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Evaluation of the Results of Pesticide Residue Analysis in Food Sampled between 2017 and 2021

Árpád Ambrus [1,*], Adrienn Vásárhelyi [2], Géza Ripka [3], Henriett Szemánné-Dobrik [4] and Júlia Szenczi-Cseh [5]

1 Doctoral School of Nutrition and Food Sciences, University of Debrecen, 4032 Debrecen, Hungary
2 Directorate of Agricultural Genetic Resources, National Food Chain Safety Office, 1118 Budapest, Hungary; vasarhelyia@nebih.gov.hu
3 Directorate of Plant Protection and Oenology, National Food Chain Safety Office, 1118 Budapest, Hungary; ripkag@nebih.gov.hu
4 Food Chain Safety Centre, Non-Profit Ltd., Pesticide Residue Analytical Laboratory, 3526 Miskolc, Hungary; szemanneh@nebih.gov.hu
5 Freelancer Food Safety Adviser, 1116 Budapest, Hungary; julia_cseh@yahoo.de
* Correspondence: ambrusadr@yahoo.co.uk

Abstract: As mandated by the EU and the national risk management duties, pesticide residues were determined by four specialized laboratories in 9924 samples taken from 119 crops of economic importance in Hungary and imported foodstuffs during 2017–2021. The screening method applied covered 622 pesticide residues as defined for enforcement purposes. The limit of detection ranged between 0.002 and 0.008 mg/kg. The 1.0% violation rate concerning all commodities was lower than in the European Union. No residue was detectable in 45.9% of the samples. For detailed analyses, six commodities (apple, cherry, grape, nectarine/peach, sweet peppers, and strawberry) were selected as they were analyzed in over 195 samples and most frequently contained residues. Besides testing their conformity with national MRLs, applying 0.3 MRL action limits for pre-export control, we found that 73% of the sampled lots would be compliant with ≥90% probability based on a second independent sampling. Multiple residues (2–23) in one sample were detected in 36–50% of the tested lots. Considering the provisions of integrated pest management, and the major pests and diseases of selected crops, normally three to four and exceptionally, seven to nine active ingredients with different modes of action should suffice for their effective and economic protection within four weeks before harvest.

Keywords: pesticides; multiple residues; plant protection; monitoring results; compliance assessment

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1. Introduction

Many cultivated plants, used as food, feed, or industrial raw material, must be protected from arthropod pests, diseases and weeds. Chemical substance-based pesticides and micro-organisms are used for their protection. The popularity of the so-called bio-products, cultivated with limited or no pesticide use, is on the rise. However, their proportion in the total production is low. According to the available information, bio-farming was conducted in 9.2% of the whole agricultural area within the European Union (in Hungary about 6%) in 2020 [1].

Because pesticides are generally toxic substances, their authorization and use are strictly regulated worldwide. In the European Union (EU) the European Parliament (EP) and the Council or the Council alone, issue regulations concerning the place of chemical and micro-biological plant protection products on the market [2–6].

Each Member State shall take a sufficient number and range of samples ensuring that they the results are representative of the market. The process should take into account, the results of previous control programs. Such sampling shall be carried out as close to the point of supply as is reasonable, to allow for subsequent enforcement action to be taken [6].

Owing to legal obligations and vested interest, many national authorities regularly monitor the pesticide residues in food and feed products. For example, extensive control is in place and the results are published by Austria, Australia, Germany, Japan and the USA [7–11]. The main objectives of the programs, for instance, are to provide data and information for testing the compliance of marketed foodstuffs with the legal limits, preventing marketing products with unacceptable residues, performing dietary exposure assessment, and managing the identified risks [12,13].

Besides the nationwide monitoring programs, researchers often determine the pesticide residues in/on specialty crops or specific groups of plant commodities [14–18], including feed [19], and fish [20,21]. They perform dietary risk assessment based on the residue levels found and corresponding food consumption data. Moreover, various non-profit activity groups, such as the Environmental Working Group (EWG) in the USA conduct surveys of pesticide residues and other toxic chemicals in food and environmental samples to provide information needed to make "smart, healthy choices" [22]. The EWG recently published the list of "dirty dozen" and "clean 15" commodities based on the frequency and concentration of detected pesticide residues. Our findings in Hungary largely agree with those of EWG.

