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Chronic dietary risk for pesticide residues in food in Brazil: an update

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The objective was to conduct a dietary risk assessment of pesticides registered in Brazil up to 14 January 2004. The theoretical maximum daily intake (TMDI) was calculated for 275 compounds using the Brazilian maximum residue limits (MRL) and food consumption. The chronic dietary risk assessment was conducted by comparing the TMDI with the Brazilian acceptable daily intake (ADI) or, when not available, with the ADI from other sources. The TMDI was higher than the ADI (%ADI > 100) in at least one Brazilian region for eight pesticides, including five organophosphorus insecticides. For these compounds, the higher TMDI (expressed as %ADI) ranged from 140 (metam sodium) to 14 000 (methyl bromide). Rice, beans, citrus and tomato were the commodities that contributed most to ingestion. Change in pesticide use patterns and the establishment of ADIs by the Brazilian government have reduced the number of compounds for which the TMDI exceeded the ADI in the last 4 years. Risk assessment methodology based on TMDI calculation, however, is conservative as it assumes that the food supply is always treated with all the registered pesticides for that crop and that one always consumes food containing residues at the tolerance level. Furthermore, for six compounds with TMDI exceeding the ADI, a more realistic estimation of the pesticide daily intake was conducted using monitoring residue data from the Brazilian National Pesticide Residue Program. For these compounds, the higher refined intakes ranged from 2% (dimethoate) to 180% (fenitrothion) of the ADIs. The implementation of a national pesticide residue monitoring programme by the government was important to allow the refinement of the risk assessment. However, adequate daily food consumption data are still needed to assess better the public health risk to Brazilian consumers from food produced from crops treated with pesticides.

Keywords: pesticide residues, chronic dietary risk assessment, TMDI

Introduction

Brazil is the third largest market for pesticides in the world and places eighth in use per cultivated area (SINDAG 2003). Up to January 2004, 396 active ingredients were registered in the country for agricultural or urban use, including insecticides, fungicides, growth regulators, synthetic pheromones, antibiotics and organisms for biological control (ANVISA 2004). According to the Brazilian Pesticide Industry Union (SINDAG), 654 formulated products are currently market in the country (SINDAG 2003).

Man is exposed to pesticides occupationally, during the handling and application of the product, or through the consumption of treated food that contains their residues. Adverse effects are found in laboratory animals dosed with pesticides for long periods (WHO 2003) and governments are concerned with human exposure to these compounds in the diet and the potential risks to health (Wiles *et al.* 1998, EC 2001). The risks from chronic dietary exposure to pesticides can be assessed by comparing the daily intake with a toxicologically acceptable level, the acceptable daily intake (ADI). Risks might exist when the intake exceeds the ADI (WHO 1997).

A previously published chronic dietary risk assessment of the pesticides registered in Brazil up to 1999 showed that the estimated intake exceeded the ADI for 23 compounds (Caldas and Souza 2000). The present study shows an update of that study, based

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on the Brazilian pesticide monographs published by the National Sanitary Agency (ANVISA) of the Ministry of Health, up to 14 January 2004 (ANVISA 2004). The conclusions of the study can be used by the Brazilian government in planning future actions concerning pesticide residues in food to prevent unsafe exposure of consumers.

Materials and methods

The chronic dietary risk assessment was conducted by comparing the theoretical maximum daily intake (TMDI), defined as the summation of all maximum residue limits (MRL, $mg\,kg^{-1}$), multiplied by the food consumptions (F, $kg\,day^{-1}$), with the acceptable daily intake (ADI, $mg\,kg^{-1}\,bw\,day^{-1}$) of the pesticide. A person body weight of 60 kg is assumed in the calculation (WHO 1997). The %ADI is the estimate of the total intake expressed as percentage of the ADI. Risk might exist when the TMDI exceeds the ADI (TMDI exceeds 100% ADI).

MRLs and ADIs were obtained from the published Brazilian monographs of pesticides (ANVISA 2004). When not available at national level, the ADI was obtained from the Codex Alimentarius (2001), the USA (IRIS 2004), the Australian government (2003), the German government (2002) or *The Pesticide Manual* (Tomlin 2001).

The TMDI of the ethylenebisdithiocarbamates (EBDCs) mancozeb, maneb and metiram was grouped as they share a common mechanism of action (US EPA 2001). Molecular conversion factors (1 mol dithiocarbamate yields 2 moles CS_2) of 1.77, 1.69, 1.9 and 1.58 were multiplied by the intake calculated as CS_2 to obtain the intake of EBDCs, metam sodium, propineb and thiram, respectively.

