Risk Assessment of Tunisian Consumers and Farm Workers Exposed to Residues after Pesticide Application in Chili Peppers and Tomatoes

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ABSTRACT

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In Tunisia, to prevent and control pests and diseases during cultivation under greenhouses, chili pepper and tomato require the use of a wide range of pesticides potentially toxic and thus presenting a possible risk for farm operators, workers or consumers. A study has been carried out in the Sahel region of Tunisia to assess the risk for farm operators and workers exposed, by contact during harvest tasks, to possible pesticide residues remaining in tomato and chili pepper crops, and for the Tunisian consumers (adults and children) after intake. A questionnaire was addressed to a group of 73 market gardeners to better understand the local professional practices and to determine the main route of exposure to pesticide. Twenty samples of cotton gloves (2 pairs / sample) were distributed to 20 volunteers who worn them for two consecutive half-days during the harvest of chili peppers or tomatoes before analysis of the dislodgeable pesticide residues which could be transferred from crops to hands. Using models, predictive exposures values were calculated for consumers and farm workers. The highest exposure of consumers was observed for chlorpyrifos residues on tomatoes (with 82% and 312% of the Acute Reference Dose (ARfD), for adults and children respectively). The systemic exposure (SE) of farm workers was estimated for the median, the 90th percentile and the maximum concentration. At the highest observed concentrations, 15 pesticide residues (active ingredients and metabolites) used in pepper greenhouses, and 9 in tomato crops, exceeded the Acceptable Operator Exposure Level (AOEL). Exposure appeared to be particularly critical for chlorothalonil sprayed in chili pepper greenhouses with SE_{MAX} values 113 times higher than the AOEL (11285%). Long task duration (8 h/day) after re-entry in greenhouse, limited access to personal protective equipment (PPE), lack of hygiene and bad habits (eating, drinking, or smoking at work) have also been observed and discussed as risk factors.

Keywords: Consumers, dermal exposure, farm workers, pesticide residues, risk assessment, Tunisia

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In Tunisia, horticulture is an important, dynamic and vast agricultural sector. Today the total area of vegetable crops (field, tunnel and greenhouse) exceeds 160,000 ha. Tunisia produces around 3.2 million tons of vegetable crops (GIL 2015), mainly tomato (39%), watermelon (15%), onion (12%), potato (11.5%) and chili pepper (10%) (APIA 2015; GIL 2015). Tomato and green pepper are a basic component of the Tunisian diet and are used almost on a daily basis as part of raw or home cooked preparations (Jeder et al. 2017).

During cultivation, chili pepper and tomato require the use of a broad range of pesticides to prevent and control pests and diseases. Fruits and vegetables are sprayed several times and up to the final harvest. Pesticides are considered necessary by farmers to provide high crop yields ensuring food security, high agriculture productivity and good quality products. Despite their popularity, pesticides are potentially toxic to humans (farm operators, workers or consumers) and can generate both acute and chronic health effects (WHO 2005), mostly in developing countries (Ortiz et al. 2002). Harvested products are often put onto the markets without consideration of the pre-harvest interval (PHI). As a consequence, the pesticide residues left on fruits can generate a potential health hazard for consumers (Chourasiya et al. 2015; Darko and Akoto 2008; Elgueta et al. 2017; Nougadère et al. 2012).

Many pesticides sprayed on tomatoes and chili peppers leave persistent, fat-soluble pesticide residues which can be dislodged from the two-sided foliar surface of a plant through contact. Workers who enter treated areas for pruning or who handle products during harvesting can easily absorb residues through their skin (EFSA 2014),

are potentially exposed daily and thereby possibly endangering their health. Potential exposure of workers through contact with treated foliage is a significant concern in greenhouse production. High humidity, temperature and poor ventilation in greenhouses promote dermal exposure during working (Hanke et al. 2004). Several studies have assessed dermal exposure to pesticides during re-entry of workers in greenhouses for various crops: cucumbers (Caffareli et al. 2004; Jurewicz et al. 2009), strawberries (Caffareli et al. 2004), tomatoes (Caffarelli et al. 2004; Kasiotis et al. 2017; Kittas et al. 2013; Ramos et al. 2010) and peppers (Kasiotis et al. 2017). Health problems have been reported for workers exposed to pesticides during re-entry activities, including reproductive problems (Abell et al. 2000a-b), genetic damage (Lander et al. 2000), neurological disorders (Baldi et al. 2011, increases in bladder cancer (Boulanger et al. 2016) and even breast cancer (Lemarchand et al. 2016).

In this context, a study has been carried out in the Sahel region of Tunisia to estimate the concentrations of pesticide residues on tomatoes and chili peppers collected in greenhouses in order to assess the potential exposure for some consumers groups and farm workers during the harvest tasks through models.

MATERIALS AND METHODS
Fruit sampling for residue analysis.

A random sampling of tomatoes (10 samples) and chili peppers (10 samples) was carried out from 25 to 28 April 2017 in Sousse governorate, according to the guidelines of the European Directive 2002/63/EC (EU Commission 2002) (sampling at the precise time of harvest and sample size of about 1 kg). Samples of 1.00 kg ± 0.04 kg

(with at least 10 units) were collected in 20 greenhouses and weighed. The average unit weight (U, the smallest discrete portion in each lot, Directive 2002/63/EC) was about 106 g for tomatoes and 70 g for chili peppers. All samples were labeled and all useful information was collected for each sample (sample number, origin, sampling date, plant protection product applied, etc.).

Analytical procedures.

