Risk ranking of residual contaminants in vegetable and fruits, on the basis of human health risks, for the purpose of risk-based monitoring

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Preface

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Abstract

Introduction Chemical plant protection products are widely used in the agricultural sector to prevent or destroy the growth of undesired plants, help control weed and harmful insects as well as plant diseases. Pesticides can be toxic to a range of other organisms as well, including animals and humans. Humans can be exposed to pesticides via consumption of crops that have been treated, or indirectly, via water or soil. Several toxicological and epidemiological studies have identified potential risks of pesticides to human health. The application of pesticides should follow good agricultural practices. In order to protect human health, limits for their maximal presence on food products have been set, and pesticides are regularly monitored to check whether they comply with these limits. However, due to limited resources not all pesticides can be included in the monitoring program.

Aim The aim of this study is to estimate the risk related to pesticides, based on their human health impacts, such to underpin the current national monitoring program.

Methods Risk rankings were performed using the risk ratio method for four horticultural products, being apples, strawberries, tomatoes and grapes. Monitoring data from both the Netherlands (period from 2004 to 2014) and the European Union (period 2013) were used, such to evaluate potential differences in the presence of the pesticide residues. The endpoint for occurrence was the percentage of pesticides which had a concentration similar to, or above the maximum residue limit (MRL). The endpoint for severity consisted of human health effects based on acceptable daily intakes (ADIs) and acute reference doses (ARfDs), with ADIs examining the chronic health effect and ARfDs the acute health effect. MRL exceedance rates were calculated per country of origin as well using Dutch monitoring data (KAP data).

Results For apples, 28 out of 73 pesticides exceeded the MRL, with diazinon, dimethoate, azinphos-methyl, carbaryl and tolylfluanid scoring the highest for chronic risk. For the risk ranking based on acute risk, 20 out of 52 pesticides exceeded the MRL, with azinphos-methyl, carbaryl, carbendazim, dimethoate and tolylfluanid having the highest scores. Pesticides currently not included in the national monitoring program with high scores in the EU risk ranking performed in this study, were flusilazole and fenvalerate. For strawberries, dichlorvos, methomyl, procymidone, oxamyl and flutolanil scored highest based on the risk ranking examining chronic risk. In total, 35 out of 95 pesticides exceeded the MRL. In the risk ranking based on acute risk, 29 out of 71 pesticides exceeded the MRL, with methomyl, oxamyl, procymidone, dichlorvos and monocrotophos scoring highest. The pesticides ethion, formetanate and fenthion scored high in the EU risk ranking (based on chronic and acute risks) and are currently not part of the national monitoring program. For tomatoes, 92 pesticides were included in the risk ranking based on chronic risk, of which 55 pesticides exceeded

the MRL. The pesticides which had the highest scores were chlorpyrifos, procymidone, oxamyl, chlorothalonil and buprofezin. Based on the risk ranking of acute risk, the pesticides oxamyl, chlorpyrifos, carbendazim, procymidone and methomyl scored highest. In total, 39 out of 63 pesticides exceeded the MRL. Pesticides scoring high in the EU risk ranking performed in this study, which are currently not part of the national monitoring program, were fenamiphos and ethephon. For the risk ranking of grapes, 125 pesticides were included in the risk ranking based on chronic risk, of which 58 exceeded the MRL. Carbofuran, procymidone, methomyl, flusilazole and omethoate were the five riskiest pesticides for grapes. For the risk ranking based on acute risk, 49 out of 91 pesticides exceeded the MRL with carbofuran methomyl, procymidone, flusilazole and triadimenol being the riskiest. All the pesticides scoring high in the EU risk ranking were also included in the national monitoring program. MRL exceedance rates for apples were the highest for pesticides from countries outside the EU, for the pesticides used on other crops also countries from within the EU had high MRL exceedance rates.

Conclusion The current national monitoring program of pesticide residues on vegetable and fruit crops is still relevant for the most part. However, some adjustments can be made to include pesticides with a high score (the riskiest pesticides) and exclude pesticides with a low score.

List of abbreviations

ADI	Acceptable Daily Intake
ARfD	Acute Reference Dose
EC	European Commission
EU	European Union
EFSA	European Food Safety Authority
GAP	Good Agricultural Practices
LOD	Limit of Detection
LOQ	Limit of Quantification
MRL	Maximum Residue Limit
NOAEL	No Observed Adverse Effect Limit
NVWA	Nederlandse Voedsel en Waren Autoriteit (Dutch Food Safety Authority)
PPP	Plant Protection Product
PPPAMS	Plant Protection Products Application Management System
RASFF	Rapid Alert System for Food and Feed
RIVM	Rijksinstituut voor Volksgezondheid en Milieu (National Institute for Public Health and the Environment)
RMS	Rapporteur Member State
zRMS	zonal Rapporteur Member State

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1 Introduction

Chemical plant protection products (PPPs) are widely used in the agricultural sector, amongst others, to prevent or destroy the growth of undesired plants or parts of plants, and thereby increasing crop yield (Fantke, Friedrich and Jolliet, 2012; EC, 2015a). They help to control hundreds of weed species, more than one million species of harmful insects and roughly 1,500 plant diseases (Karabelas, Plakas, Solomou, Drossou and Sarigiannis, 2009). PPPs also minimize vector-borne diseases by killing the vector, which otherwise might have spread the disease (Aktar, Sengupta and Chowdhury, 2009).

In 1939, the first PPP that was placed on the market was dichlorodiphenyltrichloroethane (DDT). Soon after, in the 1940s, manufacturers started producing other synthetic PPPs as well, for example, the organophosphates which - despite their toxicity - gained great popularity because of their broad spectrum efficacy and low costs (Karabelas et al. 2009). At present, pesticide use has increased 50-fold since the 1950s and there are thousands of synthetic pesticide products made up of more than 1,000 different chemicals or combinations thereof (Karabelas et al. 2009). Worldwide approximately 2.5 million tons of active ingredients[1] are used with expenditures adding up to between 25 billion and 52.4 billion euros (Fantke et al. 2012; Karabelas et al. 2009). Of these, insecticides make up almost 50 percent of expenditures, and fungicides and herbicides approximately 25 percent each (Worm & Vaupel, 2008).

According to the European Commission (EC), a pesticide can be defined as "something that prevents, destroys, or controls a harmful organism ('pest') or disease, or protects plants or plant products during production, storage and transport". It is an overarching term which includes, amongst others: fungicides, herbicides, growth regulators, insecticides, rodenticides and biocides. PPPs are 'pesticides' that protect plants and crops (EC, 2015a). The major groups of pesticides are (Kumari, Madan, Kumar and Kathpal, 2001; Green Peace, 2015):

- Organochlorines Some pesticides within this class have been banned or severely restricted, because of their toxicity in non-target organisms including human. *Examples:* carbon tetrachloride, DDT, chlordane and heptachlor;

[1] An active substance is any chemical, plant extract, pheromone or micro-organism (including viruses), that has action against 'pests' or on plants, parts of plants or plant products (EC, 2015).

- Organophosphates This class includes a wide range of chemical structures. They act by inhibiting acetylcholinesterase in the nervous system (both central and peripheral). *Examples:* acephate, diazinon, dichlorvos and chlorpyrifos;
- Carbamates Neurotoxic acetylcholinesterase inhibitors. *Examples:* aldicarb, carbaryl methiocarb, mancozeb and maneb;
- Synthetic pyrethroids This class of pesticides interfere with cell signaling ion channels. *Examples:* cyhalothrin, cypermethrin, permethrin and deltamethrin;
- Neonicotinoids Newer class of pesticides. These substances are similar to nicotine. They act by blocking certain signalling pathways. *Examples:* clothianidin, imidacloprid and thiamethoxam;
- Chloroacetamides Causes developmental abnormalities. This pesticide class is no longer approved in the EU. *Examples:* alachlor and metolachlor;
- Paraquat Neurotoxic herbicide which inhibits photosynthesis. It is no longer approved in the EU;
- Glyphosate Active constituent of Roundup. It acts by inhibiting a particular enzyme in plants.

Pesticides can contaminate water, soil, and other vegetation. Besides killing insects or weeds, pesticides can be toxic to a range of other organisms including birds, non-target insects, fish and non-target plants (Aktar et al., 2009). Besides organisms, humans can also be exposed to pesticides via consumption of crops that have been treated or, indirectly, via soil or water (Figure 1.1). When pesticides are applied to crops, fractions may also be lost from the target and be absorbed by the soil, the air, or end up in ground water (Fantke et al. 2012). Pesticides like organochloride also have the potential of bioaccumulation in adipose tissue (Sasaki, Ishizaka, Suzuki, Takeda and Uchiyama, 1991).

Several toxicological and epidemiological studies have identified potential risks of pesticides on human health. Examples of ways in which human health can be affected by pesticides are cancer, neurodevelopmental disorders, genetic malformations and damage to the immune system (Karabelas, et al. 2009; Green Peace, 2015; Wilson & Tisdell, 2001). A study conducted by Fantke et al. (2012), estimating the overall quantifiable health impact of pesticides in Europe, found that pesticide residues on fruits and vegetables had a health impact of 113.36 and 1100.58 DALYs[2] per year, respectively.

In order to protect human health, residues on crops should be as low as possible. Therefore, maximum residue limits are set by the EC, which are mostly based on good agricultural practices. Crops are monitored for the presence of the residues by EC Member States. However, Member States have limited resources and therefore they cannot sample each batch and analyze every pesticide. The pesticides that should be given priority can change over time due to, for example, new research that has shown toxic effects of a pesticide, or changes in use (increase or decrease) of a particular pesticide. The aim of this study is to estimate the risk related to pesticides, based on their human health impact. The underlying aim is to evaluate whether the current monitoring program for pesticides is still relevant based on human health risks, and if pesticides currently not monitored in the Netherlands should be included, or vice versa (pesticides can be excluded from the program due to low health implications). The study focused on selected horticultural products, including apples, strawberries, tomatoes and grapes, in the Netherlands.

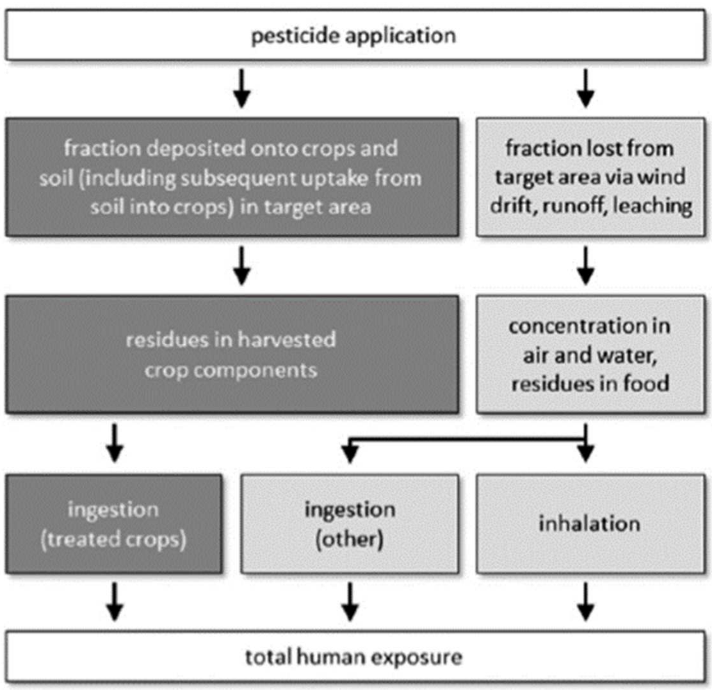

Figure 1.1. Environmental pathways of pesticides from application onto crops to exposure to humans (Fantke et al. 2012).

[2] DALY = disability adjusted life year

2 Background

The European Union (EU) has established a legislative framework defining rules for the use of plant protection products and pesticide residues in food, as well as rules for the approval of active substances (EFSA, 2015a). In Regulation EC 396/2005 rules are defined concerning MRLs in food and animal feed (EC, 2005). Since the 1st of September 2008, there is a new Regulation in place since the rules applicable before were complex and unclear. The new Regulation covers all agricultural products intended for food or feed and all the pesticides currently and formerly used on these crops, both inside and outside the EU (around 1100 pesticides). Furthermore, the roles of Member States, the Commission and EFSA are clearly outlined in the new Regulation as well (EC 2008a).

After applying pesticides on the crops, residues can remain on the crop, when used for feed or food purposes. The remaining residues have to be as low as possible in order to protect animal and human health. Therefore, maximum residue limits (MRLs) are set by the European Commission for food and animal feed. An MRL is the highest concentration of a pesticide residue that is legally tolerated in feed or food, based on the lowest consumer exposure necessary to protect vulnerable consumers, and based on good agricultural practice (GAP) (EFSA, 2016; EC, 2008a). The MRLs for all pesticides and all crops can be found in an online pesticide database established by the EC (EC, 2008a). For pesticides not explicitly mentioned, a default MRL of 0.01 mg/kg applies, which is equal to the limit of quantification (LOQ)[3].

2.1 Approval of active substances and setting of MRLs

Before new pesticides (i.e. which are not on the market yet) can be used, MRLs need to be defined. To this end, the applicant requesting approval has to submit scientific information about the active substance of the particular pesticide. Information about the minimum amounts of pesticide necessary to protect the crop and the amount of residue remaining after application have to be submitted to a Member State of choice; a so-called Rapporteur Member State (RMS). When the dossier is considered admissible, the RMS shall notify the applicant, the other Member States, the EC and EFSA. Thereafter, the RMS prepares a draft

[3] Limit of quantification is the lowest amount that can be reliably quantified with an acceptable level of precision and accuracy (EC, 2008b).

assessment report (DAR), which is sent to the EC and EFSA (EC, 2016c). EFSA will then give a peer review on the DAR by commenting on the report and, if necessary, perform an expert consultation. After EFSA circulated the conclusion about the DAR, the EC presents a review report and a draft regulation on the approval or non-approval of the active substance (EC, 2011). How and when a pesticide may be used after approval of the application within the EC, is up to the national authority, because of local differences between environmental conditions and the occurrence of plant pests. For crops grown outside the European Union, MRLs are set on request of the exporting countries (EC, 2008a).

