also a factor that may influence the level present in ready-to-eat food. For example, crops produced in southern European countries appear to contain more insecticides than countries with more humid and cold weather conditions. In these countries infections of fungi appear to be the predominant problem and therefore fungicides are the most commonly used pesticides. Due to shortage of information on these issues, at the level of both consumption and pesticide-commodity combination, these variables are not considered below.

2.1 Food as eaten in terms of raw agricultural commodities

Most analyses of pesticide residues in fruits and vegetables are conducted in raw agricultural commodities (RAC) including peels and non-edible parts. Processed or prepared foods are either not monitored or the number of samples is very small. This is due to the fact that in legislation limits of residues are mainly set for RAC. To model dietary exposure to pesticides, some countries have developed so-called recipe data banks for RAC to provide a link between residue data and food consumption data (Dooren *et al.* 1995). In the Dutch database all foods listed in the Dutch food composition table have been converted to RAC based on several sources of information (Dooren *et al.* 1995). These include among others recipes from cookbooks, and information from either the literature or label of the product. The type of processing a RAC has undergone before consumption is also recorded. E.g. apple juice may be converted to RAC 'apple' with processing type 'juiced', apple eaten peeled to RAC 'apple' with processing type 'peeled', apple eaten raw to RAC 'apple' with processing type 'none'. In this way the effect of processing on residue levels in RAC can be taken into account in exposure assessment.

2.2 Pesticide levels

Pesticide levels in RAC can be derived from monitoring programmes executed by Member States of the European Union (EU). These data are mainly used to estimate the dietary exposure to pesticide residues, although they are not widely available. The raw data are present nationally in almost all EU countries, but are not easily accessible. Furthermore, there are differences in data collection and reporting between countries (no standardisation). This problem was recognised by the EU, which has set up a co-ordinated programme aiming to 'work towards a system which makes it possible to estimate dietary pesticide exposure throughout Europe' (EC 2000) sing a standardised manner of collecting and reporting pesticide levels in agricultural products. Apart from this problem, it should also be kept in mind that monitoring data may not always be representative of the levels people are exposed to in real life. The majority of pesticide monitoring programmes in Europe was initially set up for law enforcement reasons. Because of this sampling is not always at random and often focussed on those samples suspected to contain residue levels above the limit permitted (e.g. those products produced during wet seasons, under difficult conditions, or out of season). Use of these data in exposure assessment may thus lead to overestimates of exposure. However, recently it was recognised that there is an increasing need to compare the results of pesticide monitoring between countries and to use these data for exposure assessment. Because of this, current directives on pesticide monitoring programmes include more rules on representative sample taking (Codex Alimentarius 1999).

Pesticide levels can also be derived from field trials (experiments performed to authorise the use of a pesticide in agriculture). These types of data are, however, not considered here, because they are unsuitable to estimate dietary exposure to a certain chemical in a certain population. In field trials pesticides are applied in a controlled manner, the whole crop is treated with the pesticide and a standardised period between administration and harvesting is observed. In real life, however, it is unlikely that all commodities have been treated, the circumstances of application will have been less controlled and the period between application and harvesting will vary. These factors will result in a larger variability in residue levels in real life compared to results from field trails.

When using monitoring data in exposure assessment several variables that determine the level to which consumers are exposed in real life need to be considered, namely processing, variability and level assigned to samples with residue concentrations below the reporting level. These issues will be addressed below.

2.2.1 Processing

As mentioned above, pesticide analyses are mainly performed in RAC, which includes the peel and (other) non-edible parts. These commodities are however not eaten as such, but undergo some form of food processing before actual consumption. Processing has been interpreted as any operation performed on a food, food source, or food product from the point of harvest through consumption (Ritchey 1981). For example, most vegetables are washed and cooked and non-edible parts are removed, and fruits are often washed, peeled and/or processed into juices or sauces. Processing affects pesticide levels (mainly reduction) as is evident from numerous studies (Celik *et al*. 1995, Elkins 1989, Hasegawa *et al.* 1991, Holland *et al*. 1994, Petersen *et al*. 1996, Ritchey 1981, Zabik *et al*. 2000), and from the pesticide evaluations reported yearly by the Joint

FAO/WHO Meeting on Pesticide Residues (e.g. (FAO/WHO 2000, FAO/WHO 2001)). The eventual effect of processing depends on many factors. These include the initial concentration of the residue, the inherent properties of the pesticide itself (e.g. water solubility, systemic versus non systemic), as well as the product to which it has been applied (e.g. Burchat *et al.* 1998). These processing effects on residue levels are extremely important in evaluating the risk associated with ingestion of pesticides residues. For example, Zabik et al. even stated that their study on the effect of processing on the level of four pesticides in apples provides data to alleviate recent concern for the level of pesticide residues in food, particularly those in foods eaten by children (Zabik *et al.* 2000).

Due to the amount of pesticides authorised for use in agriculture and the different forms of processing applicable to one product, little detailed information is available on the influence of food processing on a specific pesticide-commodity combination. This is important because the behaviour and fate of the chemical varies with the pesticide as well as the crop (Burchat *et al.* 1998). A manufacturer, requesting authorisation of a certain pesticide, is obliged to produce information on food processing if relevant. However, this information is mainly confidential and even if available many gaps certainly remain. Furthermore, conditions under which the effect of processing on a chemical level are evaluated may not always reflect accurately the practice in real life.

If processing influences pesticide levels, it is relevant to have information on these items from food consumption surveys. However, it is not common practice in this type of survey to inform about food processing practices. For example, if an apple has been washed or peeled before consumption. In the absence of this type of information it may be possible to make general assumptions about processing, like 50% of the population peels the apple before consumption.

2.2.2 Unit variability within composite samples

Monitoring measurements are typically performed in composite samples of RAC (e.g. peppers are analysed in samples consisting of 20 individual commodities each). Recently, it was recognised that pesticides may be unequally distributed within such a sample (Hamey and Harris 1999, Harris 2000, Harris *et al.* 2000, PSD 1998a). Studies showed that individual units within a composite sample may contain high residue levels (Ambrus 2000, Andersson 2000, Earl *et al.* 2000, PSD 1998b, PSD 1999). To account for this phenomenon, the term variability was introduced in acute exposure assessment of pesticides.

In non-probabilistic modelling, variability is defined as the ratio of the 97.5^{th} percentile (P97.5) of residue level of an individual commodity to the mean composite sample residue level (FAO/WHO 2002). Variability was only defined for products with a unit weight larger than 25 g (Crossley 2000, FAO/WHO 2002). For unit weights lower than 25 g is was assumed that the composite residue data reflect the residue levels in the food commodity as consumed. Variability is included in the equation that estimates the acute intake of a certain pesticide from one specific commodity (national or international estimate of short-intake; NESTI or IESTI). This equation was defined at the FAO/WHO Geneva Consultation in 1997 (Crossley 2000, FAO/WHO 1997) and later refined in subsequent meetings (FAO/WHO 2001, PSD 1998a):

$$\text{NESTI or IESTI} = f_{processing} \frac{\min(U, LP) \times HR \times \nu + \max(0, LP - U) \times HR}{bw_{mean}}$$

where:

- LP is the large-portion consumption of the commodity (P97.5 of consumers only), kg food per day;
- U is the unit weight of one commodity, kg;
- HR is highest reported residue level in a composite sample, mg per kg;
- bw_{mean} is the mean body weight of the chosen (sub)population, kg;
- $f_{processing}$ is a factor accounting for processing and/or edible portions;
- ν is the variability factor – the factor applied to the composite residue to estimate the residue level in a high-residue unit

With insufficient data from measurements on individual units the FAO/WHO Expert Consultation recommended to assume (conservatively), when applying a variability factor, that all residue in a composite sample is present on one unit. Under this assumption ν equals the number of units in the composite sample (FAO/WHO 1997). If Codex sampling protocols are used, then the number of units per composite sample is 5 for large crops (unit weight > 250 g) and 10 for medium crops (unit weights 25-250 g). More recently, the FAO/WHO concluded that a ν of 7 for medium sized units could be used on a temporary basis until the database was further refined. The variability factor of 7 does not apply to granular soil treatment or leafy vegetables where the factor of 10 should be retained for medium sized units (FAO/WHO 2002). Unit weights are also important in probabilistic approaches, as will become evident in next chapter.

Guidelines on how to apply variability in a probabilistic approach are not available. The US Environmental Protection Agency (US EPA) developed a method for extrapolating from pesticide residue levels in composite samples to residue levels in single units ('decomposition method'; (US EPA 1999)). This method results in a new residue data set of individual commodities that can then be incorporated into a probabilistic exposure estimation model in order to estimate exposure to pesticide residues in foods. The accuracy of this methodology depends on the number of samples collected from the same population of commodities (number should preferably exceed 30) and on the number of units in the composite sample (N) relative to the number of samples (n). If N >> n and the number of samples is small (e.g. 7 samples of 100 apples) the accuracy of the method deteriorates (US EPA 1999).

To apply the variability factor successfully in exposure assessment, this factor should be representative of the level of variability to which people in real life can be subjected when consuming fruits and vegetables. Variability studies are not standardised as yet. Studies are performed on batches of individual commodities sampled from different locations, such as wholesalers and retailers, local and central markets, points of entry (for imported products) and processing industries. Variability studies may also be performed as part of field trials. All these studies result in variability factors that may be more or less representative of variability factors applicable to ready-to-eat products. The within-batch variability obtained from field trials may be smaller than that found in batches available for sale. Field trials are normally carried out under controlled circumstances, resulting in residue levels within a batch that are likely to be more uniform than that following commercial application of pesticides. When studied at the level of

retailer or (local and central) market the individual units of a composite sample may have been sorted according to size (e.g. fruit) or colour (e.g. red, yellow and green peppers) which will increase the residue level variability within a batch. Variability studies performed at the end of the distribution process will typically be most representative of variability factors applicable to products as consumed. However, these studies are not common. In authorisation of pesticides, default factors for variability (see above) are used when no variability study is available. It is however very questionable that these factors should also be applied when using monitoring results in probabilistic exposure assessment. These factors are fixed values and can therefore not be used as such in single simulations of a probabilistic exposure analysis. All the above mentioned factors complicate the application of variability in acute dietary exposure assessment of pesticides. In the next chapter several approaches are proposed to incorporate variability in the probabilistic approach to estimate the acute dietary exposure to pesticides.

Information on the variability factor for a certain pesticide-commodity combination is expected to be very limited. Some studies have been performed as mentioned above, but information remains scarce. Nowadays, Authorisation Committees ask for variability studies when a compound is acute toxic. However, in the past this was not requested so limited data will be available from this source. And if available, as mentioned above, the use of these factors is questionable in probabilistic exposure assessment of pesticides.

2.2.3 Percentage crop treated and levels below the limit of reporting

Another important issue in exposure assessment to pesticides is the treatment of samples that are reported to contain no residues (Loftus *et al.* 1992, US EPA 2000a). These 'non-detects' (NDs) do not necessarily contain no residue, but may have levels below the level (limit of reporting, LOR) at which laboratories or monitoring authorities are obliged to report. The status of the LOR used by the laboratory is often not clear. In pesticide exposure assessment the limit is commonly indicated as LOD (limit of detection) or LOQ (limit of quantification). Unfortunately, only residue levels higher than LOD or LOQ are reported, in spite of official IUPAC (International Union for Pure and Applied Chemistry) recommendations to always report the numerical values below LOD or LOQ limits if available (Cressie 1994, Currie 1999, IUPAC 1995).

The effect of the level assigned to the NDs on the estimated chemical intake of a population depends on several factors. These include the percentage of residue levels that are NDs, the level of the LOR relative to the levels monitored above this limit, and the percentage of the crop that has been treated with the pesticides (determines the percentage of NDs that can be considered to be real zeros). This issue is important in pesticide exposure assessment because in pesticide monitoring the majority of samples has residue levels below LOR.