The residual pesticide deposits were analyzed by PRIMORIS (formerly FYTOLAB, Technologiepark 2/3, 9052 Zwijnaarde, Belgium) laboratory holding a BELAC (Belgian Accreditation Council) accreditation to ISO/CEI 17025 for pesticide residues on vegetables and herbal products in general. Food and glove samples were analyzed with a multiple-residue Quick Easy Cheap Effective Rugged Safe (QuEChERS) method validated by the laboratory for analysis of residues in foodstuffs, which will detect approximately 500 different pesticide residues (active ingredients and metabolites) in a single analysis thanks to a combination of gas chromatography (GC) and liquid chromatography (LC) according to the active ingredients to be determined (GC-MS/MS for small, thermally stable, volatile, non-polar molecules or LC-MS/MS for larger, thermolabile, non-volatile, and polar molecules). For almost all active ingredients, the quantification limit (LOQ) was ≤ 0.01 mg/kg. The extraction procedure is based on the AOAC Official Method (Lehotay 2007). Briefly, a homogenous 10.0 g sub-sample (crushed fruits or small pieces of gloves) was weighted into a 50 ml polypropylene tube. Then, 10 ml of acidified acetonitrile (1% acetic acid), 4 g of anhydrous magnesium sulfate (MgSO4), and 1 g of sodium acetate (NaOAc) were added. After shaking and sonication in an ultrasonic bath, the polypropylene tube was centrifuged. A portion of the acetonitrile phase (upper layer) was transferred to vials and further analyzed (Toumi et al. 2016a-b; Toumi et al. 2017a-b). The analytical results were corrected when necessary with the previously determined recovery rates (Toumi et al. 2017a).

Consumer risk assessment.

The human health risk was evaluated based on the concentration of pesticide residues in chili peppers and tomatoes at harvest. To evaluate the acute risk for child and adult consumers, we used a Predicted Short Term Intake (PSTI) values calculated with the general following formula: PSTI= (LP × OR × v) / bw, where **LP** is the 97.5[th] percentile of the portion size taken by people consuming tomatoes or chili peppers, in kg food per day, **OR** is the observed residue level the sample (in mg/kg), **bw** is the mean body weight for the target population subgroup (in kg) and **v** variability factor, the factor applied to the composite residue to approximate the residue level in a high-residue single unit.

For samples with pesticide residues, PSTI values were calculated with the EFSA Primo Model (RASFF, 2016; excel file version 11, 17/04/2017) and the European food consumption database was used since Tunisia has no national data for large portions. The risk level for each active ingredient was established by comparison to the Acute Reference Dose (ARfD): according to FAO (2002) a risk exists when the PSTI > ARfD. When no ARfD value was available, no calculation of the acute risk was performed.

Exposure scenario of farm operators and farm workers.

To have a better understanding of the route of exposure and professional practices, a survey through questionnaires was carried out among 73 farmers which were randomly chosen from professionals located in the Sahel region of Tunisia, more precisely in governorates of Sousse (59 market gardening farmers, i.e., 81%) and Monastir (14 market gardening farmers, i.e., 19%). Farmers were contacted with the help of the heads of Extension Territorial Cells (ETC) and met individually. The size of the group was considered large enough to be representative as all of the participants have the same activities.

The survey was conducted between February and April 2017. It consisted on face-to-face interviews with farm operators and farm workers in rural areas in the Sahel region where horticultural crops (vegetables, fruits) were mainly cultivated. A questionnaire was addressed to two professional categories: operators who are directly exposed to plant protection products (PPP) and workers who are indirectly exposed to PPP during re-entry activities (pruning, tying, leaf pulling, harvesting, etc.). They were asked to answer a detailed questionnaire (fourteen pages) on their socio-demographic data (identity, age, sex, level of education, etc.), their horticultural production, their estimated working hours, the personal protective equipment (PPE) they worn, their hygiene rules, and their perception of health problems linked to their occupation, their management of PPP, their knowledge of pesticide residues and their suggestions and recommendations related to this subject.

Assessment of workers hands exposure using cotton gloves.

Dermal exposure was determined according to a previously published procedure (Toumi et al. 2017a-b). Twenty volunteers working in tomato or chili pepper greenhouses located in Sousse governorate were chosen at random to measure the transfer of pesticide residues from treated fruits to hands and to evaluate their potential dermal exposure (PDE). Two pairs of 100% cotton gloves were distributed to each worker and worn during two consecutive half days during harvesting fruits in 10 tomato and 10 chili pepper greenhouses (from min 2 to max 3 h/day). The two pairs were collected as a single sample (4 gloves/sample), weighed, cut in small pieces, and stored in freezing bags at -18°C until transport and analysis.

Based on the determination of pesticide residues detected on gloves, the potential dermal exposure values were estimated. For each substance, a PDE value was calculated as follows: PDE (mg/kg bw per day) = (C$_T$ (mg/kg) × GW (kg) × 4)/bw (kg), where **C** is the concentration of the substance in the sub-sample (5 g), **GW** is the average weight of the cotton gloves samples (61 ± 3.27 g), **T** is the task duration (2 h during the trial; 8 h/day), and **bw** is the body weight (conventionally, 60 kg). The duration of the task used to evaluate the dermal exposure of workers is 8 h/day (EFSA 2014) and the local survey showed an average harvesting time close to 8 h.

The PDE values were then converted into systemic exposure values (SE) using an appropriate dermal absorption percentage of 75% (default value) (EFSA 2012) as follows: SE (mg/kg bw per day) = PDE (mg/kg bw per day) × 0.75. The risk characterization is obtained as the ratio of the systemic

exposure level to the reference value of each active ingredient, the AOEL (Acceptable Operator Exposure Level; in mg a.s./kg bw per day). It should not be exceeded to avoid any adverse effect to farm operators' and workers' health. To assess the risk, several prediction levels of the SE were considered: the median, 90th percentile, and the maximum (in mg/kg bw per day). Therefore, the SE values were expressed as percentage of the AOEL. It has been assumed that the most appropriate level to cover and assess the risk is the maximum value of the SE (SE_{MAX} or worst case).