2.2 Authorisation of a plant protection product

After an active substance is approved on a European level, PPPs based on these substances need to be authorised on a national level (CTBG, n.d.). Therefore, an application has to be made to the EC country where the PPP is expected to be placed on the market. This is done via the Plant Protection Products Application Management System (PPPAMS). The PPPAMS is a system developed by the EC to facilitate the authorisation for PPPs by creating applications and submitting these to Member States for evaluation. For evaluation of the application, a zonal rapporteur member state[4] (zRMS) is selected, which performs an assessment and makes a decision to authorize or to refuse the PPP. Both the assessment and the decision to authorize or refuse the PPP are also performed by the other the Member States within the same zone after the zRMS has commented and decided. If an applicant wants to place an already authorized PPP on the market of another Member State (where it has not been authorized yet), an application for mutual recognition is made. This application can only be used if it concerns the same PPP under the same agricultural circumstances (EC, 2016d).

2.3 Control and enforcement of MRLs

In Regulation EC 395/2005 it is stated that Member State authorities are responsible for control and enforcement of MRLs (EC, 2005). This Regulation outlines two control programs; one on a European level and one on a national level. The EU-coordinated control program

[4] zRMS is a rapporteur member state selected for each zone where the PPT will be authorised (EC, 2016d).

defines which and how many food products and pesticides should be monitored by the Member States. The EU-coordinated control program is established on a tri-annual basis and is revised every year (EFSA, 2014). The scope of the national control program is usually defined by the Member States themselves. In the Netherlands, the Dutch Food Safety Authority is responsible for these control programs. To ascertain that the monitoring is performed in a uniform and appropriate way, the EC has three instruments (EC, 2008a):

1) The coordinated EU multi-annual control program; Member States are obliged to share with EFSA and the other Member States the results of their official controls. The results of all the Member States will be published in an annual report compiled by EFSA.
2) Community Reference Laboratories; methods of analysis are developed to test the skills of the national control laboratories.
3) The Food and Veterinary Office; this office of the EC carries out inspections in the Member States to monitor their control activities.

3 Materials & methods

3.1 Materials

For this study, data were used on the occurrence of pesticides of the four selected food products in the Netherlands, and on their toxicological effects. Monitoring data from both the Netherlands and the European Union were used, such to evaluate potential differences in the presence of the pesticide residues. Data were collected from the national monitoring program of the Dutch Food Safety Authority and from the EFSA monitoring program. Dutch monitoring data from the period 2004-2014 were retrieved from KAP[5], a database in which these data are stored. The European monitoring data used in this study for strawberries, apples and tomatoes were obtained from an EFSA report on pesticide residues in food from 2015 (EFSA, 2015a). The European data for grapes were retrieved from an EFSA report from 2014 since these data were not covered in the report of 2015 (EFSA, 2014). European data covered data from all the Member States and Iceland and Norway (EFSA, 2014; EFSA, 2015a). Toxicological data (acceptable daily intakes[6] (ADIs) and acute reference doses[7] (ARfDs)) were obtained by the EFSA report of 2015 as well. The MRLs were downloaded from

[5] Quality Program for Agricultural Products
[6] Acceptable Daily Intake: the amount of a substance that can be ingested every day over an entire lifetime without any risk to human health.
[7] Acute Reference Dose: the amount of a substance that can be ingested on a single day without any health risk

the European Commission database (EC, 2015b); a public website where the most recent MRL data and additional pesticide information can be found. The MRLs for the crops included in this study, being apples, strawberries, tomatoes and grapes, were downloaded on the following dates respectively: February 9th, 2016, February 17th, 2016, February 10th, 2016 and February 19th, 2016. Monitoring data used in this study were compared with the most recent MRLs and do not necessarily correspond to the MRLs which were in place at the time of sampling. The crops included in this study were selected based on the number of pesticides used on the crops (EFSA, 2015a). Peppers had the largest number of pesticides used on the crop, with a total of 124 pesticides. Grapes, tomatoes, strawberries and apples had the following number of pesticides used on the crops respectively: 109 pesticides, 106 pesticides, 101 pesticides and 96 pesticides. Due to time constraints, peppers were not included in this study.

3.2 Methods

The risk on human health of residual contamination of pesticides on vegetable and fruit crops was estimated based on a risk ranking method. Several risk-ranking methods have been published (Van der Fels-Klerx, et al., 2015). The risk ranking method used in this study was selected using the decision risk ranking tool (Van der Fels-Klerx, et al., 2015). For this study, the risk ratio method turned out to be one of the best options (Annex 1). The risk ratio method is a method which takes both occurrence and severity of the compound into account. Ranking of pesticides is performed by dividing occurrence and toxicity (Figure 3.1). This method is often used to quickly screen the risk of chemicals for their human health impacts. There are multiple endpoints for both occurrence and toxicity. In the current study, the endpoint for occurrence was the percentage of samples of the particular pesticide that had a concentration similar to, or above the MRL. The endpoint for toxicity consisted of human health effects based on ADIs and ARfDs, with ADIs examining the chronic health effect and ARfDs the acute health effect. The MRL exceedance rates per country of origin were calculated for all countries as well, using the KAP data. Data were analyzed in Excel.

For the Dutch data, every pesticide had the same total number of samples since it was assumed that the multi-residue analytical methods that were used, measured all pesticides found on that crop. A multi-residue method is a method which can test 180 to 420 different pesticides at a time (NVWA, 2014). For the Dutch data, several multi-residue methods were used, varying from 10 to 17 methods per crop.

OCCURRENCE

a) (# of samples ≥ MRL / total # of samples) * 100%

TOXICITY

b) ARfD

c) ADI

SCORE = occurrence / toxicity

Figure 3.1. Equations of the risk ratio method.

4 Results

Scores were calculated for each pesticide and crop separately. Results were represented in four risk ranking tables per crop; two for the Dutch data and two for European data. In every first table the risk ranking based on the chronic health risks were calculated (ADI) and in every second table the risk ranking based on the acute risks were calculated (ARfD). MRLs were extracted from the EC database (EC, 2015b) and if no MRL was set, the default MRL of 0.01 mg/kg applied (EC, 2014). Measurement uncertainty of MRLs was not taken into account. Pesticides for which data were missing (ARfDs or ADIs), were not included in this study; a tiered approach[8] was followed. Substances which were left out in all of the crops examined in this study were piperonyl butoxide and oxamyl oxime. The first one is a synergist used in combination with pesticides (insecticides) and is not a pesticide itself, and oxamyl oxime was found to be toxicologically irrelevant according to EFSA (EC, 2015b; EFSA, 2010; PAN, 2014).

Differences between Dutch and EU data were analyzed to evaluate if the Netherlands should monitor pesticides that are currently not included in the national monitoring program. The scores and ranks for both chronic and acute risks were given to get insight in the severity of the potentially harmful effects of the pesticides. In addition, MRL exceedance rates were calculated per country of origin per crop using the KAP data. Calculations of countries based on 20 sample results or more were found to be reliable. Hence, countries with less than 20 sample results were excluded.

[8] Inclusion of pesticides was done by going through different stages. Initially, all pesticides were included (1st tier). Thereafter, only pesticides which had an ADI and/or ARfD were included (2nd tier).

In total, 193 different pesticides, which are monitored in the Netherlands, were examined in this study (6117 sample results). For apples, 1288 samples were tested of which 1139 were positive for one or more pesticides. Of these 1139 samples, 705 fell within legal limits (61.9 percent), and 434 samples results exceeded the MRL (38.1 percent). For strawberries, 1154 samples were analyzed of which 1020 samples were positive for one or more pesticides. Almost 80 percent of these samples fell within legal limits (805 samples) and over one fifth (21.1 percent) of the samples tested for strawberries, exceeded the MRL. For tomatoes 1275 samples were tested of which 1269 samples tested positive for one or more pesticides. Of these positive samples, two-third fell within legal limits and 20.1 percent exceeded the MRL. The highest number of samples were analyzed for grapes. In total, 2400 samples were tested of which 2210 sampled were positive for one or more pesticides. Of the positive samples, 1552 fell within legal limits (70.2 percent). Approximately one-third exceeded the MRL.

4.1 APPLES

4.1.1 Risk ranking Dutch data

In total, 82 different pesticides were detected (above the LOD) on apples. The MRL was most frequently exceeded for tolylfluanid, diphenylamine, DMST, propargite and azinphos-methyl (exceedances of 16.7 percent, 10.2 percent, 7.8 percent, 4.3 percent and 2.7 percent respectively). In table 4.1., the scores of the pesticides based on the chronic health impact, are listed. After obtaining the scores, the pesticides were ranked from highest to lowest score (shown in the last column). Of the 73 pesticides, 28 pesticides exceeded the MRL, with scores ranging from 0.4 for 2-phenylphenol to 1941.0 for diazinon. For the remaining 45 pesticides, all the samples tested, were below the MRL. In table 4.1., 73 pesticides are listed, because pesticides for which no ADI was established were left out in this risk ranking, being carbendazim (including benomyl), desmethyl pirimicarb, DMST, ethirimol, ethoxyquin, fenthion sulfoxide, propargite, pyridafenthion and sum of triadimefon and triadimenol.

Table 4.1. Risk ranking of pesticides sprayed on apples using the risk ratio method based on the MRL and the ADI – the Netherlands (KAP, 2004-2014).

Pesticide	Group	Total # of samples	# of samples	MRL (mg/kg)	Percentage > MRL	ADI (mg/kg/bw/d)	Score	Rank
Diazinon	Insecticides, Acaricides, Nematicides	1288	5	0.01*	0.39	0.0002	1941.0	1
Dimethoate	Bactericides and Fungicides	1288	11	0.02*	0.85	0.001	854.0	2
Azinphos-methyl	Bactericides and Fungicides	1288	35	0.05*	2.72	0.005	543.5	3
Carbaryl	Insecticides, Acaricides, Nematicides	1288	33	0.01*	2.56	0.0075	341.6	4
Tolylfluanid	Bactericides and Fungicides	1288	215	0.01*	16.69	0.1	166.9	5
Trichlorfon	Insecticides, Acaricides, Nematicides	1288	4	0.01*	0.31	0.002	155.3	6
Diphenylamine	Bactericides and Fungicides	1288	131	0.1	10.17	0.075	135.6	7
Ethion	Insecticides, Acaricides, Nematicides	1288	3	0.01*	0.23	0.002	116.5	8
Carbendazim	Bactericides and Fungicides	1288	24	0.2**	1.86	0.02	93.2	9
Indoxacarb	Insecticides, Acaricides, Nematicides	1288	7	0.5	0.54	0.006	90.6	10
Chlorpyrifos	Insecticides, Acaricides, Nematicides	1288	1	0.5	0.08	0.001	77.6	11
Dicofol	Insecticides, Acaricides, Nematicides	1288	1	0.02*	0.08	0.002	38.8	12
Triadimenol	Bactericides and Fungicides	1288	24	0.01*	1.86	0.05	37.3	13
Fenitrothion	Insecticides, Acaricides, Nematicides	1288	2	0.01*	0.16	0.005	31.1	14
Methomyl	Insecticides, Acaricides, Nematicides	1288	1	0.01*	0.08	0.0025	31.1	15
Endosulfan (alpha, beta, sulphate)	Insecticides, Acaricides, Nematicides	1288	2	0.05*	0.16	0.006	25.9	16
Famoxadone	Bactericides and Fungicides	1288	3	0.01*	0.23	0.012	19.4	17
Pyridaben	Insecticides, Acaricides, Nematicides	1288	2	0.5	0.16	0.01	15.5	18
Fenthion	Insecticides, Acaricides, Nematicides	1288	1	0.01*	0.08	0.007	11.1	19
Bromopropylate	Insecticides, Acaricides, Nematicides	1288	1	0.01*	0.08	0.03	2.6	20
Malathion	Insecticides, Acaricides, Nematicides	1288	1	0.02*	0.08	0.03	2.6	21
Dimethomorph	Bactericides and Fungicides	1288	1	0.01*	0.08	0.05	1.6	22
Azoxystrobin	Bactericides and Fungicides	1288	3	0.01*	0.23	0.2	1.2	23
Thiophanate-methyl	Bactericides and Fungicides	1288	1	0.5	0.08	0.08	1.0	24
Diflubenzuron	Bactericides and Fungicides	1288	1	5	0.08	0.1	0.8	25
Vamidothion sulfoxide	Insecticides, Acaricides, Nematicides	1288	1	0.01*	0.08	0.1	0.8	26
Dichlofluanid	Bactericides and Fungicides	1288	2	0.01*	0.16	0.3	0.5	27
2-phenylphenol	Bactericides and Fungicides	1288	2	0.05*	0.16	0.4	0.4	28
Acetamiprid	Insecticides, Acaricides, Nematicides	1288	0	0.8	0.00	0.025	0.0	29
Bifenthrin	Insecticides, Acaricides, Nematicides	1288	0	0.3	0.00	0.015	0.0	30
Boscalid	Bactericides and Fungicides	1288	0	2	0.00	0.04	0.0	31
Bupirimate	Bactericides and Fungicides	1288	0	0.2	0.00	0.05	0.0	32
Buprofezin	Insecticides, Acaricides, Nematicides	1288	0	3	0.00	0.01	0.0	33
Captan	Bactericides and Fungicides	1288	0	3	0.00	0.1	0.0	34
Chlorantraniliprole	Insecticides, Acaricides, Nematicides	1288	0	0.5	0.00	1.56	0.0	35
Chlorpyrifos-methyl	Bactericides and Fungicides	1288	0	0.5	0.00	0.01	0.0	36
Chlorothalonil	Insecticides, Acaricides, Nematicides	1288	0	2	0.00	0.015	0.0	37
Cypermethrin	Insecticides, Acaricides, Nematicides	1288	0	1	0.00	0.05	0.0	38
Cyprodinil	Bactericides and Fungicides	1288	0	1.5	0.00	0.03	0.0	39
Deltamethrin	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.01	0.0	40