The US EPA developed a method in which the percentage of NDs that are real zeros depends on the percentage of the crop that has been treated with the pesticide (US EPA 2000a). For the other NDs, that are estimated to contain residue and are therefore no real zeros, different approaches were recommended, such as assigning them either the LOR or 0.5 $\times$ LOR, or using statistical methods to estimate the values or distribution of values associated with the ND values (US EPA 2000a). In general, these statistical methods should be used only in situations where the NDs compromise less than half the data set and the rest of the data are normally or lognormally distributed. In pesticide exposure assessment, however, the number of NDs will often exceed 50% of the data set, making this approach less applicable when addressing dietary exposure to these chemicals.

3 PROBABILISTIC MODELLING OF DIETARY EXPOSURE TO PESTICIDES

3.1 Model description

This chapter describes a probabilistic model for the assessment of acute dietary exposure to pesticide residues. The model combines food consumption survey data and pesticide concentration data from monitoring programmes. The model allows for effects of food processing on residue levels between monitoring and ingestion, it can model unit variability either from available data or default assumptions, and it can use information on LOR and percentage crop treated to check whether NDs present a source of uncertainty. The model is only concerned with single-pesticide exposure modelling. The basic model is

$$y_{ij} = \frac{\sum_{k=1}^{p} x_{ijk}\ c_{ijk}}{w_i}$$

where y_{ij} is the intake of individual i on day j (in $\mu g \cdot kg^{-1} \cdot d^{-1}$), x_{ijk} is the consumption by individual i on day j of food commodity k (in g), c_{ijk} is the concentration of the pesticide in commodity k eaten by individual i on day j (in $mg \cdot kg^{-1}$), and w_i is the body weight of individual i (in kg). Finally, p is the number of food commodities accounted for in the model.

In the stochastic model the quantities x_{ijk}, w_i and c_{ijk} are assumed to arise from probability distributions for individual food consumption and body weight, $p(x_1,\ldots,x_p,w)$, and for pesticide concentrations in each food commodity, $p_k(c)$. In principle these probability distributions may be parametric (e.g. completely defined by the specification of some parameter values) or empirical (e.g. only implicitly and roughly defined by the availability of a representative sample). In this report we incorporate food consumption and body weight into the model as an empirical distribution. Pesticide residue levels will be addressed both as a parametric and empirical distribution.

3.1.1 Modelling of pesticide concentrations

Rationale

Residue concentrations in various food commodities are independent and can therefore be modelled by univariate distributions. Two approaches are possible:

1) Non-parametric modelling of residue levels

In the non-parametric approach, residue values are sampled at random from the available data and combined with the consumption data to generate a new distribution of exposure values. To assess the risk of exposure, percentiles of the exposure distribution are estimated. The disadvantage of this approach is that the results will be limited by the observations (another experiment will very likely result in other values). Therefore, in most cases the parametric approach is preferred.

2) Parametric modelling of residue levels

In the parametric approach, residue concentrations per commodity are sampled from parametric distributions. A special feature of residue data is that the large majority of measured concentrations (often more than 80%) is below LOR, resulting in skew residue concentration distributions with a large spike at zero and an extended tail to higher values. For statistical

Table 1. Number and mean pesticide residue level of samples with levels at or above the limit of reporting (LOR) for several pesticide–commodity combinations. Data are from monitoring programmes performed from 1997 till 2000 in the Netherlands.

pesticide	commodity	number of samples ≥ LOR	concentration ($mg \cdot kg^{-1}$)
bromopropylate	apple	33 (5)[1]	0.15 ± 0.12
	pear	74 (21)	0.21 ± 0.20
captan	apple	248 (38)	0.26 ± 0.44
	pear	77 (27)	0.32 ± 0.38
chlorfenvinphos	carrot	79 (39)	0.16 ± 0.26
chlorpropham	potato	257 (66)	1.71 ± 2.36
diphenylamine	apple	36 (7)	1.00 ± 1.07
iprodione	carrot	96 (46)	0.11 ± 0.15
pirimicarb	apple	71 (11)	0.07 ± 0.07
tolyfluanid	pear	128 (33)	0.21 ± 0.26

[1] Number in brackets is percentage samples at or above LOR.

modelling a two-step procedure should be applied when addressing residue levels parametrically: 1) modelling the presence of a concentration ≥ LOR on food products with a binomial distribution with a parameter *p* representing the probability of a reported residue level, and 2) modelling residue levels ≥ LOR with a parametric distribution.

Methods

To establish which type of distribution fits pesticide residue levels best we performed a study in which we modelled residue data in randomly selected foods. We chose those pesticides that were most frequently found to be ≥ LOR during 2000 in the Netherlands. Pesticide residue data were derived from the KAP-database (Quality Programme for Agricultural Products), in which annually more than 200,000 records of measurements originating from Dutch food monitoring programmes for meat, fish, dairy products, vegetables and fruit are stored. The selected pesticide-commodity combinations are listed in table 1. We used BestFit (version 2.0d, Palisade Corp., Newfield, NY) to determine the top ranking of accepted distributions for the selected combinations. BestFit is a decision tool, which can be linked to Excel to fit more than 21 distribution types to data. It performs statistical tests to compare quality of fit and ranks distributions by three goodness-of-fit statistics. In this study we used the Anderson-Darling test which is similar to the Kolmogorov-Smirnov test, but places more emphasis on the tail values. All tests are very sensitive to the number of values. The BestFit programme was used to select the number of intervals for the data classification prior to distribution fitting. The method for determining the distribution parameters was the maximum likelihood estimators (MLEs). Graphs were used to assess visually how well distributions agreed with the input data. Both test statistics as graphs should be used in interpreting the results.

Results

In total 10 pesticide-commodity combinations were tested. The distributions that were accepted four times or more are listed in table 2. Modelling lognormal distributions to residue levels was acceptable for all pesticide-commodity combinations. PearsonVI distribution, second best, was rejected for one combination, namely tolyfluanid-pear. This outcome confirms the results of a study conducted by Voet *et al* (1999). Products with at least 30 positive measurement values of iprodione were taken to explore which distributional type was suitable. The lognormal and the

Table 2. Percentage[1] of times a distribution was acceptable. Only samples with pesticide residue levels at or above the limit of reporting were used.

distribution	% accepted
Lognorm	100
Lognorm2	100
Pearson VI	90
Expon	70
Inverse Gaussian	70
Gamma	50
Pearson V	50
Weibull	50
Beta	40

[1] Only those distributions accepted four times or more are displayed.

Pearson VI turned out to have an adequate fit for iprodione content in endive, cabbage lettuce, strawberry, carrot and currant.

Conclusion

Since residue data are positive, positively skewed and originate by mechanisms generating the lognormal distribution under a variety of biological circumstances (Crow & Shimizu, 1988) the lognormal was chosen to proceed with. At least in those situations where not contradicted by the data. Of course, in future research when more data may be available, the choice of distributional form should be reinvestigated.

3.1.2 Modelling of processing

Rationale

Pesticide levels in food as eaten may differ from levels in the product as measured in monitoring programmes (typically raw product) due to processing (§2.2.1). In general we assume the following model:

$$cpos_{ijk} = f_k \cdot cr_{ijk}$$

where cr_{ijk} is the pesticide concentration in a raw agricultural commodity k (RAC), and f_k a processing factor for a specific combination of RAC and processing type with values typically between 0 and 1. Occasionally this factor may exceed 1. The user of the model should specify processing factors for each commodity – processing type combination as defined in the food consumption database. For this purpose it is advised to maintain a database of processing factors, indexed by pesticide, RAC and processing type. Before running the model it may then be necessary to specify how the processing factors are derived (e.g. from the data base entries and/or from other information). For example, if no processing factors are known for captan in pears, it may be decided to use the corresponding factors for captan in apples.

Often the information will be limited and this may be entered in the probabilistic model by specifying uncertainties. A practical proposal is to specify for each processing factor two values, namely $f_{k,nom}$ and $f_{k,upp}$, the nominal value (typically some sort of mean from an experimental study) and an upper 95% confidence limit, respectively. The $f_{k,upp}$ is typically set by an expert (even when statistical information on variability of the factor is available, there will often be uncertainty due to

appropriateness of the processing study for the target population). The upper limit should be such that experts easily agree that the limit is not too low.

A typical database entry might thus read:

pesticide	RAC	processing	$f_{k,nom}$	$f_{k,upp}$
captan	apple	washing	0.5	0.7

and, confronted with the need to have processing factors for pears in a specific exposure analysis, an expert judgement may be:

pesticide	RAC	processing	$f_{k,nom}$	$f_{k,upp}$
captan	pear	washing	0.5	0.8

In probabilistic modelling processing factors can be used in either of three ways (for each RAC k to be chosen by the user):

1. No processing factor: take $f_k = 1$. This is in most (but not all) cases a worst-case assumption. No data on processing are needed and therefore this route is useful as a first step.
2. Fixed value: use $f_k = f_{k,upp}$. Available information on processing effects is used, although in a cautionary way (in accordance to the precautionary principle). Note that $f_{k,nom}$ values need not to be specified when using this approach.
3. Distribution: The logarithms of $f_{k,nom}$ and $f_{k,upp}$ are equated to the mean and the 95% one-sided upper confidence limit of a normal distribution, respectively. This normal distribution is thus specified by a mean $\ln(f_{k,nom})$ and a standard deviation $\{\ln(f_{k,upp}) - \ln(f_{k,nom})\}/1.645$. Values are drawn from this distribution in probabilistic analysis.

Values equal to 0 are replaced by a low user-specified value (e.g. 0.01); this is useful computationally to avoid problems with logarithms.

Methods

To study the effect of processing we used residue data of captan in raw apples of the Dutch Inspectorate of 1997 till 2000. We chose this combination because captan is a pesticide often found in this product at levels ≥ LOR. In total 570 composite samples (n = 20) of apples were analysed of which 248 contained levels of captan ≥ LOR (0.01 $mg \cdot kg^{-1}$). The mean level of captan in these apples was 0.26 ± 0.44 $mg \cdot kg^{-1}$, ranging from 0.01 $mg \cdot kg^{-1}$ to 3.50 $mg \cdot kg^{-1}$. Samples with captan levels < LOR were considered to contain no residue.

The apple consumption data of the Dutch population were taken from the DNFCS 97/98 (Kistemaker *et al.* 1998). This survey sampled 6,250 respondents (1-97 years) who recorded and weighed all food consumed during two consecutive days. This resulted in 12,500 single consumption days. In the survey apple was consumed on 7,737 days (62% of all survey days). The mean consumption (including the 'non-consumption' days) was 61 g per day. To link the consumption data to the residue levels in raw apples, the consumption of food products

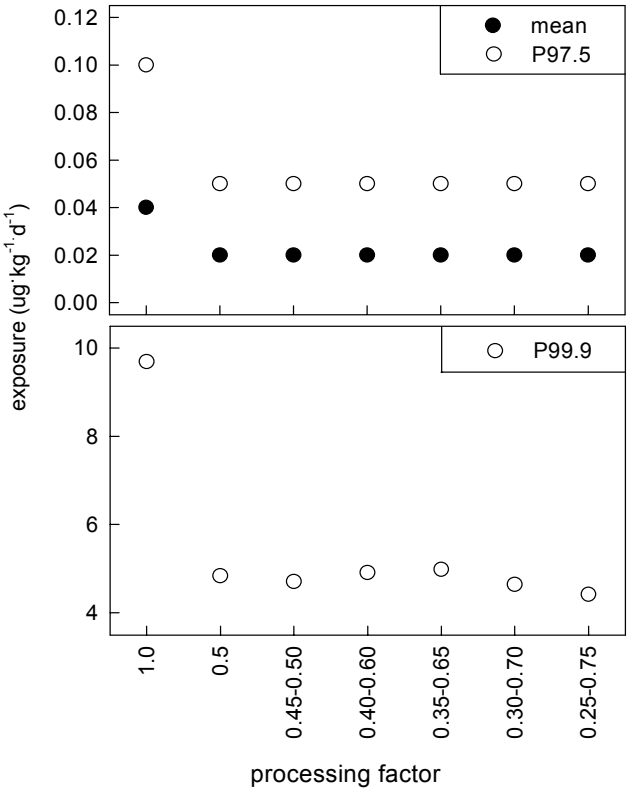


Figure 1. Effect of processing on the exposure ($\mu g \cdot kg^{-1} \cdot d^{-1}$) to captan through the consumption of apples in the Dutch population.

containing apples was converted to the consumption of raw apples using the recipe database CPAP (Conversion of food products to Primary Agricultural Products; (Dooren *et al.* 1995)).