In addition to the national control programs, the European Commission (EC) has selected certain foodstuffs that constitute major components of the diet in which pesticide residues should be monitored since 2009. The changes in residue levels are monitored within the compulsory coordinated multiannual control programs [23]. The European Food Safety Authority (EFSA) evaluates the results of the national and coordinated monitoring programs. The results show that out of 96,302 and 88,141 samples 2.3% and 3.6% were non-compliant in 2019 and 2020, respectively. Based on the acute and chronic risk assessment it was concluded that the residue levels are unlikely to pose any concern for consumer health [24,25].

Despite the low frequency of residues exceeding the MRLs, 40% of European citizens consider pesticide residues in food as a health risk [26].

To test conformity with MRLs, the residues defined for enforcement purposes [27,28] should be determined in the portion/part of the commodity to which the MRLs apply, and which is analyzed [6,29]. The test portion should represent the composite sample containing the specified minimum number of primary samples taken from the sampled material [30,31]. For the evaluation of the test results the measurement uncertainty should always be considered according to ISO Standard 17025 and Codex GL [32,33]. Where the compliance of locally marketed products is tested, the combined relative uncertainty of the within laboratory reproducibility (CV_L) [34] or the 0.25 default value introduced by the EU [35] should be used. For the control of imported products, specific import MRLs apply, if available. However, when the results of pre-export control are evaluated, the combined uncertainty of the whole process, including that of sampling, ought to be considered [36] in combination with a properly selected action limit [37–39].

The sampling uncertainty was determined based on analyses of over 10,000 duplicate supervised trial results [38,40]. The practical application of the action limit was explained in detail in a recent article [36].

The objectives of this article are to present the summary results of the Hungarian national pesticide residue monitoring conducted between 2017 and 2021, and to evaluate, as an example, the compliance of six selected commodities with the Hungarian (EU) MRLs, and to predict the potential acceptability of the sampled lots if they were exported to the EU. Furthermore, we critically review the plant protection practice that resulted in multiple residues detected in the selected crops. However, we do not discuss the analytical methods for the determination of pesticide residues.

2. Materials and Methods

2.1. Sampling

The sampling plan was prepared by the Central Office of the Hungarian National Food Chain Safety Office (NFCSO) considering, in general, the principles of risk-based monitoring programs [41] and the coordinated multi-annual control plan of the European Commission [23,42].

The plant protection or quarantine inspectors took samples at farm gates, border control points and in wholesale markets or large supermarkets over the whole country. The specified number of primary samples and the minimum mass of the composite sample were collected from randomly selected positions according to the Codex [30] and EC sampling standard/instruction [31]. Once collected, the samples were transported to the laboratories in cooled transport vans. The sampling records were directly uploaded to the central online database. The laboratory staff could download and insert the data relevant to the analyses of samples in the laboratory sample registry book [36]. The authorized officials of NFCSO undertook the necessary official control actions. The system allows authorized personnel to access records from their offices thereby enabling real-time observation of operational progress. Moreover, it eliminates the need for repeated manual data entry and potential errors.

2.2. Analyses of Pesticide Residues

Four laboratories of NFCSO were involved in the analyses of pesticide residues in plant commodities in 2017–2021. The laboratories considered the samples as having an unknown pesticide treatment history even if the pesticide applications were indicated on the sampling record sheet. Altogether, over 9000 samples comprised of fruits, vegetables, cereals, and baby food were analyzed. The scope of the screening included 465 pesticide active substances and their metabolites as defined for enforcement purposes. The limit of detection ranging from 0.002 to 0.008 mg/kg enabled detection of any unauthorized use of pesticides also. The laboratories applied different versions of the QuEChERS methodology in combination with LC–MS/MS and GC–MS/MS detection depending on the physicochemical properties of the residues [43–45]. As a function of the water content of the sample matrix, additional water is added to the 5–10 g portions of the sample material and homogenized thoroughly with dry ice and then extracted with 10 mL acetonitrile. Details of the basic procedures applied for fruits and vegetables as well as for cereal grains are given in our previous publication [46]. The pesticide residues were divided into subgroups depending on the methods and detection conditions applied. Some very polar compounds such as glyphosate and glufosinate and some others such bromide-ion and dithiocarbamates required single residue methods.

The laboratories worked in coordination and shared the tasks of method validation, performance verification, and confirmation of critical results. However, the rolling program of the recovery tests were carried out in each laboratory at the LOQ and MRL levels. The criteria for the acceptable performance parameters established by the European Commission [35] were the basis of their internal quality control. The performance of the laboratories was verified by their good results achieved in the European Proficiency tests (Table 1), similar to that reported previously [47].

Table 1. Summary of the results obtained in EP proficiency tests.