Diethofencarb	Bactericides and Fungicides	1288	0	0.5	0.00	0.43	0.0	41
Difenoconazole	Bactericides and Fungicides	1288	0	0.8	0.00	0.01	0.0	42
Dithianon	Bactericides and Fungicides	1288	0	3	0.00	0.01	0.0	43
Dithiocarbamates (As Cs2)	Bactericides and Fungicides	1288	0	5	0.00	0.003	0.0	44
Dodine	Bactericides and Fungicides	1288	0	0.9	0.00	0.1	0.0	45
Etofenprox	Insecticides, Acaricides, Nematicides	1288	0	1	0.00	0.03	0.0	46
Fenazaquin	Insecticides, Acaricides, Nematicides	1288	0	0.1	0.00	0.005	0.0	47
Fenhexamid	Bactericides and Fungicides	1288	0	0.05*	0.00	0.2	0.0	48
Fenoxycarb	Insecticides, Acaricides, Nematicides	1288	0	1	0.00	0.053	0.0	49
Fenpyroximate	Insecticides, Acaricides, Nematicides	1288	0	0.3	0.00	0.01	0.0	50
Flonicamid	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.025	0.0	51
Fludioxonil	Bactericides and Fungicides	1288	0	5	0.00	0.37	0.0	52
Flufenoxuron	Insecticides, Acaricides, Nematicides	1288	0	0.5	0.00	0.01	0.0	53
Folpet	Bactericides and Fungicides	1288	0	3	0.00	0.1	0.0	54
Hexythiazox	Insecticides, Acaricides, Nematicides	1288	0	1	0.00	0.03	0.0	55
Imazalil	Insecticides, Acaricides, Nematicides	1288	0	2	0.00	0.025	0.0	56
Imidacloprid	Insecticides, Acaricides, Nematicides	1288	0	0.5	0.00	0.06	0.0	57
Methoxyfenozide	Insecticides, Acaricides, Nematicides	1288	0	2	0.00	0.1	0.0	58
Parathion (ethyl)	Insecticides, Acaricides, Nematicides	1288	0	0.05*	0.00	0.0006	0.0	59
Penconazole	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.03	0.0	60
Pirimicarb	Insecticides, Acaricides, Nematicides	1288	0	2	0.00	0.035	0.0	61
Prochloraz	Bactericides and Fungicides	1288	0	0.05*	0.00	0.01	0.0	62
Pyraclostrobin	Bactericides and Fungicides	1288	0	0.5	0.00	0.03	0.0	63
Pyrimethanil	Bactericides and Fungicides	1288	0	15	0.00	0.17	0.0	64
Pyriproxyfen	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.1	0.0	65
Spirodiclofen	Insecticides, Acaricides, Nematicides	1288	0	0.8	0.00	0.015	0.0	66
Tebuconazole	Bactericides and Fungicides	1288	0	0.3	0.00	0.03	0.0	67
Tebufenozide	Insecticides, Acaricides, Nematicides	1288	0	1	0.00	0.02	0.0	68
Tebufenpyrad	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.01	0.0	69
Thiabendazole	Bactericides and Fungicides	1288	0	5	0.00	0.1	0.0	70
Thiacloprid	Insecticides, Acaricides, Nematicides	1288	0	0.3	0.00	0.01	0.0	71
Thiametoxam	Insecticides, Acaricides, Nematicides	1288	0	0.5	0.00	0.026	0.0	72
Trifloxystrobin	Bactericides and Fungicides	1288	0	0.5	0.00	0.1	0.0	73

* Indicates lower limit of determination
** MRL based on related compound (Carbendazim including benomyl)

In table 4.2., scores based on the acute risks are shown. This list contains 52 pesticides, since for 30 pesticides (2-phenylphenol, azoxystrobin, boscalid, bromopropylate, bupirimate, carbendazim (including benomyl), chlorantraniliprole, cyprodinil, desmethylpirmicarb, dichlofluanid, diethofencarb, diflubenzuron, diphenylamine, DMST, ethion, ethirimol, ethoxyquin, fenhexamid, fenthion sulfoxide, fludioxonil, flufenoxuron, hexythiazox, propargite, pyridaphenthion, pyrimethanil, spirodiclofen, sum of triadimefon and triadimenol, tebufenozide, trifloxystrobin and vamidothion sulfoxide) an ARfD was either not necessary due to the low acute toxicity of the substance or the ARfD was not allocated (EFSA, 2015a). The scores of the 20 pesticides exceeding the MRL vary from 0.1 to 271.7, with dimethomorph having the lowest score and azinphos-methyl the highest. The other 32 pesticides did not exceed the MRL.

Table 4.2. Risk ranking of pesticides sprayed on apples using the risk ratio method based on the MRL and the ARfD – the Netherlands (KAP, 2004-2014).

Pesticide	Group	Total # of samples	# of samples	MRL (mg/kg)	Percentage > MRL	ARfD (mg/kg/bw)	Score	Rank
Azinphos-methyl	Bactericides and Fungicides	1288	35	0.05*	2.72	0.01	271.7	1
Carbaryl	Insecticides, Acaricides, Nematicides	1288	33	0.01*	2.56	0.01	256.2	2
Carbendazim	Bactericides and Fungicides	1288	24	0.2**	1.86	0.02	93.2	3
Dimethoate	Bactericides and Fungicides	1288	11	0.02*	0.85	0.01	85.4	4
Tolylfluanid	Bactericides and Fungicides	1288	215	0.01*	16.69	0.25	66.8	5
Triadimenol	Bactericides and Fungicides	1288	24	0.01*	1.86	0.05	37.3	6
Methomyl	Insecticides, Acaricides, Nematicides	1288	1	0.01*	0.08	0.0025	31.1	7
Diazinon	Insecticides, Acaricides, Nematicides	1288	5	0.01*	0.39	0.025	15.5	8
Chlorpyrifos	Insecticides, Acaricides, Nematicides	1288	1	0.5	0.08	0.005	15.5	9
Fenitrothion	Insecticides, Acaricides, Nematicides	1288	2	0.01*	0.16	0.013	11.9	10
Endosulfan (alpha, beta, sulphate)	Insecticides, Acaricides, Nematicides	1288	2	0.05*	0.16	0.015	10.4	11
Fenthion	Insecticides, Acaricides, Nematicides	1288	1	0.01*	0.08	0.01	7.8	12
Indoxacarb	Insecticides, Acaricides, Nematicides	1288	7	0.5	0.54	0.125	4.3	13
Pyridaben	Insecticides, Acaricides, Nematicides	1288	2	0.5	0.16	0.05	3.1	14
Trichlorfon	Insecticides, Acaricides, Nematicides	1288	4	0.01*	0.31	0.1	3.1	15
Famoxadone	Bactericides and Fungicides	1288	3	0.01*	0.23	0.2	1.2	16
Dicofol	Insecticides, Acaricides, Nematicides	1288	1	0.02*	0.08	0.2	0.4	17
Thiophanate-methyl	Bactericides and Fungicides	1288	1	0.5	0.08	0.2	0.4	18
Malathion	Insecticides, Acaricides, Nematicides	1288	1	0.02*	0.08	0.3	0.3	19
Dimethomorph	Bactericides and Fungicides	1288	1	0.01*	0.08	0.6	0.1	20
Acetamiprid	Insecticides, Acaricides, Nematicides	1288	0	0.8	0.00	0.025	0.0	21
Bifenthrin	Insecticides, Acaricides, Nematicides	1288	0	0.3	0.00	0.03	0.0	22
Buprofezin	Insecticides, Acaricides, Nematicides	1288	0	3	0.00	0.5	0.0	23
Captan	Bactericides and Fungicides	1288	0	3	0.00	0.3	0.0	24
Chlorothalonil	Insecticides, Acaricides, Nematicides	1288	0	2	0.00	0.6	0.0	25
Chlorpyrifos-methyl	Bactericides and Fungicides	1288	0	0.5	0.00	0.1	0.0	26
Cypermethrin	Insecticides, Acaricides, Nematicides	1288	0	1	0.00	0.2	0.0	27
Deltamethrin	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.01	0.0	28
Difenoconazole	Bactericides and Fungicides	1288	0	0.8	0.00	0.16	0.0	29
Dithianon	Bactericides and Fungicides	1288	0	3	0.00	0.12	0.0	30
Dithiocarbamates (As Cs2)	Bactericides and Fungicides	1288	0	5	0.00	0.04	0.0	31
Dodine	Bactericides and Fungicides	1288	0	0.9	0.00	0.1	0.0	32

Etofenprox	Insecticides, Acaricides, Nematicides	1288	0	1	0.00	1	0.0	33
Fenazaquin	Insecticides, Acaricides, Nematicides	1288	0	0.1	0.00	0.1	0.0	34
Fenoxycarb	Insecticides, Acaricides, Nematicides	1288	0	1	0.00	2	0.0	35
Fenpyroximate	Insecticides, Acaricides, Nematicides	1288	0	0.3	0.00	0.02	0.0	36
Flonicamid	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.025	0.0	37
Folpet	Bactericides and Fungicides	1288	0	3	0.00	0.2	0.0	38
Imazalil	Insecticides, Acaricides, Nematicides	1288	0	2	0.00	0.05	0.0	39
Imidacloprid	Insecticides, Acaricides, Nematicides	1288	0	0.5	0.00	0.06	0.0	40
Methoxyfenozide	Insecticides, Acaricides, Nematicides	1288	0	2	0.00	0.2	0.0	41
Parathion (ethyl)	Insecticides, Acaricides, Nematicides	1288	0	0.05*	0.00	0.005	0.0	42
Penconazole	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.5	0.0	43
Pirimicarb	Insecticides, Acaricides, Nematicides	1288	0	2	0.00	0.1	0.0	44
Prochloraz	Bactericides and Fungicides	1288	0	0.05*	0.00	0.025	0.0	45
Pyraclostrobin	Bactericides and Fungicides	1288	0	0.5	0.00	0.03	0.0	46
Pyriproxyfen	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	10	0.0	47
Tebuconazole	Bactericides and Fungicides	1288	0	0.3	0.00	0.03	0.0	48
Tebufenpyrad	Insecticides, Acaricides, Nematicides	1288	0	0.2	0.00	0.02	0.0	49
Thiabendazole	Bactericides and Fungicides	1288	0	5	0.00	0.1	0.0	50
Thiacloprid	Insecticides, Acaricides, Nematicides	1288	0	0.3	0.00	0.03	0.0	51
Thiametoxam	Insecticides, Acaricides, Nematicides	1288	0	0.5	0.00	0.5	0.0	52

* Indicates lower limit of determination
** MRL based on related compound (Carbendazim including benomyl)

4.1.2 Risk ranking EU data

In total, 89 different pesticides were detected (above the LOD) on apples (table 4.3.). The five pesticides with the highest MRL exceedance rates were dimethoate, fenbutatin oxide, methomyl, fenvalerate and fenthion, with percentages of respectively 0.7, 0.2, 0.2, 0.1 and 0.1 percent. For two of the pesticides (ethirimol and propargite), the ADIs were not defined (EFSA, 2015a). Therefore, these pesticides were excluded from the EU risk ranking for apples. From the 87 pesticides included in the risk ranking based on chronic risks, nine pesticides exceeded the MRL with scores ranging from 4.2 (carbendazim) to 648.3 (dimethoate). The other 78 pesticides showed no exceedances in any of the samples tested.

Table 4.3. Risk ranking of pesticides sprayed on apples using the risk ratio method based on the MRL and the ADI – the EU (EFSA, 2015a).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ADI (mg/kg/bw/d)	Score	Rank
Dimethoate	Bactericides and Fungicides	1234	8	0.01*	0.65	0.001	648.30	1
Methomyl	Insecticides, Acaricides, Nematicides	1194	2	0.01*	0.17	0.0025	67.00	2
Chlorpyrifos	Insecticides, Acaricides, Nematicides	1543	0	0.5	0.06	0.001	64.27	3
Flusilazole	Bactericides and Fungicides	1350	1	0.01*	0.07	0.002	37.04	4
Fenazaquin	Insecticides, Acaricides, Nematicides	1311	1	0.1	0.08	0.005	15.26	5
Fenthion	Insecticides, Acaricides, Nematicides	1017	1	0.01*	0.10	0.007	14.05	6
Fenvalerate	Insecticides, Acaricides, Nematicides	754	1	0.1	0.13	0.0175	7.58	7
Fenbutatin oxide	Insecticides, Acaricides, Nematicides	472	1	2	0.21	0.05	4.24	8
Carbendazim	Bactericides and Fungicides	1185	1	0.2	0.08	0.02	4.22	9
2-phenylphenol	Bactericides and Fungicides	1078	0	0.05*	0.00	0.4	0.00	10
Acetamiprid	Insecticides, Acaricides, Nematicides	1355	0	0.8	0.00	0.025	0.00	11
Amitraz	Insecticides, Acaricides, Nematicides	704	0	0.05*	0.00	0.003	0.00	12
Azinphos-methyl	Insecticides, Acaricides, Nematicides	1457	0	0.05*	0.00	0.005	0.00	13
Azoxystrobin	Bactericides and Fungicides	1438	0	0.01*	0.00	0.2	0.00	14
Bifenthrin	Insecticides, Acaricides, Nematicides	1504	0	0.3	0.00	0.015	0.00	15
Bitertanol	Bactericides and Fungicides	1366	0	0.01*	0.00	0.003	0.00	16
Boscalid	Bactericides and Fungicides	1490	0	2	0.00	0.04	0.00	17
Bupirimate	Bactericides and Fungicides	1422	0	0.2	0.00	0.05	0.00	18
Buprofezin	Insecticides, Acaricides, Nematicides	1518	0	3	0.00	0.01	0.00	19
Captan + Folpet	Bactericides and Fungicides	742	0	0.01*	0.00	0.1	0.00	20
Carbaryl	Insecticides, Acaricides, Nematicides	1425	0	0.01*	0.00	0.0075	0.00	21
Chlorantraniliprole	Insecticides, Acaricides, Nematicides	972	0	0.5	0.00	1.56	0.00	22
Chlorothalonil	Insecticides, Acaricides, Nematicides	1271	0	2	0.00	0.015	0.00	23
Chlorpyrifos-methyl	Insecticides, Acaricides, Nematicides	1556	1	0.5	0.00	0.01	0.00	24
Clofentezine	Insecticides, Acaricides, Nematicides	1197	0	0.5	0.00	0.02	0.00	25
Cyfluthrin	Insecticides, Acaricides, Nematicides	1186	0	0.2	0.00	0.003	0.00	26
Cypermethrin	Insecticides, Acaricides, Nematicides	1406	0	1	0.00	0.05	0.00	27
Cyproconazole	Bactericides and Fungicides	1401	0	0.1	0.00	0.02	0.00	28
Cyprodinil	Bactericides and Fungicides	1505	0	1.5	0.00	0.03	0.00	29
Deltamethrin	Insecticides, Acaricides, Nematicides	1545	0	0.2	0.00	0.01	0.00	30
Difenoconazole	Bactericides and Fungicides	1383	0	0.8	0.00	0.01	0.00	31
Diflubenzuron	Bactericides and Fungicides	1037	0	5	0.00	0.1	0.00	32
Diphenylamine	Bactericides and Fungicides	1463	0	0.1	0.00	0.075	0.00	33
Dithianon	Bactericides and Fungicides	230	0	3	0.00	0.01	0.00	34
Dithiocarbamates	Bactericides and Fungicides	843	0	5	0.00	0.003	0.00	35
Dodine	Bactericides and Fungicides	750	0	0.9	0.00	0.1	0.00	36
Ethephon	Plant Growth Regulator	519	0	0.01*	0.00	0.03	0.00	37
Etofenprox	Insecticides, Acaricides, Nematicides	1301	0	1	0.00	0.03	0.00	38
Fenhexamid	Bactericides and Fungicides	1476	0	0.05*	0.00	0.2	0.00	39
Fenoxycarb	Insecticides, Acaricides, Nematicides	1340	0	1	0.00	0.053	0.00	40

