Effectiveness of household processes in the reduction of pesticide residue concentrations on fruit and vegetables

25-01-2022

Examiners: Dr. Anke Huss & Dr. Ulrike Gehring

Daily Supervisor: Dr. Daniel M. Figueiredo

Master student: Student number - Floris Pekel: 1334543

Laymen Summary

Pesticides worden op grote schaal gebruikt in de landbouw sector om gewassen te beschermen tegen pathogenen zoals schimmels of insecten. Het gebruik van pesticides is van groot belang in de sector om aan opbrengst verwachtingen en voedselzekerheid te voldoen. In de afgelopen jaren is er in toenemende mate bewezen dat blootstelling aan pesticides, via inademing, het eten van voedsel en aanraking via de huid leidt tot hogere kansen op ontwikkeling van ziektes. Eten van voedsel is voor consumenten het grootste risico om bloot gesteld te worden aan pesticides, daarom is het van groot belang om te weten wat de hoeveelheid pesticides is op voedsel op het moment van consumptie.

Een belangrijke factor die invloed heeft op de pesticideconcentratie in groente en fruit is de mate waarin producten na het oogsten worden behandeld voordat ze worden geconsumeerd. Behandelingen die veel mensen vooral thuis uitvoeren zijn het wassen, koken, blancheren, schillen of invriezen van groente en fruit voor consumptie. De effectiviteit van pesticide verwijdering van groente en fruit is afhankelijk van het type pesticide, de fruit- of groentesoort, type behandeling en wordt in de wetenschappelijke literatuur uitgedrukt als ‘Bewerkingsfactor’ (BF). De bewerkingsfactor van een zo’n unieke pesticide/type voedselproduct/type behandeling combinatie wordt berekend door de pesticide concentratie voor en na de handeling (Bijv. schillen van een appel) in het voedselproduct te meten, waarna de ratio tussen beide concentraties de BF is.

Door het grote aantal pesticides, groente- en fruitsoorten en mogelijke behandelingen is het op het moment niet duidelijk hoeveel bewerkingsfactoren al bekend zijn in de literatuur. Bovendien is er ook meer kennis informatie nodig over andere factoren die een invloed kunnen hebben op de effectiviteit van pesticideconcentratie afname. Voorbeelden hiervoor zijn het gebruiken van azijn of soda tijdens het wassen, de duur van een behandeling (lees: hoelang kook je de groentes) of de tijd tussen het aanbrengen van de pesticides en het tijdstip van de behandeling om de pesticides eraf te krijgen.

Het doel van deze studie is om de bewerkingsfactoren van de huidig pesticide/product/behandeling combinaties in kaart te brengen. Daarnaast onderzoekt deze studie wat de nieuwe inzichten over het belang van gebruikte meettechnieken en andere factoren die invloed kunnen hebben op de pesticide reductie. Om een duidelijk overzicht te krijgen van de effectiviteit van verschillende behandelingen (koken, blancheren, pellen/schillen, wassen of onderdompelen) van voedsel in het huishouden werd wetenschappelijke literatuur doorzocht voor de tien meest gegeten groentes en fruit in Nederland in combinatie met 15 veel gebruikte pesticides**.** Kortom, de ontdekte behandelingsfactoren kunnen worden gebruikt voor toekomstige risicoschattingen van pesticides voor de volksgezondheid en daarnaast .

Abstract

Widespread use of pesticides on food products and the associated risks of exposure through dietary intake require the need for precise quantitative risk assessments. Household processing of food products affect the pesticide residue concentration and plays an important role in the accurately estimating if concentrations exceed legislative limits. The mean or median reduction of such processes is expressed as processing factor (PF) and is unique for each combination of pesticide, process and type of product (PPP). Currently, many PFs for household processes are unknown and a clear overview in literature is lacking. Therefore, this study performed a systematic review on literature that researched the reduction effectiveness of specific household processes (boiling, blanching, washing, soaking, peeling) on widely used pesticides (*Azoxystrobin, Boscalid, Carbendazim, Chlorpyrifos, Cypermethrin, Cyprodinil, Difenoconazole, Fenhexamid, Fludioxonil, Imidacloprid, Iprodione, Pyraclostrobin, Pyrimethanil, Tebuconazole, Thiacloprid*) present in popular fruits and vegetables between 2011 and 2022. In total 28 out of 121 articles were included and their data extracted. High variation was found in effectiveness of PPPs with studies showing contradicting results which is possibly a result of differences in experimental set-up. In general the included studies revealed that peeling, boiling and blanching were more effective in reducing pesticide concentration. Washing and soaking showed high variability in effectiveness with indications that washing with acetic-/citric acid or sodium carbonate solvents were more effective than tap water, however this was highly dependent on the physiochemical properties of the pesticide. This review provides an overview of currently known literature on household processing factors and discusses important determinants of PF. Moreover, it discusses important challenges and factors that are present in this field of research. By doing so, this research aims to contribute to the field of pesticide risk assessments and increase the public health.

Table of content

1 - Introduction

Consumption of fruits and vegetables are crucial for a balanced and healthy lifestyle. The World Health Organisation (WHO) promotes the consumption of five fruits or vegetables on a daily basis (World Health Organisation, 2020). One of the major issues in meeting the worldwide growing demand of fruit and vegetable consumption is the presence of crop destroying pests, such as fungi, insects or other pathogens. Plant protection products, also known as pesticides, are widely used in the agricultural industry to combat these pests. Due to the high variety in pests, an equally broad spectrum of pesticides is developed which differ in their chemical properties and relative toxicity towards the environment and non-target species such as humans (Chung, 2018; Nicolopoulou-stamati et al., 2016).

Increasing evidence associates pesticide exposure, either via ingestion, inhalation or contact via the skin, to increased risks of negative health outcomes (Nicolopoulou-stamati et al., 2016). For the non-occupational population, exposure through food consumption is proposedly the major exposure pathway of pesticides. To determine if individuals ingest unacceptable quantities of pesticide, dietary risk assessments calculate the daily or yearly intake of pesticides based on estimations of pesticide concentrations and product consumption. Countries set maximum residue limits (MRL) that determine the maximum concentration of pesticides on consumer goods, based on the chronic and acute exposure estimates (e.g. Acceptable dietary intake (ADI)) (Carrasco Cabrera & Medina Pastor, 2021) .

Factors that determine residue concentration are the pesticides half-life time and the effects of commercial and household processing or cleaning steps (Bonnechère et al., 2012; Skovgaard et al., 2017; T. Yang et al., 2017). While many products undergo commercial processing steps after harvesting (e.g.) washing in ozonated or chlorinated water, canning or pasteurization), pesticide residues are often not completely degraded and can remain present on sold products. Frequently performed household processes are washing, soaking, boiling, blanching or peeling of fruits or vegetables before consumption. The reduction effectiveness of such processes mainly depend on the type of product, the physio-chemical properties of the pesticide residue and the processing technique.

The reduction of the unique combination of the product/pesticide/process (PPP) are expressed in processing factors (PF). The PF of a reduction process is calculated by taking the ratio between the residue concentration in the processed product and in the raw commodity. A PF above 1 would imply that the concentration of a pesticide has increased, whereas a PF lower than one would indicate a reduction. The time of pesticide application and general study set-up are also known to influence the reduction capabilities of different processes (Anastassiades et al., 2003; Jankowska et al., 2019; Rani et al., 2013). Unfortunately, consensus on processing factors estimates as well as a clear overview of the total literature available for household practices remains lacking. In 2011, the European Food Safety Agency (EFSA) set up a pesticide processing factor database, which mainly encompasses processing factors for commercial practises.

Therefore, the aim of this study was to gain insight in the effectiveness of pesticide reduction by household processes on fruits and vegetables after harvest and commercial processing. The study focused on a selection of products with a high consumption rate in combination with fifteen widely used pesticides for the five household processes: washing, soaking, cooking, blanching and peeling. Moreover, the report investigated which determinants of each PPP combination explained the observed PF. Finally, this reports discusses the challenges in comparing reduction effectiveness between studies due to measurement uncertainties.

2 - Methods

To obtain the required literature, a systematic search was conducted on the online database Scopus that focused on studies that could be present in the EFSA PF database. Since the database includes studies from 2011 onwards, this systematic search also focused on studies published in 2011 or later. The search syntax included search terms for household processes (washing, boaking, boiling, blanching, peeling) in combination with 16 often applied PPP's (Azoxystrobin, Benomyl, Boscalid, Carbendazim, Chlorpyrifos, Cypermethrin, Cyprodinil, Difenoconazole, Fenhexamid, Fludioxonil, Imidacloprid, Iprodione, Pyraclostrobin, Pyrimethanil, Tebuconazole, Thiacloprid). Furthermore, the search focussed on the following food products, which were chosen because of their high consumer rate in the Netherlands (Voedingscentrum Nederland, 2022): apple, broccoli, carrot, lettuce, mandarin, pear, (sweet) pepper, spinach, strawberry, tomato, orange. The complete syntax search can be found in the supplemental material.

