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Dietary exposure assessment of Danish consumers to dithiocarbamate residues in food. A comparison of the deterministic and probabilistic approach

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Dietary exposure assessment of Danish consumers to dithiocarbamate residues in food - A comparison of the deterministic and probabilistic approach.

Abstract

Probabilistic and deterministic estimates of the acute and chronic exposure of the Danish populations to dithiocarbamate residues were performed. The Monte Carlo Risk Assessment program (MCRA 4.0) was used for the probabilistic risk assessment. Food consumption data were obtained from the nationwide dietary survey conducted in 2000-2002. Residue data for 5721 samples from the monitoring program conducted in the period 1998 – 2003 were used for dithiocarbamates, which had been determined as carbon disulphide. Contributions from 26 commodities were included in the calculations. Using the probabilistic approach the daily acute intakes at the 99.9% percentile for adults and children were 11.2 µg/kg bw/day and 28.2 µg/kg bw/day, representing 5.6% and 14.1% of the ARfD for maneb. When comparing the point estimate approach with the probabilistic approach the outcome of the point estimate calculations was generally higher or comparable with the outcome of the probabilistic approach at the 99.9 percentile (consumers only). The chronic exposure for adults and children were 0.35 µg/kg bw/day and 0.76 µg/kg bw/day at the 99.9 percentile, representing 0.7% and 1.5%, respectively of the ADI for mancozeb and maneb at 50 µg/kg bw.

Introduction

Dithiocarbamates are some of the most often used pesticides in the EU. These pesticides are also some of the most frequently detected pesticides in national monitoring programs (Poulsen et al. 2005, Caldas et al. 2004, EU 2003). Dithiocarbamate residues were found in 7.1% of the samples in the monitoring period 1998-2003 in Denmark, and are among the six pesticides, which contribute most to the intake of pesticides from fruit and vegetables in the Danish market (Poulsen et al. 2005). Residues are determined as carbon disulphide (CS_2) (limit of quantification (LOQ) 0.1 mg/kg as CS_2) in fruit and vegetables (Andersen et al. 2001).

To estimate the exposure to pesticides, both deterministic and probabilistic modelling can be used. For legislation purposes the deterministic point estimate approach is used to estimate the exposure to acute toxic pesticides today in the EU. The calculations of the point estimate are performed using the equations defined by JMPR (FAO/WHO 1997, FAO/WHO 2002). The point estimate approach is conservative since one high level for both consumption and residue level is used at the same time. As described in the EFSA opinion on acute dietary assessment (EFSA 2007), the deterministic approach is not suitable to estimate the protection for the total population. This is because that this approach only considers the exposure from one commodity at a time and does not take into account the contribution of individual commodities to the total intake of a pesticide. Assessing the acute and chronic exposure using the probabilistic approach takes into account the variation in food consumption, pesticides residue levels and body weight by using a Monte Carlo simulation.

In the present work, a probabilistic and deterministic estimation of the acute and chronic exposure to dithiocarbamate residues for the Danish population is presented.

Materials and methods

Dithiocarbamate residue data

Dithiocarbamate residue data were taken from the Danish pesticide monitoring program during the period 1998-2003 (Poulsen et al.). The calculations include analyses for dithiocarbamate residues from 5721 samples of fruit and vegetables from 26 commodities (only commodities, where dithiocarbamates were found, are included).

Dithiocarbamate residues were found in 34 commodities, but aubergines, peas with pod, exotic vegetables, spring onions, herbs, figs, currants and other small fruits (355 samples) were not included because of lack of data for consumption. Findings in these commodities were few and consumptions presumedly small. No distinctions were made between imported commodities and those of Danish origin in the calculations. Residues were determined as CS₂ (LOQ = 0.1 mg/kg) in the fruit and vegetables samples (Andersen et al. 2001). Samples analysed did not include Brassica crops, as these are known to give false positives by the method used (Perz et al. 2000). Papayas (included in the group "Exotic fruits") were analysed without pulp or seeds (Lund et al. 1999). For intake calculations, residue levels below the LOQ were considered as zero.

Processing factors

There is not described much about processing factors for dithiocarbamates in the literature. Caldas et al. (2006) used 0.26 for cooking, 0.3 for washing of apple, strawberry, carrot, lettuce and tomato and 0.08 for peeling of orange, banana and papaya. Since most commodities in which dithiocarbamate residues were found in this study can be eaten raw, except for melon, which is peeled, processing factors for cooking and peeling was not applied in the calculation. In the EU washing is not considered as a processing factor for most commodities as it is assumed that they can be eaten unwashed.