Fenpyroximate	Insecticides, Acaricides, Nematicides	1147	0	0.3	0.00	0.01	0.00	41
Flonicamid	Insecticides, Acaricides, Nematicides	517	0	0.2	0.00	0.025	0.00	42
Flubendiamide	Insecticides, Acaricides, Nematicides	531	0	0.8	0.00	0.017	0.00	43
Fludioxonil	Bactericides and Fungicides	1427	0	5	0.00	0.37	0.00	44
Flufenoxuron	Insecticides, Acaricides, Nematicides	1240	0	0.5	0.00	0.01	0.00	45
Fluopyram	Bactericides and Fungicides	726	0	0.6	0.00	0.012	0.00	46
Fluquinconazole	Bactericides and Fungicides	1247	0	0.1	0.00	0.002	0.00	47
Glyphosate	Herbicides	52	0	0.1	0.00	0.3	0.00	48
Hexaconazole	Bactericides and Fungicides	1400	0	0.01*	0.00	0.005	0.00	49
Hexythiazox	Insecticides, Acaricides, Nematicides	1268	0	1	0.00	0.03	0.00	50
Imazalil	Insecticides, Acaricides, Nematicides	1453	0	2	0.00	0.025	0.00	51
Imidacloprid	Insecticides, Acaricides, Nematicides	1343	0	0.5	0.00	0.06	0.00	52
Indoxacarb	Insecticides, Acaricides, Nematicides	1387	0	0.5	0.00	0.006	0.00	53
Iprodione	Bactericides and Fungicides	1446	0	6	0.00	0.06	0.00	54
Kresoxim-methyl	Bactericides and Fungicides	1478	0	0.2	0.00	0.4	0.00	55
Lambda-cyhalothrin	Insecticides, Acaricides, Nematicides	1396	0	0.1	0.00	0.0025	0.00	56
Methidathion	Insecticides, Acaricides, Nematicides	1479	0	0.03	0.00	0.001	0.00	57
Methoxyfenozide	Insecticides, Acaricides, Nematicides	1216	0	2	0.00	0.1	0.00	58
Myclobutanil	Bactericides and Fungicides	1528	0	0.5	0.00	0.025	0.00	59
Oxadixyl	Bactericides and Fungicides	1454	0	0.01*	0.00	0.01	0.00	60
Paclobutrazol	Other pesticides	1250	0	0.5	0.00	0.022	0.00	61
Penconazole	Insecticides, Acaricides, Nematicides	1524	0	0.2	0.00	0.03	0.00	62
Pendimethalin	Herbicides	1412	0	0.05*	0.00	0.125	0.00	63
Phosalone	Insecticides, Acaricides, Nematicides	1522	0	0.01*	0.00	0.01	0.00	64
Phosmet	Insecticides, Acaricides, Nematicides	1150	0	0.5	0.00	0.01	0.00	65
Pirimicarb	Insecticides, Acaricides, Nematicides	1238	0	2	0.00	0.035	0.00	66
Pirimiphos-methyl	Insecticides, Acaricides, Nematicides	1542	0	0.05*	0.00	0.004	0.00	67
Prochloraz	Bactericides and Fungicides	903	0	0.05*	0.00	0.01	0.00	68
Procymidone	Bactericides and Fungicides	1503	0	0.01*	0.00	0.0028	0.00	69
Propiconazole	Bactericides and Fungicides	1444	0	0.01*	0.00	0.04	0.00	70
Pyraclostrobin	Bactericides and Fungicides	1338	0	0.5	0.00	0.03	0.00	71
Pyrimethanil	Bactericides and Fungicides	1550	0	15	0.00	0.17	0.00	72
Spinosad	Insecticides, Acaricides, Nematicides	1171	0	0.3	0.00	0.024	0.00	73
Spirodiclofen	Insecticides, Acaricides, Nematicides	1076	0	0.8	0.00	0.015	0.00	74
Spiroxamine	Bactericides and Fungicides	1401	0	0.05*	0.00	0.03	0.00	75
tau-Fluvalinate	Insecticides, Acaricides, Nematicides	1219	0	0.3	0.00	0.005	0.00	76
Tebuconazole	Bactericides and Fungicides	1511	0	0.3	0.00	0.03	0.00	77
Tebufenozide	Insecticides, Acaricides, Nematicides	1292	0	1	0.00	0.02	0.00	78
Tebufenpyrad	Insecticides, Acaricides, Nematicides	1377	0	0.2	0.00	0.01	0.00	79
Tetraconazole	Bactericides and Fungicides	1353	0	0.3	0.00	0.004	0.00	80
Thiabendazole	Bactericides and Fungicides	1381	0	5	0.00	0.1	0.00	81
Thiacloprid	Insecticides, Acaricides, Nematicides	1340	0	0.3	0.00	0.01	0.00	82
Thiamethoxam	Insecticides, Acaricides, Nematicides	1211	0	0.01*	0.00	0.026	0.00	83
Thiophanate-methyl	Bactericides and Fungicides	1300	0	0.5	0.00	0.08	0.00	84

Triadimenol	Bactericides and Fungicides	1205	0	0.01*	0.00	0.05	0.00	85
Trifloxystrobin	Bactericides and Fungicides	1405	0	0.5	0.00	0.1	0.00	86
Triflumuron	Insecticides, Acaricides, Nematicides	1221	0	0.5	0.00	0.014	0.00	87

* Indicates lower limit of determination

The risk ranking for apples based on acute risks is shown in table 4.4. For 28 out of 87 pesticides, there was no ARfD allocated or necessary according to EFSA (EFSA, 2015a). Hence, these pesticides (2-phenylphenol, azoxystrobin, boscalid, bupirimate, chlorantraniliprole, clofentezine, cyprodinil, diflubenzuron, diphenylamine, ethirimol, fenhexamid, fludioxonil, flufenoxuron, glyphosate, hexaconazole, hexythiazox, iprodione, kresoxim-methyl, oxadixyl, pendimethalin, propargite, pyrimethanil, spinosad, spiodiclofen, tebufenozide, trifloxystrobin and triflumuron) were excluded from the risk ranking based on the ARfD. Of the 87 pesticides listed, 9 exceeded the MRL, with scores varying from 0.6 (chlorpyrifos) to 67.0 (methomyl).

Table 4.4. Risk ranking of pesticides sprayed on apples using the risk ratio method based on the MRL and the ARfD – the EU (EFSA, 2015a).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg/bw)	Percentage > MRL	ARfD (mg/kg/bw)	Score	Rank
Methomyl	Insecticides, Acaricides, Nematicides	1194	2	0.01	0.17	0.0025	67.00	1
Dimethoate	Bactericides and Fungicides	1234	8	0.01	0.65	0.01	64.83	2
Flusilazole	Bactericides and Fungicides	1350	1	0.01	0.07	0.005	14.81	3
Fenthion	Insecticides, Acaricides, Nematicides	1017	1	0.01*	0.10	0.01	9.83	4
Fenvalerate	Insecticides, Acaricides, Nematicides	754	1	0.1	0.13	0.0175	7.58	5
Carbendazim	Bactericides and Fungicides	1185	1	0.2	0.08	0.02	4.22	6
Fenbutatin oxide	Insecticides, Acaricides, Nematicides	472	1	2	0.21	0.1	2.12	7
Fenazaquin	Insecticides, Acaricides, Nematicides	1311	1	0.1	0.08	0.1	0.76	8
Chlorpyrifos	Insecticides, Acaricides, Nematicides	1543	0	0.5	0.06	0.1	0.64	9
Acetamiprid	Insecticides, Acaricides, Nematicides	1355	0	0.8	0.00	0.025	0.00	10
Amitraz	Insecticides, Acaricides, Nematicides	704	0	0.05	0.00	0.01	0.00	11
Azinphos-methyl	Insecticides, Acaricides, Nematicides	1457	0	0.05	0.00	0.01	0.00	12
Bifenthrin	Insecticides, Acaricides, Nematicides	1504	0	0.3	0.00	0.03	0.00	13
Bitertanol	Bactericides and Fungicides	1366	0	0.01	0.00	0.01	0.00	14
Buprofezin	Insecticides, Acaricides, Nematicides	1518	0	3	0.00	0.5	0.00	15
Captan + Folpet	Bactericides and Fungicides	742	0	0.01	0.00	0.3	0.00	16
Carbaryl	Insecticides, Acaricides, Nematicides	1425	0	0.01	0.00	0.01	0.00	17
Chlorothalonil	Insecticides, Acaricides, Nematicides	1271	0	2	0.00	0.6	0.00	18
Chlorpyrifos-methyl	Insecticides, Acaricides, Nematicides	1556	1	0.5	0.00	0.005	0.00	19
Cyfluthrin	Insecticides, Acaricides, Nematicides	1186	0	0.2	0.00	0.02	0.00	20
Cypermethrin	Insecticides, Acaricides, Nematicides	1406	0	1	0.00	0.2	0.00	21
Cyproconazole	Bactericides and Fungicides	1401	0	0.1	0.00	0.02	0.00	22
Deltamethrin	Insecticides, Acaricides, Nematicides	1545	0	0.2	0.00	0.01	0.00	23
Difenoconazole	Bactericides and Fungicides	1383	0	0.8	0.00	0.16	0.00	24
Dithianon	Bactericides and Fungicides	230	0	3	0.00	0.12	0.00	25
Dithiocarbamates	Bactericides and Fungicides	843	0	5	0.00	0.04	0.00	26
Dodine	Bactericides and Fungicides	750	0	0.9	0.00	0.1	0.00	27
Ethephon	Plant Growth Regulator	519	0	0.01	0.00	0.05	0.00	28
Etofenprox	Insecticides, Acaricides, Nematicides	1301	0	1	0.00	1	0.00	29
Fenoxycarb	Insecticides, Acaricides, Nematicides	1340	0	1	0.00	2	0.00	30
Fenpyroximate	Insecticides, Acaricides, Nematicides	1147	0	0.3	0.00	0.02	0.00	31
Flonicamid	Insecticides, Acaricides, Nematicides	517	0	0.2	0.00	0.025	0.00	32
Flubendiamide	Insecticides, Acaricides, Nematicides	531	0	0.8	0.00	0.1	0.00	33
Fluopyram	Bactericides and Fungicides	726	0	0.6	0.00	0.5	0.00	34
Fluquinconazole	Bactericides and Fungicides	1247	0	0.1	0.00	0.02	0.00	35
Imazalil	Insecticides, Acaricides, Nematicides	1453	0	2	0.00	0.05	0.00	36
Imidacloprid	Insecticides, Acaricides, Nematicides	1343	0	0.5	0.00	0.06	0.00	37
Indoxacarb	Insecticides, Acaricides, Nematicides	1387	0	0.5	0.00	0.125	0.00	38
Lambda-cyhalothrin	Insecticides, Acaricides, Nematicides	1396	0	0.1	0.00	0.005	0.00	39
Methidathion	Insecticides, Acaricides, Nematicides	1479	0	0.03	0.00	0.01	0.00	40

Methoxyfenozide	Insecticides, Acaricides, Nematicides	1216	0	2	0.00	0.2	0.00	41
Myclobutanil	Bactericides and Fungicides	1528	0	0.5	0.00	0.31	0.00	42
Paclobutrazol	Other pesticides	1250	0	0.5	0.00	0.1	0.00	43
Penconazole	Insecticides, Acaricides, Nematicides	1524	0	0.2	0.00	0.5	0.00	44
Phosalone	Insecticides, Acaricides, Nematicides	1522	0	0.01	0.00	0.1	0.00	45
Phosmet	Insecticides, Acaricides, Nematicides	1150	0	0.5	0.00	0.045	0.00	46
Pirimicarb	Insecticides, Acaricides, Nematicides	1238	0	2	0.00	0.1	0.00	47
Pirimiphos-methyl	Insecticides, Acaricides, Nematicides	1542	0	0.05	0.00	0.15	0.00	48
Prochloraz	Bactericides and Fungicides	903	0	0.05	0.00	0.025	0.00	49
Procymidone	Bactericides and Fungicides	1503	0	0.01	0.00	0.012	0.00	50
Propiconazole	Bactericides and Fungicides	1444	0	0.01	0.00	0.3	0.00	51
Pyraclostrobin	Bactericides and Fungicides	1338	0	0.5	0.00	0.03	0.00	52
Spiroxamine	Bactericides and Fungicides	1401	0	0.05	0.00	2	0.00	53
tau-Fluvalinate	Insecticides, Acaricides, Nematicides	1219	0	0.3	0.00	0.05	0.00	54
Tebuconazole	Bactericides and Fungicides	1511	0	0.3	0.00	0.03	0.00	55
Tebufenpyrad	Insecticides, Acaricides, Nematicides	1377	0	0.2	0.00	0.02	0.00	56
Tetraconazole	Bactericides and Fungicides	1353	0	0.3	0.00	0.05	0.00	57
Thiabendazole	Bactericides and Fungicides	1381	0	5	0.00	0.1	0.00	58
Thiacloprid	Insecticides, Acaricides, Nematicides	1340	0	0.3	0.00	0.03	0.00	59
Thiamethoxam	Insecticides, Acaricides, Nematicides	1211	0	0.01	0.00	0.5	0.00	60
Thiophanate-methyl	Bactericides and Fungicides	1300	0	0.5	0.00	0.2	0.00	61
Triadimenol	Bactericides and Fungicides	1205	0	0.01	0.00	0.05	0.00	62

* Indicates lower limit of determination

4.1.3 Comparison Dutch and EU data

In table 4.5., 34 pesticides are listed which are currently included in the monitoring program in the EU, but not included in the Dutch monitoring program. As can be seen in the third and the last column, the scores are rather low (and thus the ranks are rather low). For both chronic and acute risk, the exceptions are flusilazole, fenvalerate and fenbutatin oxide, which ranked fourth, seventh and eighth respectively for chronic risk, and third, fifth and seventh for acute risk.