For article selection, the following criteria were set: studies needed to include one of the process/product/pesticide combination from the syntax search. Reduction processes should be applicable to a household setting (e.g. ozonated water was excluded).

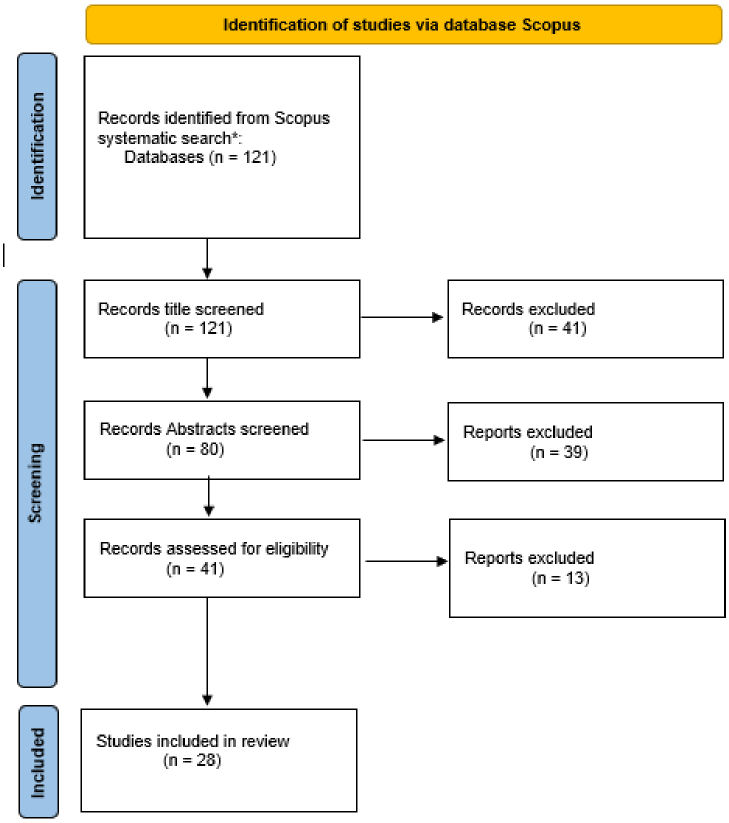

Figure 1 – PRISMA flow overview of the systematic review and the selection criteria.

Title and Abstract screening reduced the initial 121 articles to 41, after which an additional 13 articles were excluded upon reading the complete study. These final thirteen were excluded based on the exclusion criteria. This resulted in a total of 28 articles that were included in the systematic review (Tab S1).

Table 1 - Overview of physiochemical characteristics of the pesticides included in the systematic review. Log P/K_{ow} *= the octanol-water coefficient.* S_w *= Water solubility. Mode of action: sys = systemic, non-sys = non systemic. Data derived from database University of Hertfordshire. Website visited: http://sitem.herts.ac.uk/aeru/ppdb/en/Reports/75.htm*

Nr	**Pesticide**	**Active Group**	**Type**	**Mode of action**	**log P / Kow**	***Sw (mg/L)***	**Degredation point (°C)**	***Molecular mass (g/mol)***
1	Azoxystrobin	Strobilurin	fungicide	sys	2,5	6,7	345	403,4
2	Benomyl	Benzimidazole, Carbamate	fungicide	sys	1,4	2	140	290,32
3	Boscalid	Carboxamide	fungicide	sys	2,96	4,6	300	343,21
4	Carbendazim	Benzimidazole, Carbamate	fungicide	sys	1,48	8	305	191,21
5	Chlorpyrifos	Organophosphate	insecticide	non-sys	4,7	1,05	170	350,58
6	Cypermethrin-α	Pyrethroid	insecticide	non-sys	5,8	0,004	248	416,3
7	Cypermethrin-β	Pyrethroid	insecticide	non-sys	5,8	0,9	253	416,3
8	Cyprodinil	Anilinopyrimidine	fungicide	sys	4	13	-	225,29
9	Difenoconazole	Conazole	fungicide	sys	4,36	15	337	406,26
10	Fenhexamid	Hydroxyanilide	fungicide	non-sys	3,51	24	230	302,2
11	Fludioxonil	Phenylpyrrole	fungicide	non-sys	4,12	1,8	306	248,19
12	Imidacloprid	Neonicotinoid	insecticide	sys	0,57	610	230	255,66
13	Iprodione	Dicarboximide	fungicide	non-sys	3	6,8	233	330,17
14	Pyraclostrobin	Strobilurin	fungicide	sys	3,99	1,9	200	387,82
15	Pyrimethanil	Anilinopyrimidine	fungicide	non-sys	2,84	110	189,9	199,28
16	Tebuconazole	Triazole, Conazole	fungicide	sys	3,7	36	350	307,82
17	Thiacloprid	Neonicotinoid	insecticide	sys	1,26	184	250	252,72

3 –Results literature findings

3.1 - Results systematic search

Of the twenty-eight articles included, the most studies (n=14) presented results on soaking and washing (n=13), shortly followed by peeling (n=11). Six studies included results on the effects of boiling, while only three studies provided results for blanching. Finally, one study mentioned the reduction effects of prolonged freezing and another study investigated the combined effects of soaking, washing and heating in a microwave on pesticide concentrations.

Table 2 shows the amount of times each pesticide and household process, or the combination between pesticide and process, was included in a study. Overall the pesticides Chlorpyrifos, Azoxystrobin, Cypermethrin, Imidacloprid, Boscalid and Difenoconazole were studied most frequently ($n \geq 5$), whereas Carbendazim, Fenhexamid, Fludioxonil, Pyrimethanil and Thiacloprid were included the least ($n \leq 2$). Of the included products, apple, tomato and spinach were studied five, ten and seven times, respectively. The majority of the products (Broccoli, Lettuce, Orange, Pepper and Strawberries) were only included one or two times. Three of the eleven products were not included at all (Pear, Mandarin and Carrot).

One of the most important conditions of good research is reproducibility and the ability to compare results between different research groups, so that potential measurement errors are minimized (Schwab et al., 2022). Ideally, each product/process/pesticide combination should be investigated/studied at a minimum of two times. Table 2 indicates that for the majority of the combinations this is not feasible. Fortunately, comparability between pesticides remains possible if both the product and household process remain similar because this provides the opportunity to compare the effects of physiochemical properties of the pesticides.

Table 2 - Number of studies in the review that included one of the process-pesticide combination. Moreover, the total number of studies that investigated one of the included processes is given in the last row and column.

Number of studies mentioning specific pesticide/process combination							
	Boiling	**Blanching**	**Peeling**	**Washing**	**Soaking**	**Combi / Freezing**	**Total**
Azoxystrobin	2	2	3	3	4	0	7
Boscalid	2	1	2	2	2	1	5
Carbendazim	0	0	1	1	0	0	2
Chlorpyrifos	2	0	6	6	7	1	10
Cypermethrin	3	1	3	2	4	0	8
Cyprodinil	1	0	3	2	1	0	4
Difenoconazole	0	0	3	2	2	0	5
Fenhexamid	1	0	0	0	1	0	1
Fludioxonil	2	1	0	1	2	0	2
Imidacloprid	1	1	1	3	4	0	6
Iprodione	1	0	0	0	2	1	2
Pyraclostrobin	3	2	1	3	2	0	4
Pyrimethanil	0	0	1	0	0	0	1
Tebuconazole	1	0	2	2	0	0	3
Thiacloprid	0	0	1	0	0	0	1
Total	6	3	11	13	14	2	-

3.2 - Washing

The definitions of washing and soaking are interchangeably applied in many studies included in the research. Therefore, the following criteria were set to define 'soaking' : No specific specification or mention of running water, the duration of the process exceeds 5 minutes and solvents other than tap/distilled/chlorinated water were used during the washing step.

For tomatoes, only limited to intermediate reduction was observed for all pesticides tested (Table 3 & S1), with Azoxystrobin (PF = 0.32) and Boscalid (PF = 0.39) showing to be reduced the most (Jankowska et al., 2016), however this was dependent on the type of tomato. For Pyraclostrobin, Cypermethrin and Cyprodinil the mean reduction ranged between 10-29%, 27-41% and 36-41%, respectively, differing in washing duration and type of tomato (Ajeep et al., 2021; Jankowska et al., 2016; Wanwimolruk et al., 2017). Washing broccoli with chlorinated water resulted in a similar range order for Azoxystrobin (PF = 0.59), Boscalid (PF = 0.76) and Pyraclostrobin (PF = 0.69) (Lozowicka, Jankowska, & Rutkowska, 2016).