RESULTS
Lessons learnt from observation of practices and interviews.

According to the survey, the majority of the 73 interviewed people were plot owners (86%), predominantly adult male aged from 20 to 78 years (mean age: 47 ± 12 years). A vast majority of the respondents (77%) can be considered as workers (people who enter in treated areas or who handle treated crops) as well as operators (people involved in mixing, loading, spraying or emptying/cleaning operations) (categories defined by EFSA, 2014). Sixteen respondents should only be considered as workers and one as an operator applying PPP. The main crops in the Tunisian Sahel region were tomatoes (26%), chili peppers (28%) and potatoes (25%), evenly distributed between greenhouses (45%) and open fields (44%). A small part of the production is carried out under shelter (8%) or in tunnels (3%). Tomatoes and chili peppers are grown in greenhouses and are exposed to various pests (*Tuta absoluta*, whiteflies, soil nematodes, mites, psyllids and thrips) and diseases (*Phytophthora* sp. and *Botrytis* sp.). Almost all preventive and/or curative

treatments are systematic, with PPP obtained from the local authorized suppliers.

The majority of respondents (53%) have a rather low level of education but an average working experience of 30 years. Bad habits (smoking) and lack of hygiene rules (36% workers eat and 60% drink while working) observed during the survey contribute to increase the risk of exposure of operators and farm workers to pesticide residues through direct or indirect contact.

Behavioral observations of operators made during the survey showed that 42% of them do not read the labels on PPP packaging and 9% do not understand the instructions of use. More than 20% have no idea about the recommended dosage of the PPPs and use them based on their experience or according to their supplier indications. The majority harvest their products without respect of the PHI, sometimes the day after treatment. Regarding security, 57 operators never wear Plant Protection Equipment (PPE) during mixing and loading or cleaning of the spray equipment. During application, some of them wear a protective coverall (2%), gloves (23%), masks (16%), boots (12%), goggles (11%) or blouse (14%). After application, 47% wash only their hands; 19% wash their hands and arms; 28% their hands, arms, and faces. A relatively high percentage of operators (84%) take a shower when they return to home.

The survey indicates that working time in greenhouses can vary according to the season and the crop. Activities extend over the entire week, with an average daily duration of 8 ± 1 h (n = 73). The observed contact duration of workers with crops is 5 ± 2 h/day (n = 72 workers). Most workers (65%) return to plots or greenhouses immediately or a few hours

after treatment. The majority of them wear long (68%) or short (39%) sleeve shirts and long (68%) or short (14%) leg trousers, but very few wear appropriate protective equipment such as gloves (8.3%), aprons (15%) or special clothing (22%). After working, 44% of workers wash only their hands; 18% wash their hands and arms, 38% their hands, arms, and faces. Nevertheless 74% of workers made full body toilet (shower) at home.

Operators report various health problems such as: eye problems (21); respiratory problems (13); skin problems: irritation (13) and dry (5) and other symptoms: stomach cramps (5); nosebleeds (4); nausea (4); dizziness (3); headaches (2); and sweating (2); repetitive fatigue (2); fevers (1) and dry mouth (1) sneezing (1). Farm workers complain about: eye problems (8); respiratory problems (5); skin problems: irritation (7) and dry (3) and other symptoms: nausea (3); stomach cramps (2); repeated strain (1); nosebleeds (1); headaches (1) and dizziness (1). Despite all reported problems, the majority of respondents have a passive attitude regarding pesticide use and no proposal for improvement was formulated from the survey. Problems are mainly linked to regulation weaknesses, the lack of awareness and monitoring, but also to the inefficacy of some PPP leading them to increase the dosage or application frequency. Workers have proposed that PPE (including gloves) be distributed, or offered with purchased PPP, to encourage them to wear protective equipment and to improve their behavior.

Results of analysis of residual deposits in fruit samples.

Pesticide residues have been detected in almost all tomato and chili pepper samples. Only two samples (one of tomato and one of chili pepper) were free from detectable residues (concentrations below the analytical limit of quantification). Eighteen active ingredients have been detected on 10 chili pepper samples (average: 2.9 a.i./sample), with an average total pesticide load of 0.41 mg a.i./kg. Two fungicides, proquinazide and benomyl (and its metabolite, carbendazim), had the highest detection frequency (30%) (Table 1).

Fifteen different active ingredients have been detected in 10 tomato samples (average: 2.4 a.i/sample), with an average total pesticide load of 0.38 mg a.i./kg. The most frequently detected residue on tomatoes is the fungicide propamocarb (6 samples out of 10) (Table 2).

Consumer risk assessment.

Tables 1 and 2 summarize the detected active ingredients and their concentrations (in mg/kg) in the fruit samples, the concentration expressed as percentage of MRL (Maximum Residue Limit), the PSTI (in mg/kg bw/day) and the PSTI value expressed as a percentage of ARfD for both adults and children. Seven MRL exceedances were reported: six exceedances in chili pepper (Table 1) and one exceedance for chlorpyrifos ethyl (insecticide) in tomatoes (Table 2). The MRL exceedances appeared particularly critical for propargite (chili pepper) and chlorpyrifos-ethyl (tomato) with concentration values respectively 20 (2000% of MRL) and 26 (2685% of MRL) times higher than the MRL values.