Food consumption data (F) were obtained by the Brazilian Institute of Geography and Statistic between 1995 and 1996, in 11 metropolitan regions of the country (Belém, Fortaleza, Recife, Salvador, Belo Horizonte, Rio Janeiro, São Paulo, Curitiba, Porto Alegre, Distrito Federal and Goiânia) (IBGE 1999). In the survey, 10 families of different economic status were asked to register during 7 consecutive days the amount of food purchased. Factors of 0.87, 0.03, 0.1 and 0.2 were multiplied by the consumption of eggs, milk, meat and chicken, respectively, to obtain the consumption of eggs without shell, milk fat, meat fat and chicken fat (Teixeira and Luna 1996, FAO 2003) for which there are existing MRLs. The %ADI for each compound was calculated using food consumption data in each Brazilian region. The national TMDI ($TMDI_N$) and %ADI ($\%ADI_N$) were calculated using the average food consumption in the 11 regions.

The refinement of the TMDI was conducted by replacing the MRL in the calculation for the residue data from the Brazilian National Monitoring Program for Pesticide Residues in Food (PARA 2004). This programme, implemented in 2001 by the Ministry of Health through the National Sanitary Agency (ANVISA), analysed 96 pesticides in 2664 samples of lettuce, banana, potato, carrot, orange, apple, strawberry and tomato collected from June 2001 to October 2003 in 10 Brazilian capitals.

Results and discussion

Assessment of the dietary risks using the theoretical maximum daily intake (TMDI)

From the 396 active ingredients currently registered in Brazil, the chronic dietary risk was assessed for 275 compounds. One hundred and thirty-six compounds, or 49.4%, had the $TMDI_N \leqslant 1\%$ ADI, i.e. the national theoretical maximum daily intake of these compounds, considering the average national food consumption ($TMDI_N$), represented less than 1% of the toxicological intake (ADI) (figure 1). Only 13 compounds, or 4.7%, had the $TMDI_N > 50\%$ ADI. Seven compounds had the $TMDI_N$ contributing more than 100% ADI (table 1). Dimethoate ($TMDI_N = 100\%$ ADI) had an intake higher than the ADI in the metropolitan regions of Belo Horizonte (BH), São Paulo (SP), Curitiba, Distrito Federal (DF) and Goiânia. From the eight compounds with TMDI > 100% ADI in at least one region, five are organophosphorus insecticides (prothiophos, ethion, fenitrothion, methidathion and dimethoate). Citrus, tomato, rice and beans were the commodities that most contributed to the intake.

The risk assessment was not performed for 121 compounds with registered uses in Brazil. Fifty compounds are registered only for urban use, including house sanitation, gardening and public health; 23 compounds are registered only for sugar cane

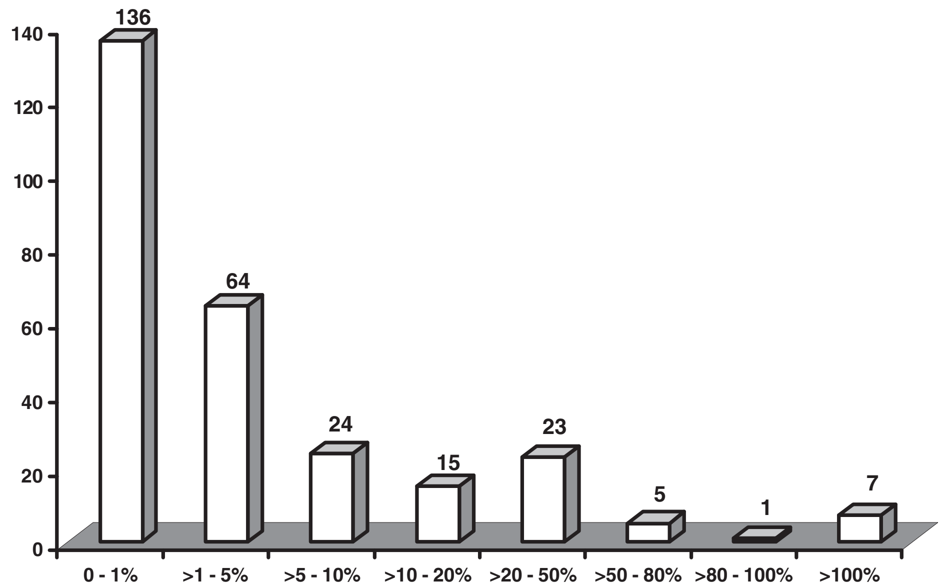

Figure 1. Distribution of the $TMDI_N$ expressed as $\%ADI_N$ for the 275 pesticides with registered uses in Brazil up to January 2004 evaluated in the present study.