Interestingly, the same pesticides were reduced significantly more effective on leave vegetables such a lettuce and spinach, with the notion that the washing duration was a five-fold longer. For Azoxystrobin and Pyraclostrobin a minimum reduction 85% was observed in spinach and lettuce, while Fludioxonil and Imidacloprid also showed good reduction percentages ranging between 86.4 – 87.3% and 83.6 – 46.8%, respectively (S. J. Yang et al., 2022).

Moreover, Rani et al (2013) and Hendawi et al (2013) illustrated that prolonged time between pesticide application and the moment of reduction processes, diminishes the effectiveness of these processes such as washing. The studies showed that a waiting step of 7 days, decreased the pesticide removal efficiency with 13.67% for Imidacloprid in strawberries and 27.66 % for Chlorpyrifos in tomatoes. Contradictory, a similar study on lettuce showed higher removal of Chlorpyrifos with increased waiting time (Akoto et al., 2016).

Table 3 – Effect of washing on pesticide reduction in fruit and vegetables. Note: This tables displays some of the highlights of the studies that were included in the systematic search. All the study results (including study details) can be found in the supplemental material.

Washing					
Product	Duration min	Pesticide	PF mean or range	Mean reduction %	Author
Tomato	NA	Cypermethrin		27	Wanwimolruk et al (2017)
Tomato (marissa)	*1*	Azoxystrobin		68	Jankowska et al (2016)
	1	Boscalid		61	
	1	Cyprodinil		41	
	1	pyraclostrobin		29	
Tomato (Harzfeuer)	*1*	Azoxystrobin		38	
	1	Boscalid		35	
	1	Cyprodinil		36	
	1	pyraclostrobin		10	
Tomato	*1*	Chlorpyrifos		41.29	Rani et al (2013)
	1	Chlorpyrifos		31.7	
	1	Chlorpyrifos		25.67	
	1	Chlorpyrifos		19.56	
	1	Chlorpyrifos		13.63	
Tomato	5	Chlorpyrifos	0.58		Ajeep et al (2021)
	5	Cypermethrin	0.59		
Spinach	*5*	Azoxystrobin		96.4	Yang et al (2022)
	5	Fludioxonil		86.4	
	5	Imidacloprid		83.6	
	5	pyraclostrobin		91.5	
Spinach	-	Azoxystrobin		100	Yang et al (2012)
Lettuce	*5*	Azoxystrobin		92.1	Yang et al (2022)
	5	Fludioxonil		87.3	
	5	Imidacloprid		46.8	
	5	pyraclostrobin		85.9	
Broccoli	-	Azoxystrobin	0.59	40.5	Lozowicka et al (2016)
	-	Boscalid	0.76	24.2	
	-	Pyraclostrobin	0.69	30.8	
Apple	1	Cyprodinil	0.63		Słowik-Borowiec et al (2020)
	1	Difenoconazole	0.62		
	1	tebuconazole	0.43		
Apple	3	Chlorpyrifos		18	Pirsaheb et al (2016)

3.3 - Soaking

Soaking proved to have varying effectiveness, depending on the type of pesticide, soaking duration, product and in some cases addition of household chemicals. Soaking with tap water or sodium bicarbonate (Na2HCO3) reduced Chlorpyrifos with approx. 90% and Imidacloprid with 70.9% in tomatoes (Table 4). Addition of acetic acid (CH3COOH) and sodium carbonate (Na2HCO3) failed to significantly increase the reduction effectiveness of Chlorpyrifos, Azoxystrobin and Imidacloprid in tomatoes (Rodrigues et al., 2017; Wasilewski et al., 2022). Furthermore, Azoxystrobin (26%) and Difenoconazole (17%) concentrations are poorly reduced by immersion in water for a long period of time (30 min) (Wasilewski et al .,2022). However, the application of acetic acid or sodium carbonate improved reduction efficiency to 43 and 32% for Azoxystrobin and 42 to 47% for Difenoconazole, respectively (Wasilewski et al .,2022).

Soaking strawberries in chlorinated water for 1 to 5 minutes, showed sharp concentration decrease with increased time for Cyprodinil (PF = 0.85 to PF = 0.46), Fenhexamid (PF = 0.73 to 0.43), Fludioxonil (PF = 0.88 to PF = 0.47), Chlorpyrifos (PF = 0.52 to PF= 0.32). In contrast, Boscalid and Pyraclostrobin concentrations appeared to be more stable with observed PFs of 0.67 to 0.61 and 0.8 to 0.69, respectively (Lozowicka, Jankowska, Hrynko, et al., 2016).

Cypermethrin concentrations in spinach, similar to the observations in the boiling process, only reduced 17% during soaking processes with tap water. However, addition of approx. 5 a 6% of acetic acid, citric aced or sodium carbonate resulted in reductions in the range of 56 to 68%, 38 to 64% and 40 to 61%, respectively (Amir et al., 2019; Hussnain et al., 2021). Moreover, multiple studies investigated the effects of adding garlic or ginger extracts to water, which resulted in observed Cypermethrin reduction in the range of 43 – 55 % for garlic and 50 – 55% for ginger extract (Amir et al., 2019; Hussnain et al., 2021). Similar experiments for Imidacloprid showed that adding acetic acid and citric acid sharply decreased the Imidacloprid concentration. Adding 2% of sodium carbonate or sodium chloride resulted in similar reduction values when only water was used, ranging between 24 to 27% (Abdullah et al., 2016).

Table 4 - Effect of soaking on pesticide reduction in fruit and vegetables. Note: This table displays some of the highlights of the studies that were included in the systematic search. All the study results (including study details) can be found in the supplemental material.

Soaking						
Product	Duration min	Process details	Pesticide	PF	Mean reduction %	Title
Tomato	15	Tap water	Chlorpyrifos		90.58	Wasilewski et al (2021)
	15	Na2HCO3			90.88	
	15	Vit C.			70.85	
Tomato	15	2% acetic acid	Chlorpyrifos	0.48		Ajeep et al (2021)
	15	2% acetic acid	Cypermethrin	0.55		
Tomato	30	distilled water (25 °C)	Azoxystrobin		26	Rodrigues et al (2017)
	30	acetic acid (0.15%)			33	
	30	acetic acid (5%)			43	
	30	sodium carbonate (1,5%)			22	
	30	sodium carbonate (5%)			32	

Tomato	30	sodium hyopchlorite (0,04%)			21	
	30	sodium hyopchlorite (1%)			30	
	30	distilled water	Difenoconazole		17	
	30	acetic acid (0,15%)			28	
	30	acetic acid (5%)			42	
	30	sodium carbonate (1,5%)			35	
	30	sodium carbonate (5%)			47	
	30	sodium hyopchlorite (0,04%)			32	
	30	sodium hyopchlorite (1%)			43	
	10	tap water	Imidacloprid		70,9	Al-Amir et al (2015)
	10	5% acetic acid			75,37	
	10	5% sodium hypochlorite			73,13	
	10	0,01 % potassium permanganate			76,86	
	10	1% Hula-San			78,36	
Spinach (Cezanne)	*3*	Tap water & 15 C + stirring	Boscalid	0.71 +- 0.10		Bonnechère et al (2012)
	3		iprodione	0.57 +- 0.13		
Spinach (SP-916)	*3*	Tap water & 15 C + stirring	Boscalid	0.53 +- 0.07		
	3		iprodione	0.52 +- 0.09		
Apple	*15*	Tap water	Chlorpyrifos		87.52	Wasilewski et al (2021)
	15	Na2HCO3			88.97	
	15	Vit C.			61.74	

3.4 - Boiling

Boiling for a prolonged duration (> 5 min) shows to be effective for almost all pesticides tested (Cypermethrin β, Chlorpyrifos, Tebuconazole, Azoxystrobin, Fludioxonil, Pyraclostrobin, Iprodione, Boscalid, Fenhexamid) and products with PFs ranging between 0.57 and 0. Interestingly, the studies by Lozowicka et al (2016) and Słowik-Borowiec et al (2020) clearly indicate that increased boiling duration results in higher pesticide reduction. In contrast, Cypermethrin α concentration increased with increased duration of boiling. Lozowicka et al (2016) showed that the PF for Cypermethrin α followed a increasing trend if strawberries were boiled for 1, 2 or 5 minutes with PF values of 1.02 ; 1.66 and 1.76, respectively. Similar results were described by Ajeep et al. (2021) with tomato samples, where PF values for Cypermethrin ranged between 1.35 and 1.38. However, as stated in the description of Table S1, the study of Ajeep et al (2021) should be interpreted with caution due to its low number of samples.