Food consumption data

The food consumption data were obtained from the nationwide dietary survey conducted in 2000-2002 by the Department of Nutrition, Technical University of Denmark (Andersen et al. 2005). The survey included 3299 adults aged 15-75 years old and 231 children aged 4-6 years old. The survey used a seven-day prospective food record with a pre-coded (semi-closed) questionnaire that included answering categories for the most commonly eaten foods and dishes in the Danish diet. The survey was distributed equally over the 7 days of the week and over the whole year to take into account possible seasonal variation in dietary habits. Holidays were excluded.

The questionnaire was organised according to a normal daily meal pattern. The body weight of the participant was noted and used in the calculations. In the calculations of the deterministic acute and chronic exposure, an average body weight of 74 kg was used for adults and 22 kg for children.

Deterministic intake calculations

Acute intake calculation. The acute dietary intake or short-term exposures (the point estimate) were estimated by using the equations for the National Estimate of Short Term Intake (NESTI) (FAO/WHO 2002). The large portion sizes were derived from the Danish National Consumptions survey from 2000-2002 (Andersen et al. 2005). In the calculations the default variability factors 7, 5 and 1 were used according to JMPR (FAO/WHO 2002). The unit weight was derived from the reference "Typical weights for food, 1996" (Johanna Haraldsdóttir et al.) and highest residue found in the monitoring program was used in the calculations.

Chronic intake calculation. The National Estimated Daily Intake (NEDI) of dithiocarbamate residues was calculated by multiplying the mean consumption for the consumer group with the mean level of dithiocarbamate residues for each commodity. The total chronic intake for dithiocarbamate residues was calculated by summing up the individual intakes from all commodities. The sum was compared to the ADI of maneb and mancozeb (see below).

Probabilistic intake calculations

The probabilistic exposure assessment was conducted using the Monte Carlo Risk Assessment (MCRA 4.0) system, an internet-based program developed by RIKILT – Institute of Food Safety, Wageningen UR, in the Netherlands (Boer et al. 2005).

The outputs from the intake distribution generated after the simulation were specified at percentiles P50, P90, P95, P99, P99.9 and P99.99. The calculations have been performed as described by Boon et al. (2005).

Acute intake calculation. For the estimation of the daily acute exposure, 100 000 randomly drawn consumption patterns selected from the consumption database were multiplied with randomly selected dithiocarbamate residues giving an empirical estimate of the intake distribution for the acute exposure of dithiocarbamate residues. The default variability factors 7, 5 and 1 were used as fixed factors in the calculations. Currently there are no guidelines on how to incorporate variability into the probabilistic approach (Boon et al. 2004).

Chronic intake calculation. The chronic intake calculations were performed by multiplying randomly drawn consumption patterns from the food consumption database with the mean residue levels of dithiocarbamate per food and then summing up over all food. The mathematical model used for estimating the chronic intake is proposed by statisticians at the Iowa State University and described by Nusser et al. (1996) and Guenther et al. (1997).

Toxicological data for dithiocarbamates

In the EU the six dithiocarbamates maneb, mancozeb, metiram, propineb, thiram and ziram are registered for use. In Denmark only mancozeb and maneb are registered for use. The residue definition for the dithiocarbamates is the sum of maneb, mancozeb, metiram, propineb, thiram, ziram expressed as CS₂. The analytical methodology applied to analyse dithiocarbamates in food did not discriminate among the compounds used on the crops. The ADI for both mancozeb and maneb is 50µg/kg/bw/day and since these substances are the most frequently used of the dithiocabamates in the EU this ADI was used in the chronic risk assessment. The ARfDs for mancozeb and maneb are 600 µg/kg bw/day and 200 µg/kg bw/day, respectively. The ARfD for maneb was used in the risk assessment in this study. Range for the six dithiocarbamates is 7- 50 µg/kg bw/day for the ADI and 80– 600 µg/kg bw/day for the ARfD (EU 2007).

Results and discussion

Residue data and food consumption data.

In Table I, the frequencies, mean levels and highest residues found in 5721 samples, accounting for contributions from 26 commodities of fruits and vegetables are shown.