The effect of processing on captan levels in apples was varied as follows: no effect of processing, processing equals 0.5 (fixed level), and processing equals 0.5 but with a certain amount of uncertainty. In this last case processing was assumed to be, with a 95% certainty, between 0.45-0.55, 0.40-0.60, 0.35-0.65, 0.30-0.70 or 0.25-0.75. For this the processing factor was modelled as a normal distribution with a mean of 0.5 and a standard deviation of 0.025, 0.05, 0.075, 0.10, and 0.125, respectively. By increasing the uncertainty of processing, we intended to show how sensitive the model was for variation in this parameter. In this example we assumed that 100% of the population consumed processed apples. The selected processing factors were never lower than 0.

The probabilistic analyses were performed with spreadsheet models written in Excel (version 7.0, Microsoft Crop., Redmond, WA) and with the @Risk add-in (version 3.5.2, Palisade Corp., Newfield, NY), using Latin Hypercube sampling. Individual consumption data and residue levels of individual composite samples of apples were used. Each Monte Carlo simulation was run for 5,000 iterations. During the simulation every captan residue level selected was multiplied with one (no processing), 0.5 (fixed level) or a number selected from a normal distribution (mean = 0.5) with varying standard deviations. The resulting captan level was then multiplied with a selected intake of apple from the consumption database. The exposure to captan was divided by the

corresponding body weight to give intake (μg) per unit body weight per day. We calculated the mean and P97.5 and P99.9 of each distribution.

Results

The mean and P97.5 and P99.9 of the intake distribution are plotted in figure 1 for the different levels of processing. It is evident that by applying a processing factor of 0.5 to all residue levels the exposure to captan, compared to the situation that no processing is applied, was halved. It is clear from this figure that the mean and P97.5 of the intake distribution were not sensitive to an increasing amount of uncertainty around the estimation of the processing effect. P99.9 fluctuated somewhat more, due to the fact that this percentile is more sensitive for extreme values occurring both in the consumption and residue database.

Conclusion

We conclude that processing has a major effect on the magnitude of exposure to pesticides, and should therefore be considered in exposure calculations if information is available, especially because in most cases the regulatory threshold risk is at the upper tail of the exposure distribution. However, it should be kept in mind that the results of any exposure assessment are dependent on the data used and assumptions made. The final effect of processing on the upper tail of the distribution will depend on the magnitude of the processing effect on the residue level and the contribution of the consumption of processed foods to the total consumption of the raw agricultural commodity.

3.1.3 Modelling of unit variability within composite samples

Rationale

Monitoring programmes analyse pesticide levels in an agricultural product cm_k typically as a homogenised composite sample (§2.2.2). Such a composite sample is composed of nu_k units with unit weight wu_k. The weight of a composite sample is therefore $wm_k = nu_k \times wu_k$. This weight is often larger than a consumer portion. For example, a typical composite sample of 20 sweet peppers weighs 3.2 kg, whereas daily consumer portion weights in the DNFCS 97/98 range from 0.08 g to 458 g.

How should monitoring data be used to estimate the individual raw commodity pesticide level cr_{ijk}? Although the mean residue level of a composite sample (cm_k) may be a fair estimate of the mean residue level an individual unit (cr_{ijk}), the variability of cm_k is not appropriate for the estimation of the variability of cr_{ijk}. In smaller portions extreme values may occur more readily, and thus acute exposures may be higher than when using composite sample residue levels.

In non-probabilistic modelling of acute dietary exposure the unit-to-unit variability has been addressed by the definition of a variability factor v (§2.2.2). Values for v can be obtained by measuring individual units within a composite sample. In practice such data are mostly available from field trials, although for estimating the exposure in real life it would be more appropriate to derive unit variability from monitoring samples (§2.2.2). We therefore advise the use of field trial values for v only when data from monitoring programmes are not available. Because, the lognormal distribution is considered an appropriate model for positive residue levels (§3.1.1), we will also assume a lognormal distribution for unit residue levels. Let this distribution be characterised by μ (mean) and σ (standard deviation), the parameters of the log-transformed

concentrations *lc*. The variability factor v can be converted into the standard deviation σ (see below). Upper-tail percentiles of this lognormal distribution are influenced in two opposing ways by the magnitude of the variability factor:

1. Because of more spread, the percentiles $c_q = e^{\mu + z_q \sigma}$ increase with σ relative to the median e^{μ} (z_q is the $100q$ percent point of the standard normal distribution);
2. However, the median e^{μ} decreases with σ relative to the expected value (mean) *E(c)* according to: $e^{\mu} = E(c) \cdot e^{-\frac{1}{2}\sigma^2}$.

Composite sample levels cm_k are estimates of *E(c)*. Percentiles of the unit distribution for a batch with expected value (mean) cm_k are therefore equal to

$$c_q = cm_k \cdot e^{-\frac{1}{2}\sigma^2 + z_q \sigma}$$

The combined influence in this simple case is that c_q increases with σ for high percentiles ($z_q > \sigma$), but decreases with σ for relatively low percentiles ($z_q < \sigma$).

The following approaches to model sample variability should be incorporated in the model:

1. Use estimated values of v
2. Use default (conservative) values of v
3. Use weight ratios to define maximum variability of residue levels in consumed portions
4. Use weight ratios in combination with a unit homogeneity assumption

These approaches are described in more detail below. In all cases we assume that the majority of residue levels is derived from a representative sample of composite samples. Alternatively, surveys may be available in which residue levels have been collected for individual units. These data can be used directly, although care is needed to reflect the structure of between-batch/within-batch variability (Hamey 2000). In the DEEM model approaches related to 1 and 4 are implemented (US EPA 2000b).

1) Use estimated values of v

In this approach it is essential to discern between-batch variability from within-batch variability. Typically, variability factors are calculated for units from one field trial or commercial batch, although such batches are not always clearly defined. Variability factors describe the variability between units within batches. The proposed approach is as follows:

- If individual unit levels are available, estimate σ as the standard deviation of the logarithmically transformed concentrations. When data from several batches or field trials are available, pool the estimates.
- If a value for v is available that can be interpreted as the ratio between P97.5 and the median of a lognormal distribution of unit residue levels in one batch, then, with μ and σ representing the mean and standard deviation of the log-transformed concentrations *lc* , we have

$$v = \frac{e^{\mu + 2\sigma}}{e^{\mu}} = e^{2\sigma}$$

or $\sigma = \frac{1}{2}\ln(v)$

- If a value for v is available that has been calculated as the ratio between the maximum and the mean of n individual values, then σ can be calculated as $\sigma = z_q - \sqrt{z_q^2 - 2\ln(v)}$, where z_q is the quantile of the standard normal distribution corresponding with the maximum of a sample of n units. According to Blom (Blom 1958) and Harter (Harter 1961) it can be approximated very accurately by $z_q = \phi^{-1}((n - \alpha)/(n - 2\alpha + 1))$, where $\phi^{-1}(\ .)$ is the inverse cumulative standard normal distribution function, and $\alpha = 0.315065 + 0.057974u - 0.009776u^2$, with $u = \log_{10}(n)$.
- For each iteration i in the Monte Carlo simulation, obtain for each commodity k a simulated intake x_{ik} and a simulated composite sample residue level cm_{ik}.
- Calculate the number of unit intakes nux_{ik} in x_{ik} (round upwards) and set weights w_{ikl} equal to wu_k, except for the last partial intake that has weight $w_{ikl} = x_{ik} - (nux_{ik} - 1)wu_k$.
- Draw nux_{ik} simulated log-concentration values lc_{ikl} from a normal distribution with mean $\mu = \ln(cm_{ik}) - \frac{1}{2}\sigma^2$ and standard deviation σ.
- Backtransform and sum to obtain the simulated concentration in the consumed portion:

$$cr_{ik} = \sum_{l=1}^{nux_{ik}} w_{ikl} e^{lc_{ikl}} \Big/ x_{ik}$$

Variability between units is often quantified with the coefficient of variation (CV) rather than the variability factor v. With v defined as the ratio between P97.5 and median, the relation between these two parameters in a lognormal distribution is $CV = \sqrt{v - 1}$ or $v = 1 + CV^2$.

2) Use default (conservative) values of v

In the absence of reliable data a default value for v (e.g. v = 5 or 7) may be used. This approach is almost equal to the approach described above. However, in order to be conservative, the variability factor is only used to obtain a larger spread in unit pesticide levels, not to lower the estimate of the median value μ. This can be interpreted as assuming that composite samples are obtained from very homogeneous sets of units (with effectively v = 1), and that this homogeneity does not apply to consumer portions. Consequently, in this approach the unit log-concentrations are drawn from a normal distribution with mean $\mu = \ln(cm_{ik})$ and is otherwise the same as described above.

3) Use weight ratios to define maximum variability of residue levels in consumed portions

When no variability factors are available another approach is possible. The conservative assumption can be made that a composite sample is constructed by combining an appropriate number of portions of size x_{ijk} where these portions are independent and random samples from the total population. If this is true the unit-to-unit variability will be larger than in the more realistic situation where composite samples consist of units with a shared history. Therefore, the use of this assumption results in a conservative estimate of exposure.

Table 3. Percentiles (%) of exposure to methamidophos via the consumption of sweet peppers.

all days	95	97.5	99	99.9	99.99
positive intakes	34.1	67.1	86.8	98.7	99.9

Under the assumption given above the variability of concentrations in consumed portions of weight x_{ik} can be related to the variability of monitoring measurements of weight wm_k . Basic statistical considerations show that standard deviations or coefficients of variation should be multiplied by a factor $\sqrt{wm_k / x_{ik}}$. If concentrations in the total population of portions of a certain size are modelled with a lognormal distribution, then simulated concentrations for a portion x_{ik} can be obtained by sampling from this distribution. The proposed approach is as follows:

- Estimate the mean μ and the standard deviation σ_{comp} in the set of log-transformed composite sample residue concentrations: $lcm_k = \ln(cm_k)$.
- Estimate the coefficient of variation at the original scale as $CV_{comp} = \sqrt{e^{\sigma_{comp}^2} - 1}$.
- For each iteration i in the probabilistic analysis, obtain for each commodity k a simulated intake x_{ik} .
- Correct the coefficient of variation for portion size: $CV = CV_{comp} \sqrt{\frac{wm_k}{x_{ik}}}$.
- Draw a simulated log-concentration lc_{ik} from a normal distribution with mean μ and standard deviation $\sigma = \sqrt{\ln\left(1 + CV^2\right)}$.
- Backtransform to obtain the simulated concentration in the consumed portion: $cr_{ik} = e^{lc_{ik}}$.

4) Use weight ratios and the assumption of homogeneous units

A variation on the previous approach is to add the assumption that units are always homogeneous. In fact, this assumption is also implicit in the first two approaches. This means that pesticide levels in portions smaller than unit weights are treated the same as concentrations in a unit portion. In that case the *CV* correction factor $\sqrt{wm_k / x_{ik}}$ is replaced by $\sqrt{wm_k / \max\left(wu_k, x_{ik}\right)}$. These four possible approaches to model variability are illustrated below.

Methods

Residue data of methamidophos in raw sweet peppers from the Dutch Health Inspectorate for 1999 were used, because a variability study was performed during that year (Schee 2000). In total 283 composite samples (consisting of 20 peppers each) were analysed. The unit weight of one pepper is 160 g, so the weight of a composite sample is 3.2 kg. In 37% of the samples (106 samples) levels of methamidophos ≥ LOR (= 0.01 mg·kg $^{-1}$). To study the unit variability, individual peppers of six composite samples were analysed. Ratios of maximum residue level in single units to the composite sample residue level were used as estimates of the variability factor *v*. The individual estimates of *v* were 3.15, 2.80, 4.14, 5.96, 7.72 and 7.42 (mean 5.20).