Year	PT Code	Test Material	No. of Components/ No. of Residues	No. of Participating Labs	AZ^2 * Range
2019	EUPT-CF13	Rye	192/19	157	0.1
	EUPT-FV21	Red cabbage	237/21	188	0.1–0.4
	EUPT-SM11	Red cabbage	not identified no./16	67	50–93% **

Table 1. *Cont.*

Year	PT Code	Test Material	No. of Components/ No. of Residues	No. of Participating Labs	AZ^2 * Range
2020	EUPT-CF14	Rice	202/20	158	0.1–1.3
	EUPT-FV22	Onion	244/19	176	0.2–1.1
	EUPT-SRM15	Rice	30/16	60	0.5–1.1
	EUPT-SM12	Onion	not identified no./17	62	76% **
2021	EUPT-CF15	Rapeseed cake	213/22	137	0.3–2.0
	EUPT-FV23	Aubergine	256/20	182	0.2–1.3
	EUPT-SRM16	Sesame	21/13	132	0.3–0.6
	EUPT-SM13	Aubergine	not identified no./18	60	78–89% **

* Average of the Squared Z-scores (Combined Z-scores). ** The range of the percentage of qualified residues.

2.3. Assessment of Compliance with Legal Limits (MRL)

For making a fair decision on the compliance of a sampled lot with the relevant MRLs, the uncertainty of measured residues should always be considered as per ISO Standard 17025 [32]. The practical application of the principles is explained in detail by Ambrus et al. [34].

There are two principally different situations:

(a) the sampled lot is intended for the local market;
(b) the lot is sampled before export.

Case (a): when a commodity is placed on the local market the average residue content of the tested composite sample (R) should be equal or lower than the corresponding MRL taking into account the expanded within laboratory reproducibility relative standard deviation (CV_L):

$$R - 2 \times R \times CV_L \leq MRL \tag{1}$$

If the residue calculated with the expanded uncertainty (Equation (1)) exceeds the MRL, the sampled lot should not be marketed.

However, the European Union only rejects an imported product if the measured residue (R′) adjusted with its combined relative uncertainty exceeds the MRL. For facilitating uniform decisions, a default among laboratories relative reproducibility of 0.25 is used within the EU [35].

$$R' - 2 \times 0.25 \times R \geq MRL \tag{2}$$

It practically means that the sampled lot would only be accepted if the measured residue, R′, is equal to or less than two times the MRL.

Applying this rule, the probability of wrongly rejecting a lot by the importing country is about 2.3–2.5% which is a fair treatment according to the principles of Codex GL on settling dispute [33].

EFSA applies the same principle and distinguishes cases of exceedance of MRL in the evaluation of monitoring data. For example, in 2020 the analyses of 88,141 samples were reported. The residues exceeded the MRL in 5.1% of the samples of which 3.6% were non-compliant after taking the expanded measurement uncertainty into account [25].

In Case (b) the compliance of the exported commodity will be decided by the importing country based on the analyses of an independently taken composite sample at the border control point. Consequently, the likely upper 95–98% tail of the distribution of the residues in repeated composite samples should be predicted and compared to the MRL of the importing county to make sure that the exported lot will be accepted. Therefore, for pre-export control the sampling uncertainty should also be accounted for in the combined uncertainty of the whole determination process (CV_R) [34,37]. For this reason, an action limit (AL) lower than the MRL should be used as the acceptance criterion.

$$AL + k \times CV_R \times AL = MRL \tag{3}$$

$$AL = \frac{MRL}{1 + k \times CVR} \tag{4}$$

The value of k is contingent upon the targeted compliance level that is typically 95–98%.

Applying an action limit for facilitating compliance with export MRLs is a relatively new approach. In addition to pesticide residues [36,48], it was recently applied for mycotoxins [49] and gluten in oat groats [39]. In view of its applicability for three different analyte–matrix combinations, its use can be generally recommended during pre-marketing control.

Based on the evaluation of over 10,000 supervised trial results, Farkas and co-workers [38,41] concluded that a default action limit should be chosen at around 0.3 MRL to assure with about 95–98% probability that the sampled product would be accepted in the EU, taking into account the decision rule specified with equation 2.

The pre-export evaluation of residues in a tested commodity is illustrated with the example of acetamiprid residues in apple (MRL = 0.4 mg/kg). Figure 1 shows the operation characteristic curves if a single sample is taken from a lot and 0.12 mg/kg, 0.15 mg/kg and 0.2 mg/kg action limits are considered. Moreover, the targeted compliance level is 98% (the probability of rejection is 2%).