Table 1. Compounds for which the national theoretical maximum daily intake ($TMDI_N$) contributed at least 50% of the acceptable daily intake ($TMDI \geqslant 50\%\ ADI_N$).

Pesticide	$\%ADI_N^{1,2}$	%ADI, low—high (region)	Food[3]	ADI (mg kg^{-1} bw day^{-1})
Methyl bromide	8000	6300 (RJ)—14 000 (SP)	citrus (65%)	0.0004[4]
Prothiofos	270	180 (Fortaleza)—340 (Goiânia)	tomato (95%)	0.0001[4]
Dicofol	210	150 (Fortaleza)—360 (SP)	citrus (84%)	0.002
Ethion	170	130 (Fortaleza)—240 (DF)	citrus (39%)	0.002
Fenitrothion	140	110 (SP)—180 (RJ)	beans (67%)	0.0005
Methidathion	140	100 (Fortaleza)—270 (SP)	citrus (99%)	0.001
Metam sodium[5]	120	70 (Fortaleza)—140 (P. Alegre)	tomato (76%)	0.001[6]
Dimethoate	100	70 (Fortaleza)—160 (SP)	citrus (73%)	0.002
Carbaryl	70	70 (Goiânia)—100 (Belém)	meat (47%)	0.003
Thiram	60	30 (Recife)—100 (DF)	rice (96%)	0.01
Pirimiphos methyl	60	34 (Recife)—90 (DF)	rice (70%)	0.03
Iminoctadine	50	30 (Fortaleza)—70 (P. Alegre)	tomato (79%)	0.0006
Propineb	50	40 (Fortaleza, Recife)—60 (DF)	tomato (37%)	0.005

[1]Using the average national consumption.
[2]Rounded to one or two significant figures.
[3]Food that most contributed to the TMDI.
[4]From the Australian database.
[5]Conversion factor from CS_2 to metam sodium was 1.69.
[6]From the German database.

(no consumption data available), cotton (no MRL for cotton seed oil), tobacco, feed items and ornamental plants, or are used as wood preservatives; 43 compounds do not have MRLs, including 28 pheromones and four organisms for biological control; four compounds are exclusively reserved for export commodities and for one compound, benzalkonium chloride, no ADI was found from national or international databases.

Benzalkonium chloride has been used mainly for sanitation and as an algacide in the urban and rural environment and as a veterinarian drug

Table 2. Compounds with TMDI exceeding 100% ADI in at least one Brazilian region, according to Caldas and Souza (2000), and the current situation with respect to use pattern and ADI.

Pesticide	Current situation
Prothiofos	no change
Dicofol, dimethoate, malathion, methamidophos, methidathion, pirimiphos methyl	establishment of a Brazilian ADI
Carbaryl, diazinon, disulfoton, ethion, fenamiphos, mevinphos, parathion methyl	change in the use pattern and establishment of the Brazilian ADI
Mancozeb, maneb	change in the use pattern and change in residue definition
Dichlorvos	urban use only
Azinphos-ethyl, benomyl, carbophenothion, dicloran, dicrotophos, ziram	use cancelled

(EMEA 1997). In Brazil, it is registered for direct foliar application in potatoes, coffee and tomatoes, with MRLs of $1\,mg\,kg^{-1}$, and in carrots, with an MRL of $2\,mg\,kg^{-1}$. However, the manufacturer recommends the product be used only on transportation vehicles and agriculture equipment as a bactericidal agent to control the citric cancro (CHEMITEC 2004). The company does not recommend direct application in any crop.

In the study conducted by Caldas and Souza (2000), based on the Brazilian monographs published until 1999, the TMDI exceeded the ADI (TMDI > 100% ADI), in at least one Brazilian region, for 23 compounds. Sixteen compounds were organophosphorus insecticides, and parathion-methyl was the compound for which the intake most exceeded the toxicological parameter (TMDI up to 13 000% ADI), followed by dichlorvos, pirimiphos-methyl and dissulton (TMDI up to 3700% ADI). When the study was conducted, no Brazilian ADI was available for the risk assessment and, for each compound, the lowest value found from other governments and the Codex Alimentarius ADI databases was used. Food consumption data used in that study are the same as currently used (IBGE 1999).

