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Risk assessment of the cumulative acute exposure of Hungarian population to organophosphorus pesticide residues with regard to consumers of plant based foods

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ABSTRACT

Based on the Hungarian pesticide residues monitoring data of the last five years and the consumption data collected within a 3-day dietary record survey in 2009 (more than 2 million pesticide residue results and almost 5000, 0-101-year-old consumers 3 non-consecutive-day personal fruit and vegetable consumption data), the cumulative acute exposure of organophosphorus pesticide residues was evaluated. The relative potency factor approach was applied, with acephate chosen as index compound. According to our conservative calculation method, applying the measured residues only, the 99.95% of the 99th percentiles of calculated daily intakes was at or below 87 μg/kgbwday, indicating that the cumulative acute exposure of the whole Hungarian population (including all age classes) to organophosphorus compounds was not a health concern.

1. Introduction

Pesticide residues potential dietary exposure is mainly calculated before MRLs are established by the Codex Alimentarius Commission, the European Union and many countries around the world. In the Codex MRL setting process, it is the JMPR (Joint Meeting on Pesticide Residues) which performs the dietary risk assessment. These calculations are based on deterministic estimates of the expected intake of the proposed chemicals. After introduction of pesticides to the market, more realistic estimates of actual intake can be made considering monitoring results, which are collected on national level. Concerns have been raised whether the simultaneous exposure to several pesticide residues could have adverse effects on human health, which was not reflected at the one-by-one evaluation of these chemicals.

The Panel on Plant Protection Products and their Residues (PPR Panel) of the European Food Safety Authority (EFSA) issued a scientific opinion concerning the suitability of methodologies and identification of new approaches to assess the cumulative and synergistic risks to pesticides (EFSA, 2008). A tiered approach was recommended for both the hazard characterization and exposure assessment steps of cumulative risk assessment, which has been subsequently (EFSA, 2009) tested on a group of triazole pesticides. There are several methods to combine toxicity from dose-additive co-exposure, of which the relative potency factor approach is a preferred one for probabilistic assessments (EFSA, 2008; Boobis et al., 2008).

Several publications have already dealt with cumulative exposure assessment of pesticide residues (Caldas et al., 2006; Boon et al., 2008; Jensen et al., 2013; Müller et al., 2009). Relative potency factor (RPF) method was used in these publications, i.e. the potencies of each pesticide in a group have been calculated relative to a chosen reference pesticide (index compound), based on their relevant benchmark doses (or NOAELs) for the common effect.

For most pesticide residue monitoring results the non-detects or non-reported values are dominant. For instance, the 2013 report on pesticide residues in food (EFSA, 2015) has included results of analyses of 685 pesticides in 80,967 samples from the European Union. Overall, 97.4% of the tested food samples fell within the legal limits and 54.6% of the samples contained not quantified residues (EFSA, 2015). The EFSA guidance on probabilistic modelling (EFSA, 2012) suggests to count residues below the LOQ/LOR as zeros in

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Abbreviations: EFSA, European Food Safety Authority; LOQ, limit of quantification; LOR, limit of reporting; OP, organophosphates; RPF, relative potency factor; NOAEL, no observed adverse effect level; MRL, maximum residue limit.

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an optimistic approach, and as LOQ/LOR in a pessimistic approach and to use parametric modelling for a refined assessment (EFSA, 2012).

Organophosphorus esters (OPs) and carbamates are known for their acetylcholinesterase inhibiting activity, resulting in acute cholinergic effects. Their mechanism of action is not completely the same, while OPs act mostly irreversibly, carbamates act reversibly, therefore cumulating exposure for both groups would probably overestimate the risk (Boon et al., 2008).

In our paper, we assess the cumulative acute exposure to acetylcholinesterase inhibiting organophosphorus insecticide residues detected in foods of plant origin utilising the Hungarian food consumption and pesticide residue monitoring data from the last 5 years.

2. Materials and methods

2.1. Consumption data

Consumption data of 84 foods of plant origin were obtained from the national dietary survey performed with 3-day dietary record method in 2009, using the sampling procedures of the Hungarian Central Statistical Office (Szeitz-Szabo et al., 2011). A total of 4992 people reported food consumption for 3 non-consecutive days including one weekend or holiday, giving 14,976 consumption days in the database, which were considered to be independent.

The final form of the consumption database mostly contained raw product equivalents of the processed products to enable direct link to residue concentrations measured in raw products. The database contained some processed foods as well, which had not yet been converted to raw equivalents such as orange juice, apple juice, wheat flour.