Table 4.5. Pesticides used on apples included in the monitoring program in the EU, but not currently included in the Dutch monitoring program.

Pesticide	Score chronic	Rank	Score acute	Rank
Amitraz	0.0	12	0.0	11
Bitertanol	0.0	16	0.0	14
Captan+Folpet	0.0	20	0.0	16
Clofentezine	0.0	25	-	-
Cyfluthrin	0.0	26	0.0	20
Cyproconazole	0.0	28	0.0	22
Ethephon	0.0	37	0.0	28
Fenbutatin oxide	4.2	8	2.1	7
Fenvalerate	7.6	7	7.6	5
Flubendiamide	0.0	43	0.0	33
Fluopyram	0.0	46	0.0	34
Fluquinconazole	0.0	47	0.0	35
Flusilazole	37.0	4	14.8	3
Glyphosate	0.0	48	-	-
Hexaconazole	0.0	49	-	-
Iprodione	0.0	54	-	-
Kresoxim-methyl	0.0	55	-	-
Lambda-cyhalothrin	0.0	56	0.0	39
Methidathion	0.0	57	0.0	40
Myclobutanil	0.0	59	0.0	42
Oxadixyl	0.0	60	-	-
Paclobutrazol	0.0	61	0.0	43
Pendimethalin	0.0	63	-	-
Phosalone	0.0	64	0.0	45
Phosmet	0.0	65	0.0	46
Pirimiphos-methyl	0.0	67	0.0	48
Procymidone	0.0	69	0.0	50
Propiconazole	0.0	70	0.0	51
Spinosad	0.0	73	-	-
Spiroxamine	0.0	75	0.0	53
tau-Fluvalinate	0.0	76	0.0	54
Tetraconazole	0.0	80	0.0	57
Thiamethoxam	0.0	83	0.0	60
Triflumuron	0.0	87	-	-

* The pesticides with a "-" behind it did not have a score, because no ARfD was allocated or necessary and subsequently no score could be obtained.

4.1.4 MRL exceedance rates per country of origin

Almost all countries from which apples were imported, exceeded the MRL for one or more pesticides (see supplementary material), except for China. The countries with the highest MRL exceedance rates were (in descending order): South Africa, Chili and France, with South Africa having an MRL exceedance rate of 80 percent, Chili 53.6 percent and France 44.7 percent. The countries with the lowest percentages of MRL exceedances were (in ascending order): China, New Zealand and Italy. China did not exceed the MRL in any of the tested samples, New Zealand exceeded the MRL in 10 percent of all samples and Italy in 16.9 percent.

4.2 STRAWBERRIES

4.2.1 Risk ranking Dutch data

In total, 103 different pesticides were detected (above the LOD) on strawberries (see supplementary material). The five pesticides which showed the highest MRL exceedance rates were tolylfluanid, triadimenol, methomyl, procymidone and DMST, with percentages of 6.7, 5.4, 2.9, 2.7 and 2.4, respectively. In this risk ranking, 32 pesticides were not included; for eight pesticides no ADI was established (DMST, ethirimol, sum of malaoxon and malathion, methiocarb sulfone, methiocarb sulfoxide, propamocarb hydrochlorid and propargite), and for another 24 pesticides no ARfDs were allocated or necessary (EFSA, 2015a), being azoxystrobin, bifenazate, boscalid, bupirimate, chromafenozide, clofentezine, clopyralid, cyprodinil, dichlofluanid, etoxazole, fenhexamid, fludioxonil, hexaconazole, hexythiazox, iprodione, kresoxim-methyl, oxadixyl, pencycuron, pyrimethanil, quinoxyfen, spinosad, spirodiclofen, sum of triadimefon and triadimenol, teflubenzuron and trifloxystrobin. Out of the 95 pesticides included in the risk ranking based on chronic risk, 35 exceeded the MRL, with scores varying from 4332.8 (dichlorvos) to 0.3 (dichlofluanid). For the risk ranking based on acute risks, 29 out of the 71 pesticides exceeded the MRL, with methomyl having the highest score of 1143.9 and chlorpropham having the lowest score (0.1). The top five of the risk ranking of chronic and acute risks are listed in tables 4.6 and 4.7 respectively.

Table 4.6. Top five risk ranking of pesticides sprayed on strawberries using the risk ratio method based on the MRL and the ADI – the Netherlands (KAP, 2004-2014).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ADI (mg/kg/bw/d)	Score	Rank
Dichlorvos	Insecticides, Acaricides, Nematocides	1154	4	0.01*	0.35	0.00008	4332.8	1
Methomyl	Insecticides, Acaricides, Nematocides	1154	33	0.01*	2.86	0.0025	1143.9	2
Procymidone	Bactericides and Fungicides	1154	31	0.01*	2.69	0.0028	959.4	3
Oxamyl	Insecticides, Acaricides, Nematocides	1154	4	0.01*	0.35	0.001	346.6	4
Flutolanil	Bactericides and Fungicides	1154	5	0.01*	0.43	0.002	216.6	5

* Indicates lower limit of determination

Table 4.7. Top five risk ranking of pesticides sprayed on strawberries using the risk ratio method based on the MRL and the ARfD – the Netherlands (KAP, 2004-2014).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ARfD (mg/kg/bw)	Score	Rank
Methomyl	Insecticides, Acaricides, Nematocides	1154	33	0.01*	2.86	0.0025	1143.9	1
Oxamyl	Insecticides, Acaricides, Nematocides	1154	4	0.01*	0.35	0.001	346.6	2
Procymidone	Bactericides and Fungicides	1154	31	0.01*	2.69	0.012	223.9	3
Dichlorvos	Insecticides, Acaricides, Nematocides	1154	4	0.01*	0.35	0.002	173.3	4
Monocrotophos	Insecticides, Acaricides, Nematocides	1154	1	0.01*	0.09	0.0005	173.3	5

* Indicates lower limit of determination

4.2.2 Risk ranking EU data

In total, 84 different pesticides were detected (above the LOD) on strawberries. The five pesticides with the highest MRL exceedance rates were carbendazim (0.4 percent), trifloxystrobin (0.4 percent), propiconazole (0.4 percent), fenbutatin oxide (0.3 percent) and procymidone (0.3 percent). However, for ethirimol and propargite no ADIs were defined. For another 24 pesticides no ARfDs were allocated or necessary. Hence, these pesticides were excluded from the list (EFSA, 2015a): azoxystrobin, boscalid, bupirimate, chlorantraniliprole, clofentezine, cyprodinil, dichlofluanid, diethofencarb, ethion, fenhexamid, fludioxonil, hexythiazox, iprodione, kresoxim-methyl, lufenuron, pendimethalin, propyzamide, pyrimethanil, quinoxyfen, spinosad and spirdodiclofen (no ARfD). Of the 82 pesticides included in the risk ranking based on chronic risks, 18 exceeded the MRL. Of the pesticides exceeding the MRL based on acute risks, dimethoate had the highest score of 106.3 and triadimenol had the lowest score of 2.1 (the top five is listed in table 4.8). For the risk ranking based on acute risks, 15 out of 60 pesticides exceeded the MRL; formetanate (33.2) had the highest score and mepanipyrim (0.4) the lowest (the top five is listed in table 4.9).

Table 4.8. Top five risk ranking of pesticides sprayed on strawberries using the risk ratio method based on the MRL and the ADI – the EU (EFSA, 2015a).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ADI (mg/kg/bw/d)	Score	RANK
Dimethoate	Insecticides, Acaricides, Nematicides	941	1	0.01	0.11	0.001	106.3	1
Procymidone	Bactericides and Fungicides	1095	3	0.01	0.27	0.0028	97.9	2
Flusilazole	Bactericides and Fungicides	1043	1	0.01	0.10	0.002	47.9	3
Ethion	Insecticides, Acaricides, Nematicides	1099	1	0.01	0.09	0.002	45.5	4
Formetanate	Insecticides, Acaricides, Nematicides	603	1	0.4	0.17	0.004	41.5	5

Table 4.9. Top five risk ranking of pesticides sprayed on strawberries using the risk ratio method based on the MRL and the ARfD – the EU (EFSA, 2015a).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ARfD (mg/kg/bw)	Score	Rank
Formetanate	Insecticides, Acaricides, Nematicides	603	1	0.4	0.17	0.005	33.2	1
Procymidone	Bactericides and Fungicides	1095	3	0.01	0.27	0.012	22.8	2
Carbendazim	Bactericides and Fungicides	928	4	0.1	0.43	0.02	21.6	3
Flusilazole	Bactericides and Fungicides	1043	1	0.01	0.10	0.005	19.2	4
Fenthion	Insecticides, Acaricides, Nematicides	855	1	0.01	0.12	0.01	11.7	5

4.2.3 Comparison Dutch and EU data

In table 4.10., the 18 pesticides currently not included in the Dutch monitoring program for strawberries are listed. For chronic risks, the scores are quite high, except for ethion, formetanate and fenthion, which ranked fourth, fifth and eighth, respectively, in the EU risk ranking. For acute risks, formetanate and fenthion ranked first and fifth, respectively, in the EU risk ranking.

Table 4.10. Pesticides used on strawberries included in the monitoring program in the EU, but not currently included in the Dutch monitoring program.

Pesticide	Score chronic	Rank	Score acute	Rank
Abamectin	0.0	19	0.0	16
Captan+Folpet	0.0	24	0.0	19
Chlorantraniliprole	0.0	25	-	-
Diethofencarb	0.0	35	-	-
Dodine	0.0	39	0.0	29
Ethion	45.5	4	-	-
Etofenprox	0.0	41	0.0	31
Fenbutatin oxide	5.7	14	2.9	10
Fenthion	16.7	8	11.7	5
Fluquinconazole	0.0	49	0.0	37
Formetanate	41.5	5	33.2	1
Lufenuron	0.0	57	-	-
Pendimethalin	0.0	64	-	-
Propamocarb	0.0	67	0.0	50
Propyzamide	0.0	68	-	-
tau-Fluvalinate	0.0	77	0.0	55
Terbuthylazine	0.0	79	0.0	57
Thiamethoxam	0.0	81	0.0	59

* The pesticides with a "-" behind it did not have a score, because no ARfD was allocated or necessary and subsequently no score could be obtained.

4.2.4 MRL exceedance rates per country of origin

Sample results from strawberries from five different countries were reported (see supplementary material). The countries with the highest MRL exceedance rates were Spain and Morocco, with exceedances of 36.7 and 36.0 percent respectively. The countries with pesticides which exceeded the MRL the least for strawberries, were Belgium and the Netherlands, having exceeded the MRL in 9.1 and 9.2 percent of all samples respectively.

4.3 TOMATOES

4.3.1 Risk ranking Dutch data

In total, 102 different pesticides were detected (above the LOD) on tomatoes in the Netherlands. The five pesticides with the highest MRL exceedances were chlorothalonil (3.0 percent), cyprodinil (2.3 percent), methoxyfenozide (2.2 percent), pyrimethanil (1.7 percent) and iprodione (1.7 percent). For 10 pesticides an ADI was not established. Hence, these pesticides (i.e. DMST, EPN, ethirimol, fluvalinate, isocarbophos, nuarimol, propargite, sum of triadimenol and triadimefon, tetramethrin and triflumizole) were excluded from this risk ranking. For another 29 pesticides an ARfD was either not necessary or not allocated (EFSA, 2015a). Therefore, these pesticides (i.e. azoxystrobin, bifenazate, boscalid, bromopropylate, bupirimate, chlorantraniliprole, clofentezine, cyazofamid, cyprodinil, diethofencarb, ethion, fenamidone, fenhexamid, fludioxonil, hexaconazole, hexythiazox, iprodione, kresoxim-methyl, metrafenone, oxadixyl, pyridalyl, pyrimethanil, spinosad, tebufenozide, teflubenzuron, tetradifon, toclophos-methyl, trifloxystrobin and zoxamide) were excluded as well. For the risk ranking based on chronic risks, 55 out of 92 pesticides exceeded the MRL. From these 55 pesticides, chlorpyrifos scored the highest, having a score of 470.6, and chlorantraniliprole the lowest, having a score of 0.1. For the risk ranking based on acute risks, 39 out of 63 exceeded the MRL, with oxamyl scoring the highest (235.3) and pyriproxifen the lowest (0.1). The five pesticides scoring highest are listed in table 4.11 (chronic risks) and 4.12 (acute risks).

