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Chronic cumulative risk assessment of the exposure to organophosphorus, carbamate and pyrethroid and pyrethrin pesticides through fruit and vegetables consumption in the region of Valencia (Spain)

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ABSTRACT

In the present study, the chronic cumulative exposure to organophosphorus (OPs), carbamates (CBs) and pyrethroid and pyrethrin (PPs) pesticides in the region of Valencia through fruit and vegetables consumption is presented. A total of 752 samples and 84 pesticides were studied of which, 52 were OPs, 23 CBs and 9 PPs. Residue data were derived from the Valencia Region monitoring program 2007–2011 and food consumption levels from a questionnaire-based dietary survey conducted in 2010 in the same area. The relative potency factor (RPFs) approach was used to estimate chronic cumulative dietary exposure to OPs, CBs and PPs using acephate, oxamyl and deltamethrin as index compounds, respectively. The exposure was estimated using a deterministic approach and two scenarios were assumed for left-censored results: the lower-bound (LB) scenario, in which unquantified results (below the limit of quantification (LOQ)) were set to zero and the upper-bound (UB) scenario, in which unquantified results were set to the LOQ. Results demonstrate that the chronic exposure of the young (<16 years) and adult (≥16 years) population to pesticides through fruits and vegetables is under control (even at high or frequent consumption of fruits and vegetables), for the three groups of pesticides.

1. Introduction

Pesticides are used before and after harvest to protect crops from infestation by pest and plant diseases. The use of pesticides in the European Union (EU) is intensive. Only in 2010, pesticide-active ingredient consumption in the EU was about 208,000 tons (including insecticides, fungicides and herbicides), of which 19% (about 39,000 tones) had their origin in Spain (AEPLA, 2009). Valencia is the second most important agricultural area in Spain and one of the regions with the highest pesticide use, 12.1% of the national total in 2009 (ECPA, 2010). One possible consequence of pesticide use is the presence of their residues in treated commodities and eventually in the food chain. It is therefore necessary to ensure that such residues in food do not present an unacceptable risk for consumers, due to the fact that pesticides are biologically active compounds with a component-specific inherent toxicity. Although pesticide exposure can occur through different routes, such as inhalation, dermal exposure and dietary and non-dietary intake, food has been recognized as the main exposure route to pesticide residues for consumers not working with pesticides (Juraske et al., 2009). According to the World Health Organization (WHO), food consumption consists on average of 30% of fruits and vegetables (WHO, 2003) and it is well known that fruits and vegetables are more contaminated by pesticides than products of animal origin (Claeys et al., 2011). In addition, fruits and vegetables are frequently consumed raw or semi-processed and as a consequence it is expected that they contain higher pesticide-residue levels than other food groups of plant origin such as cereals.

To evaluate food safety, exposure needs to be assessed and compared to health safety limits or toxicological endpoint values.

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For quantitative exposure assessment, data on food consumption and chemical occurrence are usually combined using either a deterministic (Jensen et al., 2003) or probabilistic approach (Nougadère et al., 2012). The Scientific Panel on Plant Protection products and their Residues (PPR) recommended (EFSA, 2010), for chronic exposure assessments, a tiered approach for exposure estimations, starting with different deterministic methodologies and ending with probabilistic approaches. If a lower tier assessment (such as the deterministic assessment) does not give adequate reassurance of safety, the risk assessor should progress to higher tier method (including probabilistic methodologies). For this reason, a deterministic approach was selected to carry out this chronic cumulative risk assessment. Dietary assessment of the exposure to pesticide residues is traditionally performed for single compounds. However, people are daily exposed to more than one pesticide through diet, because a food may contain more than one residue or people consume combinations of foods containing different pesticides. If these compounds have the same toxicologically endpoint and the same mechanism of action, the conventional way of assessing the dietary risk of exposure to pesticides separately may lead to an underestimation of the health risk (Boon et al., 2008). Therefore, to address the risk of exposure to such compounds, the individual exposures should be addressed simultaneously. In this respect, cumulative exposure can be defined as simultaneous or subsequent exposures to various chemicals that contribute to a cumulative effect.