Table 4. Exposure to methamidophos ($\mu g \cdot kg^{-1} \cdot d^{-1}$) via the consumption of sweet peppers for different approaches to model variability[1].

% of days	P95	P97.5	P99	P99.9	P99.99
0. no correction for variability	0.010	0.064	0.23	1.3	8
1. ν (= 5.2) from data	0.007	0.050	0.21	1.2	9
2. ν = 10 (conservative)	0.010	0.070	0.31	3.1	13
3. weight ratio	0.006	0.098	0.71	23.6	177
4. weight ratio + hom. units	0.008	0.076	0.43	6.1	21

[1]For more details see text.

The consumption data consisted of sweet pepper intake on 12,500 consumption days (DNFCS 97/98; (Kistemaker *et al.* 1998)). Sweet peppers were consumed in portions ranging from 0.08 g to 458 g on 2,533 days (20%). Thus positive intakes of methamidophos via sweet peppers were expected only in 7.4% of all cases (37% of 20%). This means that percentiles in the distribution of all days will correspond with much lower percentiles in the distribution of only the positive intakes of methamidophos (table 3). For example, P95 for 'all days' corresponded with P34.1 of the exposure distribution when only the positive intakes were considered (table 3).

Probabilistic analyses were performed with a general-purpose statistical programming language, GenStat (GenStat 2002; see §3.3). For this positive levels were simulated for each of the 2,533 positive consumption days, and the conversion table was applied (table 3) to obtain percentiles for the total population (including non-consumers). Concentration values were simulated without regard for unit variability, and via the four approaches mentioned above. Individual data for both residue and consumption levels were used. The exposure to methamidophos was divided by the corresponding body weight to give exposure (μg) per unit body weight per day. We calculated different percentiles of each exposure distribution.

Results

Results are listed in table 4. It is evident that with approach 1 (ν based on real data) moderate percentiles tended to get lower, while very high percentiles (P99.99 in this case) were higher than the uncorrected value. In approach 2 (assumes homogeneous composite samples, but a large default ν = 10) all percentiles > 50^{th} of the positive intakes (P97.5 and higher percentiles for the whole population; table 4) were higher than the uncorrected values, but the differences were small. Approach 3 (correction based on the weight ratio between composite sample and consumed portion) resulted in very high percentiles, mainly due to the large number of very low consumption levels of sweet peppers. Incorporating the unit homogeneity assumption in this method (approach 4) lowered the percentiles considerably, although values remained more conservative than those of approaches 1 and 2.

Conclusion

Incorporating variability into the probabilistic exposure assessment of pesticides influences the outcome. However, more experience with these approaches to include unit variability effects in stochastic exposure is necessary.

3.1.4 Modelling of levels below the limit of reporting

Rationale

Most monitoring measurements of pesticides are nondetects (NDs), i.e. no quantitative measurements are reported. When a pesticide enters the food chain only via crop treatment, and when the percentage of crop treated is (approximately) known to be $100p_{crop\text{-}treated}$, then this knowledge may be used to infer that $100(1\text{-}p_{crop\text{-}treated})$ % of the monitoring measurements should be real zeroes, contributing nothing to pesticide intake, whereas other NDs in the monitoring data could have any value between zero and the reporting level (LOR). For $100(p_{nondetect} - p_{crop\text{-}treated})$ % of the monitoring measurements, 0 and LOR represent best-case and worst-case estimates, respectively. A simple way (first step) to consider the uncertainty associated with NDs is to compare exposure distributions for these two situations.

Methods

To study the effect of residue levels assigned to NDs, we performed two studies. In the first study, we applied 0 or LOR = 0.01 $mg{\cdot}kg^{-1}$ (two most extreme possibilities) to the NDs of captan in apples as a function of percentage NDs in the residue data set (see §3.1.2). The percentage samples below LOR was varied from 0% up to 90%. The amount of samples with levels above LOR was unchanged throughout the analyses (n = 248). For example, 10% NDs meant 27 samples below LOR, 20% NDs equalled 62 samples below LOR, etc. The percentage crop treated was set at 100%, which represents the 'worst-case situation'. The probabilistic analyses were performed with spreadsheet models written in Excel (version 7.0, Microsoft Crop., Redmond, WA) and with the @Risk add-in (version 3.5.2, Palisade Corp., Newfield, NY) using individual consumption data and residue levels of individual composite samples of apples as described in §3.1.2. Each simulation was run with 5,000 iterations, using Latin Hypercube sampling. The exposure to captan was divided by the corresponding body weight to give intake (μg) per unit body weight per day. We calculated the mean, P97.5 and P99.9 of each distribution.

In a second study, the sensitivity of exposure percentiles to the treatment of NDs was investigated further using data of methamidophos in sweet peppers (§3.1.3). Initially we used 106 methamidophos levels in sweet peppers (range = 0.02-2.75 $mg{\cdot}kg^{-1}$; median = 0.28 $mg{\cdot}kg^{-1}$) ≥ LOR (= 0.01 $mg{\cdot}kg^{-1}$; 37% of all analyses). To check the importance of the level assigned to NDs these data were reduced to 79 levels (median = 0.37 $mg{\cdot}kg^{-1}$) ≥ LOR (= 0.1 $mg{\cdot}kg^{-1}$) and 13 levels (median = 1.4 $mg{\cdot}kg^{-1}$) ≥ LOR (= 1 $mg{\cdot}kg^{-1}$). The percentage of NDs was varied from 63% (real situation) to 90% and 99%. Probabilistic analyses were performed with GenStat (GenStat 2002) by simulating positive exposure days for each of the 2,533 positive consumption days, and applying table 3 to obtain percentiles for the whole population. The same set of random values was used for all situations. Individual consumption data and residue levels of composite samples of peppers were used.

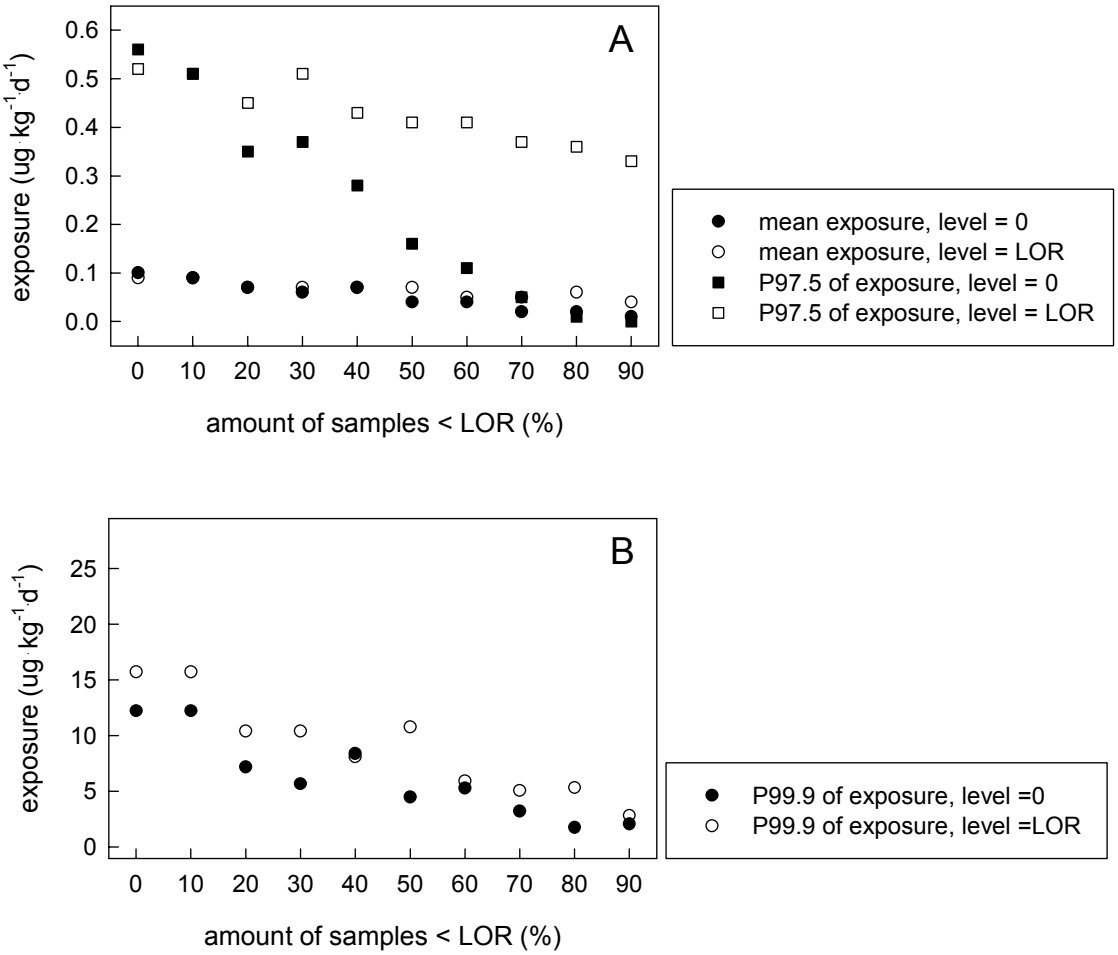


Figure 2. Effect of % samples with levels below the limit of reporting (LOR) and the level assigned to the non-detects (LOR or zero) on the exposure ($\mu g \cdot kg^{-1} \cdot d^{-1}$) to captan through the consumption of apples in the Dutch population. A: mean and P97.5 of exposure; B: P99.9 of exposure.

Results

The results of the first study are plotted in figure 2. It is evident that the level assigned to NDs (0 or LOR) affected the upper percentile of the distribution when the percentage of NDs was 20% or more. This was most clear for P97.5 (upper panel). The mean and P99.9 of the intake distribution were fairly insensitive to the level assigned to the NDs (fluctuations were due to the fact that different simulations were performed). When 70% or more of the samples were below LOR, P97.5 approached zero when level assigned to NDs was zero.

Figure 3 shows the estimated percentiles for nine combinations of LOR and percentage of values ≥ LOR (37%, 10% and 1%; %det). Each panel shows percentiles from simulations where NDs were replaced by zero (symbol 0) and LOR (symbol 1). It can be seen (for example in panel LOR = 0.10 $mg \cdot kg^{-1}$, %det = 1) that intermediate percentiles (e.g. P95, P97.5) were influenced most, whereas lower percentiles (e.g. P70) and very high percentiles (e.g. P99.99) were not or less affected.

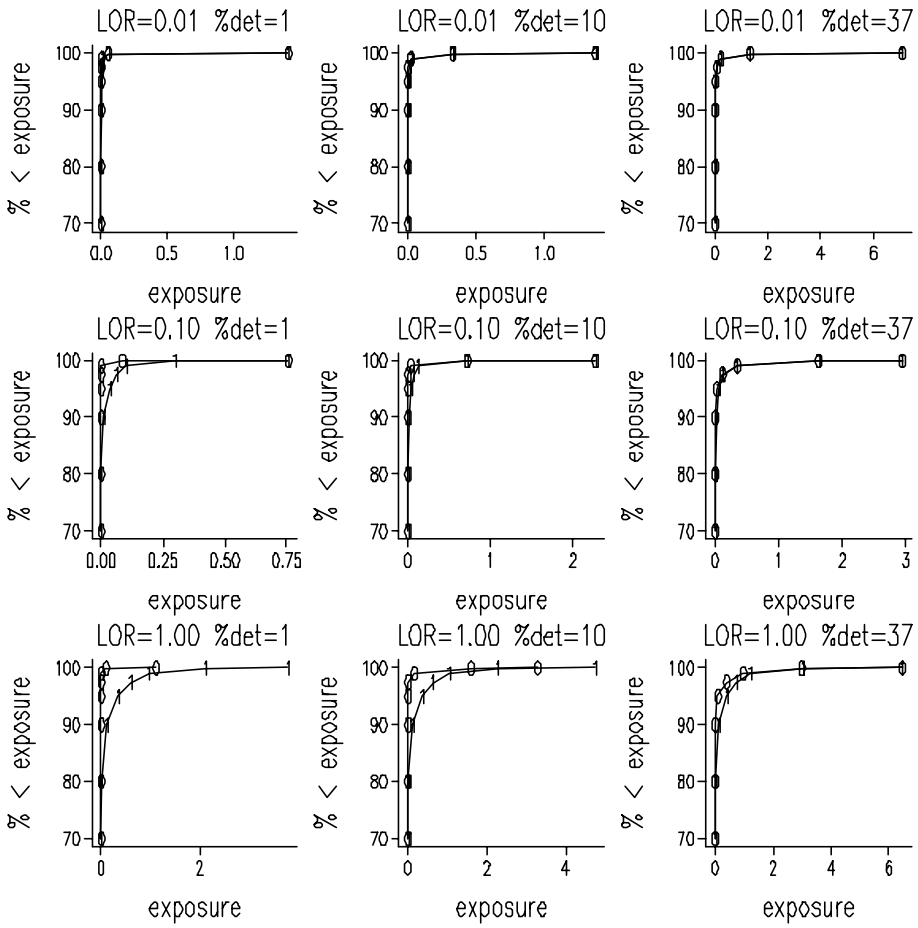


Figure 3. Residue levels of methamidophos in sweet peppers. Exposure percentiles with nondetects replaced by zero (0) or LOR (1) for different combinations of LOR and % detect concentrations. Right upper graph corresponds with real LOR and %detect.