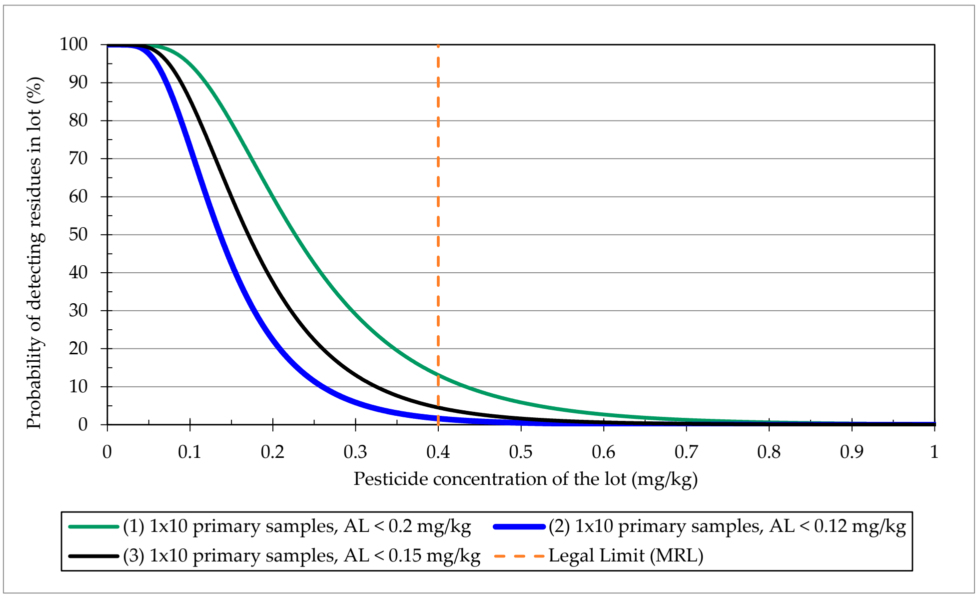

Figure 1. Operation characteristic curves indicating the probability of detection of acetamiprid residues when single samples containing ten apples each are analyzed.

The figure shows the probability of detection of pesticide residues in composite samples taken from the tested lot. The probabilities of finding $\geq$ 0.4 mg/kg residue in repeated samples are 2, 4.5 and 12.5% if the samples did not contain residues above the action limits of 0.12, 0.15 and 0.2 mg/kg, respectively. Moreover, the figure indicates that the probability of finding residues $\geq$ 0.8 mg/kg decision limit (Equation (2)) is practically zero if action limits 0.12 and 0.15 mg/kg were applied at the time of pre-export sampling of the apple lot. On the other hand, residues above 0.8 mg/kg may occur at low probability if an AL of 0.2 mg/kg was considered.

The relative sampling uncertainty (CV_S) varies between 1.2 and 1.7 in the case of fruits and vegetables [38,40]. Therefore, a default action limit of 0.3 MRL is recommended for general use to account for the sampling uncertainty.

Refined action limit can be chosen based on the CV_S values determined by Farkas and co-workers.

3. Results

3.1. Summary of the Results of Pesticide Residue Monitoring during 2017–2021

During the period of 2017–2021 pesticide residues were determined in 9924 samples taken from 119 crops. Altogether, over 2.6 million analyte-sample combinations were tested. In view of the very large database, the results obtained by the analyses of six commodities containing the most frequently detected residues were selected, as an example, for their evaluation in this article. Table 2 shows the main parameters and results of the tests carried out.

Table 2. Summary information on the pesticide residue analyses carried out during 2017–2021.

Commodity	No. of					
	Samples [2]	Analytes [3]	Tests [4]	R > MRL [5]	MRL ≥ R ≥ LOQ [6]	R < LOQ [7]
All commodities [1]	9924	622	2,652,560	102	5261	4560
Apples	803	459	227,571	1	587	215
Cherries, sour	122	441	32,962	1	95	26
Grapes, table	783	459	113,132	1	703	79
Peaches/nectarines	468	445	90,851	1	350	117
Peppers, green, red	616	459	165,388	4	298	314
Strawberries	349	447	60,458	3	291	55

Notes: [1]: Commodities included in the sampling program. [2]: Number of samples investigated. [3]: Number of active substances and their metabolites screened in the samples. All residue components defined by the relevant EC Regulations were included in the scope of methods and analyzed. The reported results are calculated from the measured residue components. However, it does not mean that all samples were tested for all active substances. [4]: Number of tests = number of samples multiplied by the number of residues analyzed. [5]: Number of samples containing residues above the MRL. [6]: Number of samples containing detectable residues lower than the MRL. [7]: Number of samples with nondetectable residues (residues were below the limit of quantification).