Table 1. Results of 10 chili pepper analyzed samples: detected active ingredients; concentrations expressed as a percentage of MRL; PSTI values; PSTI expressed as a percentage of ARfD, for adults and children

Chili pepper sample	Active ingredient	Concentration (mg/kg)	Concentration (% MRL)	PSTI (mg/kg bw/day)		ARfD (%)	
				Adults	Children	Adults	Children
Sample 1	Bifenazate	0.1800	6.0	0.00025	0.00029	n.a	n.a
	Proquinazid	0.0100	50.0	0.00001	0.00002	0.01	0.01
Sample 2	Acetamiprid	0.0649	21.6	0.00009	0.00011	0.09	0.11
	Carbendazim and benomyl	0.0121	12.1	0.000017	0.00002	0.08	0.10
	Indoxacarb	0.0453	15.1	0.00006	0.00007	0.05	0.06
	Proquinazid	0.0682	341.0	0.00009	0.00011	0.05	0.06
	Thiophanate methyl	0.2983	298.3	0.00004	0.00005	0.21	0.24
Sample 3	Spiromesifen	0.0139	2.8	0.00002	0.00002	0.00	0.00
	Carbendazim and benomyl	0.0455	45.5	0.00006	0.00007	0.32	0.37
	Fluopicolide	0.0349	3.5	0.00005	0.00006	0.03	0.03
	Myclobutanil	0.4776	95.5	0.00066	0.00078	0.21	0.25
	Propamocarb	0.1548	5.2	0.00021	0.00025	0.02	0.03
	Proquinazid	0.0901	450.5	0.00013	0.00015	0.06	0.07
	Thiophanate methyl	0.3235	323.5	0.00004	0.00005	0.22	0.26
Sample 4	Imidacloprid	0.0549	5.5	0.00008	0.00009	0.10	0.11
	Tebuconazole	0.0809	13.5	0.00011	0.00013	0.37	0.44
	Tebufenpyrad	0.1865	37.3	0.00026	0.00030	1.29	1.52
Sample 5	-	-	-	-	-	-	-
Sample 6	Acetamiprid	0.4668	155.6	0.00065	0.00076	0.65	0.76
Sample 7	Carbendazim and benomyl	0.0655	65.5	0.00009	0.00011	0.45	0.53
	Indoxacarb	0.0103	3.4	0.00001	0.00002	0.01	0.01
	Spirotetramat	0.0170	0.9	0.00002	0.00003	0.00	0.00
Sample 8	Tebuconazole	0.3706	61.8	0.00051	0.00060	1.71	2.01
Sample 9	Cyproconazole	0.0218	43.6	0.00003	0.00004	0.15	0.18
	Spinosad	0.0630	3.2	0.00009	0.00010	n.a	n.a
Sample 10	Propagite	0.2000	2000.0	0.00028	0.00033	0.93	1.08
	Bupirimate	0.3411	17.1	0.00047	0.00055	n.a	n.a

n.a.: not available; MRL and ARfD values from EU Pesticides database.

Table 2. Results of 10 tomato analyzed samples: detected active ingredients; concentrations expressed as a percentage of MRL; PSTI values; PSTI expressed as a percentage of ARfD, for adults and children

Tomato sample	Active ingredient	Concentration (mg/kg)	Concentration (% MRL)	PSTI (mg/kg bw/day)		ARfD (%)	
				Adults	Children	Adults	Children
Sample 1	Chlorantraniliprole	0.0147	2.5	0.00022	0.00085	n.a	n.a
	Propamocarb	0.2629	6.6	0.00400	0.01529	0.40	1.53
Sample 2	Propamocarb	0.0475	1.2	0.00072	0.00276	0.07	0.28
Sample 3	Propamocarb	0.1003	2.5	0.00153	0.00583	0.15	0.58
Sample 4	Indoxacarb	0.0295	5.9	0.00045	0.00172	0.36	1.37
Sample 5	Acetamiprid	0.1058	21.2	0.00161	0.00615	1.61	6.15
	Flubendiamide	0.0260	13.0	0.00040	0.00151	0.40	1.51
	Myclobutanil	0.0141	4.7	0.00021	0.00082	0.07	0.26
	Pirimicarb	0.0232	4.6	0.00035	0.00135	0.35	1.35
	Propamocarb	0.0135	0.3	0.00021	0.00078	0.02	0.08
Sample 6	Propamocarb	0.5206	13.0	0.00793	0.03027	0.79	3.03
Sample 7	-	-	-	-	-	-	-
Sample 8	Azoxystrobin	0.0226	0.8	0.00034	0.00131	n.a	n.a
	Chlorantrraniliprole	0.0311	5.2	0.00047	0.00181	n.a	n.a
	Difenoconazole	0.1231	6.2	0.00187	0.00716	1.17	4.47
	Indoxacarb	0.1020	20.4	0.00155	0.00593	1.24	4.74
Sample 9	Chlorpyrifos-ethyl	0.2685	2685.0	0.00409	0.01561	81.75	312.24
	Carbendazim and benomyl	0.0649	21.6	0.00099	0.00377	4.94	18.87
	Propamocarb	0.0326	0.8	0.00050	0.00190	0.05	0.19
	Pyrimethanil	0.5877	58.8	0.00895	0.03417	n.a	n.a
	Thiophanate methyl	0.9763	97.	0,00149	0.00568	7.43	28.38
Sample 10	Boscalid	0.0242	0.8	0.00037	0.00141	n.a	n.a
	Spinosad	0.0360	5.1	0.00055	0.00209	n.a	n.a

n.a.: not available; MRL and ARfD values from EU Pesticides database.

Results of analyses of residual deposits in glove samples.