Table 4.11. Top five risk ranking of pesticides sprayed on tomatoes using the risk ratio method based on the MRL and the ADI – the Netherlands (KAP, 2004-2014).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ADI (mg/kg/bw/d)	Score	Rank
Chlorpyrifos	Insecticides, Acaricides, Nematicides	1275	6	0.5	0.47	0.001	470.6	1
Procymidone	Bactericides and Fungicides	1275	10	0.01*	0.78	0.0028	280.1	2
Oxamyl	Insecticides, Acaricides, Nematicides	1275	3	0.01*	0.24	0.001	235.3	3
Chlorothalonil	Bactericides and Fungicides	1275	38	6	2.98	0.015	198.7	4
Buprofezin	Insecticides, Acaricides, Nematicides	1275	19	1	1.49	0.01	149.0	5

* Indicates lower limit of determination

Table 4.12. Top five risk ranking of pesticides sprayed on tomatoes using the risk ratio method based on the MRL and the ARfD – the Netherlands (KAP, 2004-2014).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ARfD (mg/kg/bw)	Score	Rank
Oxamyl	Insecticides, Acaricides, Nematicides	1275	3	0.01*	0.24	0.001	235.3	1
Chlorpyrifos	Insecticides, Acaricides, Nematicides	1275	6	0.5	0.47	0.005	94.1	2
Carbendazim (including Benomyl)	Bactericides and Fungicides	1275	19	0.3	1.49	0.02	74.5	3
Procymidone	Bactericides and Fungicides	1275	10	0.01*	0.78	0.012	65.4	4
Methomyl	Insecticides, Acaricides, Nematicides	1275	2	0.02*	0.16	0.0025	62.8	5

* Indicates lower limit of determination

4.3.2 Risk ranking EU data

In total, 82 different pesticides were detected (above the LOD) on tomatoes. The MRL was most frequently exceeded for the following five pesticides: ethephon, procymidone, fenamiphos, diflubenzuron and carbendazim. All these pesticides had a prevalence below one percent. For two pesticides, ethirimol and propargite, an ADI was not defined. Therefore, they were excluded from the risk ranking. For the following 24 pesticides the ARfD was either not allocated or not necessary (EFSA, 2015a): clofentezine, azoxystrobin, boscalid, bromide ion bupirimate, chlorantraniliprole, cyprodinil, diethofencarb, diflubenzuron, ethirimol, fenhexamid, fludioxonil, hexythiazox, iprodione, iprovalicarb, kresoxim-methyl, lufenuron, mandipropamid, propargite, pyrimethanil, spinosad, teflubenzuron, trifloxystrobin and zoxamide. From the 80 pesticides included in the risk ranking based on chronic risks, 10 exceeded the MRL. From these 10 pesticides, fenamiphos had the highest score (142.2) and thiabendazole the lowest (0.8). For the risk ranking based on acute risks, 9 out of 63 pesticides exceeded the MRL, with fenamiphos having the highest score (45.5) and chlorpyrifos methyl the lowest (0.7). The top five of pesticides scoring highest for chronic and acute risks are shown in table 4.13 and 4.14 respectively.

Table 4.13. Top five risk ranking of pesticides sprayed on tomatoes using the risk ratio method based on the MRL and the ADI – the EU (EFSA, 2015a).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ADI (mg/kg/bw/d)	Score	Rank
Fenamiphos	Insecticides, Acaricides, Nematocides	879	1	0.04	0.11	0.0008	142.2	1
Chlorpyrifos-methyl	Bactericides and Fungicides	1406	1	0.5	0.07	0.001	71.1	2
Procymidone	Bactericides and Fungicides	1384	2	0.01	0.14	0.0028	51.6	3
Endosulfan	Insecticides, Acaricides, Nematocides	1183	1	0.05	0.08	0.006	14.1	4
Ethephon	Plant Growth Regulators	458	1	1	0.22	0.03	7.3	5

Table 4.14. Top five risk ranking of pesticides sprayed on tomatoes using the risk ratio method based on the MRL and the ARfD – the EU (EFSA, 2015a).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ARfD (mg/kg/bw)	Score	Rank
Fenamiphos	Insecticides, Acaricides, Nematocides	879	1	0.04	0.11	0.0025	45.5	1
Procymidone	Bactericides and Fungocides	1384	2	0.01	0.14	0.012	12.0	2
Endosulfan	Insecticides, Acaricides, Nematocides	1183	1	0.05	0.08	0.015	5.6	3
Carbendazim	Bactericides and Fungocides	1057	1	0.3	0.09	0.02	4.7	4
Ethephon	Plant Growth Regocides	458	1	1	0.22	0.05	4.4	5

4.3.3 Comparison Dutch and EU data

For tomatoes, 16 different pesticides were included in the EU monitoring program, which were not included in the Dutch monitoring program. As shown in table 4.15., fenamiphos and ethephon rank respectively first and second in the EU risk rankings for tomatoes based on both chronic and acute risks. For the risk ranking based on chronic risk, diflubenzuron ranked high as well in the EU risk ranking (ranked ninth).

Table 4.15. Pesticides used on tomatoes included in the monitoring program in the EU, but not currently included in the Dutch monitoring program.

Pesticide	Score chronic	Rank	Score acute	Rank
Bromide ion	0.0	15	-	-
Chlorfenapyr	0.0	19	0.0	13
Chlormequat	0.0	20	0.0	14
Cymoxanil	0.0	25	0.0	18
Cyromazine	0.0	28	0.0	20
Diflubenzuron	1.1	9	-	-
Dodine	0.0	34	0.0	25
Ethephon	7.3	5	4.4	5
Fenamiphos	142.2	1	45.5	1
Fenbutatinoxide	0.0	39	0.0	30
Iprovalicarb	0.0	51	-	-
Lufenuron	0.0	54	-	-
Mandipropamid	0.0	55	-	-
Propamocarb	0.0	61	0.0	44
tau-Fluvalinate	0.0	70	0.0	51
Thiamethoxam	0.0	76	0.0	56

* The pesticides with a "-" behind it did not have a score, because no ARfD was allocated or necessary and subsequently no score could be obtained.

4.3.4 MRL exceedance rates per country of origin

Sample results from tomatoes from six different countries were reported (see supplementary material). The country having the highest number of pesticides that exceeded the MRL was Spain, with a percentage of 40.4 of all pesticides tested. Belgium and the Netherlands scored the lowest, both having MRL exceedance rates below 20 percent. The remaining countries (Israel, Morocco and Turkey) had percentages of MRL exceedances varying between 22 and 23 percent.

4.4 GRAPES

4.4.1 Risk ranking Dutch data

On grapes, in total 137 different pesticides were detected (above the LOD). The five pesticides which exceeded the MRL most frequently were triadimenol, procymidone, methomyl, tolylfluanid and flusilazole, with the following prevalences respectively: 13.8, 4.8, 2.8, 2.3 and 2.0 percent. For the risk ranking based on chronic risks, 12 pesticides were excluded (i.e. 3-hydroxy-carbofuran, diniconazole, DMSA, DMST, ethirimol, fluvalinate, sum of malaoxon and malathion, methiocarb sulfoxide, propargite, pyrifenox, quinalfos and sum of triadimefon and triadimenol), because no ADI was defined, leaving 125 pesticides for analysis of chronic risks. For 46 pesticides, no ARfD was allocated or necessary: 2-phenylphenol, 3-hydroxy carbofuran, ametoctradin, azoxystrobin, benalaxyl, boscalid, bromopropylate, bupirimate, chlorantraniliprole, clofentezine, cyazofamid, cyprodinil, dichlofluanid, diethofencarb, diniconazole, DMSA, DMST, ethion, ethirimol, fenamidone, fenhexamid, fludioxonil, flufenoxuron, fluvalinate, hexaconazole, hexythiazox, iprodione, iprovalicarb, kresoxim-methyl, lufenuron, sum of malaoxon and malathion, methiocar sulfoxide, metrafenone, oxadixyl, pencycuron, propargite, pyrifenox, pyrimethanil, quinalfos, quinoxyfen, spinosad, spiridiclofen, sum of triadimefon and triadimenol, tebufenozide and trifloxystrobin. For the risk ranking based on chronic risk, 58 out of 125 pesticides exceeded the MRL. Carbofuran had the highest score of 3333.3 and kresoxim-methyl the lowest (0.2). For the risk ranking based on acute risk, 91 pesticides were included in the risk ranking. Of these 91 pesticides, 49 exceeded the MRL, with carbofuran scoring highest (3333.3) and dodemorph scoring lowest (0.1). In table 4.16 and 4.17 the top five of pesticides scoring highest based on chronic and acute risks respectively, are listed.

Table 4.16. Top five risk ranking of pesticides sprayed on grapes using the risk ratio method based on the MRL and the ADI – the Netherlands (KAP, 2004-2014).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ADI (mg/kg/bw/d)	Score	Rank
Carbofuran	Insecticides, Acaricides, Nematicides	2400	12	0.002*	0.50	0.00015	3333.3	1
Procymidone	Bactericides and Fungicides	2400	115	0.01*	4.79	0.0028	1711.3	2
Methomyl	Insecticides, Acaricides, Nematicides	2400	66	0.01*	2.75	0.0025	1100.0	3
Flusilazole	Bactericides and Fungicides	2400	47	0.01*	1.96	0.002	979.2	4
Omethoate	Insecticides, Acaricides, Nematicides	2400	6	0.01*	0.25	0.0003	833.3	5

* Indicates lower limit of determination

Table 4.17. Top five risk ranking of pesticides sprayed on grapes using the risk ratio method based on the MRL and the ARfD – the Netherlands (KAP, 2004-2014).

Pesticide	Group	Total # of samples	# of positive samples	MRL (mg/kg)	Percentage > MRL	ARfD (mg/kg/bw)	Score	Rank
Carbofuran	Insecticides, Acaricides, Nematicides	2400	12	0.002*	0.50	0.00015	3333.3	1
Methomyl	Insecticides, Acaricides, Nematicides	2400	66	0.01*	2.75	0.0025	1100.0	2
Procymidone	Bactericides and Fungicides	2400	115	0.01*	4.79	0.012	399.3	3
Flusilazole	Bactericides and Fungicides	2400	47	0.01*	1.96	0.005	391.7	4
Triadimenol	Bactericides and Fungicides	2400	330	0.01*	13.75	0.05	275.0	5

* Indicates lower limit of determination

4.4.2 Risk ranking EU data

In total, 90 different pesticides were detected (above the LOD) on grapes. The five pesticides which exceeded the MRL most frequently were chlormequat, ethephon, procymidone, mandipropamid and dithiocarbamates, with prevalences all below 1 percent. Three pesticides (diniconazole, ethirimol and propargite) were excluded, because no ADI was defined. The following 27 were excluded for not having an ARfD (either not allocated or not necessary): 2-phenylphenol, azoxystrobin, boscalid, chlorantraniliprole, cyprodinil, diniconazole, diphenylamine, ethion, ethirimol, fenamidone, fenhexamid, fludioxonil, flufenoxuron, hexythiazox, iprodione, iprovalicarb, kresoxim-methyl, mandipropamid, pendimethalin, propargite, pyrimethanil, quinoxyfen, spinosad, spiroiclofen, tebufenozide, trifloxystrobin and zoxamide. Monocrotophos (161.7) scored the highest in the risk ranking based on chronic risks, and diphenylamine scored the lowest (1.3) of the 15 pesticides exceeding the MRL (out of 87 pesticides included). For the risk ranking based on acute risk, 63 pesticides were included, of which 13 exceeded the MRL to some extent. Monocrotophos had the highest score (48.5) and cypermethrin the lowest (0.5). The top five of pesticides scoring highest based on chronic risks is shown in table 4.18 and the top five based on acute risks is shown in table 4.19.

Table 4.18. Top five risk ranking of pesticides sprayed on grapes using the risk ratio method based on the MRL and the ADI – the EU (EFSA, 2014).

Pesticide	Group	Total # of samples	# of positives	MRL (mg/kg)	Percentage > MRL	ADI (mg/kg bw p d)	Score	Rank
Monocrotophos	Insecticides, Acaricides, Nematicides	1031	1	0.01	0.10	0.0006	161.7	1
Procymidone	Bactericides and Fungicides	1161	3	0.01	0.26	0.0028	92.3	2
Ethion	Bactericides and Fungicides	1096	1	0.01	0.09	0.002	45.6	3
Dithiocarbamates	Plant Growth Regulators	752	1	5	0.13	0.004	33.2	4
Chlormequat	Insecticides, Acaricides, Nematicides	496	3	0.05	0.60	0.031	19.5	5

Table 4.19. Top five risk ranking of pesticides sprayed on grapes using the risk ratio method based on the MRL and the ARfD – the EU (EFSA, 2014).

Pesticide	Group	Total # of samples	# of positives	MRL (mg/kg)	Percentage > MRL	ARfD (mg/kg bw)	Score	Rank
Monocrotophos	Insecticides, Acaricides, Nematicides	1031	1	0.01	0.10	0.002	48.5	1
Procymidone	Bactericides and Fungicides	1161	3	0.01	0.26	0.012	21.5	2
Acrinathrin	Insecticides, Acaricides, Nematicides	981	1	0.05	0.10	0.01	10.2	3
Ethephon	Insecticides, Acaricides, Nematicides	393	2	1	0.51	0.05	10.2	4
Azinphos-methyl	Insecticides, Acaricides, Nematicides	1102	1	0.05	0.09	0.01	9.1	5

4.4.3 Comparison Dutch and EU data

For grapes, 10 different pesticides were included in the EU monitoring program, which are currently not included in the Dutch monitoring program. In table 4.8, it can be seen that for both chronic and acute risk, all pesticides have rather low scores.

Table 4.20. Pesticides used on grapes included in the monitoring program in the EU, but not currently included in the Dutch monitoring program.

Pesticide	Score chronic	Rank	Score acute	Rank
Bromuconazole	0.0	22	0.0	17
Diphenylamine	1.3	15	-	-
Esfenvalerate	0.0	37	0.0	30
Fenbutatinoxide	0.0	43	0.0	35
Fenpropathrin	0.0	46	0.0	37
Flutriafol	0.0	51	0.0	39
Mandipropamid	1.4	13	-	-
Pendimethalin	0.0	68	-	-
Propamocarb	0.0	70	0.0	53
Thiamethoxam	0.0	84	0.0	62

* The pesticides with a "-" behind it did not have a score, because no ARfD was allocated or necessary and subsequently no score could be obtained.

4.4.4 MRL exceedance rates per country of origin

For grapes, sample results from 13 countries were reported. Each of these countries showed exceedance of the MRL for one or more pesticides. The countries having the highest MRL exceedance rates are (in descending order): Turkey (67.5 percent), Italy (61.8 percent), Argentina (41.7 percent) and India (40.9 percent). Namibia, South Africa and Brazil had the lowest MRL exceedance rates, with percentages of 3.7, 7.6 and 7.9 respectively.

5 Discussion

This study estimated the risks related to pesticides based on their human health impact. The underlying aim of the study was to evaluate whether the current monitoring program for pesticides is still relevant based on human health risks, and if pesticides currently not monitored in the Netherlands should be included, or vice versa (pesticides can be excluded from the program due to low health implications). The assessment of the human health impact was performed by applying the risk ratio method for which occurrence and toxicity data were necessary. The underlying aim was addressed by comparing the national data (KAP) with the EU data. Pesticides not included in the national monitoring programs could not be examined in this research due to lack of data, and no statements about these pesticides could be made.