The physiochemical properties of Cypermethrin α in comparison to the other pesticides could explain these observations. In general, the concentration can increase if the mass of the product decreased during the boiling (or other thermal processes) process, while the pesticide amount remains equal if their degradation point lies above the temperature of the thermal process. However, the degradation point of Cypermethrin α is lower than that of Boscalid (D_p = 300 °C) and Tebuconazole (D_p = 350 °C), both of which did show clear concentration decreases (Table 5). Lozowicka et al (2016) argue that, besides the degradation point, the location of the pesticide is equally important for the degradation process. The water solubility (S_w) determines to which extend a pesticide resides in the aqueous environment in comparison to the more non-aqueous skin of fruits and vegetables. This could explain how the non water-soluble Cypermethrin α (S_w = 0.004 mg/L) was not degraded, while the more soluble Boscalid (S_w = 4.6 mg/L) and Tebuconazole (S_w = 36 mg/L) were. In this case, it is more likely that the soaking of the products, an additional process that occurs during boiling, was the main cause for the decreased concentration of Boscalid and Tebuconazole.

Table 5 - Effect of boiling on pesticide reduction in fruit and vegetables. Note: This tables displays some of the highlights of the studies that were included in the systematic search. All the study results (including study details) can be found in the supplemental material.

Boiling

Product	Duration min	Pesticide	PF mean or range	Mean reduction %	Author
Tomato (Roma)	10	Cypermethrin α	0.851	14.96	Arowolo et al (2022)
	10	Cypermethrin β	0.534	46.61	
Tomato (Roma VF)	10	Cypermethrin α	0	100	
	10	Cypermethrin β	0	100	
Tomato	5	Cypermethrin	1.38 - 1.35		Ajeep et al (2021)
	5	Chlorpyrifos	0.4		
Apple (Gala)	1	Tebuconazole	0.45	55	Słowik-Borowiec et al (2020)
	5	Tebuconazole	0.27	73	
	15	Tebuconazole	0.22	78	
Spinach	5	Azoxystrobin		85	Yang et al (2022)
	5	Fludioxonil		70.9	
	5	Imidacloprid		94.5	
	5	Pyraclostrobin		42.7	
Lettuce	5	Azoxystrobin		87.3	
	5	Fludioxonil		71.2	
	5	Imidacloprid		92.9	
	5	Pyraclostrobin		45.7	
Brocoli	20	Azoxystrobin	0.19	81.2	Lozowicka et al (2016)
	20	Boscalid	0.31	69.4	
	20	Pyraclostrobin	0.29	70.6	

3.5 - Blanching

Blanching is a thermal process whereby vegetables are immersed in high temperature water for a short duration. In this systematic review, only three studies reported on the effects of blanching and pesticide concentrations on tomatoes, spinach, lettuce and broccoli. All pesticides under investigation showed percentual reduction, varying between the 100 and 32.9 %. No noticeable additional decrease was observed for Azoxystrobin and Pyraclostrobin concentrations with higher water temperature (85 vs 100 °C and a doubling of blanching duration from 30 seconds to 1 minute (Table 6). Interestingly, Yang et al. (2022) showed that reduction of Azoxystrobin (81.3% to 67.8%) , Fludioxonil (67.8% to 45%), Imidacloprid (88.5% to 81.4%) and Pyraclostrobin (51.4% to 32.9%) was more acute in spinach than in lettuce samples. Overall, Imidacloprid was reduced the best in both types of products with a decrease between 88.5 and 81.4 % (Yang et al., 2022). Moreover, Pyraclostrobin levels decreased the least with mean reduction levels ranging between 32.9% to 51.5%, which was similar to the reduction range of Pyraclostrobin during boiling processes (Table 5).

Arowolo et al (2022) reported the highest reduction of Cypermethrin (α and β) for blanching with 100 % reduction in Roma VF tomatoes. However, the duration in of the process (4 min) was significantly longer than the experiments conducted by the other studies (Lozowicka, Jankowska, & Rutkowska, 2016; S. J. Yang et al., 2022) (1 min & 30 s). Moreover, Cypermethrin α concentration were more reduced during blanching than during the boiling process, which is contradictory to the general understanding that exposure to higher temperatures for a longer duration would result in more degradation of a pesticide.

Table 6 - Effect of blanching on pesticide reduction in fruit and vegetables. Note: This tables displays some of the highlights of the studies that were included in the systematic search. All the study results (including study details) can be found in the supplemental material.

Blanching					
Product	Duration min	Pesticide	PF mean or range	Mean reduction %	Author
Tomato (Roma)	4	Cypermethrin α	0	100	Arowolo et al (2022)
	4	Cypermethrin β	0.518	48.23	
Tomato (Roma VF)	4	Cypermethrin α	0	100	
	4	Cypermethrin β	0	100	
Spinach	30 s	Azoxystrobin		67.8	Yang et al (2022)
	30 s	Fludioxonil		45	
	30 s	Imidacloprid		81,4	
	30 s	Pyraclostrobin		32.9	
Lettuce	30 s	Azoxystrobin		81.3	
	30 s	Fludioxonil		67.8	
	30 s	Imidacloprid		88.5	
	30 s	Pyraclostrobin		51.4	
Brocoli	1	Azoxystrobin	0.49	51.2	Lozowicka et al (2016)
	1	Boscalid	0.46	53.8	
	1	Pyraclostrobin	0.49	51.5	

3.6 - Peeling

Varying results were reported for the effect of peeling on pesticide concentrations in tomatoes, oranges and apples (Table 7). In oranges, El-Sayed et al (2021) illustrated that peeling almost completely decreased the Chlorpyrifos levels, with PF = 0.02. In both apples and tomatoes, the effectiveness of peeling was over 50 % for Azoxystrobin , Difenoconazole, Cyprodinil, Pyraclostrobin (Andrade et al., 2015; Jankowska et al., 2016; Kowalska et al., 2022; Pirsaheb et al., 2016; Rodrigues et al., 2017; Słowik-Borowiec & Szpyrka, 2020). Concentration reduction for Chlorpyrifos showed some discrepancies, with mean reduction in apples ranging between 100% to 30% (Kowalska et al., 2022; Pirsaheb et al., 2016).

Noteworthy are the extreme results mentioned by Arowolo et al (2022) for the decrease of Cypermethrin α and β in tomatoes (Roma VF), with PF = 0 & PF = 1.107 respectively. Especially a PF = 1.107 for Cypermethrin β is interesting to discuss, because this would imply that all pesticide residues would have relocated from the tomato surface into the aqueous part of the product, after which peeling would have only reduced the mass of the product. While not specified in the article, it could be possible that the concentration of Cypermethrin fell below the limit of detection (LOD) and was therefore set to zero.

Interestingly, Rani et al (2013) showed that the effect of peeling diminished if the period between pesticide application and the peeling process increased. One hour after applying Chlorpyrifos peeling resulted in a reduction of 62.58%, whereas a reduction of 20% was observed after a waiting period of 10 days. Pesticide residues can relocate from the surface into the skin or deeper into the fruit or vegetable (Holland et al., 1994). The penetration speed of pesticides is depending on the properties of both the product and the pesticide. Reduction strategies such as peeling, washing and soaking have less effect if pesticide are located inside a product (Xu et al., 2017; T. Yang et al., 2016, 2017).

*Table 7 - Effect of Peeling on pesticide reduction in fruit and vegetables. Note: This tables displays some of the highlights of the studies that were included in the systematic search. All the study results (including study details) can be found in the supplemental material. ND = Not detected. * - The study of Kowalska et al (2022) did not provide direct reduction quantities, but only showed a reduction graphic. The numbers shown in this table are extracted from this figure, therefore the percentages are given as an estimation.*

Peeling					
Product	Duration min	Pesticide	PF mean or range	Mean reduction %	Author
Tomato (Roma)		Cypermethrin α	0.849	15.14	Arowolo et al (2022)
		Cypermethrin β	0.514	48.55	
Tomato (Roma VF)		Cypermethrin α	0	100	
		Cypermethrin β	1.107	+ 10.7	
Tomato		Azoxystrobin		68	Rodrigues et al (2017)
		Difenoconazole		79	
Tomato		Azoxystrobin		66 +- 0.83	Andrade et al (2015)
		Imidacloprid		17 +- 1.30	
Tomato		Chlorpyrifos	ND		Ajeep et al (2021)
		Cypermethrin	ND		
Tomato		Chlorpyrifos		62.58	Rani et al (2013)
		Chlorpyrifos		52.43	
		Chlorpyrifos		41.89	
		Chlorpyrifos		30.43	
		Chlorpyrifos		27.27	
		Chlorpyrifos		20	
		Chlorpyrifos		ND	
Tomato (marissa)		Azoxystrobin		88	Jankowska et al (2016)
		Boscalid		88	
		Cyprodinil		81	
		pyraclostrobin		73	