All active ingredients detected on vegetables were also measured at rather high concentrations on cotton gloves. For people working in chili pepper greenhouses, 63 a.i. were identified (average: 18 a.i./sample), with an average total concentration of 148 ± 285 mg/kg. Four main active ingredients were identified: thiophanate-methyl (100%), benomyl (and its metabolite carbendazim) (90%), acetamiprid (70%) and propamocarb (70%)). A total of 57 a.i. were detected on all the gloves worn by people working in tomato greenhouses

(average: 18 a.i./sample), with an average total concentration of 111 ± 193 mg/kg. Propamocarb was detected in all samples, followed by diafenthiuron (90%) and thiophanate methyl (80%). DEET (N, N-diethyl-3-methylbenzamide) was also detected on all glove samples as it is used as a biocide in textile sector/industry.

Risk characterization for farm operators and farm workers.

Tables 3 and 4 present the systemic exposure values (SE median, 90th percentile, and maximum values, in mg/kg bw per day) and the systemic exposure expressed as a percentage of the AOEL for all active ingredients detected on the cotton gloves worn by workers in chili pepper (Table 3) and tomato greenhouses (Table 4) and having a SE exceeding their respective AOEL value.

DISCUSSION

Among all vegetable analyzed samples, only two (one tomato sample and one chili pepper sample) have residue levels below the limit of quantification (0.01 mg/kg). Most often multiple residues were detected in the samples (up to seven pesticides). These results are a direct consequence of local poor practices and bad pest management, as reported for many other countries over the world (Arias et al. 2014; Murcia and Stashenko 2008).

Chili peppers appear to be slightly more contaminated than tomatoes (higher number of different residues and more MRL exceedances). Even though the two vegetables belong to the Solanaceae and are produced according to similar practices, the difference may result from the physiological characteristics of each species and the difference in composition of each cuticle. It is known that the lipophilicity of the cuticle can help some pesticides to enter into the plant (Trapp 2004). Stronger and thicker cuticle of chili peppers could better retain the residues, and the bigger surface area could intercept more pesticide drift than tomatoes fruits (Riederer and Schönherr 1984). However, it is difficult to predict the cuticle absorption and the degradation of chemical ingredient as they depend on many factors such as the physicochemical characteristics of the chemical, the contact area, the cuticle composition and its surface (Bonmatin et al. 2015). Four samples of chili peppers (40%) had pesticide residues above the maximum residue limits (MRLs). A total of 6 MRL exceedances were observed for a single collection of 10 samples. Residues of proquinazid, thiophanate-methyl, acetamiprid and propargite exceeded dramatically the MRLs (for 156% up to 2000%). A study conducted in Egypt in 2015 showed that only one of 31 pepper samples had acetamiprid residue levels higher than the MRL value (Alla et al. 2015).

Only one sample of tomato reported a MRL violation for the insecticide chlorpyrifos-ethyl by 2685%. Similar trends in results was also reported by Bojacà et al. (2011) that conducted a monitoring study for tomatoes in Colombia and indicated that almost all the samples of greenhouse tomatoes positive for acephate, cymoxanil, hexaconazole or thiocyclam exceeded the MRLs, on average, by 356, 525, 606 and 1375%, respectively. Chlorpyrifos-ethyl in tomatoes has been detected in different countries around the world including India and Ghana (Essumang et al. 2008; Singh 2012). In contrast, any pesticide residue exceeded the MRL on 19 tomato samples (Alla et al. 2015).

Table 3. Active ingredients detected on gloves worn by workers in chili pepper greenhouses and having a SE exceeding their AOEL values, the corresponding systemic exposure (median, 90th percentile, and maximum values) in mg/kg bw per day, the systemic exposure as a percentage of the AOEL and their toxicological properties (AOEL values, and CLP classification according the EU Pesticides database)

Active ingredient		AOEL (mg/kg bw/day)	SE (Median) (mg/kg bw per day) (AOEL%)	SE (90th P) (mg/kg bw per day) (AOEL%)	SE (Maximum) (mg/kg bw per day) (SE in AOEL%)	CLP classification
Acetamiprid		0.07	0.0006 (1%)	0.0307 (44%)	0.0700 (100%)	H302
Bifenazate		0.0028	0.0044 (159%)	0.0163 (583%)	0.0195 (697%)	H317, H373
Benomyl and carbendazim		0.02	0.0055 (27%)	0.0939 (470%)	0.2745 (1373%)	H315, H317, H335, H340, H360FD
Chlorothalonil		0.009	0.5079 (5644%)	0.9141 (10157%)	1.0157 (11285%)	H317, H318, H330, H335, H351
Cyhalothrin*	Gamma	0.0003	0.0005 (181%)	0.0005 (181%)	0.0005 (181%)	-
	Lambda	0.00063	0.0005 (86%)	0.0005 (86%)	0.0005 (86%)	H301, H312, H330
Cypermethrin		0.06	0.0002 (0%)	0.0572 (95%)	0.0949 (158%)	H302, H332, H335
Dimethoate		0.001	0.0046 (456%)	0.0082 (817%)	0.0091 (907%)	H302, H312
Flubendiamide		0.006	0.0342 (569%)	0.0614 (1024%)	0.0683 (1138%)	-
Indoxacarb		0.004	0.0005 (13%)	0.0679 (1699%)	0.0883 (2208%)	H301,H317, H332, H372
Omethoate		0.0003	0.0004 (131 %)	0.0004 (131%)	0.0004 (131%)	H301, H312
Proquinazid		0.02	0.0043 (21%)	0.0241 (120%)	0.0315 (158%)	H351
Spiromesifen		0.015	0.0092 (61%)	0.0996 (664%)	0.1551 (1034%)	-
Tebuconazole		0.03	0.0053(18%)	0.0326 (109%)	0.0421 (140%)	H302, H361d
Tebufenpyrad		0.01	0.0038 (38%)	0.0592 (592%)	0.0961 (961%)	H301, H317, H332,H373
Thiophanate-methyl		0.08	0.0044 (6%)	0.4392 (549%)	1.6470 (2059%)	H317, H332, H341

