Science of the Total Environment 527–528 (2015) 262–269

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

The influence of tomato processing on residues of organochlorine and organophosphate insecticides and their associated dietary risk

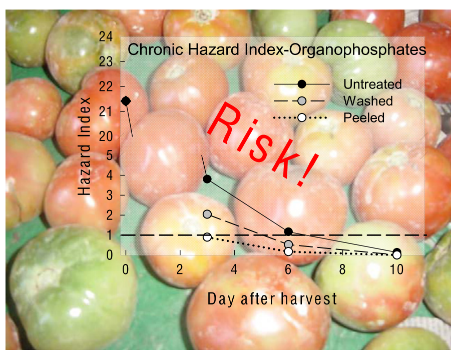
Emilie Reiler [a], Erik Jørs [b], Jesper Bælum [b], Omar Huici [c], Maria Mercedes Alvarez Caero [d], Nina Cedergreen [a,*]

[a] Department of Plant and Environmental Sciences, University of Copenhagen, Thorvaldsensvej 40, 1871 Frederiksberg, Denmark
[b] Department of Occupational and Environmental Medicine, Odense University Hospital, University of Southern Denmark, Sdr. Boulevard 29, 5000 Odense C, Denmark
[c] Fundacion Plagbol, Calle Fernando Guachalla 705, Sopocachi, La paz, Bolivia
[d] Center of Water and Environmental Sanitation, Faculty of Science and Technology, University Major of San Simón, Sucre street in front of Park La Torre, Cochabamba, Bolivia

HIGHLIGHTS

- Six organochlorines and five organophosphates were analyzed in 54 tomato samples.
- All insecticides were detected in the µg/kg range (OC) and mg/kg range (OP).
- Storage, washing and peeling reduced the concentrations.
- A cumulative risk assessment showed elevated risk for up to 6 days of the OPs.
- Farmer education and introduction of less hazardous pesticides are urgently needed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 16 January 2015
Received in revised form 19 April 2015
Accepted 21 April 2015
Available online xxxx

Editor: D. Barcelo

Keywords:
Dietary risk assessment
Consumer health
Pesticides
Organochlorines
Organophosphates
Hazard index

ABSTRACT

Due to the increasing food demand, the use of pesticides in agriculture is increasing. Particularly in low income countries poor training among farmers, combined with the use of obsolete pesticides may result in a high risk for the consumers. In this study six organochlorines and five organophosphates were analyzed in 54 samples of tomatoes from small scale farmers in Bolivia. The analyses were done on unprocessed, stored, washed and peeled tomatoes. The cumulated risk associated with consumption of the tomatoes after different storage times and processing treatments was evaluated using the Hazard Index (HI) for acute risk assessment. All 11 pesticides were detected in the analyses although several of them are obsolete and included in the Stockholm convention ratified by Bolivia. The organochlorines were found in the µg pesticide/kg tomato range and below the HI, while the organophosphates were present in the mg pesticide/kg tomato range and most often above the HI. The low organochlorine concentrations were not significantly affected by time or treatment, but storage significantly decreased the concentrations of organophosphates. Washing decreased the initial concentrations to between 53% (malathion) down to 2% (ethyl parathion), while peeling had a larger effect reducing the initial concentrations to between 33% (malathion) and 0.7% (chlorpyriphos). Both the acute and chronic cumulative risk assessment of organophosphates showed a dietary risk for unprocessed tomatoes three days after harvest. For children, also the consumption of washed tomatoes constituted a dietary risk. To reduce the dietary risk of pesticide residues in Bolivia, there is an urgent need of farmer education and introduction of less hazardous pesticides as well as resources for surveillance and enforcement of legislation in order to ensure public health.

* Corresponding author.

http://dx.doi.org/10.1016/j.scitotenv.2015.04.081

1. Introduction

There is an ever-increasing degree of public awareness and concern surrounding the issue of pesticides. Although the most commonly reported cases of adverse pesticide effects on human health are due to self-harm and occupational exposure, pesticide residues in food may add to exposures and/or in themselves cause a potential risk in the general population.

The use of pesticides in crops and the levels of residues in food are regulated in high income countries. Thus pesticide residues in food are generally low and not considered to cause severe adverse effects in the consumers (Jensen et al., 2003, 2009; Lozowicke, 2015; Nougadère et al., 2012). A lack of regulation and knowledge in low income countries, however, results in the farmer's practices and perceptions of pesticide risk being very different compared to standards in high income countries (Ecobichon, 2001). In low income countries pesticides are usually bought from agricultural supply stores, general shops, pharmacies or markets and are often dispensed in smaller quantities and in unlabelled containers, thus making it impossible for users to get information of optimal use practice, toxicity and safety measures needed (Dasgupta et al., 2007; Ngowi et al., 2007; Snelder et al., 2008). Use of instructions provided on the pesticide packaging is low, even when available, and the majority of farmers are functional illiterates and do seldom read the instructions for use, although it is an important factor for prevention of poisoning (Jørs et al., 2006). Instead the learning methods of spraying and tank filling are obtained by imitating relatives or neighbors (Ecobichon, 2001).