Tomato (Harzfeuer)		Azoxystrobin		82	
		Boscalid		55	
		Cyprodinil		93	
		pyraclostrobin		72	
Orange		Chlorpyrifos	0.02 +- 0.002		El-Sayed et al (2021)
Apple (2012)		Boscalid		~61	Kowalska et al (2022)*
		Chlorpyrifos		~ 52	
		Difenoconazole		~ 74	
		Pyrimethanil		~ 58	
		thiacloprid		~ 49	
		Cyprodinil		~ 53	
Apple (2020)		Boscalid		~ 43	
		Tebuconazole		~ 100	
Apple		Chlorpyrifos		30	Kong et al (2012)
		Cypermethrin β		66	
		Tebuconazole		34	
		Carbendazim		30	
Apple (Gala)		Cyprodinil	0.19		Słowik-Borowiec et al (2020)
		Difenoconazole	0.41		
		Tebuconazole	0.19		
Apple		Chlorpyrifos		100	Pirsaheb et al (2016)

3.7 - Remaining Thermal processes

3.7.1- Freezing

One study mentioned the effect of freezing on pesticide residues concentrations in apples (Pirsaheb et al., 2016). The study showed that Chlorpyrifos reduced by 66% after 48 hours at 4 °C (Tab S1). While this is not of the most used household processes, it is interesting because many household are costumed to store food for a period of time before consumption. Moreover, some studies in this systematic review mention the storing of products after harvest at low temperatures before pesticide analysis takes place. It is important to understand (and report) that storage of freezing will likely affect the concentrations of these experiments and could influence the interpretability of these studies.

3.7.2 - Microwave heating

Bonnechère et al (2012) investigated the combined effects of soaking, blanching and microwave heating on Boscalid and Iprodione concentrations on spinach samples. A clear increase in pesticide concentration was observed after microwave heating for Boscalid (PF = 1.06 – 1.12) and Iprodione (PF = 0.84 – 0.84) in comparison to only soaking and blanching spinach samples ($PF_{boscalid}$ = 0.71 – 0.55 & $PF_{Iprodione}$ = 0.42 – 0.63) (Tab S1). This observations was attributed to the evaporation of water, which resulted in mass reduction of the samples.

4 - Factors influencing pesticide reduction

The systematic review results indicate that the effectiveness of pesticide reduction is highly differential between types of pesticide, product and household process. Moreover, multiple studies mention contradictive results while investigating similar PPP combination, which illustrates the challenge of reaching trustworthy PF values. Differences in study protocols (sample size, waiting period, amount of pesticide applied, measurement technique) could provide a possible explanation and will be discussed here.

4.1 - Octanol-water coefficient (K_{ow})

In Table 1, the pesticides physio-chemical characteristics that are known to influence the removing efficiency of pesticide residues are shown, among which the octanol water coefficient (K_{ow}). The K_{ow} measures the ratio of a chemical's concentration in octanol and water and is also described as log P. The K_{ow} is a widely used characteristic that describes the hydrophobicity/hydrophilicity of a chemical (Cumming & Rücker, 2017). In general, chemicals with a high K_{ow} would accumulate more in octanol, which implies a higher lipophilicity.

Holland et al (1994) argued that the ability to penetrate a product's lipid wax or cuticle will enhance the difficulty to wash off the pesticide. Therefore, it is expected that pesticides with high K_{ow} values, also defined as 'hydrophobic', will have lower observed reduction values after washing or soaking. Studies in this review acknowledge this assumption in general, however it should be noted that the period between pesticide application and the washing process plays an equally important role (Holland et al., 1994; Keikotlhaile et al., 2010). Chlorpyrifos, a hydrophobic pesticide (K_{ow} = log 4,7), was reduced 35% less efficiently from tomatoes 10 days after pesticide application (Rani et al., 2013). However, on lettuce samples no significant decreased in removal was noticed for Chlorpyrifos after a longer waiting period (Akoto et al., 2016).

A solution for pesticides with high octanol-water coefficients could be by adding certain detergents for soaking/washing processes. Increasing the salinity of water will decrease the K_{ow} of pesticides and make them more water soluble (Saranjampour et al., 2017). Unfortunately, only few studies in this review investigated the effects of a salt solution on the reduction of pesticides. Soaking lettuce in a salt solution showed to be far more efficient in removing Chlorpyrifos in comparison to washing with tap water, with mean reductions ranging between approx. 26 – 39% and 9 – 14%, respectively (Akoto et al., 2016). For spinach samples, similar reduction results were found for Chlorpyrifos and Cypermethrin (approx. 26 and 23% reduction, respectively) (Amir et al., 2019).

4.2 - Systemic vs non-systemic

Besides hydrophobicity, the mode of action of a pesticide determines the location of a pesticide in fruits or vegetables. Pesticides can be categorized in two groups based on their mode of action: systemic and non-systemic (Hrynko et al., 2023). Systemic pesticides may be taken up by the plant and reside deeper into the plant or product, whereas non-systemic pesticides remain on the surface of the plant or product (Hou et al., 2016; Łozowicka et al., 2020). In general, it is assumed that systemic pesticides are less exposed and, therefore, less susceptible to surface reduction practices. However, caution is advised with over generalizing this assumption. Łozowicka et al (2020) calculated penetration factors for multiple pesticides, whereby some non-systematic pesticides showed better penetration capabilities than systematic ones.

As previously mentioned, the period between pesticide application and reduction process plays an important role for the effectiveness of removal strategies for both systemic and non-systemic pesticides. Imidacloprid (systemic pesticide) concentration was reduced by approx. 30% one hour after pesticide application, whereas only approx. 9% reduction was measured 14 days after the pesticide application (Hendawi et al., 2013).

4.3 - Product characteristics

Besides the different physio-chemical properties of the pesticide, the characteristics of fruits and vegetables matter. The major characteristics that influence the removal efficiency of pesticide are a) the surface area and the type of wax-layer of the fruits/vegetable skin c) the general cleaning practices per product.

Firstly, Yang et al (2022) showed that reduction in lettuce was more efficient (67.4%) than in spinach samples (55.1%) for the same pesticides. A previous study found a similar removal efficiency differences between the leafy vegetables kumquat (25%) and spinach (11%) (Wu et al., 2019). They hypothesize that leafy vegetables with a larger surface area (e.g. lettuce) have more contact points/area to the washing solution and therefore show higher reduction percentages.

Secondly, fruits and vegetables contain a protective lipid-dense layer around the inner parts called a cuticle or wax-layer. Dependent on the hydrophobicity of a pesticide, this determines the ability to bind/penetrate into the wax layer and decrease the removal efficiency of washing and thermal processes. A comparison study between types of lettuce showed that Mancozeb, a water-soluble fungicide (K_{ow} = 2.3), was easier removed from lettuce with more waxy surfaces (López-Fernández et al., 2013). Other studies comparisons confirm this reasoning, for example the washing efficiency (same duration) of Pyraclostrobin (K_{ow} = 4.0, which is defined as hydrophobic) for tomatoes and spinach were approx. 10-29% and 91.5%, respectively (Jankowska et al., 2016; S. J. Yang et al., 2022). Fruits like tomatoes or apples have a thick wax layer or cuticle to protect their inner tissues, which is in general is thicker than the protective layer around leaves (Liu et al., 2014; Schönherr, 2006).

Finally, studies mention washing and soaking steps above five minutes for certain fruits and vegetables. While individual behaviour can vary substantially, the likelihood of people washing products (e.g. tomatoes) for such a duration on a daily basis should be investigated. Another example of a study that likely investigates ‘unrealistic’ washing periods is the study of Wasilewski et al (2021), where apples are soaked for 30 minutes. While their results are interesting, the likelihood that individuals will undertake such long washing steps in daily life seem unlikely (based on the authors personal experience and rationale) and are worth taking into account in future research.

4.4 - Degradation mechanisms: Thermal processes & solvents

While thermal processes showed good results and washing/soaking with additives showed to often have higher degradation levels of pesticides than with tap water, harmful metabolites/by-products could be formed during the processes. Increased temperature and the presence of additives such like organic acids (increase pH) or alkaline solutions (decreased pH) can increase hydrolysis reactions or pesticide degradation (Amir et al., 2019; Holland et al., 1994; Keikotlhaile et al., 2010; Polat & Tiryaki, 2020). It is important to understand which mechanisms are involved between solvents and the pesticide or whether the solvent increases the solvability of the pesticide without creating metabolites.

A metabolite of Chlorpyrifos, 3,5,6-trichloro-2-pyridinol, was found to be more toxic than the pesticide itself, while also synergistically interacting with Chlorpyrifos (Ling et al., 2011). Another examples shows that hydrolysis of Imidacloprid results in desnitro-imidacloprid, which is also deemed more toxic for mammals than its predecessor (Klarich Wong et al., 2019). Several other examples mention the formation of (more toxic) metabolites due to degradation reactions (hydrolysis, chlorination, etc.) of pesticides (Chen et al., 2021; Rutkowska et al., 2023).