H301: Toxic if swallowed; H302: Harmful if swallowed; H312: Harmful in contact with skin; H315: Causes skin irritation; H317: May cause an allergic skin reaction; H318: Causes serious eye damage; H330: Fatal if inhaled; H332: Harmful if inhaled; H335: May cause respiratory irritation; H340: May cause genetic defects; H341: Suspected of causing genetic defects; H351: Suspected of causing cancer; H360FD: May damage fertility; May damage the unborn child; H361d: Suspected of damaging the unborn child; H372: Causes damage to organs through prolonged or repeated exposure; H373: May cause damage to organs through prolonged or repeated exposure.* The analytical method is unable to identify cyhalothrin (lambda or gamma), therefore the risk assessment was performed for both cases.

Table 4. Active ingredients detected on gloves worn by workers in tomato greenhouses and having a SE exceeding their AOEL values, the corresponding systemic exposure (median, 90th percentile, and maximum values) in mg/kg bw per day, the systemic exposure as a percentage of the AOEL and their toxicological properties (AOEL values, and CLP classification according the EU Pesticides database)

Active ingredient	AOEL (mg/kg bw/day)	SE (Median) (mg/kg bw per day) (AOEL%)	SE (90th P) (mg/kg bw per day) (AOEL%)	SE (Maximum) (mg/kg bw per day) (SE in AOEL%)	CLP classification

Bifenazate	0.0028	0.0060 (215%)	0.0097 (348%)	0.0107 (381%)	H317, H373
Benomyl and carbendazim	0.02	0.0005 (2%)	0.0621 (311%)	0.1220 (610%)	H315, H317, H335, H340, H360FD
Chlorothalonil	0.009	0.0150 (167%)	0.0431 (479%)	0.0488 (542%)	H317, H318, H330, H335, H351
Chlorpyrifos-ethyl	0.001	0.0001 (8%)	0.0052 (524%)	0.0104 (1036%)	H301
Cyhalothrin* **Gamma**	0.0003	0.0001 (40%)	0.0009 (291%)	0.0012 (398%)	-
Lambda	0.00063	0.0001 (19%)	0.0009 (138%)	0.0012 (189%)	H301, H312, H330
Flubendiamide	0.006	0.0390 (651%)	0.0702 (1170%)	0.0780 (1300%)	-
Indoxacarb	0.004	0.0163 (408%)	0.0938 (2344%)	0.1019 (2548%)	H301, H317, H332, H372
Spinosad	0.012	0.0106 (88%)	0.0182 (152%)	0.0201 (168%)	-
Thiophanate-methyl	0.08	0.0005 (1%)	0.5716 (714%)	1.6989 (2124%)	H317, H332, H341

H301: Toxic if swallowed; H312: Harmful in contact with skin; H315: Causes skin irritation; H317: May cause an allergic skin reaction; H318: Causes serious eye damage; H330: Fatal if inhaled; H331:Toxic if inhaled; H332: Harmful if inhaled; H335: May cause respiratory irritation; H340: May cause genetic defects; H341: Suspected of causing genetic defects; H351: Suspected of causing cancer; H372: Causes damage to organs through prolonged or repeated exposure; H373: May cause damage to organs through prolonged or repeated exposure.
* The analytical method is unable to identify cyhalothrin (lambda or gamma), therefore the risk assessment was performed for both cases.

The active ingredients detected on vegetables, with higher concentrations above their respective MRL values (e.g. chlorpyrifos-ethyl, acetamiprid, thiophanate-methyl, propagite or proquinazid), are known for their potential detrimental effects to health; therefore vegetables should be considered as non-compliant for the market. Nevertheless, PSTI calculation consists to estimate the actual risk to consumer group (adults and children) and whether an observed violation of an MRL can lead to a risk to the consumers (Łozowicka 2012). As shown in Table 1, no values were above the ARfD for chili pepper samples. The PSTI values in chili-pepper samples were in the range of 0.00-1.71% and 0.00-2.01% ARfD for adults and children, respectively (Table 1). Only in one tomato sample, the PSTI of the insecticide chlorpyrifos-ethyl exceeds the ARfD with a factor of 3.1 times (312%). This exceedance of the ARfD was observed for children but not confirmed for adults. The PSTI values in tomato samples were in the range of 0.02-82% and 0.08-312%ARfD for adults and children, respectively (Table 2).

These results demonstrate that despite the high level of residues in some samples of vegetables, the Tunisian consumers do not face a serious acute risk, except with insecticides such as chlorpyrifos-ethyl. Nevertheless considering the number of detected residues, their chronic exposure through the consumption of raw vegetables could be associated with a health risk. Moreover, it should be borne in mind that dietary pesticide exposure estimated in this study, considered only exposures through consumption of chili peppers and tomatoes, and did not include other food

products such as fruits, other vegetables, grains, dairy products, fish and meat. Furthermore, the estimated risk assessment is based on toxicological evaluation of single compounds and not based on an evaluation of cumulative exposure to multiple pesticide residues in crops. In addition, these horticultural commodities are also essential ingredients in Tunisian diet, more consumed than in Europe. As a result, the global consumer exposure should be higher than in the present evaluation.