[Insert Table I about here]

Dithiocarbamate residues were found in 7.1% of all the samples analyzed in the monitoring period 1998-2003 (Poulsen et al. 2005). In a Brazilian study (Caldas et al. 2004) dithiocarbamate residues were found in 60.8% of the samples. Nine commodities were analysed by Caldas; five of these (papaya, banana, apple, strawberry, tomato) were included in the Danish program for dithiocarbamate residues but much lower residue frequencies were found. In Denmark apricot (26.6%), pear (20.6%), spinach (12.9%), exotic fruits including papaya (12.6%) and apple (12.1%)

had the highest frequency of samples containing residues at or above the LOQ of 0.1 mg/kg. Samples of spinach, cucumber, melon, apricot, pear, table grape, sweet pepper, lettuce, beans with pods and carrots had contents above the EU-MRLs (Poulsen et al. 2005).

[Insert Table II about here]

Table II shows a summary of the food consumption data obtained from the nation-wide survey. Apple followed by carrot, banana, tomato, cucumber and pear showed the highest mean consumption levels for all consumers (total population; both consumers and non-consumers), while melon followed by apple, pear, banana, table grapes and peaches and nectarine had the highest mean consumption on consumption days only (only days with consumption included). Exotic fruit followed by tomato, cucumber, sweet pepper and carrots had the highest percentage of consumption days for both adults and children. That exotic fruit had the highest percentage of consumption days is probably due to that this group includes different fruits.

Intake distribution. Figure 1 shows percentiles of the acute intake distribution, summed for all commodities, calculated by the probabilistic approach. It is seen that 0.1% of the adults and children have an estimated intake higher than 11 $\mu\text{g/kg bw/day}$ and 28 $\mu\text{g/kg bw/day}$, respectively. The maximum intake is 86 $\mu\text{g/kg bw/day}$ and 138 $\mu\text{g/kg bw/day}$ for adults and children, respectively. Thus, the acute intake is below the ARfD of 200 $\mu\text{g/kg bw/day}$ for maneb, indicating that there is no acute risk for the Danish consumers from dithiocarbamate residues.

[Insert Figure 1 about here]

Acute intake calculation

Total intake. In Figure 2 the contribution (%) per commodity to the total acute intake as estimated by the probabilistic approach is shown. Nine commodities out of 26 contributed to 94 % of the total intake of dithiocarbamate residues. Apples followed by pear and cucumbers contributed most for both adults and children.

[Insert Figure 2 about here]

Comparison of the deterministic point estimate and the probabilistic approach. The point estimate approach was compared with the probabilistic approach following the same approach as Boon et al. (2004). The results are shown in Table III. Since the point estimate approach is calculated for consumers only the probabilistic approach is also performed for consumers only. For comparison, calculations for the total population (including consumers and non-consumers) are also presented. The large portion size expressed as the 97.5% percentile used in the point estimate calculation is derived from the same survey (Andersen et al. 2005) as the consumption data used in the probabilistic calculation. To compare the outcome from the point estimate approach with the probabilistic approach, the 99.9% percentile (P99.9) of the exposure was used. In the USA this percentile is used by the US EPA (US EPA 2000c). In the work example performed by (Boon et al. 2004) this percentile was also chosen for comparison.

[Insert Table III about here]

Point estimates vs. probabilistic modelling, consumers only. Table III shows that the exposures calculated by the point estimate approach in general were higher or comparable to the exposures at P99.9 for consumers only. For apricot however at the P99.9 the exposure was much higher (x3) for children than that calculated with the point estimate approach. Apricot has only 0.2 % (n=4) consumption days for children which included 3 persons. One of the persons had an intake of 150 g and a body weight of 16 kg, and the other two children had consumptions from 10-30 g and body weights of 22 kg and 19 kg, respectively. The fact that one child out of the three children had such a high intake combined with a bodyweight, which is lower than the mean body weight of 22 kg can explain the much higher exposure level calculated by the probabilistic approach than by the deterministic approach. In both cases, however, the exposure was still below 100 % of the ARfD of 200 µg/kg bw/day. Compared to the exposure at P99.99 the point estimates were generally lower. The exposure levels for children were higher than for adults.

Probabilistic modelling, consumers only approach vs. total population. Exposure levels calculated with probabilistic approach for the total population (including consumers and non-consumers) were always lower than the corresponding levels for the consumers only (Table III). As mentioned before, this is due to the fact that the total population

includes both consumers and non-consumers of a certain crop. Because a person can be consumer of a certain crop containing the pesticide of interest and non-consumer of other crops containing the pesticide of interest the consumers only approach is not suitable when considering the exposure levels for a pesticide via the consumption of more than one crop. Only the total population approach is suitable for this (Boon et al. 2004), (EFSA 2007). For rarely eaten crops as e.g. apricot, which was only consumed by 0.2% (n=4) of the child population, the exposure for the total population was 0.0% at P99.9, while it was 56% of the ARfD using the consumers only approach (Table III). Due to the risk of dilution for rarely eaten crops and to protect the small number of consumers both (Boon et al, 2004) and the US EPA (US EPA 2000c) recommend also to perform a consumers only or a point estimate calculation for rarely eaten crops.