The question arises whether the observed high pesticide removal results via these degradation reactions outweighs the formation of these metabolites or if less reactive processes such as washing with tap water or peeling of the skin (removal instead of degradation) should be preferred.

Studies that investigate the risk of exposure to pesticides should start measuring the possible metabolite concentrations before and after the reduction processes. There are studies that already include metabolite measurements, which could provide information on the processing factors for the pesticide of interest, while also determining the mean reduction/formation behaviour of the new metabolite/product/pesticide combination Rutkowska et al (2023). Needless to say, such studies would need beforehand knowledge on the possible metabolite formation.

5 - Comparability studies

In Table S1 all study results included in this systematic review are displayed and compared to one another when similar PPP combinations were investigated. Pesticide measurements are subject to multiple factors that can differ per study protocol. This could influence the observed outcome and thereby hamper comparability between studies. A general stepwise experimental protocol can be observed in the type of studies mentioned in this review; 1) obtaining food samples, 2) application of pesticides, 3) processing of sample and 4) analysis of pesticide residues. The latter step can be further divided into four major steps: (4.a) preparing the sample, (4.b) liquid extraction of analytical portion, (4.c) extract purification and (4.d) extract quantification, as described by da Silva & Camões (2010). In this section, possible factors in the overall study protocol that is characterized by steps 1 to 3 and the factors that are of interest in the analysis of pesticide residues (characterized by steps 4a to 4d) will be further discussed.

5.1 - Study protocol

The majority of the studies mentioned the size and number of samples that are included to determine a mean concentration, while some studies solely mentioned the time between pesticide application and reduction processing step (Table S1). Comparability of study results becomes difficult when it is unclear for how long pesticides have had the time to penetrate the products wax layer, which is proven to influence the reduction efficiency, as previously discussed in this paper (Akoto et al., 2016; Hendawi et al., 2013). Sample size was often referred to as the mass of sample that was used or the number of gas/liquid chromatography (GC or LC) measurements that were performed. The latter provides an indication on how representable and trustworthy the mean and standard deviation (SD) values are, while only mentioning the sample mass does give little information by itself.

Other factors such as the dosage of pesticide application could affect the relative reduction of processing steps. Hypothetically, the increase of pesticide dose on a product could influence the number of measurements that fall below the limit of quantification (LOQ), thereby decreasing the measurement certainty because it is not clear how far the concentrations lies below the LOQ. Multiple studies mentioned that they apply a dose that is double the amount of the recommended government thresholds (Ahammed Shabeer et al., 2023; Hrynko et al., 2023; Lozowicka, Jankowska, & Rutkowska, 2016; Rani et al., 2013). Interestingly, Rani et al (2013) showed that reduction of single and double dose applied Chlorpyrifos followed almost identical reduction percentages after washing procedures which argues that the effect of pesticide dosage is of limited importance. However, Chlorpyrifos is a non-systemic pesticide and is not taken up by the plant. If a systemic pesticide, such as Difenoconazole, would be sprayed/applied to a field with double or triple dosage, the extra surplus could be taken up or relocate from the stem or roots to the inside tissue of the fruit or (vegetable) leaf (Chen et al., 2021; T. Yang et al., 2016).

5.2 - Analysis uncertainties

Currently, the standard method to extract pesticide residues from fruit and vegetables for sample analysis is the QuEChERS method (Lehotay, 2011). This method was developed by Anastassiades et al (2003) and has shown to produce high recovery rates for pesticides, while also being more cost- and time efficient than previous extraction techniques (Kim et al., 2019). The QuEChERS is often used together with gas or liquid tandem-mass chromatography (GC- or LC- MS/MS) which allows for the measurement of multiple residues in one sample (Kim et al., 2019). However, within the QuEChERS method there can be a variety in types of extraction solvents (Acetonitrile (MeCN) is the most commonly used solvent), centrifugation settings or further clean-up steps based on the pesticide and matrices that are investigated with the technique (Lehotay, 2011). In certain situations, additional clean-up is needed to increase the selectivity and sensitivity during the LC-MS/MS or GC-MS/MS analysis, for example when complex fruits/matrixes contain a high number of co-expressing compounds (Rutkowska & Kaczy, 2019).

A returning challenge in GC (or LC)-MS/MS is the occurring of matrix effects, that influence the quantification and reliability of residue measurements. Matrix effect (ME) are caused by compounds that block active sites and cause less analytes to be measured, thereby affecting the signal of the GC or LC, as described by Rutkowska & Kaczy (2019). ME are affected by a) the chemical characteristics of the analytes (pesticide) such as pH-sensitivity, b) compounds present in the matrix which can be a multitude of molecules in the homogenized fruits or vegetables, c) The number of active sites present in the GC instrument and d) the concentration of the pesticide itself. These factors would argue in favour of applying a higher dose of pesticides, since an initial high concentration of pesticide in the sample would allow for a dilution of the sample to decrease the amount of interfering compounds (Zhou et al., 2017).

Matrix effects are most often measured with the help of calibration curves, whereby unexposed matrices are injected into the GC or LC column (SANTE, 2019). According to the guidelines of the European Commission, matrix effects should remain within the range of -20% – 20% to keep the analysis acceptable. Moreover, other parameters concerning the sensitivity, recovery and precision should be met before observed pesticide concentrations can be deemed true. To test the recovery and precision of the samples and study set-up, a recovery experiment is conducted beforehand of the real samples in which spike concentration of the analyte of interest are measured. The mean recovery value should be within the range of 70 to 120%. The experiment should be repeated at least 3 times to test the precision of the set-up, where the RSD should be lower or equal to 20% (SANTE, 2019) .

The majority of studies discuss and prove that they meet these criteria by conducting evaluation studies, however there are individual pesticides where criteria were barely met (e.g. RSD = 0.9 - 19.4%, with mean recoveries between 70% - 115%) (Lozowicka, Rutkowska, Jankowska, et al., 2016). While these cases meet the guidelines set by the European Committee (SANTE, 2019), there remains some uncertainty in the measured concentration that can influence the Processing Factors and make study comparability more difficult.

Finally, interpretation of residue concentrations that fall under the limit of detection (LOD) or limit of quantification (LOQ) differs between studies. The majority of studies denote such concentrations as '< LOQ ' without calculating a PF or assume that the residue is the closest concentration under the LOD or LOQ (Ajeep et al., 2021; Kowalska et al., 2022). In contrast, a single study assumed that the concentration would be equal to zero and reported reduction percentages accordingly (A. Yang et al., 2012). Calculating with assumed/predicted residue concentrations will have significant implications on the PF and should be critically interpreted by the authors and reviewer before it can be compared to other studies.

6 - Conclusion

This systematic review aimed to establish a clear overview of the effectiveness of different household processing factors in reducing pesticide concentrations on fruits and vegetables. Moreover, it discussed the multiple factors and determinants that play a role in pesticide reduction effectiveness. Overall, peeling and boiling/blanching showed to be effective reduction strategies in most of the PPP combinations that were found in de studies included in the review. Washing and soaking processes showed less consistent reduction results, which could be mainly explained by the type of solvent, hydrophobicity and mode of action of the pesticide and the duration of the process. Besides these factors, the degradation point of pesticides showed to influence the effectiveness of boiling processes and in some instances resulted in an increased pesticide concentration.

A often overlooked factor was the duration time between pesticide application and processing moment. Additionally, only few studies included the formation of metabolites out of pesticide degradation which is crucially important to fully comprehend the risk reduction/increase of the observed processing factors.

Noteworthy were the lack of studies, with similar protocols, which made it difficult to interpret and compare processing factors for all different process/product/pesticide combinations, which underlines the need for further research on household processing factors for pesticides.