Workers who come into contact with the crop or handle treated products will be contaminated through contact with pesticides that are still available on the crop after application (Dong and Beauvais 2013; Krol et al. 2005). Previous studies (Nigg et al. 1984; Zweig et al. 1985) demonstrated the relationship between the levels of residues on the crops and the dermal exposure of workers during harvesting activities. Similarly, in this study, all pesticide residues measured in chili pepper and tomato samples were also detected in glove samples worn by farm workers during harvesting. Contact with contaminated vegetable samples resulted in the transfer of pesticide residues to gloves worn by the workers allowing their measurement. All glove samples appeared to be highly contaminated by many different pesticide residues (63 active ingredients detected with an average of about 18 a.i. per sample and an average total concentration per glove sample of 148 mg/kg for chili peppers and 57 active ingredients detected with an average of about 18 a.i. per sample and an average total concentration per glove sample of 111 mg/kg for tomato). These concentrations are 1000 times higher than the concentrations which are usually detected on foodstuffs. The systemic exposures of

workers were estimated for the median, for P90, and for the maximum concentration of residues in samples (Tables 3 and 4).

For chili pepper samples, six, thirteen and fifteen active ingredients exceed the AOEL respectively at the median, the P90 and maximum values of SE indicating risk situations. However, for tomato samples, four active ingredients exceed the AOEL at the SE median values. At P90 and the maximum (or worst case), nine active ingredients exceed the AOEL indicating potential risk situations.

A recent study was conducted in Greece to assess a worker dermal exposure during re-entry activities in greenhouses reported that the total worker PDE levels ranged from 0.16 to 0.72 mg/kg bw per day and from 0.09 to 0.17 mg/kg bw per day for tomato and chili pepper crops, respectively (Kasiotis et al. 2017).

Exposure could be particularly critical for chlorothalonil, with SE_{MAX} values 113 times higher than the AOEL (11285%) for chili peppers, followed by indoxacarb and thiophanate methyl which are above 20 times higher than the AOEL: 2208 and 2059%, for chili peppers and 2548% and 2124% for tomatoes, respectively. At SE_{MAX}, five active ingredients are above 10 times higher than the AOEL: benomyl and carbendazim (1373%), flubendiamide (1138%) and spiromesifen (1034%) for chili peppers and chlorpyrifos-ethyl (1036%) and flubendiamide (1300%) for tomatoes. Even when wearing personal protection equipment that will minimize exposure by 90%, SE values will always exceed the AOEL at the worst case for these active ingredients. The systemic exposure values are in accordance with the results of a study conducted in Italy to

evaluate the risk of pesticide dermal exposure where the highest absorbed doses for workers re-entering in tomato greenhouse in % of AOEL are 288 and 7959% for azoxystrobin and chlorpyrifos-ethyl, respectively (Cafferli et al. 2004). Several reasons are behind these violations of the health based guidance values including, applying higher dose than the recommended ones, not respecting the pre-harvest interval, etc.

According to the CLP classification (Tables 3 and 4), the majority of the active ingredients detected in chili pepper and tomato samples and having a SE exceeding their AOEL value, have potential hazardous acute and/or chronic effects. The results of the observed levels of dermal exposure after re-entry of greenhouses led to the conclusion that a health hazard may exist, especially after application of high rates of relatively toxic pesticides which easily penetrate the skin.

From the survey of 73 farmers, it is concluded that workers and operators may be exposed during usual pesticide handling and re-entry activities. The task duration for harvesting for farm workers, which is an important factor to consider when building exposure scenarios for a group of workers, is equal to the default value of 8 h proposed in the EFSA Guidance Document 2014 (EFSA 2014). A considerable number of the farmers reported not using protective equipment on a regular basis. The obtained results were agreeing with those reported in Nepal (Shrestha et al. 2010), Palestine (Sa'ed et al. 2010), Lesotho (Mokhele et al. 2011), Iran (Hashemi et al. 2012), Tanzania (Lekei et al. 2014), Uganda (Oesterlund et al. 2014), Indonesia (Yuantari et al. 2015), Ghana (Okoffo et al. 2016), Gambia (Idowu et al. 2017), and Burkina Faso (Son et al. 2017). Bad

personal behavioral habits (eating, drinking, or smoking at work) were reported by many farmers (operators and workers). Thus, oral exposure may occur secondarily to dermal exposure, through hand to mouth transfer. Health risks can be due to mishandling and habits exhibited during pesticide application and re-entry activities. According to their answers in the survey, workers seem to be affected by many health problems, while it was not possible to conclude only on the basis of personal feelings and declarations, analytical results and the estimations of exposure confirmed that Tunisian farm operators and workers in the study area are at high risk.

In conclusion, observations completed by analytical results indicate multiple pesticide applications leading to MRL exceedances and probable acute risk for Tunisian consumers. It is a pity that exposure was assessed using a European food consumption database while chili peppers and tomatoes are among the staple foods in Tunisia with a consumption significantly higher than in Europe. Thus, these results stress the need for a national consumption survey and continuous monitoring programs that cover all food commodities consumed locally, especially fruits and vegetables. According to systemic exposure values, workers who spend several hours on a daily basis in greenhouses are at risk during re-entry activities, with potential effects on their health. It appears that lack of awareness, bad habits and absence of personal protective equipment increase their exposure level and their health risks. There is an urgent need for awareness raising amongst professionals' and training on good practices and hygiene rules to avoid their excessive exposure. This survey should be completed later by a bio-monitoring of the operators during

spraying and workers during re-entry activities, with analysis of blood, urine and hair samples. Moreover, considering that the concentration of pesticides in the air is of high concern in greenhouses, the evaluation of the inhalation exposure is highly recommended in the future.

