Risk assessment of agrochemical residues in food: a systematic review and meta-analysis

Avaliação de risco de resíduos de agrotóxicos em alimentos: uma revisão sistemática e metanálise

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Abstract

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The worldwide databases PubMed, Scopus and Web of Science were searched for pesticide residues with subsequent meta-analysis using the software Open Meta-Analyst. Of the total of 29 selected papers, five identified active ingredients at concentrations posing a conceivable and unacceptable risk for pesticide residues. The average of active ingredients with an unacceptable risk was of 0.004 and a 95% confidence interval (C.I.) between 0.000 – 0.007. Papers originated from developed countries presented an average of 0.002 and a 95% C.I. of 0.002 and 0.006. Papers out of developing countries presented an average of 0.009 and a 95% C.I. ranging from 0.002 to 0.016. In papers that reported active ingredients at an unacceptable risk, the average was of 0.110 and a 95% C.I. between 0.050 and 0.171. Even though the averages might be considered as very low it is important to emphasize that only papers dealing with produce were scrutinized and other eatables and exposure routes to potential toxic substances were disregarded. Standardization of risk assessment procedures as well as gathering individual food consumption data inherent to each location are aspects that might allow more appropriate comparisons amongst studies.

Keywords: active ingredient residue, dietary risk, residual intake, risk evaluation.

Resumo

As bases de dados PubMed, Scopus e Web of Science foram pesquisadas para artigos sobre análises de resíduos de pesticidas. Uma subsequente meta-análise foi conduzida utilizando *software* Open Meta-Analyst. De um total de 29 artigos selecionados, cinco identificaram ingredientes ativos em concentrações que se constituem em risco inaceitável para presença de resíduos. A média para ingredientes ativos com risco inaceitável foi de 0.004 em um intervalo de confiança (IC) de 95% de 0.000 a 0.007. Artigos científicos originários de países desenvolvidos apresentaram risco inaceitável de 0.002 em um IC de 0.002 a 0.0016. Artigos científicos de países em desenvolvimento tem média de risco de 0.009 com um IC de 0.002 a 0.016. Os artigos que reportaram ingredientes ativos com um risco inaceitável a média foi de 0.110 com um IC de 0.050 a 0.171. Mesmo que as medias de risco possam ser consideradas baixas é importante salientar que apenas artigos tratando de produtos hortícolas foram analisados. Outros consumíveis e rotas de exposição a potenciais substâncias tóxicas foram desconsideradas Uma padronização de procedimentos para análise de risco e a coleção de dados de consumo individuais características de localidades devem permitir um melhor comparativo entre os vários estudos de análise de risco.

Palavras-chave: resíduo de ingrediente ativo, risco dietético, ingestão de resíduo, análise de risco.

Introduction

Agrochemicals are the main resource to control diseases and pests on agricultural crops to warrant yield increases (Minut *et al*., 2020). Nonetheless, residues and traces of active ingredients and their metabolites might persist on food calling for a continuous monitoring and risk assessments of residual intakes (Liu *et al*., 2020; Marete *et al*., 2020; Sakthiselvi *et al*., 2020).

The United States National Research Council (NRC) introduced evaluation processes for chemical risks on food in the 80´s. Over the years, risk evaluation adjustments were made, especially with regard to toxicological and methodological characteristics. The first attempts in Brazil considered that all food items contained residues equivalent to the Maximum Residue Limit (MRL). The MRL was used to proceed with estimates of the Maximum Theoretical Daily Intake (MTDI) being the risk characterized via comparison of the MTDI with the Acceptable Daily Intake (ADI) of various countries and of the Codex Alimentarius (Caldas & Souza, 2000).

These attempts guided monitoring activities in Brazil indicating which pesticides posed highest potential risks and which products should have priority in monitoring programs such as the Program of Residue Analysis of Agrochemicals in Food (acronym in Portuguese = PARA) coordinated by the Brazilian National Agency for Sanitary Vigilance (ANVISA). However, that methodology infers that all agricultural crops contain residues equal to the MRL and that assumption, though, might either underestimate or overestimate the real amount of pesticide residues in a specific product. And beyond, it is a conservative point of view while pondering that each agricultural crop is only sprayed with molecules registered for that crop (Gad-Alla *et al*., 2015).