To assess the pesticides as a mixture the first step is to identify the substances with a common mechanism of toxicity (Gallagher et al., 2015). Some organophosphorus (OPs), carbamate (CBs) and pyrethrin and pyrethroid (PPs) pesticides causing a common neurotoxic effect (cholinesterase inhibition for OPs and carbamate and behavioral changes for PPs) have been identified by United States Environmental Protection Agency (USEPA) as members of the same cumulative assessment group (USEPA, 2006; USEPA, 2007; USEPA, 2011). To assess the cumulative dietary exposure, the relative potency factor (RPF) approach, based on dose addition is believed to be appropriate to estimate the cumulative effects of common mechanism chemicals (Jensen et al., 2013; Kennedy et al., 2015). This approach assumes doses of component chemicals that act in a toxicologically similar manner can be added together after scaling the doses by their potencies, relative to a selected index chemical, to generate exposure equivalents. By comparing the cumulative dietary exposure with a toxicological reference dose (in the case of chronic exposure, acceptable daily intake (ADI)) of the chosen index compound, the risk can be evaluated.

The aim of this study is to calculate, under a deterministic approach, the chronic cumulative chronic exposure to OPs, CBs and PPs pesticides through fruits and vegetables consumed by local inhabitants of the region of Valencia, and to characterize the associated risk. For the chronic cumulative risk assessment, acephate, oxamyl and deltamethrin were selected as index compounds for OPs, CBs and PPs, respectively.

2. Materials and methods

2.1. Food sampling and residue data

Residue data were obtained from the Valencia Region monitoring program 2007–2011.

A total of 752 individual samples of fruits and vegetables (see Table 1) were collected in different areas (covering rural and urban areas and different geographic location) and seasons to take into account potential variability. Sampling was carried out by inspectors from the Public Health Department and it was performed according to Directive 2002/63/EC on sampling for official control of pesticide residues (European Commission, 2002) at wholesalers. Although the sampling plan was set up to ensure compliance with the maximum residue limits set in the legislation, the most frequently consumed commodities in the Region of Valencia were considered based on risk assessment.

Only *pesticide-food* pairs with at least one quantified residue above the limit of reporting (LOR), corresponding to the LOQ value of 0.01 mg/kg, were included in the calculations. Analytical concentrations were normalised to wet sample weights and results were corrected by the recovery rate.

Left-censored results (data below the quantification limit) were processed according EFSA recommendations (EFSA, 2012). Residues below the LOR were treated as true zeros in the optimistic or lower-bound (LB) scenario assessment and were set to LOR in the pessimistic or upper-bound (UB) scenario assessment.

2.2. Analysis of pesticide residues

Analyzes were conducted by the Laboratorio Agroalimentario de Valencia (Burjassot), a European Union Reference Laboratory, accredited following the ISO/IEC 17025 standard. Multiresidue methods based on either gas chromatography (GC) or high performance liquid chromatography (HPLC) coupled with mass spectrometry (MS) were used to screen the 84 pesticides (52 OPs, 23 CBs and 9 PPs, see Table 1).

2.3. Consumption data

Intake estimates were based on consumption data obtained from a questionnaire-based dietary survey that was conducted and validated in 2010–2011 by the Valencia Public Health Directorate. Dietary data were collected through a 24-h recall in which 1478 subjects (195 young people from 6 to 15 years and mean 43.5 kg body weight and 1281 adults from 16 to 95 years and 71.2 kg mean body weight) were asked in a face-to-face interview to recall and describe the kinds and amounts of all foods and beverages ingested during the previous24-h period. The initial sample of individuals was divided in three waves or groups in order to take into account variations in consumption patterns according to season; the first wave was conducted between the months of June and July 2010, the second between September and November, and the third between November and February of 2011. Self-reported body weight was also collected in the face-to-face interview and used in exposure calculations.

In dietary surveys, the consumption is given for food eaten, while the residues of pesticides are usually determined in raw agricultural commodities (RACs). The approach used to convert “food as eaten” to edible RACs was that of P. E. Boon et al. (2008) using the Dutch conversion table.

2.4. Chronic cumulative risk assessment and risk characterization

The first step for the calculation of the chronic cumulative exposure was to obtain the individual pesticide intakes (or the intake for each single compound), from the consumption data in our dietary survey and the analytical data from the present study. A deterministic approach, based on single point estimates that were used for each variable within the model (such as an average value or the 95th percentile) was used. The individual estimated daily intake (iEDI) in a pair pesticide-commodity was calculated for young children (6–15 years) and for adults (16–95 years) according to the following equation:

iEDI (mg/mean kg body weight/day) = $C_i \times F_i$where

C_i: Residue level for food commodity