Table 2 shows the current situation concerning use pattern and ADI of the compounds found to be of potential risk to consumers by Caldas and Souza (2000). With the exception of methyl bromide, fenitrothion and metam sodium, all compounds found in the present study to have TMDI > 100% ADI in at least one Brazilian region (table 1), had the same situation in the previous study (table 2).

Methyl bromide is registered in the country to eliminate pests for import and export quarantine purposes in fruits and coffee (MRL between 5 and $75\,mg\,kg^{-1}$), and in Brazil nuts and cashew nuts (MRL of $200\,mg\,kg^{-1}$). It is unlikely that Brazilian consumers will eat tropical fruits (including avocado, citrus and papaya), coffee and nuts treated with the compound, as these crops are produced nationally and are exported. When only the fruits with registered uses (plums, apples, melons, pears, grapes and peaches), which might be imported by the country, are considered in the calculation, the $TMDI_N$ dropped significantly, from 8000 (table 1) to 570% ADI. Many studies have demonstrated that most of the methyl bromide applied for quarantine treatment is dissipated during storage and processing, and no residues are expected in the food at consumption (FAO 1980). Furthermore, it is likely that the real exposure and risks for Brazilian consumers to methyl bromide through the diet are not significant.

There were no changes in the last 4 years in the use pattern and ADI for prothiofos, and the result of the assessment in the present study ($TMDI_N = 270\%$ ADI) is the same as previously found (Caldas and Souza 2000). The calculated intake of prothiofos through the consumption of tomato represented 95% of the total intake, mainly due to the higher MRL of this crop ($1\,mg\,kg^{-1}$) among the five crops with registered use. Citrus fruits, which have the highest consumption by the population among the crops that can be treated with prothiofos, have an MRL of $0.01\,mg\,kg^{-1}$. According to the company responsible for the compound in the country (personal communication) and SINDAG (2003), no prothiofos product is presently being marketed in Brazil. According to the same sources, many other compounds with registered use are not or never have been marketed in the country, including the insecticides fluvalinate, terbupirinphos, etoprophos, pirazophos and alanicarb, and the fungicide ftalide.

Dicofol, ethion, methidathion and dimethoate had ADIs established by the Brazilian government in recent years (table 1). The national methidathion ADI confirmed the Codex Alimentarius value, which was previously used (Caldas and Souza 2000), and as no change in use pattern or MRL occurred, the $\%ADI_N$ remained the same for this compound. For dicofol, ethion and dimethoate, however, the ADIs established nationally (table 1) are two to four times higher than the ones used in the previous study obtained from US EPA and the Australian government database. Additionally, the use of ethion was cancelled in 11 of the 27 crops registered up to 1999. Furthermore, for these last compounds, there was a significant decrease of the TMDI in the present study (table 1), reaching 80% in the case of ethion (870 in the previous study to 180% ADI). The risk assessment conducted previously for fenitrothion indicated a $TMDI_N < 50\%$ ADI. A change in the MRL for bean, from 0.5 to 10 $mg\,kg^{-1}$ presently, was the main reason for the increase of the $TMDI_N$ in the present study, which contributed with 140% of the ADI.

In 1999, Brazilian legislation for the dithiocarbamate fungicides established MRLs individually for each compound, defined as itself, and the TMDI for mancozeb, maneb and ziram calculated by Caldas and Souza (2000) reached up to 1300% of the ADI in some regions of the country. In August 2003, the residue definition was changed and, for each crop, an MRL for the dithiocarbamate group, defined as CS_2, was established. This change was necessary to allow MRL enforcement with the available analytical methodology for these compounds in food (Keppel 1971, Caldas *et al.* 2001) and to harmonize the residue definition at international level (Codex Alimentarius 2001, EC 2001). In the present study, the TMDI calculation for the EBDCs (ethylenebisdithiocarbamates) mancozeb, maneb and metiram was grouped as they possess a common mechanism of toxicity (US EPA 2001). The change in residue definition and intake calculation, in addition to withdrawing uses on some crops and of ziram and the inclusion of propineb, lead to different conclusions over the potential risk that the daily intake of dithiocarbamates pose to Brazilian consumers. In the present study, only the metam sodium TMDI exceeded the ADI (table 1).

The current national ADI for carbaryl and mevinphos confirmed the Codex ADI used previously, but the withdrawal of some crop uses reduced significantly the TMDI for these compounds in this study. Cancelling the use of carbaryl in citrus and rice, which have a high consumption by the Brazilian population, and of mevinphos in potato and citrus had the most impact in the intakes. The higher TMDI of carbaryl decreased from 910% ADI in the study of Caldas and Souza to 100% presently (table 1). For mevinphos, the decrease was from 120 to 30% ADI.