In Bolivia tomatoes are a major economic crop for small scale farmers living close to the big markets in the cities (Alverez et al., 2010). In most areas the tomato is grown conventionally implying a high use of external inputs such as chemical fertilizers and pesticides, but without the knowledge of how to use these inputs optimally and safely (Jørs et al., 2006). In addition the prescribed pre-harvest interval is generally not respected (Jørs et al., 2006). Using a higher dose and not respecting the prescribed time between the last spray and harvest make the probability of accumulation of residues in crops greater, thus potentially jeopardizing the health of the consumer.

With today's complexity in the mixture of chemicals present in our environment, their possible combination effects have attracted increasing amounts of attention (Reffstrup et al., 2010). Particular emphasis has been put on the shortcomings in the current mainstay of risk assessment. No matter how precautionary it may seem for the separate pesticides, risk assessment today widely disregards how different chemicals may add up or even act together to produce an effect (Kortenkamp, 2007; Kortenkamp et al., 2009; Reffstrup et al., 2010) and only certain chemicals sharing a common mode of action are subject to cumulative risk assessment (US EPA, 1999). Mixtures of chemicals in concentrations below their respective No Observable Adverse Effect Concentration (NOAEL) for a particular endpoint have been shown to be able to produce adverse effects simply by adding them up (Kortenkamp et al., 2007).

The aim of this study was to investigate whether dietary exposure of pesticides through a diet rich in tomatoes could pose a risk to human health using a cumulative risk assessment approach. In addition we wished to quantify the degree of risk reduction obtained by storage and treatment of the tomatoes. Therefore, Bolivian tomatoes were analyzed for two large groups of insecticides; the organochlorines and the organophosphates, both known for having relatively high human toxicities. The organochlorines are of special interest due to their remarkable persistence in the environment and bioaccumulation in organisms, which makes many of them listed in the Stockholm Convention of banned or restricted persistent organic pollutants (Stockholm Convention on Persistent Organic Pollutants, 2001). Organophosphates, on the other hand, are less persistent but acutely toxic and widely used (Boobis et al., 2008; Buratti et al., 2007; Marrs, 1993).

To make a cumulative risk assessment of pesticide residues in food we used the approach described by Jensen et al. (2003). For the risk assessment the acute reference dose (ARfD) and the acceptable daily intake (ADI) were used as predicted no effect levels for acute and chronic exposures, respectively. Both measures define the maximum dose which, according to all known facts at the time, will result in no harm to human health. The ARfD is the limit for consumption in one meal or in one day (acute toxicity), and the ADI is the amount that can be consumed every day for a lifetime (chronic toxicity) (IPCS, 2009). In the case of oral exposure, exposure assessment equals intake assessment. This can be made using different models, ranging from worst-case scenarios to more probabilistic models based on surveillance data (Boobis et al., 2008). In this study we use the country based estimations of intake of tomatoes for children and adults determined by the World Health Organisation (EFSA/FAO/WHO, 2011). Risk is then defined by the ratio of pesticide intake to ARfD or ADI yielding a hazard quotient (HQ) (see Eq. (1a) and (1b)). If the HQ has a value higher than 1.0 (>100% of ARfD or ADI), this indicates that the intake exceeds the value believed to be safe, hence, there is a risk (Boobis et al., 2008; Wilkinson et al., 2000).

$$HQ_{Chronic} = \frac{\text{Estimated Daily Intake}}{\text{ADI}} \tag{1a}$$

$$HQ_{Acute} = \frac{\text{Estimated Daily Intake}}{\text{ARfD}} \tag{1b}$$

In order to assess the cumulative effect of chemicals with the same mode of action, individual Hazard Quotients were summed to yield a hazard index (HI) (Eq. (2)) representing cumulative toxicity for chemicals with a common mode of action. The method is frequently applied to organophosphates (Jensen et al., 2003; Wilkinson et al., 2000).

$$HI = \sum\nolimits_{\mathrm{i}}^{n} \mathrm{HQ}_{\mathrm{i}} \tag{2}$$

More sophisticated ways of assessing cumulative risk exist (Boobis et al., 2008), however, in this study, we will use the HI based on cumulative groups as described in Jensen et al. (2003).

2. Materials and methods

2.1. Study area

The study was conducted in 2008 in 17 villages in the Municipalities of Omereque and Rio Chico placed relatively close north and south of Bolivia's Capital Sucre. In both regions the tomato is of growing economic interest for export to the markets in the big cities, such as Sucre. The municipality of Omereque has a population of 5148 inhabitants and is situated at an altitude of 1550 m above sea level having an annual average temperature of 23.0 °C and an average precipitation of 641 mm. Rio Chico has 10,630 inhabitants, is situated at an altitude of 1860 m and has an average annual temperature and precipitation of 22.8 °C and 506 mm, respectively.

2.2. Sampling

Samples of tomatoes were collected at harvest time by simple random sampling at 18 producers in 17 villages in the two municipalities. Tomatoes from the 18 producers were pooled in six groups representing well defined areas in the two municipalities. From each group three 2 kg sub-samples were selected and stored in paper bags for later treatments and pesticide analyses at day 3, day 6, and day 10 after harvest (Fig. 1).

Each sub sample was then either: left untreated (1), one was washed by cleaning with a soft brush under running water for approximately 1 min per tomato (2) and one was peeled with a knife (3). A total of 54 samples were prepared for analysis (18 samples per sampling time). All safety precautions, including use of special gloves to protect from organic compounds, were taken in the laboratory.