Tables A1 and A2 indicate the number of samples in which the residues of active substances were detected by the laboratories. The residue components included in the definition of residues for enforcement purposes were analyzed with the methods applied, but they are not listed separately in the tables. Nevertheless, the active substance concentration reported, was calculated from their measured concentrations and expressed in the reported active substance equivalent.

The table indicates the frequency of occurrence of various residues and provides guidance for the relevance of their inclusion in the scope of the screening method(s) applied. It is especially important if the selected ion monitoring detection mode is used. Moreover, it should be emphasized that the 0.01 * mg/kg default limit is applicable for all substances for which MRL has not been established.

3.2. Assessment of Compliance of Residues with MRLs

3.2.1. Commodities Marketed in Hungary

The authorizations of several active substances were withdrawn by the European Commission during 2017–2021. After the grace period, these substances must not be used, and their residues should not be present in detectable concentrations in/on food and feed commodities. The R > MRL cases indicated in Table 1 for the selected six commodities resulted from the unauthorized use of these substances.

This was the case in other Member Countries of the EU [24,25] where multiple residues were detected in many samples at varying concentrations below the corresponding MRLs. The summary of findings related to the selected crops is given in Tables 2 and 3. A few samples contained residues above the corresponding MRLs: one sour cherry (dimethoate (0.052 mg/kg) + omethoate (0.101 mg/kg) in 2018); two peppers (chlorpyrifos (0.058 mg/kg and 0.036 mg/kg) in 2020 and 2021); three strawberries (flonicamid (0.32 mg/kg), tebuconazole (0.17 mg/kg) in 2019 and propiconazole (0.064 mg/kg) in 2020). The residue concentrations were generally low indicating that the pesticides were likely applied within the four weeks period before harvest and the pre-harvest intervals were considered. Moreover, we consider in Section 3.3 if the presence of multiple residues reflects good plant protection practice.

Table 3. Summary of compliance of exported lots with EU MRLs.

Commodity	No. of Tested		No. of Lots Complied [1]	No. of Lots and Proportion of Their Compliance due to Residues Detected [1]		
	Lots	As-S				
Apple	1944	50	1545 > 92%	9 tau-fluvalinate (89%)	8 folpet (88%)	21 lambda-cyhalothrin (81%)
Cherries	195	21	162 > 96%	23 (dithiocarbamates 87%)	6 thiamethoxam (83%)	4 deltamethrin (50%)
Grape	986	62	869 > 90%	8 buprofezin [2] (0%)	4 pyraclostrobin (78%)	36 acetamiprid (86%)
Peach	521	34	465 > 95%	acetamiprid (87.5%)	prochloraz (83.3%)	carbendazim (70%)
Peppers, sweet	631	48	460 > 90%	# [3]		
Strawberry	588	40	444 > 90%	# [4]		

Notes: The proportion of tested lots that would comply with the indicated probability. [1]: There are many cases where the number of measured residues was ≤5. The compliance of these lots cannot be realistically evaluated. Therefore, they are not included in the table. [2]: Buprofezin MRL was reduced to 0.01 * mg/kg. It was detected in eight lots (0.012–0.066 mg/kg). None of them would comply; Lower compliance was found in case of 16 and 12 lots because of lambda-cyhalothrin (81%), carbendazim (75%), respectively. # [3]: methomyl, pymetrozine, acetamiprid, clothianidin, spirodiclofen (0%) flonicamid (44%), acetamiprid (69%), tebuconazole (75%), cyflufenamid (80%), lambda-cyhalothrin (81%), spinosad (83%), indoxacarb (84%). # [4]: emamectin benzoate, cyflufenamid (sum), formetanate (0%), ethirimol (64%), etoxazole (60%), bupirimate (79%), abamectin (75%), spinosad (82%), thiamethoxam (83%), thiacloprid (89%).

3.2.2. Prediction of Potential Compliance with MRLs if the Sampled Products Were Exported

We postulate that the tested lots might have been exported to the EU and subjected to repeated sampling by the importing country as part of the border control. To verify compliance with export MRLs, the sampling uncertainty shall also be included in the combined uncertainty of the results.

Taking the recommended 0.3 MRL action limit, we evaluated the potential compliance of the tested lots considering the residues of all active substances detected in the samples taken from the selected commodities.

The results, shown in Table 3, indicate the number of lots that would comply with the given high probability if any of the active substances analyzed were applied to them, except those which are listed individually.