For diazinon, disulfoton, fenamiphos, malathion, methamidophos, parathion-methyl and pirimiphos-methyl, the current national ADI is up to 150 times higher than the previously used value from US EPA and Australia, leading to a significant decrease in the TMDI expressed as %ADI. The $\%ADI_N$ found in the present study for pirimiphos methyl was 60 (table 1) and for the other compounds ranged from 3 (disulfoton) to 35 (diazinon).

The establishment of an ADI depends on the critical studies (performed on laboratory animals or humans), critical effects, the safety factors applied to the critical end point (no observable adverse effect level, NOAEL) and also on the quality of the studies (WHO 2003). The data are supplied mostly by the companies for government evaluation and different studies might be used, leading to the establishment of different ADIs. On the other hand, one government might choose to be more conservative, using higher safety factors or requiring a higher number and or more complex toxicological studies, which might also lead to different ADIs. In a review of 230 ADIs assessed by WHO and adopted by the Codex Alimentarius, Lu (1995) reported safety factors ranging from 6 to 1000. In addition to the species in which the study is conducted, safety factors can vary according to the uncertainties on the toxicological studies. Furthermore, the ADI is not an absolute parameter of toxicity, but expresses the availability of experimental results at the moment of the evaluation. In the USA, for example, the ADI, referred to as the reference dose, for pirimiphos-methyl in 1998 was 0.0002 $mg\,kg^{-1}$ body weight (US EPA 1998) but currently is 0.01 $mg\,kg^{-1}$ body weight (IRIS 2004).

The per capita food consumption data used in the studies are the only and the most up-to-date available in the country. They were obtained from the information provided by 10 families in each of 11 metropolitan regions of Brazil about the amount of food purchased over 7 consecutive days (IBGE 1999). There is a large difference in per capita income among these regions, and, in addition to regional particularities in diet, the consumption reflects the economic status of the families. In 11 of the 13 compounds

Table 3. Refinement of the dietary chronic risk assessment for the compounds with a TMDI exceeding 100% ADI using residue data from the Brazilian pesticide residues monitoring programme in food (PARA 2004).

Pesticide	Number of samples analysed[1]/detected[2]	Residues[3] ($mg\,kg^{-1}$)	MRL ($mg\,kg^{-1}$)[4]	$\%ADI_N$ refined[5] (%ADI low — high)
Dicofol	orange: 242/2	< 0.05–0.81	5.0	2 (2–4)
	apple: 208/9	< 0.05–0.40	5.0	
Ethion	orange: 242/0	< 0.01	2.0	60 (50–90)
	apple: 208/3	< 0.01–0.26	2.0	
	tomato: 380/0	< 0.01	2.0	
Fenitrothion	beans: 0/–	–	10	140 (100–180)
	orange: 242/1	< 0.01–0.50	0.5	
Methidathion	orange: 242/5	< 0.02–0.50	2.0	2 (1–3)
	apple: 139/1	< 0.02–0.04	0.02	
Metam sodium[6,7]	tomato: 380/129	< 0.05–2.2	2.0	7 (4–9)
	strawberry: 284/110	< 0.05–1.1	0.2	
	potato: 361/0	< 0.05	0.2	
	carrot: 261/0	< 0.05	0.3	
Dimethoate	orange: 242/0	< 0.02	2.0	1 (< 1–2)
	apple: 178/31	< 0.02–0.80	2.0	
	tomato: 380/0	< 0.02	1.0	

[1]Only crops for which there is existing MRL.
[2] > LOQ.
[3]LOQ—higher residues.
[4]ANVISA (2004).
[5]Using LOQ as the median residues.
[6]Residues found in the samples, as CS_2, from the use of any dithiocarbamate.
[7]Assuming all CS_2 coming from the use of metam sodium.

identified in table 1, the highest TMDIs were found in the richest regions of the country, including São Paulo (SP) and the Distrito Federal (DF). For example, the consumption of citrus in São Paulo is 29.1 $kg\,year^{-1}$, against a national average of 15.3 $kg\,year^{-1}$. Citrus is one of the commodities that contributed most to the estimated intake of many compounds (table 1). On the other hand, Fortaleza and Recife, from the less economically developed region of the country, had the lowest TMDIs for the most part of the compounds with potential risk (table 1).