As the topic gained publicity, more work was undertaken and methodologies improved coming to the present-day methods grounded on the real amount of residues determined in multi-residue analyzes on ready-to-eat food and on the amounts of ingested food. Nowadays, the number of risk assessment studies on the consumption of produce containing pesticide residues is remarkable (Caldas; Velde-Koerts, 2017; Ibrahim *et al*., 2018; Kumari; John, 2019; Vetter, 2019; Galani *et al*., 2020).

Systematic review and meta-analysis are powerful tools to synthesize and summarize existing evidence on a particular research question (Cleophas; Zwinderman, 2017a). Both enable researchers to draw conclusions based on a comprehensive and rigorous evaluation of the available evidence, while also identifying gaps in the current knowledge that may require further investigation (Higgins *et al*., 2003; Cleophas; Zwinderman, 2017b).

For that reason, a systematic review of scientific papers on risk evaluations based on data of pesticide residue concentrations on produce was carried out. Data of those papers were retrieved to run meta-analysis to synthesize the results and to contribute for improvements in the awareness of the dietetic risk of consumption of produce with pesticide residues and to identify the uncertainties of risk evaluations.

Material and Methods

A literature search in PubMed, Scopus and Web of Science was conducted on February of 2022 applying the index terms *pesticide* AND *food* AND *dietary risk assessment*. No time interval restrictions were imposed and Mendeley Reference Management software (https://www.mendeley.com/) was used handle publications and to check for duplicates. The search focused on work on data of produce ready for consumption at retail or directly from growers. Publications written in Spanish, Portuguese and English were selected when the above index terms also showed up in the abstract. Review articles, book chapters and papers dealing with evaluations of new active ingredients were disregarded. Also, papers were omitted when on produce not consumed freshly, food of animal origin or when the study addressed active ingredient degradation or when the MRL was used instead of the real concentration of pesticide residues.

Full text articles were accessed whenever possible and when not available for download, the title and abstract were evaluated for its relevance. After all efforts to full text access were unsuccessful, the considered relevant articles were excluded.

Data on the tolerability to human health risk through risk evaluation via pesticide residue records on produce samples were extracted from the systematic literature search and included in a database by one reviewer and validated by a second reviewer. The retrieved data included the country where the study was conducted, the origin of the samples, the evaluated risk either acute, chronic or cumulative, the risk evaluation method (either probabilistic or deterministic), the amount of analyzed active ingredients, the amounts of quantified active ingredients and the concentrations of active ingredients presenting an unacceptable health risk.

The *Forest Plotting of Risk* and estimates of the effects in the socioeconomic subgroups were processed via Open Meta-Analyst software (available at: http://www.cebm.brown.edu/openmeta/index.html). The data were analyzed in binary models of aleatory effects via the Der Simonian and Laird method with a confidence interval of 95%.

The heterogeneity amongst the selected papers in the aleatory effects model was quantified using the inverse index of variance (I2). Values of I2 of 25%, 50% or 75% were considered as of low, moderate or of high heterogeneity, respectively (Higgins *et al.*, 2003).

The analyses were performed in subgroups separated by socioeconomic situation and country of origin. Per capita income, life expectancy and literacy (Human Development Index = HDI) were considered, dividing into two groups: developing countries and developed countries, either lower HDI or higher HDI, respectively.

Results and Discussion

To the discussion of health risks deriving from consumption of produce, only papers that presented data on concentration of pesticides resulting from residue analyses on horticultural crops were selected in the systematic review. Additional qualifications to select papers were data on individual consumption, consumer's body weight, toxicological parameters and consumption as fresh product. Reports from official monitoring programs were not taken in account.

The systematic review returned 444 papers by the use of the mentioned index terms to search the databases. After sorting out duplicates, 233 papers remained, from which 29 papers were selected after applying the indicated prerequisites (Figure 1).