Of the detected residues in the selected commodities, the grace period is over for several active substances. They should not be present in detectable concentration (MRL = 0.01 *) in the samples:

- apple: chlorothalonil, chlorpyrifos, chlorpyrifos-methyl, fenhexamid, imidacloprid and methoxyfenozide;
- grape: chlorpyrifos, chlorpyrifos-methyl, diflubenzuron, dimethoate/omethoate, famoxadone, iprodione, pirimicarb and thiophanate-methyl;
- cherry: chlorpyrifos, dimethoate, omethoate, prochloraz;
- peach: chlorpyrifos, chlorpyrifos-methyl, diflubenzuron, fenbuconazole, imazalil, imidacloprid and propamocarb;
- peppers: buprofezin, chlorpyrifos-methyl, napropamid, triadimefon, triadimenol.

In addition, the residues of glyphosate (0.1 *), captan and THPI (0.03 *), thiophanate-methyl (0.1 *) should not be present in detectable concentrations in the commodities listed in Table 2.

The test results obtained during the grace period hold no relevance for the present assessment and, thus, were not considered. The restricted substances should be included in the scope of screening methods with LOD lower than the MRLs (LOQ values) indicated with an asterisk.

Moreover, those lots exhibiting detectable concentrations of these substances must not be exported or marketed in Hungary either.

3.3. Evaluation of Plant Protection Practice

Multiple residues were detected in many samples at varying concentrations below the corresponding MRLs. Based on their residue levels, most of the detected active substances were likely applied in the period of four weeks before harvest.

The summary of findings related to the selected crops is given in Tables 4 and 5.

Table 4. Summary of samples containing multiple residues.

Year	No. of Samples Analyzed	Samples w. Multiple Residues	Max. no. of AS	No. of Samples Containing Multiple Residues [1]					
				Apples	Cherries, Sour	Grapes, Table	Peaches and Nectarines	Peppers Sweet	Strawberries
2017	1902	761	23	75	16	57	45	35	33
2018	1995	820	13	101	16	53	51	44	35
2019	1842	916	15	107	10	49	59	45	36
2020	1750	625	16	89	8	42	39	45	3
2021	1666	719	11	103	9	32	37	43	20

[1]: The minimum number of active substances in samples was two in each commodity and year. The maximum and average number of AS detected in the selected commodities together with the relevant pest and disease groups are shown in Table 4.

Table 5. Number of AS detected in individual samples.

Commodity	Max (Average) No. of AS Found in One Sample					Relevant Groups of	
	2017	2018	2019	2020	2021	Diseases	Arthropod Pests
Apples	23 (3.9)	13 (3.9)	8 (3.8)	9 (3.7)	11 (3.5)	3	5
Cherries, sour	8 (3.4)	7 (3.7)	6 (3.6)	6 (3.5)	6 (3.8)	3	5
Grapes, table	12 (4)	11 (4.1)	11 (3.8)	11 (4.1)	7 (3.3)	3	3
Peaches and nectarines	6 (2.7)	7 (3.4)	9 (3.1)	9 (4.1)	5 (2.9)	4	4
Peppers, sweet	10 (3.7)	11 (3.2)	15 (3.8)	15 (3.6)	10 (3.1)	3–4	4
Strawberries	7 (3.4)	9 (4.3)	11 (4.8)	7 (4.3)	9 (5.0)	3	4

At first sight the number of active substances look surprisingly high. However, one of the most important tools for avoiding pest resistance to pesticides is to use alternate or tank-mix substances of different chemical structures and modes of actions, and limiting the number of applications of the chemicals with site-specific modes of action, and avoidance of their eradicant use. It is the general recommendation for resistance management in agriculture. Pesticide resistance has been documented in a large number of key diseases and arthropod pests of the selected crops, e.g., apple scab, powdery mildews, downy mildew, gray mold, brown rot of stone fruits, codling moth, cotton bollworm, white flies, several aphid and spider mite species, etc. In the last decade the authorization of several broad-spectrum insecticides was withdrawn (e.g., organophosphates, several synthetic pyrethroids and zoocide carbamates). Both plant pathogens and arthropod pest species differ significantly, for this reason there is no possibility to control all with only one or two active substances. Therefore, the growers must combine and apply different plant protection products to provide high quality crops to the consumer.