Furthermore, changes became more pronounced when LOR increased (from top to bottom in figure 3) and when the percentage of values ≥ LOR became lower (or percentage of NDs larger; right to left). The panel in the right upper corner (LOR = 0.01 $mg \cdot kg^{-1}$, %det = 37) corresponds with the real situation, and therefore little problems with NDs are expected for methamidophos in sweet peppers.

Conclusion

We conclude from the results that the effect of levels assigned to NDs on the dietary exposure to pesticides depends on the percentage of samples below LOR in the residue database and the level of LOR assigned. In pesticide exposure assessment the amount of NDs is almost always substantial. Apart from the amount of samples below LOR, the effect of the level assigned to the NDs on the exposure to a pesticide also depends on the relative magnitude of LOR versus residue levels found in the monitoring programme and the percentage of crop treated.

3.1.5 Specification of model inputs and uncertainty analysis

We distinguish between choices on the model and those on model input.

1. Model: choices once made on the model are considered fixed, i.e. they add no uncertainty to the model outcomes. The following choices are relevant:
   a. consumer population: total or a subset of certain ages
   b. type of exposure calculation: acute (daily intakes) or chronic (usual intakes)
   c. type of distribution of residue data: empirical or parametric
   d. for parametric models: pooling of parameters over products (yes or no)
   e. approach to incorporate unit variability and processing
2. Model input: model inputs represent the numeric data that enter the model. In general they will have an associated uncertainty. In order to allow future extensions of the model to evaluate the uncertainty of model outcomes it is necessary that something is known about these uncertainties. Model inputs are:
   a. Food consumption: this data is considered to be a representative sample of the relevant population; uncertainty is implicit in the sample, and can be evaluated with re-sampling procedures (e.g. bootstrap)
   b. Pesticide monitoring levels: in case of modelling empirical data re-sampling procedures can be used to assess the uncertainty; in case of parametric modelling the uncertainty can be expressed as standard errors of the parameters.
   c. Percentage crop treated
   d. NDs: in a simple first approach the maximal uncertainty from NDs can be estimated from a comparison of simulations with substitution of 0 and LOR for NDs.
   e. Variability factors and unit weights (approach 1 or 2) or the weight of (mixture) samples and unit weights in the monitoring programme (approach 3 or 4)
   f. Processing factors to describe the effect of processing on pesticide residue levels.

Model inputs 3, 5 and 6 can be specified in general (i.e. applicable for all products) or specific values for products can be given. For inputs 3-6 one should specify either conservative values, or nominal values in connection with information on the uncertainty in these values. In order to make this as practical as possible this information should be requested in the form of a limit (either upper or lower), which should be considered conceptually as a one-sided 97.5 % confidence limit. The programme will translate the nominal and limit values into a normal uncertainty distribution on an appropriate scale (logistic for factors restricted to the interval (0,1), lognormal for non-negative inputs such as sample weight).

Figure 4 shows an example of a possible main input sheet of a Monte Carlo Exposure Analysis model (not yet all relevant choices are included). On a specification sheet additional inputs can be specified in a format as follows:

% crop treated

pesticide	RAC	$p_{crop\ treated,\ nom}$	$p_{crop\ treated,\ upp}$
captan	apple	0.25	0.75

Monte Carlo Risk Analysis				Pesticide:			Ipro					
Inputs				Model				Output				
Concentration data				Process								Print?
Weights of monitoring samples: see				see specification sheet				Summary of database				no
specification sheet								Summary of simulated intakes				no
Limit of reporting ('LOD'):				Monte Carlo model				Percentiles				yes
	LOR (ppm):		0.02					Summary upper quantile distributio				no
Summary data:			no	# simulations			10000	if yes:	Concentration:			*
if yes, data on file xxxx_summ.xls									Quantile (if conc=*):			98
				empirical concentration data:			yes	Summary total distribution				no
Food consumption data				if no, then (parametric modelling)				Top 10 intake				no
				pooling of means/variances:			yes	Top 10 consumption				no
Age restrictions:			no	if yes, then				Top 10 residues				no
if yes:				automatic pooling			no	Median 9 intake				no
	min. age:		0	if no, then choose:				Median 9 consumption				no
	max. age:		4	step of process (1/2/3):			1	Median 9 residues				no
								Program settings				yes
Seq. day restrictions:			no	seeds for pseudo-random sampling								
if yes, restrict to consumption data				(choose 0 for time-based values)								
of day (1 or 2):			1	days			0					
				persons			0					
				concentrations			0					

Figure 4. Possible main input sheet of a Monte Carlo Exposure Analysis model.

variability

pesticide	RAC	unit weight *(wu)*		# units in comp. sample	variability *v*	
		nom	upp		nom	upp
captan	apple	150	160	20	5	7

processing factors

pesticide	RAC	processing	$f_{k,nom}$	$f_{k,upp}$
captan	apple	washing	0.5	0.7

In principle, uncertainty analyses take a model as given and calculate the contribution of uncertainties in the model inputs to the uncertainty of specified model outputs. Stochastic exposure analysis itself is already a kind of uncertainty analysis, where uncertainties about food consumption and pesticide levels are translated to uncertainties in pesticide exposure. It is therefore natural to extend this basic model with other uncertainties relevant for pesticide exposure assessment, such as uncertainties on processing effects, on pesticide levels < LOR, due to limited amount of data, etc. As relevant model outputs, in pesticide exposure assessment usually properties of the upper tail of the pesticide intake distribution are specified, e.g. P95 or the proportion of daily intakes exceeding a reference value. Basically, uncertainties in the inputs can be specified in three ways:

1. By a direct specification of an input distribution, e.g. a normal distribution with mean and standard deviation.
2. By specifying alternative conditions that may apply, e.g. ND measurements may all have a pesticide level equal to zero or LOR.
3. Implicitly, by giving uncertainties if the data necessary for the model are limited (e.g. only a few measurements on a certain product). Re-sampling procedures (e.g. bootstrap) can be used to quantify the uncertainty in the model outputs due to this factor.

3.2 How to deal with limited information

In a probabilistic model, a distribution of both food consumption data and residue level data are used. When residue data are scarce parametric modelling becomes important.

3.2.1 The choice between a parametric and non-parametric approach

How many residue data are required for a sensible calculation of upper-tail percentiles in the exposure distribution based on a non-parametric approach? The rule of thumb can be used that the chosen percentile should be contained directly in the data. For example, at least 20 measurements are needed to estimate P95 and at least 100 measurements to estimate P99. More generally, the number of measurements per food commodity (*n*) should at least equal $1 \div (1-P/100)$ to allow a rough estimate of P of the residue concentration distribution to be made. Of course, exposure assessments are only coarse with this minimum amount of data and larger sample sizes per food commodity are certainly worthwhile.

In situations where the number of measurements poses a problem, an appropriate exposure analysis should be based on further modelling. Essentially, lack of data is compensated by *a priori* assumptions. Assuming a simple distributional form for residue data, the number of measurements can be smaller (at least 10). Because NDs provide no information about variability, we should count the number of positive measurements. Figure 5 shows which approach can be used best depending on the total number of measurements and the number of measurements $\geq$ LOR. In principle, such a choice can be made separately for each food commodity.

3.2.2 Grouping of products

Rationale

When data are limited, the parametric approach may have potential. The distributional form of pesticide residue level data is modelled lognormal (see §3.1.1) with parameters μ (mean) and σ (standard deviation). However, estimation of these parameters for all products is often difficult because data on residue levels in specific products are scarce or even missing. In those cases, grouping of products into product groups consisting of 'comparable products' increases the number of measurements per group and may give sufficient data to estimate both parameters. For this we assume that residue distributions are the same for the grouped products. A related question is the reliability of estimates based on a small number of degrees of freedom (df). The following procedure is designed to cope with the above problems (figure 6).

1. *Step 1.* For each product μ and σ are estimated. Then, products are assigned to product groups composed of related products (e.g. product groups consisting of cabbages or all kinds of berries). The homogeneity of variances in different product groups can be assessed using Bartlett's test (Snedecor and Cochran 1980). The test statistic determines whether variances are to be pooled automatically ($p>0.05$) or not ($p\leq 0.05$). For homogeneous groups, variances are pooled within product groups. For non-homogeneous groups, products are assigned to subgroups (within product groups) manually and the homogeneity of variances is tested again. This process of assigning products to subgroups is repeated until all groups have homogeneous variances. After pooling the variances, an overall test for

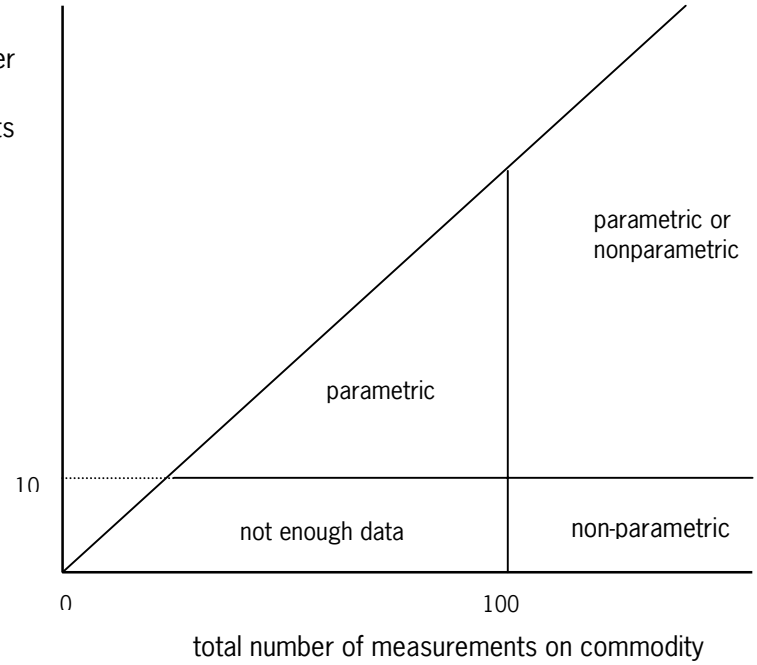


Figure 5. Use of non-parametric or parametric modelling to estimate the P99 in relation to sample size and number of positive measurements.

differences in means is performed, based on analysis of variance. Means are pooled automatically if the probability p>0.05. If not, manual pooling is performed. In figure 6 the steps on the right side require a manual assignment of products to product groups before variances are pooled. This manual step may be considered optional: when it is decided to leave this step out, all original variances and means are maintained.