Chronic dietary risk assessment

To compare the deterministic approach with the probabilistic approach estimates have been calculated for both children (4-6 years, 231 persons) and adults (15-75 years, 3299 persons). Figure 3 shows the mean chronic intake calculated by the deterministic approach as well as the mean and percentiles calculated by the probabilistic approach.

[Insert Figure 3 about here]

Using the deterministic approach, the chronic intake for adults and children was respectively 0.08 µg/kg bw/day and 0.22 µg/kg bw/day corresponding to 0.16% and 0.44% of the ADI of 50 µg/kg bw. The mean intake calculation in the probabilistic approach was nearly similar to the mean intake calculated by the deterministic approach. At the P99.9% percentile, the intake for adults and children were 0.35 µg/kg bw/day (0.70% of the ADI) and 0.76 µg/kg bw/day (1.5% of the ADI), respectively. Using the probabilistic approach instead of the deterministic approach in the calculation of the chronic intake gives insight of the exposure at different percentiles. Furthermore the uncertainties are quantified and the variability in the population is taken into account. The highest intake was estimated to 2.35 µg/kg bw/day (4.7% of the ADI) for both adults and children. Thus, the chronic exposure for Danish consumers to dithiocarbamate residues is far below the acceptable intake exposure using the ADI for mancozeb or maneb in the risk assessment.

Caldas et al. (2006) has also performed a probabilistic risk assessment of dithiocarbamates to the Brazilian population. They found that the general population and children was exposed to 6.9 and 13.5 µg/kg bw/day at the P99.9% percentile, which were higher than the exposure, calculated for the Danish consumers, but still acceptable compared to the ADI. In the study ½ LOQ was used to correct for undetected residues in the calculation, which could increase the calculated exposure as the LOQ is relatively high. When the number of samples without detected residues is high, the contribution to the intake will also be relatively high. In addition, the frequencies of dithiocarbamate residues were higher in the Brazilian study than in the present Danish study.

Conclusion

The aim of this study was to compare the outcome of probabilistic risk assessment and deterministic risk assessment to dithiocarbamates for both acute and chronic intake estimations and to see if the comparison between the point estimate approach and the probabilistic approach for the acute intake showed the same reported by Boon et al (2004).

For the acute intake calculations the comparison showed that the point estimate approach generally was higher or comparable to the exposure at the P99.9 as calculated with the probabilistic approach. Since the point estimate approach is very easy to perform and understand this approach could be used as first choice in a tiered process (US EPA 2000c). If this calculation indicates that there might be a problem with the acute intake, the probabilistic approach could then be applied to refine the calculations. Neither the point estimate, nor the probabilistic, consumers only approach can be used for estimating exposure to a pesticide from more than one crop. In this case the probabilistic total population approach should be used. On the other hand, the risk of dilution for rarely eaten crops could be masked, unless a probabilistic consumer only calculation or a point estimate calculation is performed. These conclusions are in accordance with those reported by Boon et al (2004).

Comparison of the chronic intake using both the deterministic and the probabilistic approach showed nearly the same estimate of the mean value. However, when using the probabilistic approach it is possible to estimate the exposure at different percentiles

as e.g. P99.9 and P99.99. The variability in the populations eating habits are taken into account and uncertainties is quantified.

In both calculation methods it was shown that there is no health risk for the Danish consumers to the dithiocarbamate residue levels found in the Danish monitoring program.

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Figure 1

Figure 2.