Refinement of the intake using Brazilian monitoring residue data

The methodology that uses MRL as a parameter of residue concentration in the intake calculation is very conservative as it assumes situations that are unlikely always to occur. These assumptions include a life daily consumption of all the food items to which a MRL has been set; 100% of the daily food consumption will be derived from treated crops; 100% of the treated crop will contain pesticide residues at the MRL level; no dissipation or degradation of the pesticide will occur during storage, transport, preparation, commercial processing and cooking, and that all the residues are present in the edible portion. Furthermore, the refinement of the intake calculation with more realistic residue data will lead to a better assessment of the risks to consumers.

Results from the Brazilian National Monitoring Program for Pesticide Residues in Food (PARA 2004) showed that the dithiocarbamate fungicides, as CS_2, were the pesticides most detected in the samples analysed (21% of positive samples). These compounds also are the most prevalent pesticide in monitoring programmes in other countries (EC 2001). With the exception of metam sodium (dithiocarbamates, as CS_2) in tomato and strawberry and of dimethoate in apple, less than 5% of the analysed samples had detectable residue levels of the pesticides with potential risk (table 3).

Table 3 shows the result of the refinement of the intake calculation for the compounds found to have the TMDI higher than the ADI in the present study (table 1). In this refinement, the daily intake is calculated using the median residues found in the samples analysed in the PARA programme, replacing the MRL in the TMDI calculation. For each pesticide/crop combination, the median is the limit of quantitation of the method (LOQ), as $<50\%$ of the analysed samples had detectable residues ($<$LOQ) (table 3). The refinement of the study was not conducted for methyl bromide and prothiofos, as these compounds were not analysed in the programme.

The refined %ADIs were significantly lower than the TMDIs (expressed as %ADI) (table 1) for all the compounds, except for fenitrothion. Orange was the only crop registered for fenitrothion (as citrus) that was also analysed by the PARA programme. The consumption of citrus, however, represents less than 5% of total intake of fenitrothion in the Brazilian diet and the impact of the refinement was insignificant. For this compound, residue data on beans (responsible for 67% of fenitrothion intake) would be essential to refine the intake calculation.

Monitoring programmes, however, provide residue data in food at the point of marketing and knowledge on the changes in residue concentration during food processing such as washing, peeling, boiling and salting are important to generate residue data that reflect the concentration closer to the point of consumption. Jensen *et al.* (2003) evaluated the potential cumulative effects of 35 organophosphorus and carbamate pesticides in Denmark using monitoring data and food processing factors. Cumulative chronic exposure, in methamidophos equivalents, was 0.8–2% of ADI for adults and 0.07–27% of ADI for children. In Korea, the TMDI exceeded the ADI for 16% of the 262 pesticides evaluated (Chun and Kang 2003). The dietary oncogenic risk for three compounds with known oncogenic potencies was estimated to be, on average, 7.3×10^{-4} on the basis of TMDI and 1.8×10^{-7} on the basis of the refined intake, calculated with monitoring residue data and food processing factors.

Conclusions

The actions taken by the Brazilian government in the last years, including the withdrawal of some compounds, the changes in use patterns and the establishment of ADIs, reduced the number of compounds with potential risks to Brazilian consumers compared with a previous study conducted in 1999. The organophosphate insecticides, however, are still the compounds of most concern, with the intake coming mainly from the consumption of rice, beans, tomatoes and citrus. Dietary exposure to organophosphate and carbamate insecticides is of health concern due to potential cumulative effects of these pesticides that act through a common mechanism of toxicity, the inhibition of acetyl cholinesterase (Ecobichon 1995).

The implementation of the national pesticide residues monitoring programme was important to allow a better estimation of the exposure to pesticides and the risks to consumers. The present study has shown the need to include the analysis of rice and beans in the programme, as they are important in the Brazilian diet and contribute significantly for the estimated intake of some compounds.

The country, however, still needs an adequate food consumption database that would allow a more sound dietary risk assessment of pesticides and other contaminants. Food consumption data should include specific information for special groups in the population, such as children and pregnant women. The current database reflects the amount of food purchased by the general population, not consumed, and probably over- or underestimates the consumption of certain foods by certain groups.

Estimation of acute exposure and acute reference dose of pesticides have been added to the agenda of national and international authorities in the last 5–6 years (FAO 2003, Jensen *et al.* 2003, Hamilton *et al.* 2004). It is essential that in addition to improving the database for chronic risk assessment, the Brazilian government initiates projects to generate data for the assessment of risks to consumers from acute exposure after consumption of one food portion containing pesticide residues.

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