2. *Step 2.* Estimates of variances based on less than 10 df are not considered very reliable. Therefore, variances based on less than 10 df are compared to the overall variance (pooled over all products except the tested product itself, i.c. corrected) and tested for equality. Variances are replaced by the overall variance (uncorrected) whenever the hypothesis of equality of variances is not rejected or, if rejected, the original variances are maintained. If the variance is replaced for (sub)groups consisting of two or more products, a test for differences of means is performed. Means are pooled automatically if p>0.05. If not, the original means are maintained.
3. After performing the above pooling process, there may still be products with less than 10 df. These products are considered again. The variances are judged visually and assigned by hand to one or more of the products with approximately the same value for the (pooled) variance. After testing the variances, they are pooled again, replacing the variance based on < 10 df with the pooled one. A test for differences of means is performed, and for those cases where p>0.05, means are also pooled.
4. Finally, we are left with those cases where variances are pooled, but means are not considered again. The products may be rearranged into (sub)product groups based on similarity of their means. Then, pooled means are calculated replacing the original ones. This last pooling step is optional and not indicated in the figure 6.

Once it is decided to perform a parametric exposure assessment, rearrangement of products into (sub)groups to estimate necessary parameters is needed. Therefore, it is not possible to compare

results of a non-parametric exposure assessment with a parametric one as such, because nearly always some form of pooling has preceded the estimation.

Methods

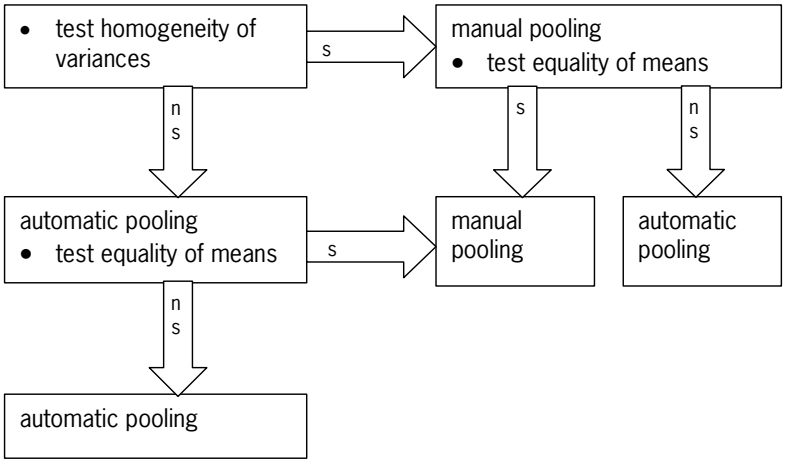
In this study, results of the parametric approach are compared with the non-parametric approach using grouping of pesticide residue levels over products. This was done for iprodione, a pesticide that has annually a high percentage of detects. Residue levels were derived from the KAP-database (§3.1.1). The average level of 55 food products for only those samples with levels above LOR (14%) was 0.83 $mg \cdot kg^{-1}$. The average of all values (including NDs) was 0.13 $mg \cdot kg^{-1}$. The highest average residue concentrations were found in oakleaf lettuce, lamb's lettuce, turnip tops/greens and a left-over category 'other agricultural/horticultural products' (table 5). Most averages were considerably lower. Products for which use of iprodione is registered are indicated with an asterix (table 5).

The lognormal distribution was selected to model positive concentrations (see §3.1.1). As mentioned above, frequently data on residue levels in specific commodities are scarce or even missing. In those cases, data on similar products may provide the necessary information on which to base the parameters. Pooling of products in product groups to allow joint estimates of parameters was applied to the iprodione residue data according to the procedure outlined above. Table 5 summarises parameter values at various stages of the pooling process:

1. The variance and mean for each product was estimated, giving sigma1, mu1 and df1. In some cases the variance is missing because only one measurement was available (e.g. "ROODLOF", 10801, sigma1 = *). Products were assigned to product groups and within each group products were marked to indicate whether the use of the pesticide was registered or not. Homogeneity of variances in different (marked) product groups was tested. For homogeneous groups, variances were pooled within product groups. This process of assigning products to subgroups was repeated until all groups had homogeneous variances. After pooling the variances, an overall test for differences of means was performed, based on analysis of variance. Means were pooled automatically if the probability $p>0.05$. If not, the original means were maintained. Table 5 shows the above procedure. The variances of product group 10701* (table 5) were pooled automatically: sigma2 = 1.31, df2 = 12 (= 5 + 7). The probability of the test for differences of means exceeded 0.05, so means were pooled automatically as well: mu2 = -1.66. The variances of product group 10801* were pooled automatically: sigma2 = 1.48, df2 = 439 (= 3 + 91 + … + 16), but here means significantly differed. The variances for product group 10904* were heterogeneous, so this group was rearranged by hand into two new subgroups (between brackets value of sigma1): strawberry (1.14) and blackberry (1.15), and secondly raspberry (1.73), blue berry (1.83) and currant (1.87). Now, variances within subgroups were homogeneous and pooled, yielding sigma2 = 1.14 for the first and 1.84 for the second subgroup. The means for the second group were pooled automatically, mu2 = -0.76. The means for strawberry and blackberry were maintained: mu2 = –1.57 and –0.89, respectively. Missing variances, e.g. for "roodlof", were replaced by the pooled variances of the product groups that the product belonged to. The missing variance of "kouseband" remained missing, because no (pooled) variance was available in the product group to which this food product belonged. Step 4 is optional.

Step 1:

- Calculate variances and means for each product
- Classify products into groups
- Test homogeneity of variances and equality of means within groups of products. Results are: not significant (ns, $p > 0.05$) or significant (s, $p \leq 0.05$)



Step 2:

- Take products(-groups) with df < 10
- Compare variance with overall variance (corrected). Replace variance with overall variance (uncorrected) for non-significant testresults.

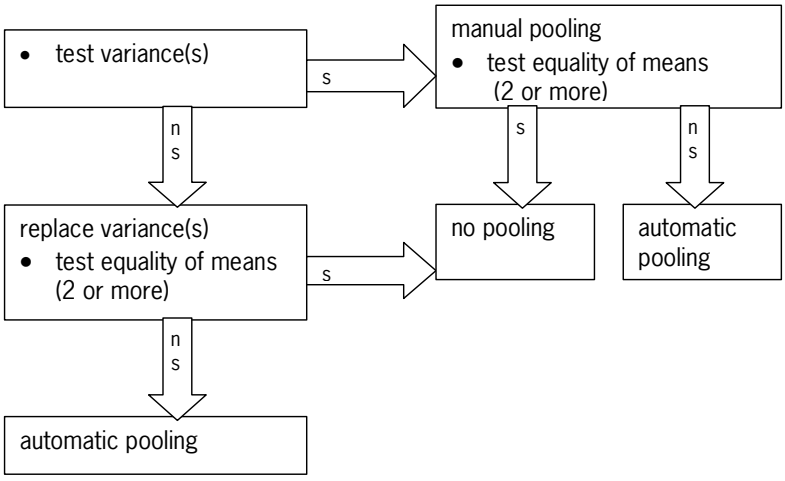


Figure 6. Schematic outline of grouping of products into (sub)product groups when the number of available data on residue levels is limited.

Table 5. Standard deviation (sigma), mean (mu) and degrees of freedom (df) in different pooling steps. The asterisk indicates that the use of iprodione on the product is allowed.

Product	group	sigma1	mu1	df1	sigma2	mu2	df2	sigma3	mu3	mu4	df3
BEAN	10701*	1.60	-1.17	7	1.31	-1.66	12	1.31	-1.66	-1.66	12
"SPERZIEBOON"	10701*	0.75	-2.33	5	1.31	-1.66	12	1.31	-1.66	-1.66	12
CHICORY	10801*	1.38	-2.69	3	1.48	-2.69	439	1.48	-2.69	-1.48	439
"ROODLOF"	10801	*	-2.30	0	1.28	-0.58	16	1.28	-0.58	-0.58	16
ENDIVE	10801*	1.52	-0.91	91	1.48	-0.91	439	1.48	-0.91	-0.83	439
ICEBERG LETTUC	10801*	1.65	-1.92	7	1.48	-1.92	439	1.48	-1.92	-1.48	439
CABBAGE LETTUCE	10801*	1.46	-1.44	285	1.48	-1.44	439	1.48	-1.44	-1.48	439
CURLY LETTUCE	10801*	1.08	-2.14	3	1.48	-2.14	439	1.48	-2.14	-1.48	439
LOLLO ROSSA	10801*	1.53	-0.99	21	1.48	-0.99	439	1.48	-0.99	-0.83	439
LEAF LETTUCE	10801	*	-1.56	0	1.28	-0.58	16	1.28	-0.58	-0.58	16
CELERY	10801	1.76	-1.27	2	1.28	-0.58	16	1.28	-0.58	-0.58	16
SPINACH	10801	1.18	-0.57	9	1.28	-0.58	16	1.28	-0.58	-0.58	16
CHERVIL	10801	*	-1.24	0	1.28	-0.58	16	1.28	-0.58	-0.58	16
PURSLANE	10801	*	0.41	0	1.28	-0.58	16	1.28	-0.58	-0.58	16
RADICCHIO ROSSO	10801*	*	-0.36	0	1.48	-0.36	439	1.48	-0.36	-0.83	439
OAKLEAF LETTUCE	10801*	1.65	-0.06	13	1.48	-0.06	439	1.48	-0.06	-0.83	439
LAMB'S LETTUCE	10801*	1.25	0.80	16	1.48	0.80	439	1.48	0.80	0.80	439
TURNIP TOPS/GREE	10801	1.19	1.30	2	1.28	-0.58	16	1.28	-0.58	-0.58	16
BLEACH-CELERY	10801	1.23	-0.88	3	1.28	-0.58	16	1.28	-0.58	-0.58	16
CAULIFLOWER	10802*	*	-1.83	0	1.62	-1.83	20	1.62	-1.83	-1.83	20
BRUSSELS SPROUT	10802	1.14	-2.70	1	1.14	-2.70	1	1.36	-2.57	-2.57	882
CHINESE CABBAGE	10802*	1.62	-2.32	20	1.62	-2.32	20	1.62	-2.32	-2.32	20
OXHEART/CONICAL	10802	*	-2.30	0	1.14	-2.30	1	1.36	-2.57	-2.57	882
ONION (SMALL)	10803*	0.07	-1.66	1	0.14	-1.66	3	0.14	-1.66	-2.09	3
FENNEL	10803*	0.16	-2.38	2	0.14	-2.38	3	0.14	-2.38	-2.09	3
POTATO	10804	0.62	0.19	1	0.59	0.19	50	0.59	0.19	0.19	50
WINTER CARROT	10804	0.62	-2.55	13	0.59	-2.55	50	0.59	-2.55	-2.64	50
CARROT	10804	0.54	-2.71	35	0.59	-2.71	50	0.59	-2.71	-2.64	50
RADISH	10804*	1.52	-2.91	5	1.52	-2.91	5	1.36	-2.91	-2.91	882
CELERIAC	10804	1.31	-2.07	1	0.59	-2.07	50	0.59	-2.07	-2.64	50
CUCUMBER	10805*	0.80	-1.55	7	0.99	-2.22	26	0.99	-2.22	-2.22	26
TOMATO	10805*	0.88	-2.50	13	0.99	-2.22	26	0.99	-2.22	-2.22	26
SWEET PEPPER	10805*	1.33	-2.19	6	0.99	-2.22	26	0.99	-2.22	-2.22	26
PUMPKIN,	10805*	*	-2.53	0	0.99	-2.22	26	0.99	-2.22	-2.22	26
PEPPER	10805	1.23	-0.94	4	1.23	-0.94	4	1.36	-0.94	-0.94	882
GHERKIN/PICKLE	10805*	*	-3.51	0	0.99	-2.22	26	0.99	-2.22	-2.22	26
"KOUSEBAND"	10889	*	-1.27	0	*	-1.27	0	1.36	-1.27	-1.27	882
PAC-CHOY	10889*	1.29	-0.48	7	1.29	-0.48	7	1.28	-0.48	-0.48	37
MIXED VEGETABLE	10890	1.37	0.09	2	2.22	-0.45	6	1.36	-0.45	-0.45	882
OTHER AGR./HORTI	10890	2.54	-0.77	4	2.22	-0.45	6	1.36	-0.45	-0.45	882
KIWI FRUIT	10901	1.96	-0.96	2	1.96	-0.96	2	1.36	-0.96	-0.96	882
APPLE	10902	2.02	-1.59	3	2.02	-1.59	3	1.36	-1.97	-1.97	882
PEAR	10902	*	-3.51	0	2.02	-3.51	3	1.36	-1.97	-1.97	882
APRICOT	10903	1.55	-1.71	1	1.26	-0.83	30	1.26	-0.83	-0.83	30
NECTARIN	10903	1.08	-0.97	8	1.26	-0.83	30	1.26	-0.83	-0.83	30
PEACH	10903	1.18	-0.74	5	1.26	-0.83	30	1.26	-0.83	-0.83	30
PLUM, INCLUDING	10903	1.34	-0.69	16	1.26	-0.83	30	1.26	-0.83	-0.83	30
SWEET CHERRY	10903*	0.89	-0.69	11	0.89	-0.69	11	0.89	-0.69	-0.69	11
GRAPE	10904	1.14	-1.06	24	1.14	-1.06	24	1.14	-1.06	-1.06	24
STRAWBERRY	10904*	1.14	-1.57	168	1.14	-1.57	184	1.14	-1.57	-1.51	184
RASPBERRY	10904*	1.73	-1.04	8	1.84	-0.76	39	1.84	-0.76	-0.76	39
BLACKBERRY	10904*	1.15	-0.89	16	1.14	-0.89	184	1.14	-0.89	-1.51	184
BLUE BERRY	10904*	1.83	-1.24	2	1.84	-0.76	39	1.84	-0.76	-0.76	39
CURRANT	10904*	1.87	-0.62	29	1.84	-0.76	39	1.84	-0.76	-0.76	39
OTHER FRUIT, NUT	10990	*	-1.51	0	*	-1.51	0	1.36	-1.51	-1.51	882