Figure 3

new Only

Table I

Commodity	Samples analyzed	Samples detected*	Mean concentration [†] (mg CS ₂ /kg)	Highest residue (mg CS ₂ /kg)
Apple	621	75 (12.1%)	0.034	0.79
Apricot	64	17 (26.6%)	0.173	3.25
Banana	386	1 (0.3%)	0.001	0.26
Bean without pod	82	4 (4.9%)	0.035	1.27
Beetroot	65	1 (1.5%)	0.008	0.5
Carrot	412	1 (0.2%)	0.001	0.27
Celery	105	4 (3.8%)	0.007	0.26
Cherry	55	5 (9.1%)	0.018	0.4
Cucumber	392	35 (8.9%)	0.025	1.17
Dill	20	2 (10.0%)	0.037	0.63
Fruit, exotic	389	49 (12.6%)	0.081	2.7
Grape, table	569	67 (11.8%)	0.039	2.49
Leek	110	9 (8.2%)	0.043	1.36
Lettuce	246	16 (6.5%)	0.083	5.3
Melon	171	12 (7.0%)	0.033	2.92
Parsley	71	5 (7.0%)	0.045	1.88
Peach, nectarines	303	35 (11.6%)	0.027	0.9
Pear	344	71 (20.6%)	0.066	2.09
Plum	121	8 (6.6%)	0.012	0.36
Raspberry	34	3 (8.8%)	0.028	0.43
Spinach	70	9 (12.9%)	0.441	21.7
Squash	153	2 (1.3%)	0.002	0.25
Strawberry	186	3 (1.6%)	0.011	0.99
Sweet pepper	271	20 (7.4%)	0.025	2.06
Tomato	450	17 (3.8%)	0.011	1.73
Watermelon	31	1 (3.2%)	0.006	0.17

* $\geq$ LOQ (0.1 mg CS₂/kg). Number of samples with detected dithiocarbamate, followed by frequency in parenthesis

[†] Levels < LOQ were considered as zero

Table II

Commodity	Mean consumption (g/day of total survey days)*		% consumption days†		Mean consumption (g/day, consumption days only)	
	Children	Adults	Children	Adults	Children	Adults
Apple	50.2	58.03	37	35.5	135.5	163.4
Apricot	0.14	0.13	0.2	0.4	55	29
Banana	25.13	27.56	27	20.4	93.2	135
Bean with pod	0.09	0.12	5.4	5	1.6	2.5
Beetroot	0.21	0.59	0.6	1.3	33.7	45.7
Carrot	30.18	32.9	43.8	43.4	68.8	75.7
Celery	0.19	0.59	0.5	1	37.5	61.7
Cherry	0.44	0.31	0.4	0.4	101.1	77.4
Cucumber	35.79	19.89	59.2	51.7	60.4	38.5
Dill	0.01	0.03	0.8	2	1	1.3
Fruit exotic	0.37	0.43	89.1	81	0.4	0.5
Grape, table	3.84	5.64	4.9	6	78.6	94
Leek	0.99	2.38	6.4	11.7	15.3	20.3
Lettuce	2.95	9.56	20.7	35	14.2	27.3
Melon	5.84	6.87	3.6	3.1	162.7	219.2
Parsley	0.02	0.05	1.9	4	1.2	1.2
Peach, nectarines	2.44	3.69	3.3	3.9	74.3	95.1
Pear	14.41	15.11	11.4	9.5	126.6	158.9
Plum	1.27	1.5	3.3	2.3	38	65
Raspberry	0.14	0.18	1	1.7	14.6	10.5
Spinach	0.5	1.08	6.9	7.3	7.2	14.8
Squash	0.22	0.94	0.9	2.5	25.3	37.7
Strawberry	2.27	2.85	3.2	5.3	70.6	54
Sweet pepper	4.87	7.91	36.3	48.9	13.4	16.2
Tomato	11.51	22.42	47.4	61.9	24.4	36.2
Watermelon	1.65	1.94	3.6	3.1	45.9	61.9

* Included zero and non-zero consumption days.
† % of non-zero consumption days for consumers.

Table III

Population, commodity	Point estimate	Probabilistic approach			Probabilistic approach		
		Consumers only			Total population		
		P99.0	P99.9	P99.99	P99.0	P99.9	P99.99
<i>Children (4-6 years)</i>							
Apple	16	5.3	20	41.3	2	13	34
Apricot	22.9	33	57	73.8	0	0	2.3
Cucumber	26.6	1.7	8.2	20.5	0	6.5	18
Lettuce	28	0.9	10	41.8	0	3	21
Peach, nectarine	12.8	2.3	9.3	21.4	0	0.9	5
Pear	42	9.7	47	59.5	0	11	35
Sweet pepper	18.7	3.4	24	60	1	11	44
Tomato	22.4	2.9	18	56.8	1	12	46
<i>Adults (15-75 years)</i>							
Apple	6	2.1	6.8	12.2	1	4.6	11
Apricot	10.2	2.3	2.6	3.8	0	0	0.8
Cucumber	7.2	0.3	2.1	5.4	0	1.4	4.6
Lettuce	14	0.5	6.8	21	0	3.1	13
Peach, nectarine	7.9	0.9	2.2	9.2	0	0.4	1.9
Pear	15	3.2	12	23	0	3.3	11
Sweet pepper	6.4	1.3	6	18.8	1	3.8	15
Tomato	7.5	1.3	8.9	21.2	1	6.4	20