Nevertheless, the residues of 23, 15, 12 and 11 different active ingredients detected in apple, pepper, grape and strawberry, respectively, are considered high. In an average year diseases and pests can be effectively controlled with a lower number of applications. Depending on the weather conditions and the pest situation in the given orchard, 2–2 combined applications are justified against plant pathogens and pests in apple, cherry, peach and nectarine within the period of four weeks before harvest. In the case of peppers and probably strawberries, a greater number of applications are reasonable in this period. There is no general rule for the number of treatments, this depends on the life cycles and flight activity of the pests, the developmental stages of the crops, the weather conditions during the growing season (temperature, precipitation, humidity), the variety, the training system, and the presence of insect pollinators, among others. For choosing the compounds to be applied, besides the pest communities present in the orchard and vineyard, it is very important to take into account the mode of action of the active substances. To carry out integrated pest management, continuous and precise pest forecasting (monitoring, scouting, pheromone trapping) in the orchard is necessary.

In apples the most important diseases and arthropod pests are apple scab, powdery mildew, codling moths, leaf miner moths, aphids and woolly aphids. In certain years fire blight, tortrix moths, spider mites and apple clearwing can cause problems, too. On average, the applications of three to four active substances (Table 5), is well justified. As many as eight or nine active substances may be required, because of the need for resistance management.

During the four week period before harvest, pesticide treatments are required to control codling moth and tortrix moths (acetamiprid, etofenprox, indoxacarb, chlorantraniliprole, thiacloprid), spider mites (etoxazole, spirodiclofen), apple scab and powdery mildew (difenoconazole, dithianon, fluopyram, pyraclostrobin, pyrimethanil, tebuconazole) and the storage diseases (cyprodinil, fludioxonil, fluopyram, pyraclostrobin).

In sour cherries the pesticides used for the control of the most important diseases and insect pests were as follows: cherry fruit flies and black cherry aphid (acetamiprid, deltamethrin, lambda-cyhalothrin, pirimicarb and thiacloprid), brown rot and anthracnose (boscalid, captan, cyprodinil, dithiocarbamates, fenhexamid, fludioxonil, fluopyram, penconazole, prochloraz and tebuconazole). The period from last decade of May until the middle of June is of crucial importance in pest management of this stone fruit in Hungary. An average of three to four active substances sprayed per growing season is not a high number given the numerous diseases and arthropod pests.

In table grapes the growers must effectively control several key diseases and arthropod pests which infest both leaves and berries, such as powdery mildew, downy mildew, gray mold (botrytis blight), grape berry moths, Northern American grapevine leafhopper (*Scaphoideus titanus*) and phytophagous mites during the growing season. The majority of the active substances were applied against diseases caused by fungi. Usually, three to four active substances applied per growing season is not a high number. The number of target pests and diseases and the number of applications are closely related. Because of the different fungal pathogen species, different active substances must be applied against powdery mildew and gray mold. Similarly, for the control of grape berry moths an acaricide which is efficacious against spider mites is not suitable.

In the period of flowering and fruit development the effective control of powdery mildew (azoxystrobin, fluopyram, metrafenone, myclobutanil, penconazole, pyraclostrobin, spiroxamine, tebuconazole), downy mildew (cyazofamid, dimethomorph, dithiocarbamates, fluopicolide, folpet, mandipropamid, metalaxyl), gray mold (boscalid, cyprodinil, fenhexamid, fenpyrazamin, fludioxonil, fluopyram, folpet, iprodione, pyrimethanil), grapevine leafhopper (chlorpyrifos, imidacloprid, lambda-cyhalothrin, spinosad, spirotetramat, thiamethoxam) and grape berry moth (chlorantraniliprol, chlorpyrifos, lambda-cyhalothrin, spinosad, tau-fluvalinate) is essential.

In the case of nectarine and peach the relevant diseases are: peach leaf curl, peach shot hole, bacterial dieback, Cytospora canker, brown rot, peach twig borer, Oriental fruit moth, aphids, scale insects and mites. Therefore, spraying is necessary to control peach twig borer and Oriental fruit moth (acetamiprid, indoxacarb, lambda-cyhalothrin), aphids (acetamiprid, flonicamid, pirimicarb) and brown rot (boscalid, captan, cyprodinil, fenhexamid, fenpyrazamine, fluopyram, penconazole, tebuconazole). On the average, treatments with three to four, and even seven to nine active substances per year are justified.