7 - References

Abdullah, Randhawa, A., Akhtar, S., Asghar, A., Sohaib, M., Muhammad, R., & Ahmar, M. (2016). Assessment of different washing treatments to mitigate imidacloprid and acetamaprid residues in spinach. *Journal of Food Science Agriculture*, *96*(December 2015), 3749–3754. https://doi.org/10.1002/jsfa.7563

Ahammed Shabeer, T. P., Somkuwar, R., Sharma, A. K., Deshmukh, U., & Hingmire, S. (2023). Multi-residue method validation, processing factor and monitoring of thirteen targeted fungicide residues in the process of wine making. *Journal of Food Composition and Analysis*, *115*(September 2022). https://doi.org/10.1016/j.jfca.2022.104912

Ajeep, L., Alnaser, Z., & Tahla, M. K. (2021). Effect of household processing on removal of multi-classes of pesticides from tomatoes. *Journal of Microbiology, Biotechnology and Food Sciences*, *10*(5), 1–8. https://doi.org/10.15414/jmbfs.2015

Akoto, O., Addai-mensah, F., & Abavare, E. K. K. (2016). Effects of per-household processes on the levels of chlorpyrifos residues in lettuce (Lactuca sativa). *International Journal of Food Contamination*, 2–7. https://doi.org/10.1186/s40550-016-0037-3

Amir, R. M., Randhawa, M. A., Nadeem, M., Ahmed, A., Ahmad, A., Khan, M. R., Khan, M. A., & Kausar, R. (2019). Assessing and Reporting Household Chemicals as a Novel Tool to Mitigate Pesticide Residues in Spinach (Spinacia oleracea). *Scientific Reports*, *9*(1), 1–6. https://doi.org/10.1038/s41598-018-37936-2

Anastassiades, M., Lehotay, S. J., Stajnbaher, D., & Schenck, F. J. (2003). Fast and easy multiresidue method employing acetonitrile extraction/partitioning and "dispersive solid-phase extraction" for the determination of pesticide residues in produce. *Journal of AOAC International*, *86*(2), 412–431.

Andrade, G. C. R. M., Francisco, J. G., Rocha, A. A., & Tornisielo, V. L. (2015). Effects of Types of Washing and Peeling in Relation to Pesticide Residues in Tomatoes. *Journal of the Brazilian Chemical Society*, *26*(10), 1994–2002. https://doi.org/http://dx.doi.org/10.5935/0103-5053.20150179 J.

Bonnechère, A., Hanot, V., Jolie, R., Hendrickx, M., Bragard, C., Bedoret, T., & Loco, J. Van. (2012). Effect of household and industrial processing on levels of five pesticide residues and two degradation products in spinach. *Food Control*, *25*(1), 397–406. https://doi.org/10.1016/j.foodcont.2011.11.010

Carrasco Cabrera, L., & Medina Pastor, P. (2021). The 2019 European Union report on pesticide residues in food. In *EFSA Journal* (Vol. 19, Issue 4). https://doi.org/https://doi.org/10.2903/j.efsa.2021.6491

Chen, S., Cai, L., Zhang, H., Zhang, Q., Song, J., Zhang, Z., Deng, Y., Liu, Y., Wang, X., & Fang, H. (2021). Deposition distribution, metabolism characteristics, and reduced application dose of difenoconazole in the open field and greenhouse pepper ecosystem. In *Agriculture, Ecosystems and Environment* (Vol. 313). https://doi.org/10.1016/j.agee.2021.107370

Chung, S. W. C. (2018). How effective are common household preparations on removing pesticide residues from fruit and vegetables ? A review. *Journal of Science, Food and Agriculture*, *98*, 2857–2870. https://doi.org/10.1002/jsfa.8821

Cumming, H., & Rücker, C. (2017). Octanol-Water Partition Coefficient Measurement by a Simple 1H NMR Method. *ACS Omega*, *2*(9), 6244–6249. https://doi.org/10.1021/acsomega.7b01102

da Silva, R. J. N. B., & Camões, M. F. G. F. C. (2010). Comparability of measurement results for pesticide residues in foodstuffs: An open issue? *Accreditation and Quality Assurance*, *15*(12), 691–704. https://doi.org/10.1007/s00769-010-0725-2

El-Sayed, E., Hassan, H., El-Raouf, A. A., & Salman, S. N. (2021). Investigation of the effects of household processing on the reduction rate of chlorpyrifos, metalaxyl and diazinon residues in orange fruit. *Hellenic Plant Protection Journal*, *14*(2), 65–76. https://doi.org/10.2478/hppj-2021-0007

Hendawi, M. Y., Romeh, A. A., & Mekky, T. M. (2013). Effect of Food Processing on Residue of Imidacloprid in Strawberry Fruits. *Journal of Agricultural and Science Technology*, *15*, 951–959.

Holland, P. T., Hamilton, D., Ohlin, B., & Skidmore, M. W. (1994). Effects of storage and processing on pesticide residues in plant products (Technical Report). *Pure and Applied Chemistry*, *66*(2), 335–356.

Hou, R., Zhang, Z., Pang, S., Yang, T., Clark, J. M., & He, L. (2016). Alteration of the Nonsystemic Behavior of the Pesticide Ferbam on Tea Leaves by Engineered Gold Nanoparticles. *Environmental Science and Technology*, *50*(12), 6216–6223. https://doi.org/10.1021/acs.est.6b01336

Hrynko, I., Kaczyński, P., Pietruszyńska, M., & Łozowicka, B. (2023). The effect of food thermal processes on the residue concentration of systemic and non-systemic pesticides in apples. *Food Control*, *143*(March 2022). https://doi.org/10.1016/j.foodcont.2022.109267

Hussnain, A., Amir, R. M., Khan, M. A., Ahmad, A., Ali, S. W., Nadeem, M., Ameer, K., Khan, M. A., Mahmood, S., & Hayat, I. (2021). Mitigating the impact of organochlorine and pyrethroid residues in fresh and chemically washed spinach. *Food Science and Technology (Brazil)*, *41*(1), 59–64. https://doi.org/10.1590/fst.37019

Jankowska, M., Kaczynski, P., Hrynko, I., & Lozowicka, B. (2016). Dissipation of six fungicides in greenhouse-grown tomatoes with processing and health risk. *Environmental Science and Pollution Research*, *23*, 11885–11900. https://doi.org/10.1007/s11356-016-6260-x

Jankowska, M., Łozowicka, B., & Kaczyński, P. (2019). Comprehensive toxicological study over 160 processing factors of pesticides in selected fruit and vegetables after water, mechanical and thermal processing treatments and their application to human health risk assessment. *Science of the Total Environment*, *652*, 1156–1167. https://doi.org/10.1016/j.scitotenv.2018.10.324

Keikotlhaile, B. M., Spanoghe, P., & Steurbaut, W. (2010). Effects of food processing on pesticide residues in fruits and vegetables: A meta-analysis approach. *Food and Chemical Toxicology*, *48*(1), 1–6. https://doi.org/10.1016/j.fct.2009.10.031

Kim, L., Lee, D., Cho, H., & Choi, S. (2019). Review of the QuEChERS method for the analysis of organic pollutants : Persistent organic pollutants , polycyclic aromatic hydrocarbons , and pharmaceuticals. *Trends in Environmental Analytical Chemistry*, *22*(e00063). https://doi.org/https://doi.org/10.1016/j.teac.2019.e00063

Klarich Wong, K. L., Webb, D. T., Nagorzanski, M. R., Kolpin, D. W., Hladik, M. L., Cwiertny, D. M., & Lefevre, G. H. (2019). Chlorinated Byproducts of Neonicotinoids and Their Metabolites: An Unrecognized Human Exposure Potential? *Environmental Science and Technology Letters*, *6*(2), 98–105. https://doi.org/10.1021/acs.estlett.8b00706

Kowalska, G., Pankiewicz, U., & Kowalski, R. (2022). Assessment of Pesticide Content in Apples and Selected Citrus Fruits Subjected to Simple Culinary Processing. *Applied Sciences (Switzerland)*, *12*(3). https://doi.org/10.3390/app12031417

Lehotay, S. J. (2011). Chapter 4 QuEChERS Sample Preparation Approach for Mass Spectrometric Analysis of Pesticide Residues in Foods. In Z. Jerry (Ed.), *Mass spectrometry in food safety - Methods and protocols* (Vol. 747, Issue 1, pp. 65–82). https://doi.org/10.1007/978-1-61779-136-9

Ling, Y., Wang, H., Yong, W., Zhang, F., Sun, L., Yang, M. L., Wu, Y. N., & Chu, X. G. (2011). The effects of washing and cooking on chlorpyrifos and its toxic metabolites in vegetables. *Food Control*, *22*(1), 54–58. https://doi.org/10.1016/j.foodcont.2010.06.009

Liu, N., Dong, F., Liu, X., Xu, J., Li, Y., Han, Y., Zhu, Y., Cheng, Y., Chen, Z., Tao, Y., & Zheng, Y. (2014). Effect of household canning on the distribution and reduction of thiophanate-methyl and its metabolite carbendazim residues in tomato. *Food Control*, *43*, 115–120.

https://doi.org/10.1016/j.foodcont.2014.03.003

López-Fernández, O., Rial-Otero, R., & Simal-Gándara, J. (2013). Factors governing the removal of mancozeb residues from lettuces with washing solutions. *Food Control*, *34*(2), 530–538. https://doi.org/10.1016/j.foodcont.2013.05.022

Lozowicka, B., Jankowska, M., Hrynko, I., & Kaczynski, P. (2016). Removal of 16 pesticide residues from strawberries by washing with tap and ozone water , ultrasonic cleaning and boiling. *Environmental Monitoring and Assessment*, *188*(51). https://doi.org/10.1007/s10661-015-4850-6