RESUME

Toumi K., Joly L., Tarchoun N., Souabni L., Bouaziz M., Vleminckx C. et Schiffers B. 2018. Evaluation des risques pour les consommateurs et les travailleurs agricoles tunisiens exposés aux résidus après l'application de pesticides sur les piments et les tomates. Tunisian Journal of Plant Protection 13 (1): 127-143.

En Tunisie, pour prévenir et contrôler les ravageurs et les maladies sur les cultures sous serres, le piment et la tomate nécessitent l'emploi d'une large gamme de pesticides potentiellement toxiques et pouvant donc présenter un risque pour les exploitants agricoles, travailleurs ou consommateurs. Une étude a été menée en Tunisie dans la région du Sahel pour évaluer le risque pour les exploitants agricoles et les travailleurs exposés, lors des récoltes, aux éventuels résidus de pesticides restant dans les cultures de tomates et de piments et pour les consommateurs tunisiens (adultes et enfants) après ingestion de ces légumes. Un questionnaire a été adressé à un groupe de 73 maraîchers pour mieux comprendre les pratiques professionnelles locales et déterminer la principale voie d'exposition. Vingt échantillons de gants en coton (2 paires / échantillon) ont été distribués à 20 volontaires qui les ont portés pendant deux demi-journées consécutives lors de la récolte de piments ou de tomates avant l'analyse des résidus de pesticides délogeables qui pourraient être transférés des cultures aux mains. En utilisant des modèles d'exposition prédictive, les valeurs ont été calculées pour les consommateurs et les travailleurs agricoles. L'exposition la plus élevée des consommateurs a été observée pour les résidus de chlorpyrifos-éthyl dans les tomates (avec 82% et 312% de l'ARfD (dose de référence aiguë), respectivement pour les adultes et les enfants). L'exposition systémique (SE) des travailleurs agricoles a été estimée pour la médiane, le 90ème centile et la concentration maximum. Aux concentrations observées les plus élevées, 15 résidus de pesticides (substances actives et métabolites) utilisées dans les serres de piment, et 9 dans les cultures de tomates, dépassent le niveau d'exposition acceptable pour l'opérateur (AOEL). L'exposition semble particulièrement critique pour le chlorothalonil pulvérisé dans des serres de piment avec des valeurs de SE$_{MAX}$ 113 fois plus élevées que l'AOEL (11285%). La durée prolongée du travail (8 h/ jour) après rentrée dans la serre, l'accès limité aux équipements de protection individuels (EPI), le manque d'hygiène et les mauvaises habitudes (manger, boire ou fumer au travail) ont également été observés et discutés en tant que facteurs de risque.

Mots clés: Consommateurs, évaluation des risques, exposition cutanée, résidus de pesticides, travailleurs agricoles, Tunisie

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ملخص

تومي، ك. وة وور جوي وناجي طرشون وةيلى صوابني ومروى بوعزيز وكريستاين فيلمانكس وبرونو شيفارس. 2018. تقييم المخاطر على المستهلكين وعمال المزارع التونسيين للبقايا بعد استخدام المبيدات في زراعتيْ الفلفل والطماطم. Tunisian Journal of Plant Protection 13 (1): 127-143.

في تونس، بهدف منع مكافحة الآفات والأمراض عند الزراعة في البيوت المحمية، يتطلب الفلفل والطماطم استخدام مجموعة واسعة من المبيدات التي يحتمل أن تكون سامة، مما يشكل خطرا محتملا على المزارعين والعمال أو المستهلكين. وقد أجريت دراسة في منطقة الساحل بتونس لتقييم المخاطر التي يتعرض لها المزارعون والعمال المعرضون لمخلفات مبيدات الآفات عن طريق اللمس خلال مهام جني الطماطم والفلفل، والمستهلكين التونسيين (البالغين والأطفال) بعد الاستهلاك. تم توجيه استبيان إلى مجموعة متكونة من 73 مزارع خضر لفهم أفضل الممارسات المهنية

المحلية تحديد الطريق الرئيسي للتعرض. تم توزيع عشرين عينة من القفازات القطنية (زوجين / عينة) على 20 متطوعا كانوا يلبسونها لمدة نصفي يومين متتاليين خلال موسم جني الفلفل أو الطماطم قبل تحليل بقايا مبيدات الآفات القابلة للتنقل من المحاصيل إلى اليدين باستخدام نماذج التعرض التنبئية، تم حساب قيم المستهلكين والعمال الزراعيين. لوحظ أن أعلى تعرض للمستهلكين لمخلفات كلوربيريفوس إيثيل في الطماطم (82%-312% من الجرعة المرجعية الحادة على التوالي للبالغين والأطفال). تم تقدير التعرض المنهجي (SE) للعمال الزراعيين للمتوسط، 90 مئوي وأقصى التركيز. في أعلى التركيزات الملحوظة، تستخدم15 بقايا مبيدات الآفات (المادة الفعالة والمستقلبات) في البيوت المحمية للفلفل و 9 بقايا مبيدات الآفات (المادة الفعالة والمستقلبات) للطماطم تجاوزا مستوى التعرض المقبول للمشغل (AOEL). يبدي التعرض حاسما بشكل خاص لكلوروثالونيل المرشوش على الفلفل مع قيمة SE 113 مرة أعلى من مستوى التعرض المقبول للمشغل (11285٪). لوحظت ونوقشت عوامل الخطر مثل ساعات العمل الطويلة (8 ساعات / يوم) بعد العودة إلى البيوت المحمية، محدودية فرص الحصول على معدات الحماية الشخصية (PPE) وانعدام النظافة والعادات السيئة (الأكل والشرب أو التدخين في العمل) أيضا.

كلمات مفتاحية: بقايا مبيدات الآفات، تعرض الجلد، تقييم المخاطر، عمال المزارع، مستهلكون

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