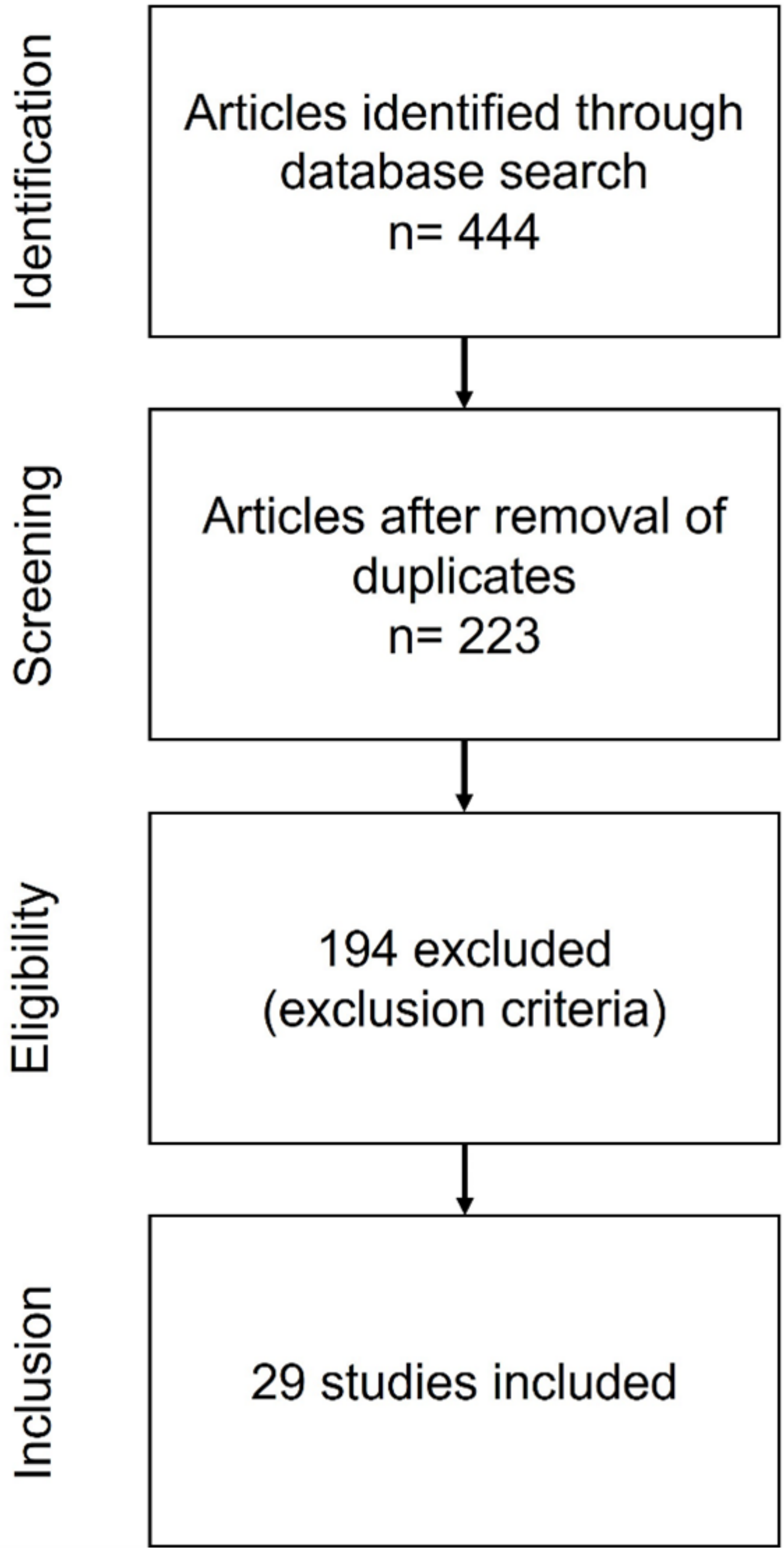
Identification
Articles identified through database search n= 444
Screening
Articles after removal of duplicates n= 223
Eligibility
194 excluded (exclusion criteria)
Inclusion
29 studies included

Figure 1

Flowchart of the literature search for papers on health risks deriving from pesticide residue analyses.