Table 6. Estimates of percentiles of exposure to iprodione using the parametric and non-parametric approach. Simulations were performed with GenStat: 50,000 iterations, repeated three times.

	percentiles (%)					
iprodione	P95	P98	P99	P99.5	P99.9	P99.99
parametric	0.68	2.4	4.6	7.7	23	66
	0.71	2.3	4.6	7.7	20	55
	0.71	2.1	4.1	7.1	20	58
non-parametric	0.66	2.1	4.3	7.0	18	64
	0.62	2.2	4.5	8.4	22	74
	0.64	2.0	4.0	6.6	16	44

2. Variances based on less than 10 df were compared to the overall variance (pooled over all products except the tested product itself) and tested for equality. Variances were replaced by the overall variance (uncorrected; sigma3 = 1.36) whenever the hypothesis of equality of variances was not rejected. If rejected, the original variances were maintained. If the variance was replaced for (sub)groups with two or more members, a test for differences of means was performed. Means were pooled automatically ($p>0.05$) or not ($p\leq0.05$). Table 5 shows how the above was implemented. E.g. brussels sprouts and oxheart/conical cabbage (10802) have less than 10 df with sigma2 = 1.14. The variances were tested against the corrected overall variance. The probability exceeded 0.05, so their variances were replaced: sigma3 = 1.36 and df3 = 882. The means, -2.70 and –2.30 were tested ($p>0.05$) and therefore pooled automatically: mu3 = -2.57. Conversely, the variances of onion (small) and fennel (10803*) were not replaced. The missing variance of "kouseband" (10889) was replaced: sigma3 = 1.36 with df3 = 882.
3. After carrying out the above pooling process, there were still products with less than 10 df. These products were considered again. The variances were judged visually and assigned by hand to one or more of the products with approximately the same value for the (pooled) variance. After testing the variances, the variances were pooled again, replacing the variance based on less than 10 df with the pooled one. Testing for differences of means was performed and for those cases where $p>0.05$, means were also pooled. E.g. pac-choy (10889*) had less than 10 df and was assigned to 10903. The pooled variance: sigma3 = 1.28 with 3 df was 37 (= 30+7), the original mean was maintained. Onion (small) and venkel were not assigned to any group, so the original variance was kept (sigma3 = 0.14).
4. Step 4 is optional for those cases where variances were pooled, but means not. Those products may be rearranged into (sub)product groups based on similarity of their means. Then, pooled means can be calculated replacing the original ones. For example, product group 10801* had a pooled variance of 1.48 (sigma3) but the means were original. Visually, with between brackets the estimate of the mean, chicory (-2.69), iceberg lettuce (-1.92), cabbage lettuce (-1.44) and curly lettuce (-2.14) were assigned to one subgroup, endive (-0.91), lollo rossa (-0.99), radicchio rosso (-0.36) and oakleaf lettuce (-0.06) to a second group, and lamb's lettuce (0.80) formed a single group. After pooling, the new means, mu4, for the three subgroups were –1.48, -0.83 and 0.80, respectively.

The simulations were performed using GenStat (GenStat 2002). The number of iterations was 50,000. The estimates are based on the parameter estimates of table 5, produced applying step 1, 2 and 3 and the optional step 4 of the pooling procedure.

Results

Table 6 summarises the results of the simulations using parametric and non-parametric distributions. It is evident that the non-parametric percentiles yielded estimates of exposure that were in the same range as the parametric ones. Although vaguely, the parametric approach seemed to give slightly less variable results and provided somewhat higher estimates of P95.

Conclusion

If the amount of information is ample, e.g. more than 20 measurements ≥ LOR, a non-parametric approach is preferable. In situations where the number of measurements is limited, an appropriate risk analysis should be based on further modelling. Two options can be chosen or combined: 1) assume a simple distributional form for the residue data, and/or 2) group products to enlarge the number of measurements per group, assuming that residue distributions are the same for the grouped products. In the case of parametric modelling the assumptions of equality can be restricted to a subset of the parameters (in the chosen binomial-lognormal model: detect probability, lognormal mean, lognormal standard deviation).

3.2.3 Estimation of pesticide exposure using histogram data

Rationale

In the EU residue data are mostly reported in a tabulated (histogram) form. The parameters of the lognormal distribution can then be estimated by fitting normal distributions to a set of observations or counts. Statistics are $n_1 \ldots n_k$, representing the number of counts in k classes. The group limits are logtransformed and a normal distribution is fitted to standardized normal probabilities based on group limits and the numbers $n_1 \ldots n_k$. Parameters μ and σ are estimated. Group limits c_k are given, with c_1 = LOR. An example of this procedure is reported below.

Methods

For the purpose of this study full data of iprodione (§3.2.2) were classified into groups using class limits as proposed by EU–standards. Table 7 summarises the results. LOR of these data is 0.02 $mg \cdot kg^{-1}$. Normal distributions were fitted to count data using logtransformed class limits.

Results

In table 8 the estimates for the means (mu) and standard deviations are listed for both the histogram data and the full data approach, which fits normal distributions to (quantifiable) logtransformed concentration levels. In figure 7 the estimates of the means and standard deviations based on the histogram data versus the full data approach are plotted. The diagonal indicates that all standard deviations based on histogram data were systematically lower than estimates based on the full approach. No systematic effect was found for the means.

A parametric simulation was performed using the standard deviations and means listed in table 8. Standard deviations and means were pooled automatically and manually (step 1, figure 6). Table 9 summarises the results. All percentiles using histogram data were comparable to percentiles based on a parametric approach using full data.

Table 7. Histogram data for iprodione (limit of reporting (LOR) = 0.02 mg·kg^{-1}).

product	Samples with quantifiable residues in classes up to and including (in mg·kg^{-1})											
	LOR											
	0.02	0.05	0.1	0.2	0.5	1	2	5	10	20	50	>50
BEAN	0	1	1	1	2	0	2	1	0	0	0	0
"SPERZIEBOON"	0	0	4	1	1	0	0	0	0	0	0	0
CHICORY	1	0	1	1	1	0	0	0	0	0	0	0
"ROODLOF"	0	0	0	1	0	0	0	0	0	0	0	0
ENDIVE	0	5	16	10	18	17	10	12	3	1	0	0
ICEBERG LETTUC	0	3	1	1	0	1	2	0	0	0	0	0
CABBAGE LETTUCE	1	35	54	47	59	38	29	16	4	2	1	0
CURLY LETTUCE	0	0	2	1	0	1	0	0	0	0	0	0
OLLO ROSSA	0	1	5	2	1	9	2	1	1	0	0	0
LEAF LETTUCE	0	0	0	0	1	0	0	0	0	0	0	0
CELERY	0	1	0	0	1	0	1	0	0	0	0	0
SPINACH	0	0	0	3	2	2	0	3	0	0	0	0
CHERVIL	0	0	0	0	1	0	0	0	0	0	0	0
PURSLANE	0	0	0	0	0	0	1	0	0	0	0	0
RADICCHIO ROSSO	0	0	0	0	0	1	0	0	0	0	0	0
OAKLEAF LETTUCE	0	0	1	2	2	1	3	3	1	1	0	0
LAMB'S LETTUCE	0	0	0	0	2	2	4	4	4	0	1	0
TURNIP TOPS/GREE	0	0	0	0	0	0	1	1	0	1	0	0
BLEACH-CELERY	0	0	1	0	0	2	1	0	0	0	0	0
CAULIFLOWER	0	0	0	1	0	0	0	0	0	0	0	0
BRUSSELS SPROUT	0	1	0	1	0	0	0	0	0	0	0	0
CHINESE CABBAGE	1	8	1	3	6	0	1	1	0	0	0	0
OXHEART/CONICAL	0	0	0	1	0	0	0	0	0	0	0	0
ONION (SMALL)	0	0	0	1	1	0	0	0	0	0	0	0
FENNEL	0	0	2	1	0	0	0	0	0	0	0	0
POTATO	0	0	0	0	0	1	1	0	0	0	0	0
WINTER CARROT	0	3	5	5	1	0	0	0	0	0	0	0
CARROT	0	9	15	12	0	0	0	0	0	0	0	0
RADISH	1	3	0	1	0	1	0	0	0	0	0	0
CELERIAC	0	0	1	0	1	0	0	0	0	0	0	0
CUCUMBER	0	0	1	3	3	1	0	0	0	0	0	0
TOMATO	0	4	3	4	3	0	0	0	0	0	0	0
SWEET PEPPER	0	1	4	0	1	0	1	0	0	0	0	0
PUMPKIN,	0	0	1	0	0	0	0	0	0	0	0	0
PEPPER	0	0	1	0	1	2	1	0	0	0	0	0
GHERKIN/PICKLE	0	1	0	0	0	0	0	0	0	0	0	0
"KOUSEBAND"	0	0	0	0	1	0	0	0	0	0	0	0
PAC-CHOY	0	0	0	2	0	3	1	2	0	0	0	0
MIXED VEGETABLE	0	0	0	0	1	0	1	1	0	0	0	0
OTHER AGR./HORTI	0	0	2	0	1	1	0	0	0	0	1	0
KIWI FRUIT	0	1	0	0	0	0	2	0	0	0	0	0
APPLE	1	0	0	0	1	2	0	0	0	0	0	0
PEAR	0	1	0	0	0	0	0	0	0	0	0	0
APRICOT	0	0	1	0	0	1	0	0	0	0	0	0
NECTARIN	0	0	2	0	3	2	2	0	0	0	0	0
PEACH	0	0	1	0	1	2	2	0	0	0	0	0
PLUM, INCLUDING	0	1	1	1	5	5	2	2	0	0	0	0
SWEET CHERRY	0	0	1	0	5	5	0	1	0	0	0	0
GRAPE	0	1	2	4	7	7	2	2	0	0	0	0
STRAWBERRY	1	17	20	46	49	19	14	3	0	0	0	0
RASPBERRY	0	2	0	1	1	3	1	1	0	0	0	0
BLACKBERRY	0	0	2	3	4	4	3	1	0	0	0	0
BLUE BERRY	0	1	0	0	1	0	1	0	0	0	0	0
CURRANT	0	4	3	3	2	4	5	6	3	0	0	0
OTHER FRUIT, NUT	0	0	0	0	1	0	0	0	0	0	0	0

Table 8. Standard deviation (sd), mean (mu) and degrees of freedom (df) based on full data and histogram data. Lognormal distributions have been fitted.