Lozowicka, B., Jankowska, M., & Rutkowska, E. (2016). Investigations on fungicide removal from broccoli by various processing methods. *Desalination and Water Treatment*, *3994*, 1–9. https://doi.org/10.1080/19443994.2014.988408

Łozowicka, B., Kaczyński, P., Mojsak, P., Rusiłowska, J., Beknazarova, Z., Ilyasova, G., & Absatarova, D. (2020). Systemic and non-systemic pesticides in apples from Kazakhstan and their impact on human health. *Journal of Food Composition and Analysis*, *90*(April). https://doi.org/10.1016/j.jfca.2020.103494

Lozowicka, B., Rutkowska, E., Jankowska, M., & Hrynko, I. (2016). Toxicological evaluation of multi-class pesticide residues in vegetables and associated human health risk study for adults and children. *Human and Ecological Risk Assessment*, *22*(7), 1480–1505. https://doi.org/10.1080/10807039.2016.1185690

Nicolopoulou-stamati, P., Maipas, S., Kotampasi, C., Stamatis, P., & Hens, L. (2016). Chemical Pesticides and Human Health : The Urgent Need for a New Concept in Agriculture. *Frontiers in Public Health*, *4*(July), 1–8. https://doi.org/10.3389/fpubh.2016.00148

Pirsaheb, M., Rezaei, M., & Sharafi, K. (2016). Evaluating the effect of peeling, washing and storing in the refrigerator processes on reducing the Diazinon, Chlorpyrifos and Abamectin pesticide residue in apple. *International Journal of Pharmacy & Technology*, *8*(2), 12858–12873. https://doi.org/ISSN: 0975-766X

Polat, B., & Tiryaki, O. (2020). Assessing washing methods for reduction of pesticide residues in Capia pepper with LC-MS/MS. In *Journal of Environmental Science and Health - Part B Pesticides, Food Contaminants, and Agricultural Wastes* (Vol. 55, Issue 1, pp. 1–10). https://doi.org/10.1080/03601234.2019.1660563

Rani, M., Saini, S., & Kumari, B. (2013). Persistence and effect of processing on chlorpyriphos residues in tomato (Lycopersicon esculantum Mill .). *Ecotoxicology and Environmental Safety*, *95*, 247–252. https://doi.org/10.1016/j.ecoenv.2013.04.028

Rodrigues, A. A. Z., Queiroz, M. E. L. R. De, Fernando, A., Oliveira, D., Neves, A. A., Heleno, F. F., Zambolim, L., Freitas, J. F., Morais, E. H. C., Oliveira, D., Neves, A. A., Heleno, F. F., Zambolim, L., Freitas, J. F., & Heleno, F. F. (2017). Pesticide residue removal in classic domestic processing of tomato and its effects on product quality. *Journal of Environmental Science and Health, Part B*, *52*(12), 850–857. https://doi.org/10.1080/03601234.2017.1359049

Rutkowska, E., & Kaczy, P. (2019). Three approaches to minimize matrix e ff ects in residue analysis of multiclass pesticides in dried complex matrices using gas chromatography tandem mass spectrometry. *Food Chemistry*, *279*(November 2018), 20–29. https://doi.org/10.1016/j.foodchem.2018.11.130

Rutkowska, E., Wołejko, E., Kaczyński, P., Łuniewski, S., & Łozowicka, B. (2023). High and low temperature processing: Effective tool reducing pesticides in/on apple used in a risk assessment of dietary intake protocol. In *Chemosphere* (Vol. 313). https://doi.org/10.1016/j.chemosphere.2022.137498

SANTE. (2019). *ANALYTICAL QUALITY CONTROL AND METHOD VALIDATION PROCEDURES FOR PESTICIDE RESIDUES ANALYSIS IN FOOD AND FEED - European Commission Document No SANTE 12682/2019*.

Saranjampour, P., Vebrosky, E. N., & Armbrust, K. L. (2017). Salinity impacts on water solubility and n-octanol/water partition coefficients of selected pesticides and oil constituents. *Environmental Toxicology and Chemistry*, *36*(9), 2274–2280. https://doi.org/10.1002/etc.3784

Schönherr, J. (2006). Characterization of aqueous pores in plant cuticles and permeation of ionic solutes. *Journal of Experimental Botany*, *57*(11), 2471–2491. https://doi.org/10.1093/jxb/erj217

Schwab, S., Janiaud, P., Dayan, M., Amrhein, V., Panczak, R., Palagi, P. M., Hemkens, L. G., Ramon, M., Rothen, N., Senn, S., Furrer, E., & Held, L. (2022). Ten simple rules for good research practice. *PLoS Computational Biology*, *18*(6), 1–14. https://doi.org/10.1371/journal.pcbi.1010139

Skovgaard, M., Encinas, S. R., Chresten, O., Andersen, J. H., Condarco, G., & Jørs, E. (2017). *Pesticide Residues in Commercial Lettuce , Onion , and Potato Samples From Bolivia — A Threat to Public Health ?* 18–22. https://doi.org/10.1177/1178630217704194

Słowik-Borowiec, M., & Szpyrka, E. (2020). Selected food processing techniques as a factor for pesticide residue removal in apple fruit. *Environmental Science and Pollution Research*, *27*(2), 2361–2373. https://doi.org/10.1007/s11356-019-06943-9

Voedingscentrum Nederland. (2022). *Groente Voedingscentrum*. https://www.voedingscentrum.nl/encyclopedie/groente.aspx

Wanwimolruk, S., Duangsuwan, W., Phopin, K., & Boonpangrak, S. (2017). Food safety in Thailand 5 : the effect of washing pesticide residues found in cabbages and tomatoes. *Journal of Consumer Protection and Food Safety*, *12*, 209–221. https://doi.org/10.1007/s00003-017-1116-y

Wasilewski, T., Hordyjewicz-Baran, Z., Zarębska, M., Zajszły-Turko, E., Zimoch, J., Kanios, A., & De Barros Sanches, M. (2022). Effect of Talc Particle Size in Detergents for Fruits and Vegetables on the Ability to Remove Pesticide Residues. *ACS Omega*, *7*(29), 25046–25054. https://doi.org/10.1021/acsomega.2c01029

World Health Organisation. (2020). *Healthy diet [Fact Sheet]*. https://www.who.int/news-room/fact-sheets/detail/healthy-diet

Wu, Y., An, Q., Li, D., Wu, J., & Pan, C. (2019). Comparison of Different Home / Commercial Washing Strategies for Ten Typical Pesticide Residue Removal Effects in Kumquat , Spinach and Cucumber. *International Journal of Environmental Research and Public Health*, *16*(472), 1–20. https://doi.org/10.3390/ijerph16030472

Xu, M., Gao, Y., Han, X. X., & Zhao, B. (2017). Detection of Pesticide Residues in Food Using Surface-Enhanced Raman Spectroscopy: A Review. *Journal of Agricultural and Food Chemistry*, *65*, 6719–6726. https://doi.org/10.1021/acs.jafc.7b02504

Yang, A., Park, J., El-aty, A. M. A., Choi, J., Oh, J., Do, J., Kwon, K., Shim, K., Choi, O., & Shim, J. (2012). Synergistic effect of washing and cooking on the removal of multi-classes of pesticides from various food samples. *Food Control*, *28*(1), 99–105. https://doi.org/10.1016/j.foodcont.2012.04.018

Yang, S. J., Mun, S., Kim, H. J., Han, S. J., Kim, D. W., Cho, B. S., Kim, A. G., & Park, D. W. (2022). Effectiveness of Different Washing Strategies on Pesticide Residue Removal: The First

Comparative Study on Leafy Vegetables. *Foods*, *11*(18). https://doi.org/10.3390/foods11182916

Yang, T., Zhao, B., Hou, R., Zhang, Z., Kinchla, A. J., Clark, J. M., & He, L. (2016). Evaluation of the Penetration of Multiple Classes of Pesticides in Fresh Produce Using Surface-Enhanced Raman Scattering Mapping. *Journal of Food Science*, *81*(11). https://doi.org/10.1111/1750-3841.13520

Yang, T., Zhao, B., Kinchla, A. J., Clark, J. M., & He, L. (2017). Effectiveness of Commercial and Homemade Washing Agents in Removing Pesticide Residues on and in Apples. *Journal of Agricultural and Food Chemistry*, *65*, 9744–9752. https://doi.org/10.1021/acs.jafc.7b03118

Zhou, W., Yang, S., & Wang, P. G. (2017). Matrix effects and application of matrix effect factor. *Bioanalysis*, *9*(23), 1839–1844. https://doi.org/10.4155/bio-2017-0214 C?