The selected papers were published in between 2004 and 2022 and are out of 14 countries. China, 12 papers, and Brazil, five papers are the countries with the highest number of published articles. Evidencing the worldwide concern with health risks deriving from pesticide residues there are publications issued in Africa (three papers), Americas (eight papers), Asia (13) and Europe (five papers).

Produce samples evaluated in the health risk studies had been collected from retailers or directly from production units. The exception is the work of Caldas *et al* (2011) in which the samples were retrieved from a university cafeteria. All the samples were ready to eat so that the determined pesticide residues correspond to the theoretically ingested amount of residues as there was no afterwards processing before intake. The exception is a publication by Tsoutsi *et al*. (2008). The authors evaluated Greek olive oil samples (Table 1).

Table 1

Number of samples, searched active ingredients and risk condition retrieved from publications selected by a systematic review and meta-analysis to determine dietetic risk deriving from intakes of produce with presence of pesticide residues.

Authors	Year	Country	Nº of samples	Active Ingredients			
				No. of searched pesticides	Quantified	Unacceptable	Risk
Atuhaire et al.	2017	Uganda	225	1 chemical group (Dithiocarbamate)	11	0	Acceptable
Caldas et al.	2004	Brazil	540	5	5	0	Acceptable
Caldas et al.	2011	Brazil	175	10	10	0	Acceptable
Chen et al.	2021	China	800	14	6	0	Acceptable
Chu et al.	2020	China	440	98	26	0	Acceptable
Dülger and Tiryaki	2021	Turkey	360	3	3	0	Acceptable
Elgueta et al.	2021	Chile	57	180	14	0	Acceptable
Fatunsin et al.	2020	Nigeria	- /1	2 chemical groups (Organophosphates and Carbamates)	2	0	Acceptable
Gu et al.	2021	China	40	141	43	0	Acceptable
Hero et al.	2018	Bosnia - Herzegovina	10	8	4	1	**Unacceptable** (Acute risk for Carbofuran)

All the analyses in the listed publications quantified the active ingredients and in 24 papers (82.8%), the health risks determined were considered acceptable. That means that in the majority of the selected work the estimated ingestion of pesticide residues was below the safe limit for maximum daily intake of residues for every quantified active ingredient. In five papers (17.2%) were listed residues of pesticides posing a potential health risk to consumers.

The number of determined active ingredients varied considerably: papers related to one single active ingredient (Xu *et al*., 2018) and papers with more than 200 determined active ingredients (Jardim *et al*., 2018a; Jardim *et al.*, 2018b; Li *et al.*, 2021; Nougadére *et al.*, 2014; Sieke, 2018; Zhang *et al.*, 2021). Similarly, the number of evaluated species varied also extensively. The paper of Jing *et al.* (2021) presents the lowest number (9) of evaluated samples. On the other hand, in the work of Jardim *et al*. (2018a; 2018b) 31,024 samples were evaluated. Xu *et al*. (2018) examined 20,496 samples. In total, 65,582 produce samples were examined for presence of residues and in the work of Jardim *et al.* (2018a; 2018b) the samples were tested for residue presence of distinct chemical groups.

The average occurrence of active ingredient residues of pesticides in agricultural crops suggesting a potential health risk in the reviewed papers is of 0.004 and a 95% confidence interval (95% C.I.) in between 0.000 and 0.007. These figures indicate that out of 100 active ingredients tested for, 0.4% might pose an unacceptable risk for human health (Figure 2). The zero percent 95% C.I. (*p*=<0.684) indicates low heterogeneity, which means that all the evaluated papers presented similar results with reference to the residues of active ingredients classified as of an unacceptable risk.