		full data		histogram data		
product	group	sd	mu	sd	mu	df
BEAN	10701	1.60	-1.17	1.48	-1.06	7
"SPERZIEBOON"	10701	0.75	-2.33	0.50	-2.30	5
CHICORY	10801	1.38	-2.69	1.30	-2.61	3
"ROODLOF"	10801	*	-2.30	0.04	-1.95	0
ENDIVE	10801	1.52	-0.91	1.46	-0.87	91
ICEBERG LETTUC	10801	1.65	-1.92	1.58	-1.82	7
CABBAGE LETTUCE	10801	1.46	-1.44	1.42	-1.39	285
CURLY LETTUCE	10801	1.08	-2.14	0.92	-1.90	3
OLLO ROSSA	10801	1.53	-0.99	1.37	-0.96	21
LEAF LETTUCE	10801	*	-1.56	0.06	-1.15	0
CELERY	10801	1.76	-1.27	1.55	-1.41	2
SPINACH	10801	1.18	-0.57	1.22	-0.55	9
CHERVIL	10801	*	-1.24	0.06	-1.15	0
PURSLANE	10801	*	0.41	0.04	0.35	0
RADICCHIO ROSSO	10801	*	-0.36	0.04	-0.34	0
OAKLEAF LETTUCE	10801	1.65	-0.06	1.51	-0.01	13
LAMB'S LETTUCE	10801	1.25	0.80	1.16	0.84	16
TURNIP TOPS/GREE	10801	1.19	1.30	0.94	1.39	2
BLEACH-CELERY	10801	1.23	-0.88	1.12	-0.75	3
CAULIFLOWER	10802	*	-1.83	0.04	-1.95	0
BRUSSELS SPROUT	10802	1.14	-2.70	0.71	-2.68	1
CHINESE CABBAGE	10802	1.62	-2.32	1.42	-2.20	20
OXHEART/CONICAL	10802	*	-2.30	0.04	-1.95	0
ONION (SMALL)	10803	0.07	-1.66	0.11	-1.61	1
FENNEL	10803	0.16	-2.38	0.09	-2.34	2
POTATO	10804	0.62	0.19	0.10	0.00	1
WINTER CARROT	10804	0.62	-2.55	0.60	-2.46	13
CARROT	10804	0.54	-2.71	0.50	-2.60	35
RADISH	10804	1.52	-2.91	1.42	-2.90	5
CELERIAC	10804	1.31	-2.07	0.71	-1.92	1
CUCUMBER	10805	0.80	-1.55	0.64	-1.55	7
TOMATO	10805	0.88	-2.50	0.80	-2.36	13
SWEET PEPPER	10805	1.33	-2.19	1.17	-2.12	6
PUMPKIN,	10805	*	-2.53	0.04	-2.65	0
PEPPER	10805	1.23	-0.94	1.01	-0.83	4
GHERKIN/PICKLE	10805	*	-3.51	0.06	-3.45	0
"KOUSEBAND"	10889	*	-1.27	0.06	-1.15	0
PAC-CHOY	10889	1.29	-0.48	1.10	-0.30	7
MIXED VEGETABLE	10890	1.37	0.09	0.91	0.12	2
OTHER AGR./HORTI	10890	2.54	-0.77	2.23	-0.67	4
KIWI FRUIT	10901	1.96	-0.96	1.77	-0.91	2
APPLE	10902	2.02	-1.59	1.91	-1.69	3
PEAR	10902	*	-3.51	0.06	-3.45	0
APRICOT	10903	1.55	-1.71	1.13	-1.50	1
NECTARIN	10903	1.08	-0.97	1.04	-0.97	8
PEACH	10903	1.18	-0.74	1.02	-0.63	5
PLUM, INCLUDING	10903	1.34	-0.69	1.15	-0.74	16
SWEET CHERRY	10903	0.89	-0.69	0.83	-0.75	11
GRAPE	10904	1.14	-1.06	1.09	-0.96	24
STRAWBERRY	10904	1.14	-1.57	1.09	-1.54	168
RASPBERRY	10904	1.73	-1.04	1.49	-1.06	8
BLACKBERRY	10904	1.15	-0.89	1.06	-0.88	16
BLUE BERRY	10904	1.83	-1.24	1.55	-1.41	2
CURRANT	10904	1.87	-0.62	1.75	-0.56	29
OTHER FRUIT, NUT	10990	*	-1.51	0.06	-1.15	0

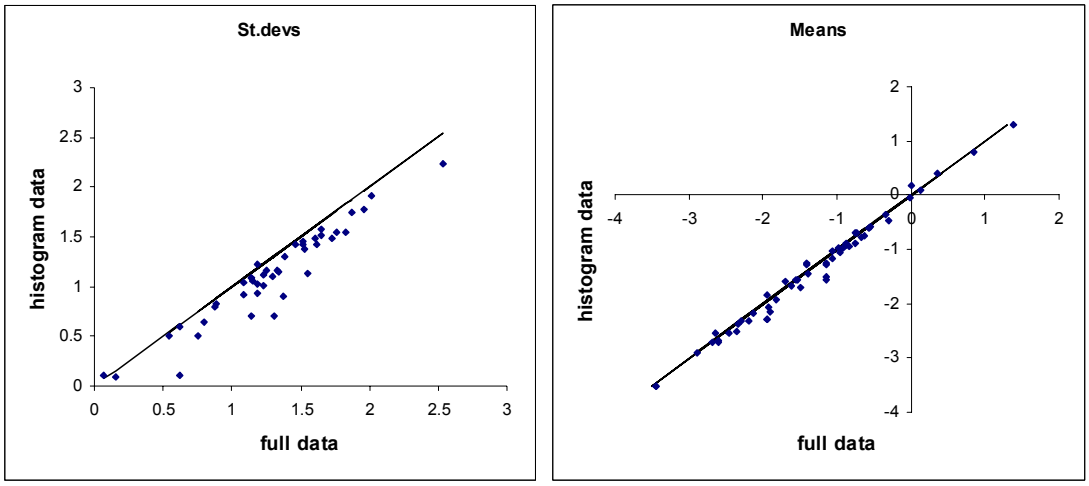


Figure 7. Estimates of standard deviations and means based on histogram data versus full data approach.

Conclusion

The use of histogram data as reported by the EU can be used to estimate the dietary exposure to pesticides.

3.3 GenStat versus @Risk

Rationale

To perform some of the studies described above we used the computer programme 'Monte Carlo Risk Analysis', developed at the RIKILT to assess the acute and chronic exposure to chemicals through the diet (Voet *et al.* 2002). In an early stage implementations of a simple non-parametric model were made both in a general-purpose statistical programming language, GenStat (GenStat 2002), and in the currently popular management decision tool @Risk, which is an add-in module in Excel. A comparison was made between these two implementations.

Methods

The data used were residue levels of iprodione in 55 products (§3.2.2) and the consumption data of the second Dutch National Food Consumption Survey (DNFCS; Kistemaker *et al.* 1998). This survey was carried out among a large number of representative Dutch households in 1992. On two successive days 6,218 respondents reported their daily consumption of food commodities. These data were transformed into consumed amounts of raw agricultural commodities (Dooren *et al.* 1995).

GenStat implementation

The GenStat programme is fast, due to sampling in parallel data structures. Let *n* be the chosen number of simulations, and *k* the number of food products. The programme selects a simple random sample of *n* individuals and a simple random sample of *n* day numbers (1 or 2). The selection of individuals may be restricted to a specified age range (e.g. only children from 0 to 6 years). A typical value of *n* may be 100,000. Note that each of the 6,218 individuals is likely to

Table 9. Estimates of percentiles of dietary exposure to iprodione using a parametric approach based on histogram data and full data. Simulations were performed with GenStat (50,000 iterations).

data	P95	P98	P99	P99.5	P99.9	P99.99
histogram	0.75	2.3	4.3	7.2	21	64
full	0.68	2.4	4.6	7.7	23	66
	0.71	2.3	4.6	7.7	20	55
	0.71	2.1	4.1	7.1	20	58

occur many times in the sample. For each juxtaposed combination of sampled individual and sampled day the consumption data are retrieved from an ASCII file, and stored in an $n \times k$ matrix. Another $n \times k$ matrix is constructed to contain simulated concentration data. For all k products the total number of measurements (t) and the number of non-zero measurements (w) is determined. Then random index numbers (i) between 1 and t are sampled for each cell of the matrix. If $i \leq w$, then the i^{th} value for this product is selected from an ASCII-file containing non-zero residue data. If $i > w$, then a value 0 is inserted in the concentration matrix. Both $n \times k$ matrices are now multiplied per element. Summing over the k products and dividing by the n body weights corresponding with the n selected individuals will give the simulated exposure distribution as a vector of n values. Relevant percentiles can be obtained from this vector.

@Risk implementation

@Risk is a simulation add-in for Excel and adds Monte Carlo simulations to spreadsheets. Uncertain values in the spreadsheet are replaced by @Risk or user-defined probability distribution functions. Spreadsheets are recalculated sequentially 10,000 – 50,000 times, each time sampling random values from the @Risk functions. The sequential nature of the spreadsheet recalculations makes the @Risk implementation much slower than GenStat (hours instead of minutes), thereby limiting the practical number of simulations. The result is a distribution of possible outcomes, which again can be investigated for relevant percentiles. The Monte Carlo simulation in @Risk can be carried out either by simple random sampling or by Latin Hypercube sampling. The latter method is in theory more efficient, and was therefore used in this study. The practical implementation of risk analysis in Excel and @Risk is an Excel worksheet. This worksheet contains references to all necessary input files (ASCII). The worksheet makes calculations involving the @Risk functions *RiskDuniform* and *RiskDiscrete*, which are recognised by @Risk during the simulation, and used for sampling the data in the other sheets. At any time the worksheet shows the results of one simulation.

Results

Table 10 summarises the percentiles of dietary exposure to iprodione of the distributions using GenStat or @Risk, applying 50,000 iterations. Of the 6,915 samples analysed only 937 samples had levels at or above the limit of reporting (14%). Because 6218 persons were surveyed during two days, the incidence matrix contained 87,117 values. A simulation run with @Risk took 2h.9', while GenStat completed the task within 2 min. Between GenStat and @Risk only minor differences occurred, no more than between repeated simulations with any one of the programmes. Results were relatively stable for the estimates of P95, P98, and P99 of exposure to iprodione. Discrepancies occured at the higher percentiles, e.g. estimates of P99.99 ranged from 44 $\mu g \cdot kg^{-1} \cdot d^{-1}$ to 74 $\mu g \cdot kg^{-1} \cdot d^{-1}$.

Table 10. Dietary exposure to iprodione ($\mu g \cdot kg^{-1} \cdot d^{-1}$) calculated with GenStat and @Risk using a non-parametric approach (50,000 iterations).

	percentile					
method	P95	P98	P99	P99.5	P99.9	P99.99
GenStat	0.66	2.1	4.3	7.0	18	64
	0.62	2.2	4.5	8.4	22	74
	0.64	2.0	4.0	6.6	16	44
@Risk	0.64	2.1	4.2	7.0	17	47

Conclusion

For Monte Carlo calculations based on realistic amounts of data the use of a general statistical package is more practical than the use of @Risk.

4 CONCLUSIONS

The following conclusions can be drawn:

1. The lognormal distribution was shown to give an adequate fit for several pesticide levels at or above the limit of reporting in several agricultural products. We therefore considered the lognormal distribution an appropriate model to present pesticides levels above the limit of reporting. This together with the fact that residue data are often skewed to the left and originate from mechanisms that generate a lognormal distribution under a variety of biological circumstances.

2. Processing has a major effect on the magnitude of exposure to pesticides and should therefore be included in the exposure calculations if information is available.

3. Four approaches are described to incorporate variability between units within a composite sample in a probabilistic exposure assessment. It is clear that this factor influences the outcome of exposure assessment. However, more experience with the four approaches to include unit variability in probabilistic models is necessary.

4. When only a limited amount of data regarding pesticide levels in food commodities is available, grouping of products may be considered, assuming that residue distributions are the same for the grouped products. In this way the number of measurements per (food) group is increased.

5. The use of histogram data as reported by the EU can be used to estimate the exposure to pesticides through the diet.

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