Which pesticides are most harmful to human health based on the KAP data, is based on several aspects. Firstly, the pesticides most hazardous differ per crop and per endpoint of toxicity (either acute or chronic risk). In addition, pesticides scoring high included in the EU monitoring program, but currently not included in the national monitoring program, are also risky pesticides which should be included in the national monitoring program. For apples, almost all pesticides included in the EU top five of the risk ranking based on chronic risks are monitored in the Netherlands as well, except for flusilazole. For the EU risk ranking based on acute risks, flusilazole and fenvalerate scored high, and are currently not part of the national monitoring program. Why these pesticides are currently not included in the national monitoring program, is not known. No data were available about pesticides not included in the monitoring program. It could be due to low occurrence rates or low exposure in the Netherlands. The toxicity of these pesticides (and the pesticides used on the other crops which are currently not included in the national monitoring program) are quite high, so that could not have been a reason for exclusion. The pesticides included in the EU monitoring program for strawberries, but currently not included the national monitoring program, were ethion (ranked fourth for chronic risk in the EU), formetanate and fenthion (ranked second and fifth for acute risk respectively). In tomatoes, pesticides which turned out to be risky based on the EU monitoring data, which are currently not monitored in the Netherlands, were fenamiphos and ethephon. For grapes, all pesticides in the top five of the risk rankings based on EU data were also included in the national monitoring program.

The riskiest pesticides (ranking fifth or higher) for all the crops examined are listed in The Black List (see supplementary material), established by Greenpeace Germany. This list comprises 451 active substances (39 percent of the 1150 substances analyzed) which are heavily toxic to human health and/or the environment. In 2008, the first Black List was issued and even though since then much progress has been made (a new regulation concerning authorization of pesticides was issued by the EU in November 2009), still about one-third of the pesticides on the Black List are approved in the EU (Worm & Vaupel, 2008; Reuter, Neumeister and Krautter, 2010). In addition, the number of pesticides ending up on the Black List is increasing. In 2008, 29 percent out of the 1134 pesticide ingredients analyzed (among which all relevant pesticides currently authorised for agricultural use worldwide) was classified as being hazardous based on several categories (e.g. carcinogenic, mutagenic, etc.), while in 2010, 39 percent out of 1150 active substances tested ended up on this list. On the other hand, the percentage of pesticides ending up on the “yellow list” have decreased since 2008. This list consists of active substances which could not be evaluated because of a lack of data from publicly available databases (Reuter, et al., 2010). The pesticides included in the top five of most hazardous pesticides for human health, which score particularly high (within the top 50) on the Black List are carbaryl, azinphos-methyl, carbendazim and dimethoate used on apples, methomyl used on strawberries, carbendazim used on tomatoes, and carbofuran, methomyl and omethoate used on grapes. Most of these pesticides are also used on other crops, but did not end up in the top five of those crops: carbaryl and azinphos-methyl are also used on grapes, chlorpyrifos is used on all crops examined, methomyl is also used on tomatoes, and dimethoate is used on strawberries as well. Of these pesticides, carbaryl, azinphos-methyl, carbofuran, omethoate and carbendazim are not approved in the EU (Annex 2).

The Draft List of Candidates for Substitution (Annex 3) is a list established by the European Commission, which consists of pesticides approved in the EU that have certain negative properties. Such pesticides are eligible for substitution if they meet one of the criteria set in Annex II, point 4 of Regulation (EC) No 1107/2009. Examples of these criteria are that the ADI, AOEL[9] or ARfD is considerably lower than other pesticides; that the substance is (very) bio-accumulative and/or persistent; or has mutagenic-, carcinogenic- or endocrine disrupting properties (EC, 2015). Several pesticides in this list are also listed in the risk rankings of one or more of the crops examined; carbendazim and methomyl (strawberries, tomatoes and grapes), and dimethoate (apples, strawberries and grapes), are candidates for substitution.

Which pesticides could be eliminated from the monitoring program, is difficult to determine. Pesticides which scored lowest are not necessarily the ones that should be eliminated. The following pesticides, for example, have a very high acute and/or chronic toxicity (concentration of ≤ 0.001 mg/kg/bw (per day)): omethoate (used on apples and grapes), parathion ethyl and diazinon (used on apples), monocrotophos (used on strawberries and grapes), dichlorvos, haloxyfop and methamidophos (used on strawberries), dimethoate (used on apples, strawberries and grapes), oxamyl (used on strawberries and tomatoes), carbofuran and fipronil (used on grapes) and chlorpyrifos (used on all crops examined). However, not all of these pesticides had high MRL exceedance rates in the crops studied and subsequently did not have high scores. However, due to their high toxicity, they are relevant to human health and should thus remain to be monitored. In addition, pesticide use has shown to differ significantly over a period of three years **(**Regulation (EU) No 788/2012). So only pesticides which have a low toxicity score and a low occurrence rate could be eligible for elimination. However, besides high acute toxicity, there are other factors which contribute to the toxicity of pesticides. Azoxystrobin (used on all crops examined), for example, has a low ADI and no ARfD, but it is very persistent and can remain in the environment for a long time. Pesticides which are persistent as well are imidacloprid, chlorpyrifos, chlorpyrifos-methyl and endosulfan (used on all crops examined). Endosulfan and chlorpyrifos are bio-accumulative as well, meaning that the pesticide accumulates in

[9] Acceptable operator exposure level: the maximum amount of an active substance to which an operator may be exposed without adverse health effects.

organisms (Reuter & Neumeister, 2010). Not all of these pesticides ranked high in the risk rankings because only the ADI and the ARfD were used as parameters for toxicity and not the persistence and bioaccumulation, which are also important factors, but out of the scope of the current study. Even though determining pesticides that could be eliminated is difficult, since not all factors influencing the toxicity of the pesticide were taken into account in this study, some cautious statements could be made based on the (relatively) low toxicity scores (> 1 mg/kg /bw (per day)) in combination with low occurrence rates (below 1 percent). For apples, chlorantraniliprole, etofenprox, fenoxycarb and pyriproxyfen met up to these criteria, since none of the mentioned pesticides exceeded the MRL. In addition, the toxicity scores were above 1 mg/kg/bw (per day). For the other crops, the following pesticides met up to the criteria: permethrin and spiromesifen for strawberries, chlorantraniliprole, pyriproxyfen, spiromesifen, permethrin, propamocarb hydrochlorid and etofenprox for tomatoes, and ametoctradin, chlorantraniliprole, profenofos, spiromesifen, etofenprox, fenoxycarb, permethrin, spirotetramat and pyriproxyfen for grapes.

Regarding the MRL exceedance rates in general, it can be said that they were quite high. Especially in comparison to the EU: for apples the MRL exceedance rate was 33.7 percent based on KAP data, while none of the EU apple samples exceeded the MRL, for strawberries it was 18.6 percent based on KAP data versus 2.9 percent in the EU, for tomatoes 16.2 percent versus 6.8 percent and for grapes 27.4 percent versus 3.5 percent (EFSA, 2015a). When comparing the MRL exceedance rates per crop between the Dutch and EU monitoring data, it can be concluded that for apples and tomatoes there are no similarities in the pesticides that exceed the MRL the most. For strawberries and grapes, only procymidone was amongst the pesticides which have the highest MRL exceedance rates in both the Netherlands and the EU (Annex 4). What might have caused these differences, is that many pesticides did not have an MRL and thus the default MRL of 0.01 mg/kg was used. This value is equal to the limit of quantification (LOQ), which means that if a certain pesticide is detected, it immediately exceeds the MRL. This has affected the Dutch data more than the EU data as can be seen in the risk ranking tables; for the pesticides ranking highest in the Netherlands there was often no MRL set and thus the default MRL was used. It could have also been the other way around; because default MRLs were used, the toxicity was relatively high, therefore these pesticides ended up in the top five of riskiest pesticides. This has probably also caused the overall scores to be higher for the Dutch data than for the EU data. For the pesticides in the EU risk ranking, there was an MRL set in most cases. One of the reasons for the fact that the Netherlands had more pesticides included for which no MRL was set, was because the national monitoring program analyzed on average 20 pesticides more per crop than the EU monitoring program. Another factor that might have caused differences between the Dutch and EU scores, is that MRLs have to be continually monitored, and changed whenever new data about risks become available, which prove an active substance to be more (or less) dangerous than previously thought (EC, 2005). However, KAP data was used of 2004 to 2014, while using MRLs of the year this study was conducted. For EU data the EFSA report of 2015 was consulted, which comprised data of 2013 and MRLs corresponding to these data. Thus, the Dutch monitoring data used in the current study are compared with the most recent MRLs and do not correspond to the MRLs which were in place at the time of sampling. This might have caused a slight change in the MRL exceedance rates, causing the exceeding rates to be either higher or lower than they actually are. Another factor that could have led to the differences between the Dutch and EU scores, is the assumption that the multi-methods used, tested for the presence of every pesticide. Although it was known in advance not to be the case, it was found to be too complicated to examine which multi-method tested for which distinct pesticides. The total number of samples, which are now the same for all pesticides used on a particular crop, should have possibly been lower for most pesticides. Hence, the percentages similar to, or above the MRL are most likely higher than the results show. However, since this assumption was made for all the Dutch data, it still gives a good indication of what pesticides most frequently exceed the MRL and which pesticides are the riskiest to human health.

Regarding the MRL exceedances per country, it can be concluded that for apples the highest MRL exceedances[10] can be found in apples imported from countries outside the EU; the samples with the highest exceedances came from South Africa, Chili and Brazil. For the other four crops, however, countries within the EU belonged to the countries having the highest exceedances as well. This is in contrast with what is stated in the EFSA pesticide report of 2015 and the Dutch Food Safety report of 2014, where the results showed that

[10] MRL exceedance is based on the total level of exceedance and not based on the number of pesticides exceeding the MRL.

MRL exceedances were more common in non-EU countries (EFSA, 2015a; NVWA, 2014). What might have caused this discrepancy, is unclear. Countries exceeding the MRL the most, get stricter import controls, which lead to better compliance with the requirements. However, this does not explain the differences between the Dutch and EU MRL exceedance rates (NVWA, 2014). Spain was the country having the highest MRL exceedance for tomatoes and Turkey and Italy exceeded the MRL the most for grapes. On the other hand, the countries exceeding the MRL the least also consisted of EU countries as well as countries outside the EU. Where Italy had the highest MRL exceedance for grapes, it had, together with New Zealand, the lowest MRL exceedance for apples. And where Chili, Brazil and South Africa had the highest MRL exceedance for strawberries, they had, as well as Namibia, the lowest MRL exceedance for grapes. The countries exceeding the MRL the least for strawberries and tomatoes were Belgium and the Netherlands. Thus, the exceedances are not necessarily related to a country, but they are rather crop specific.

In some countries, the MRL exceedance rates were quite high (up to 80 percent) and thus a reason for concern. Even though the MRL is a measure of good agricultural practice (GAP) and not necessarily a safety limit, exceeding this limit to a large extent can be harmful to human health (FAO, z.j.). Furthermore, the pesticides that fell within legal limits (not exceeding the MRL) could be a threat as well, due to the fact that often a diverse range of pesticides is used on a crop (Greenpeace, 2015a). We are exposed to a mixture of pesticides every day. Not only can one crop be treated with multiple pesticides (which is the case for roughly one-third of all food commodities[11]), we also eat more than for example just strawberries in a day. Lots of fruits and vegetables are consumed, which are (if not organic) all treated with (multiple) synthetic chemical pesticides (EFSA, 2015b). All these pesticides could have left residues on these products, which are consumed on a daily basis. According to the EFSA pesticide residue report of 2014 and 2015 crops containing multiple residues (ranging from two to more than six residues) were mostly found on, among others, strawberries (12 percent of the samples tested contained more 6 residues), table grapes and apples, with percentages of 63.0, 59.6 and 46.0 respectively (EFSA, 2015a; EFSA, 2014). A Greenpeace study about pesticide residues on apples showed that 60 percent of the 126

[11] Food commodities tested by EFSA: Apples, head cabbage, leek, lettuce, peaches, strawberries, tomatoes, oats rye, wine, cow’s milk and swine meat.

apples tested, contained two or more pesticides of which some were regarded as being highly persistent and likely to bio-accumulate (Greenpeace, 2015b). The accumulative and synergistic impacts of these pesticides together on the human body (or the environment for that matter) are still unknown. It is difficult to determine the impact since there is a great variety of pesticides used and a great variety of diets (not only between countries but also between the population within a country) as well. In addition, the residues are usually too small to cause acute health effects, but could lead to chronic effects, which are more difficult to examine.

Although the risk ranking gives a good indication of what pesticides are risky to human health, the problem might be larger than what is portrayed here. A report published by Greenpeace showed that there are so-called "hidden" pesticides as well; pesticides which cannot be detected with the routine methods commonly used. Laboratories use both single- and multi-methods to test for pesticides, but only a selection thereof is used, due to capacity or resource limitations or because of the fact that it is not practicable for routine testing. The number of pesticides that can be tested by a multi-method by top laboratories lies between 400 and 616 pesticides. However, because of the abovementioned reasons, the number that can be realistically tested lies between 250 and 500 pesticides. For average laboratories, this number is even lower; they can cover up to 200 pesticides (EFSA, 2015a; Worm & Vaupel, 2008). Thus, more pesticides could be detected, but because of limitations, this is not the case. And even when all available analytical possibilities are considered, only 30 to 46 percent of the 1350 pesticides known and used worldwide can be detected, even by top laboratories. Moreover, a great number of the pesticide components (around 16 percent) cannot be assessed due to a lack of publicly available toxicity data. This implies that next to the dangers to not only human health, but also the environment, there are possible dangers of pesticide contamination of food, ground water and soil, that cannot be identified thoroughly (Worm & Vaupel, 2008).

6 Conclusion

In conclusion, it can be said that the national monitoring program is still relevant for the most part. A large part of pesticides currently included have either high chronic or acute toxicity. In addition, many of the pesticides which turned out to be amongst the riskiest pesticides, are listed on the Black List as well, meaning that they should be monitored. For apples, diazinon, dimethoate, azinphos-methyl, carbaryl, tolylfluanid and carbendazim scored the highest. For strawberries, dichlorvos, methomyl, procymidone, oxamyl, flutolanil and monocrotophos were listed in the top five of riskiest pesticides. Chlorpyrifos, procymidone, oxamyl, chlorothalonil, buprofezin, carbendazim and methomyl were the riskiest pesticides in tomatoes. And for grapes, carbofuran, procymidone, methomyl, flusilazole, omethoate and tridimenol were the riskiest for human health. Pesticides which are currently not included in the national monitoring program and turned out to be risky based on the EU risk ranking performed in this study, based on both chronic and acute risks, were flusilazole and fenvalerate (used on apples), ethion, formetanate and fenthion (used on strawberries), and ethephon and fenamiphos (used on tomatoes). Pesticides that could possibly be excluded from the national monitoring program are pesticides with both a low toxicity score and a low occurrence rate, being chlorantraniliprole, etofenprox, fenoxycarb and pyriproxyfen for apples, permethrin and spiromesifen for strawberries, chlorantraniliprole, pyriproxyfen, spiromesifen, permethrin, propamocarb hydrochlorid and etofenprox for tomatoes, and ametoctradin, chlorantraniliprole, profenofos, spiromesifen, etofenprox, fenoxycarb, permethrin, spirotetramat and pyriproxyfen for grapes.