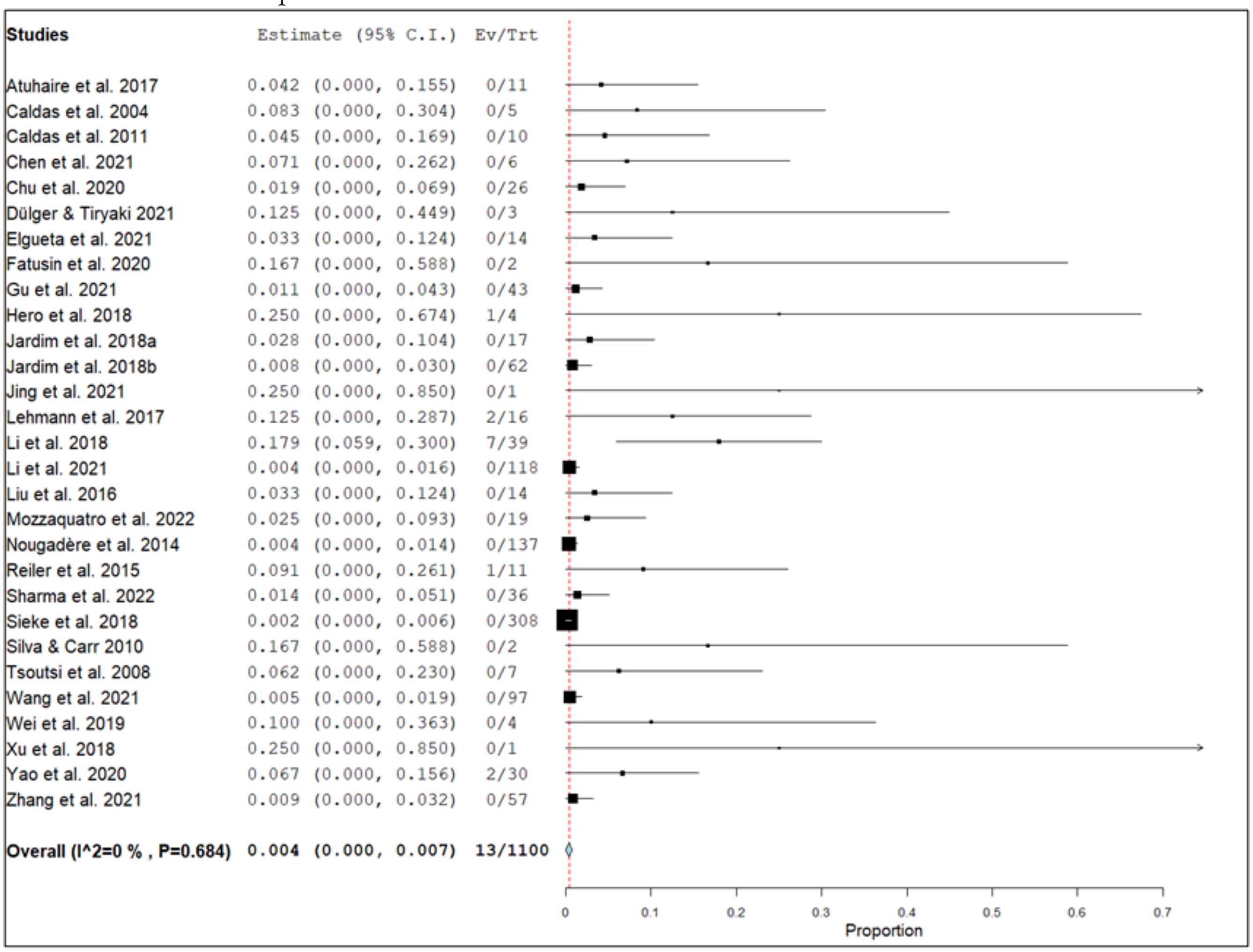

Figure 2

Averages of active ingredient residues of food samples with an unacceptable risk in studies selected through a systematic search procedure.

In developed countries, the average occurrence of active ingredients with an unacceptable risk was of 0.002 and a 95% C. I. in between (-)0.002 and 0.006. The average occurrence in less developed countries was 0.009 and a 95% C. I. in between 0.002 and 0.016 (Figure 3). In both subgroups no data heterogeneity was determined (zero percent). A *p* ≤0.688 for developed countries and a *p* ≤0.702 for undeveloped countries.

Values of *p* are a probability measure in which values above 0.05 indicate that there is no significant statistical heterogeneity amongst the studies in every category of countries.