2.2. Residue data

Pesticide residue data were derived from the Hungarian Pesticide Monitoring Programme, which covered all fruits and vegetables with daily consumption >0.12 mg/kgbw/day. The number of samples to be taken is usually decided annually taking into account the results of previous years.

The residue database included 64 foods of plant origin containing the residues of one or more of 24 substances that were measured from the organophosphate group. Food of animal origin was not considered as no residue was detected in any of the samples analysed. The laboratories in most cases applied the QuEChERs multi-residue procedure (Anastassiades et al., 2003; CEN, 2008) optimised for their available GC or GC/MS instruments. The LOQs ranged from 0.005 mgkg^{-1} to 0.1 mgkg^{-1} depending on the sensitivity of the detection system and the pesticide residue/matrix combinations analysed. The residue data from the evaluated five years amounted to about 2,218,000 and 225,251 specifically for the relevant pesticide residues. For most of the crop–residue combinations, the proportion of non-detects was larger than 80–90%. It was not possible to reliably estimate the proportion of true zeroes, therefore, as a frequently used conservative approach (EFSA, 2010), we decided to omit non-detects from the calculations (chosen as our basic approach). This basic approach is an acceptable compromise for acute intake calculations, when the high percentile daily intakes are of interest. It is also in line with our previous findings (Zentai et al., 2013) which showed that considering non-detects at LOQ or 0 residue level reduced the upper percentiles of exposure estimates due to the much larger data population containing the non-detects.

To validate our basic approach, we also decided to study the effect of inclusion of non-detects on the final exposure estimates calculated for a random subpopulation of 1000 consumption days. To model this effect, zeros or LOQ values (0 scenario and LOQ scenario) were added to the measured positive results in twice the amount, assuming the ratio of non-detects was 66% which is about a middle value between the European average and the Hungarian results. When different LOQ values belonged to a pesticide-crop combination, the values were approximately proportionally selected.

Processing factors were not taken into account in our calculations, given that the majority of consumption data were expressed in raw commodities. Furthermore, residues were directly measured in fruit juice and flour. Cooking and baking raw products would reduce the residue levels. Since there is very little information available on the effect of such processes, we have no alternative than using the residue levels in raw food items.

Application of variability factors for residues and unit weights of crops (which affect the calculation of medium-sized crops intake) was omitted as well, in order to reduce the runtime of modelling, and because the majority (68%) of consumed food items was not medium sized.

2.3. Methods

All the foods (n = 64) in which the organophosphorus compounds were analysed were matched to consumed items (n = 84) from the consumption database. Thus occasionally more than one (2 or 3) commodities, in which residues were measured, could be considered together with corresponding food items from the consumption database. For example, residues measured in wine-grapes, grapes (unspecified) and table grapes were combined and used with the table grape consumption figures for calculation of residue intake because wine-grapes are also consumed occasionally. The combined residue values were linked to the respective consumed commodities.

The residue values of the twenty-four organophosphorus pesticides, having the same mechanism of toxicological action, were expressed in acephate equivalents as the reference compound according to Caldas et al. (2006) and Boon et al. (2008). In case different RPF values were reported by Caldas and Boon for the same chemical, the higher one was used. Table 1 presents the pesticide residues and their relative potency factors involved in the calculation. Altogether 1450 positive residue data were available for intake calculations. Concentration data of different residues measured in the same sample were all converted to the index compound taking into account their RPFs. Cumulative residue content of a sample expressed as acephate equivalent was calculated by summing up the adjusted residue values (see equation (1)).

$$\mathrm{Req}_{\mathrm{acephate}} = \mathrm{R}_{\mathrm{acephate}}*1 + \mathrm{R}_{\mathrm{azynphos-methyl}}*\mathrm{RPF}_{\mathrm{azinphos-methyl}} + \mathrm{R}_{\mathrm{chlorpyrifos}}*\mathrm{RPF}_{\mathrm{chlorpyrifos}} + \ldots.. \tag{1}$$

To calculate the cumulative exposure for a consumption day, each food consumption data of that day were multiplied by all relevant acephate equivalent residue concentrations, and the calculated intakes from the different foods were summed up in all possible combinations. The following example is an illustration of the procedure: a person consumed apples, pears and grapes on a given day, and residues were detected in 6, 4, and 7 samples, respectively. The apple consumption is multiplied by the 6 residue values, the pear consumption is multiplied by the 4 residue values and these results are added in all combinations (24). Then the grape consumption is multiplied by the 7 residue values and added to the