The MRL exceedance rates per country of origin for pesticides used on apples were higher for countries outside the EU than inside the EU. However, for the other crops, also countries within Europe belonged to the countries with the highest MRL exceedance rates for the total amount of pesticides used on the crops. Thus, the MRL exceedance rates seem to be rather crop specific instead of country specific.

6.1 Recommendations

In future research, risk rankings could be performed for other crops (for example peppers) in which factors like persistence of pesticides in the environment and bioaccumulation of pesticides could be taken into account as well. Furthermore, monitoring data should be compared to the MRLs which were in place at the time of sampling, and which multi-method tests for the presence of which pesticides should also be considered in order to gain more accurate results.

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ANNEX 1. Decision tool to assist the choice of risk ranking methodology for multiple hazards in food

Question	Answer	Indicate options	Risk Assessment'	Comparative Risk Assessment'	Ratio (Exposure/ Effect)'	Scoring method	Cost of Illness'	DALY/QALY'	VTP'	MCDA'	Risk Matrix'	Flow charts /Decision trees'	Expert Judgment'
RISK MANAGER PREREQUISITIVES (Several answers per question acceptable)							**MODEL CAPACITY**						
Amount of person hours resources available?	High		1	1	1	1		1	1	1	1	1	1
	Moderate	1			1	1	1	1		1	1	1	1
	Low										1	1	1
Level of ouput needed	Quantitative	1	1	1			1	1	1				
	Semi-quantitative	1			1	1	1	1	1	1	1		
	Qualitative										1	1	1
Is it important that the methodology can be easily communicated to stakeholders (laymen)?	Yes	1			1	1	1			1	1		1
	No		1	1	1	1	1	1	1	1	1	1	1
Should stakeholder perception be included?	Yes					1			1	1		1	1
	No	1	1	1	1	1	1	1	1	1	1	1	1
Should uncertainty be included?	Yes		1	1	1	1	1	1	1	1			1
	No	1	1	1	1	1	1	1	1	1	1	1	1
Should weights be included for the various risk ranking criteria?	Yes					1				1			1
	No	1	1	1	1	1	1	1	1	1	1	1	1
Should human incidences be included?	Yes		1	1			1	1	1	1		1	1
	No	1	1		1	1			1	1	1	1	1
Should economic impact be included?	Yes						1		1	1		1	1
	No	1	1	1	1	1		1		1	1	1	1
Preferred method of communication	Graph		1	1			1	1	1		1		
	Tabulation	1	1	1	1	1	1	1	1	1			1
	Decision Tree											1	

DATA AVAILABILITY (only one answer per question acceptable)			MODEL REQUIREMENTS										
Human incidence data available?	Yes		1	1	1	1	1	1	1	1	1	1	1
	No	1			1	1			1	1	1	1	1
Dose-response data available?	Yes	1	1	1	1	1	1	1	1	1	1	1	1
	No				1	1	1	1	1	1	1	1	1
Occurrence data (concentration, prevalence, dose) available?	Yes	1	1	1	1	1	1	1	1	1	1	1	1
	No						1	1	1	1	1	1	1
Food consumption data available?	Yes	1	1	1	1	1	1	1	1	1	1	1	1
	No						1	1	1	1	1	1	1
Growth models available? (if no microbiological hazards are ranked, please indicate: not applicable)	Yes		1	1	1	1	1	1	1	1	1	1	1
	No						1	1	1				
	Not applicable	1											
Toxicological reference values (ADI, TDI, etc.) available? (if no chemical hazards are ranked, please indicate: not applicable)	Yes	1	1	1	1	1	1	1	1	1	1	1	1
	No					1	1	1	1	1	1	1	1
	Not applicable												
EVALUATION			**Risk Assessment***	**Comparative Risk Assessment***	**Ratio (Exposure/ Effect)***	**Scoring method**	**Cost of Illness***	**DALY/QALY***	**WTP***	**MCDA***	**Risk Matrix***	**Flow charts /Decision trees***	**Expert Judgment***
			Prerequisites not fulfilled Data available	Prerequisites not fulfilled Data available	Prerequisites fulfilled Data available	Prerequisites fulfilled Data available	Prerequisites not fulfilled Data available	Prerequisites not fulfilled Data available	Prerequisites not fulfilled Data available	Prerequisites fulfilled Data available	Prerequisites not fulfilled Data available	Prerequisites not fulfilled Data available	Prerequisites not fulfilled Data available
METHOD COMMENTS					**Possibly appropiate method**	**Possibly appropiate method**				**Possibly appropiate method**			
					Resources: High Moderate Accuracy: Semi-quantitative Uncertainty included Outputs: Tabulation	Resources: High Moderate Accuracy: Semi-quantitative Uncertainty included Outputs: Tabulation				Resources: High Moderate Accuracy: Semi-quantitative Uncertainty included Outputs: Tabulation User-friendly tools may be availabe			

ANNEX 2. List of approved and non-approved substances in the EU (European Commission, 2015b)

2-Phenylphenol	Approved	Clopyralid	Approved
3-Hydroxy-Carbofuran	?	Clothianidin	Approved
Acephate	Not approved	Cyazofamid	Approved
Acetamiprid	Approved	Cyfluthrin	Not approved
Acrinathrin	Approved	Cymoxanil	Approved
Ametoctradin	Approved	Cypermethrin	Approved
Azinphos-Methyl	Not approved	Cyproconazole	Approved
Azoxystrobin	Approved	Cyprodinil	Approved
Benalaxyl	Approved	Deltamethrin	Approved
Bifenazate	Approved	Desmethylpirimicarb	Approved
Bifenthrin	Approved	Diazinon	Not approved
Bitertanol	Not approved	Dichlofluanid	Not approved
Boscalid	Approved	Dichlorvos	Not approved
Bromopropylate	Not approved	Dicloran	Not approved
Bromuconazole	Approved	Dicofol	Not approved
Bupirimate	Approved	Dicrotophos	Not approved
Buprofezin	Approved	Diethofencarb	Approved
Captan	Approved	Difenoconazole	Approved
Carbaryl	Not approved	Diflubenzuron	Approved
Carbendazim	Not approved	Dimethoate	Approved
Carbendazim (including Benomyl)	?	Dimethomorph	Approved
Carbofuran	Not approved	Diniconazole	Approved
Carbosulfan	Not approved	Diphenylamine	Not approved
Chlorantraniliprole	Approved	Dithianon	Approved
Chlorfenapyr	Not approved	Dithiocarbamates	Approved
Chlormequat	Approved	DMSA	?
Chlorothalonil	Approved	DMST	Approved
Chlorpropham	Approved	Dodemorph	Approved
Chlorpyrifos	Approved	Dodine	Approved
Chlorpyrifos-Methyl	Approved	Endosulfan	Not approved
Chromafenozide	Approved	Endosulfan-Alpha	?
Clofentezine	Approved	Endosulfan-Sulphate	?

EPN	Not approved	Flutolanil	Approved
Epoxiconazole	Approved	Flutriafol	Approved
Ethephon	Approved	Fluvalinate	Approved
Ethion	Approved	Folpet	Approved
Ethirimol	Not approved	Haloxyfop	Not approved
Ethoprophos	Approved	Hexaconazole	Not approved
Ethoxyquin	Not approved	Hexythiazox	Approved
Etofenprox	Approved	Imazalil	Approved
Etoxazole	Approved	Imidacloprid	Approved
Famoxadone	Approved	Indoxacarb	Approved
Fenamidone	Approved	Iprodione	Approved
Fenarimol	Not approved	Iprovalicarb	Approved
Fenazaquin	Approved	Isocarbophos	Approved
Fenbuconazole	Approved	Isoprothiolane	Not approved
Fenhexamid	Approved	Kresoxim-Methyl	Approved
Fenitrothion	Not approved	Lambda Cyhalothrin	Approved
Fenobucarb	Not approved	Lufenuron	Approved
Fenoxycarb	Approved	Malaoxon/Malathion (sum)	?
Fenpropathrin	Not approved	Malathion	Approved
Fenpropimorph	Approved	Mepanipyrim	Approved
Fenpyroximate	Approved	Metalaxyl	Approved
Fenthion	Not approved	Methamidophos	Not approved
Fenthion Sulfoxide	?	Methidathion	Not approved
Fenvalerate	Not approved	Methiocarb	Approved
Fipronil	Approved	Methiocarb Sulfone	?
Flonicamid	Approved	Methiocarb Sulfoxide	Approved
Fluazifop-Butyl	Approved	Methomyl	Approved
Flubendiamide	Approved	Methoxyfenozide	Approved
Fludioxonil	Approved	Metrafenone	Approved
Flufenoxuron	Not approved	Monocrotophos	Not approved
Fluopicolide	Approved	Myclobutanil	Approved
Fluopyram	Approved	Nuarimol	Not approved
Flusilazole	Not approved	Omethoate	Not approved

Oxadixyl	Not approved
Oxamyl	Approved
Oxamyl Oxime	Approved
Parathion	Not approved
Parathion-Methyl	Not approved
Penconazole	Approved
Pencycuron	Approved
Permethrin	Not approved
Phenthoate	Not approved
Phosalone	Not approved
Phosmet	Approved
Piperonyl Butoxide	Approved
Pirimicarb	Approved
Pirimicarb-Desmethyl-Formamido	?
Pirimiphos-Methyl	Approved
Prochloraz	Approved
Procymidone	Not approved
Profenofos	Not approved
Propamocarb Hydrochlorid	Approved
Propargite	Not approved
Propiconazole	Approved
Proquinazid	Approved
Pymetrozine	Approved
Pyraclostrobin	Approved
Pyridaben	Approved
Pyridalyl	Approved
Pyridaphenthion	Not approved
Pyrifenox	Not approved
Pyrimethanil	Approved
Pyriproxyfen	Approved
Quinalfos	Approved
Quinoxyfen	Approved
Spinetoram	Approved
Spinosad	Approved
Spirodiclofen	Approved
Spiromesifen	Approved
Spirotetramat	Approved
Spiroxamine	Approved
Sum of Triadimefon and Triadimenol	Approved
Tebuconazole	Approved
Tebufenozide	Approved
Tebufenpyrad	Approved
Teflubenzuron	Approved
Tetraconazole	Approved
Tetradifon	Not approved
Tetramethrin	Not approved
Thiabendazole	Approved
Thiacloprid	Approved
Thiametoxam	Approved
Thiophanate-Methyl	Approved
Tolclofos-Methyl	Approved
Tolylfluanid	Not approved
Triadimefon	Not approved
Triadimenol	Approved
Triazophos	Not approved
Trichlorfon	Not approved
Tricyclazole	Pending
Trifloxystrobin	Approved
Triflumizole	Approved
Vamidothion Sulfoxide	Approved
Vinclozolin	Not approved
Zoxamide	Approved

ANNEX 3. The Draft List of Candidates for Substitution, European Commission January 2015

(1) 1-methylcyclopropene

(2) aclonifen

(3) amitrole

(4) bifenthrin

(5) bromadiolone

(6) bromuconazole

(7) carbendazim

(8) chlorotoluron (unstated stereochemistry)

(9) copper compounds (variants copper hydroxide, copper oxychloride, copper oxide, Bordeaux mixture and tribasic copper sulphate)

(10) cyproconazole

(11) cyprodinil

(12) diclofop

(13) difenacoum

(14) difenoconazole

(15) diflufenican

(16) dimethoate

(17) dimoxystrobin

(18) diquat

(19) epoxiconazole

(20) esfenvalerate

(21) ethoprophos

(22) etofenprox

(23) etoxazole

(24) famoxadone

(25) fenamiphos

(26) fenbutatin oxide

(27) fipronil

(28) fludioxonil

(29) flufenacet

(30) flumioxazine

(31) fluometuron

(32) fluopicolide

(33) fluquinconazole

(34) glufosinate

(35) haloxyfop-P

(36) imazamox

(37) imazosulfuron

(38) isoproturon

(39) isopyrazam

(40) lambda-cyhalothrin

(41) lenacil

(42) linuron

(43) lufenuron

(44) mecoprop

(45) metalaxyl

(46) metam

(47) metconazole

(48) methomyl

(49) metribuzin

(50) metsulfuron-methyl

(51) molinate

(52) myclobutanil

(53) nicosulfuron

(54) oxadiargyl

(55) oxadiazon

(56) oxamyl

(57) oxyfluorfen

(58) paclobutrazol

(59) pendimethalin

(60) pirimicarb

(61) prochloraz

(62) profoxydim

(63) propiconazole

(64) propoxycarbazone

(65) prosulfuron

(66) quinoxyfen

(67) quizalofop-P (variant quizalofop-P-tefuryl)

(68) sulcotrione

(69) tebuconazole

(70) tebufenpyrad

(71) tepraloxydim

(72) thiacloprid

(73) tri-allate

(74) triasulfuron

(75) triazoxide

(76) warfarin

(77) ziram

ANNEX 4. Comparison EU and NL of highly risky pesticides based on MRL exceedance rates

	EU	**The Netherlands**
Apples		
	Methomyl	Tolylfluanid
	Dimethoate	Diphenylamine
		DMST
		Propargite
		Azinphos-methyl
Strawberries		
	Carbendazim	Tolylfluanid
	Propiconazole	Triadimenol
	Trifloxystrobin	Methomyl
	Procymidone	Procymidone
		DMST
Tomatoes		
	Procymidone	Chlorothalonil
		Cyprodinil
		Methoxyfenozide
		Pyrimethanil
		Iprodione
Grapes		
	Folpet	Triadimenol
	Chlormequat	Procymidone
	Ethephon	Methomyl
	Procymidone	Tolylfluanid
		Flusilazole