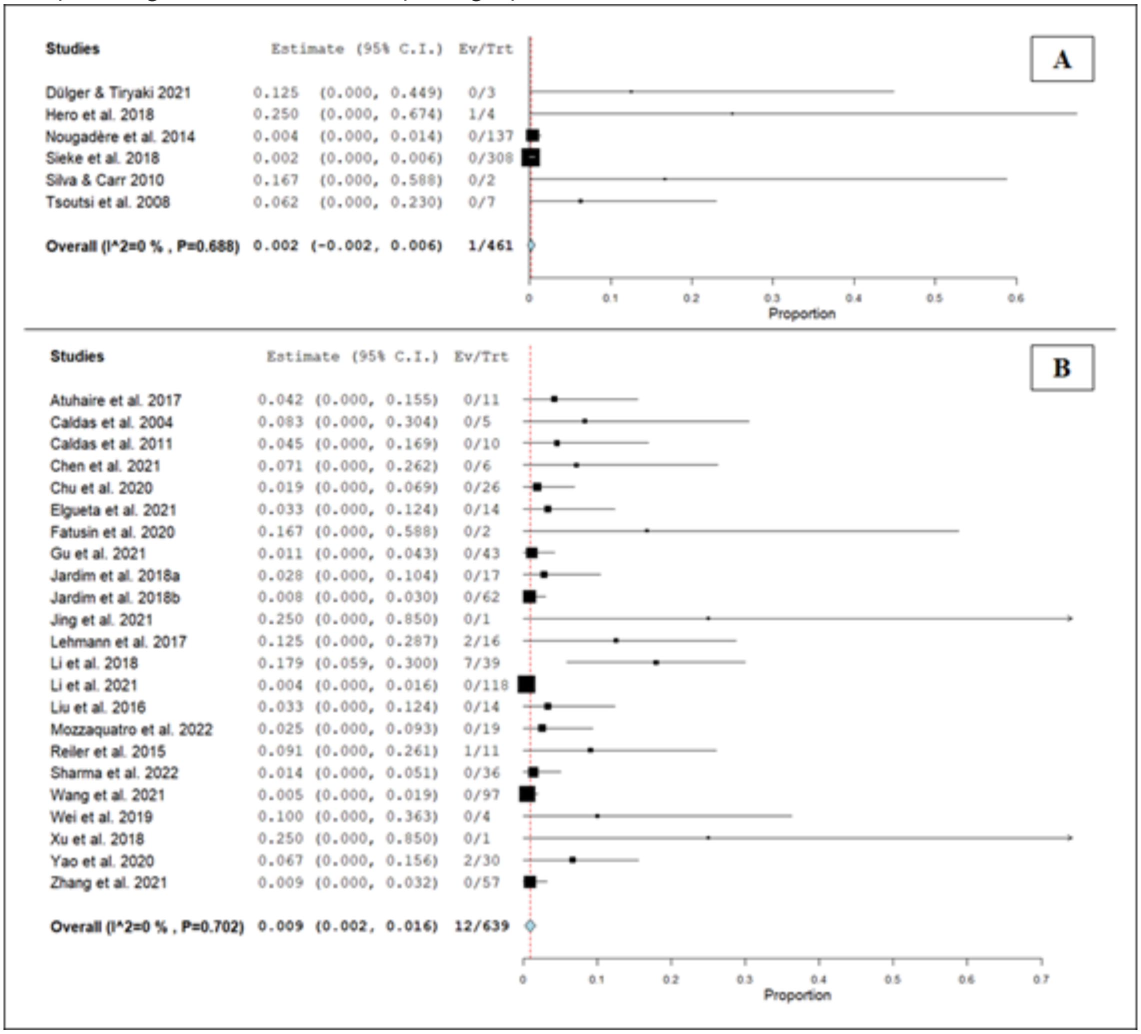

Figure 3

Averages of active ingredient residues in food samples posing an unacceptable risk reported in studies from developed countries (A) and undeveloped countries (B) that were selected through a systematic review.

Papers from developed countries presented lower averages and a lower number of selected publications. Out of six papers in only one was reported an unacceptable risk. On the other hand, coming from undeveloped countries, 25 papers met the selection criteria and five reported unacceptable risks for at least one active ingredient.

A lower occurrence of studies from developed countries reporting unacceptable risks might be a result of higher compliance to Good Agricultural Practices and growers' awareness. As evidence for that statement, the European Union determines that all countries under its jurisdiction should have a grower's advisory system to assist in the implementation of the European legislation on environmental, agricultural, public health, animal health and animal welfare specifications.

The occurrence averages of active ingredients indicating an unacceptable risk were in the range 0.110 with a 95% C.I. of 0.050 and 0.171 (Figure 4). These figures signal that out of 100 quantified active ingredients, in average, 11 are likely at concentrations that characterize an unbearable human risk.

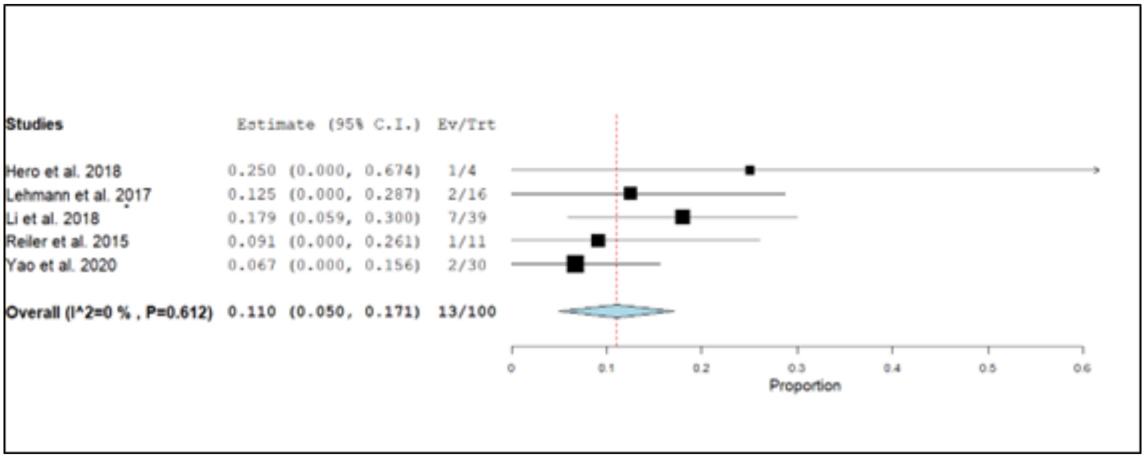

Figure 4

Averages of active ingredients residues in food samples at an unacceptable risk reported in selected studies through a systematic review procedure.

In all the publications reporting unacceptable risk only the food samples analyzed by Hero *et al*. (2018) were collected at vending points. In the remaining papers, samples were retrieved from production sites and before reaching retailer shelves. Retrieval directly from growers might result in the reduction of the interval between the last product application and the ingestion of that product.

Reiler *et al.* (2015) evaluated the cumulative risk of the presence organophosphates in tomatoes after three, six or 10 days after harvest and concluded that the risks become significantly lower already after three days after harvest. Therefore, the reduction of the interval between the last pesticide spray and the consumption might aggravate the risk. And moreover, when the growers do not comply with good agricultural practices guidance's. Siqueira *et al.* (2013) observed that the grace period, the interval in between the last pesticide spray and harvest, is unknown for as much of 28.3% and not abided by 3.8% of interviewed field workers.

The majority of active ingredients listed in papers retrieved from the systematic review with an unacceptable risk are used as insecticides and belong to the organophosphate chemical group (Table 2). That result is noticeable bearing in mind that organophosphate pesticides act by inhibiting cholinesterases, especially acetylcholinesterase (AchE; EC 3.1.1.7) increasing acetylcholine levels in the synapses. Lachrymation, salivation, sweating, diarrhea, tremors and cardiorespiratory disorders are the main symptoms of organophosphate intoxication in mammals (Cavaliere *et al.*, 1996).

Table 2

Active ingredients and their respective chemical groups and agronomic use retrieved from papers selected via a systematic review process and meta-analysis to determine dietetic risk of consumption of produce with presence of pesticide residues.

Author	Year	Unacceptable active ingredients	Chemical group	Agronomic use
Hero et al.	2018	Carbofuran	Carbamate	Insecticide
Lehmann et al.	2017	Chlorpyrifos	Organophosphate	Insecticide
		Lambda-cyhalothrin	Pyrethroid	Insecticide
Li et al.	2018	Carbendazim	Benzimidazol	Fungicide
		Chlorpyrifos	Organophosphate	Insecticide
		Cyhalothrin	Pyrethroid	Insecticide
		Cypermethrin	Pyrethroid	Insecticide
		Lambda-cyhalothrin	Pyrethroid	Insecticide
		Pyridaben	Piridazinone	Acaricide and Insecticide
		Triazophos	Organophosphate	Insecticide, Acaricide and Nematicide
Reiler et al.	2015	Chlorpyrifos	Organophosphate	Insecticide
		Dimethoate	Organophosphate	Insecticide
		Malathion	Organophosphate	Insecticide
		Ethyl-parathion	Organophosphate	Insecticide
		Methyl- parathion	Organophosphate	Insecticide
Yao et al.	2020	Carbofuran	Carbamate	Insecticide
		Methamidophos	Organophosphate	Insecticide

Both, chlorpyriphos and carbofuran are the most frequent active ingredients determined in the papers retrieved by the systematic review. Massive use of chlorpyriphos has caused soil, air and water contamination in many places of the world by way of industrial and agricultural processes release (Lakshmi *et al*., 2008). Carbofuran is highly toxic to humans and wildlife and, even though being easily degraded, might induce deleterious effects to non-target species before its environmental dissipation (Moreira *et al.*, 2004).

Another point to highlight are the methods used to conduct risk assessments, which may vary depending on the objectives of the assessment and data availability. In the deterministic model fixed values either the average or certain percentile of concentration and intake per body weight are used to calculate ingestion (Caldas & Jardim, 2012). In the probabilistic model, the variables concentration and consumption are described as distributions and a simulation model such as the Monte Carlo computational algorithm is used to build an intake distribution and to characterize its variability and uncertainty (van Klaveren; Boon, 2009). Cumulative exposure to various chemicals with same mode of action might be estimated on the basis of relative potency to a one single reference compound (Boobis *et al.*, 2008).

In the review process, papers were included independently of the method reported for risk analysis. However, the more standardized and detailed the information used, the more accurate the risk assessments and the comparisons. Overall, the averages of active ingredients at concentrations pointing to an unacceptable risk were low. It is important emphasize, though, that in the systematic review only papers evaluating produce were included. Nonetheless, the human diet is based on a miscellaneous food assortment that might, as well, carry pesticide residues or even residues of other chemical substances. Medicaments, veterinary drugs, food additives, microbiological contaminants or even toxic compounds formed during processing, such as acrylamides, polycyclic aromatic hydrocarbons, acroleins, nitrosamines, biogenic amines are examples of residues that need to undergo risk analysis (Moura *et al.*, 2020).

Exposure to substances of conceivable risks to human health might also befall by polluted air or contaminated waters. Sieke (2018) calls attention to the fact that the Acceptable Daily Intake (ADI) of copper was almost entirely fulfilled by food intakes sprayed with copper-based pesticides and yet, potable water is considered the main source of that element.

Despite the appointed negative aspects, healthy and diversified diets consist also of fresh produce consumption. The benefits outweigh the risks. Reiss *et al.* (2012) estimated the effects of daily ingestions of a portion of fruits and vegetables on the risk of cancer upsurges because of pesticide residues. The authors concluded that, yearly, 20 thousand cancer occurrences could be prevented via consumption increments of fruits and vegetables while only 10 new cancer events could be attributed to the additional pesticide residues intakes.

Conclusions

Meta-analysis of scientific publications might be used to identify and quantify the associated risk of fresh produce consumption in future risk evaluation procedures. A systematic compilation of available evidences is decidedly important for decision-making by risk managers and to the outline of public health policies and measures.

Conflict of interest statement

The authors declare no conflicts of interest.

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