For the successful production of peppers, the efficacious control of the following key diseases and insect pests is essential, i.e., root rots, bacterial spots, powdery mildew, soil-dwelling insects, thrips species and cotton bollworm. In the period of flowering and fruit development the effective control of thrips species (abamectin, acetamiprid, spinosad, thiamethoxam), aphids (acetamiprid, flonicamid, pirimicarb, thiacloprid, thiamethoxam), cotton bollworm (chlorantraniliprol, lambda-cyhalothrin, spinosad) and powdery mildew (azoxystrobin, boscalid, difenoconazole, penconazole, pyraclostrobin) is necessary. Besides fungicides and zoocides, in peppers herbicides were also used and detected in some samples (napropamid, pendimethalin).

The strawberry growers must effectively control several key diseases and arthropod pests, such as soil pathogens, leaf diseases, gray mold (fruit rot), strawberry blossom weevil, strawberry rhynchites, strawberry root weevil, aphids and strawberry mite. The number of target pests and diseases and the number of applications are closely related. An average of three to five active substances applied per growing season is not a high number because different pesticides have to be used to control, for instance, soil pathogens and leaf diseases or gray mold, or aphids and mites.

In the period of flowering and fruit development the control of gray mold (boscalid, cyprodinil, fenhexamid, fenpyrazamin, fludioxonil, fluopyram), strawberry blossom weevil and aphids (lambda-cyhalothrin, thiacloprid, thiamethoxam) and strawberry mite (abamectin, bifenazate, hexythiazox) is very important.

4. Discussion and Conclusions

Altogether the residues of 622 pesticide active ingredients were analyzed in 9924 samples taken mostly from 119 fruits and vegetables of economic importance grown in Hungary as well as imported during 2017–2021. The pesticide residue–sample combinations amounted to over 2.6 million. The risk-based sampling plan was developed by the NFCSO. It also incorporated the samples specified by the multi-annual control program of the European Commission [42].

The analyses were performed in laboratories accredited according to the ISO 17025 Standard [32]. The accuracy of their results and in general the technical level of laboratory analyses was demonstrated with the successful participation in EU proficiency tests covering fruits, vegetables and cereals.

Considering the very large number of results, six crops having the largest frequency of detectable pesticide residues were selected to illustrate the results and our evaluation methods.

Out of the 9924 samples/lots 102 (1.0%) contained residues above the Hungarian (EU) MRLs. The violation rate was lower than that reported by EU Member countries. In Hungary, the violation of the MRLs resulted from the use of unauthorized pesticides which were applied after the grace period expired. Such a situation requires action from the regulatory agency. The growers who misused pesticides were fined and advised on the changed authorization status of these substances to reduce the chance of placing plant commodities containing unauthorized residues on the market in the future.

The very low MRL violation rate and the fact that about 10–50% of the samples did not contain detectable residues provide broad confidence that, under current pesticide regulations, the food supply is broadly safe for consumption.

In addition to assessing compliance to legal MRLs of commodities marketed in Hungary, we examined the fictive situation of their potential export to the EU. For making a decision on whether the tested lot would contain residues below the corresponding MRLs upon the border control in the importing country, we used an action limit of 0.3 MRL for the evaluation of detected residue concentrations. In view of its applicability for three different analyte–matrix combinations [37,39,48], we recommend its use generally for pre-marketing control.

In the evaluation of residue data, the proportion of lots that contained residues $\leq$ 0.3 MRL was considered compliant. It was found that all tested residues in 79% of apple, 83% of cherry, 88% of grape, 89% of peach/nectarine, 73% of pepper and 76% of strawberry lots would comply with the import MRLs with >90% probability. The residues of active substances that would lead to a lower level of probability of compliance were identified.

Our results draw attention to a very important practical situation. Notwithstanding that the residues in tested lots conformed with the EU MRLs based on the first sampling, it cannot be excluded that a certain proportion of these lots would contain higher residues and be rejected, based on the results of repeated independent sampling, even if both sampling was representative, and the analyses provided accurate results. The inevitable variation in the results of repeated random sampling is caused by the very heterogeneous distribution of residues in primary samples [50,51] and consequently in the composite samples, too. Therefore, to avoid rejection of export shipments, the lots to be exported should be selected based on pre-export sampling and analyses. Their results should be evaluated applying the appropriate action limit.

The wide scope of the screening methods and low LOD values enabled the detection of all residues present even in trace concentrations. As a result, we found that 36–50% of samples of selected crops contained multiple residues ranging from 2 to 23. The frequency of multiple residues was within the same range in European countries.

The residue levels in the samples analyzed in Hungary were typically low, indicating that some of the pesticides were applied well before the harvest of the crops. Since the residue levels are compared to the corresponding MRLs [6,52] individually, the samples containing multiple residues complied with MRLs.

Given the high number of pesticide residues present in some samples, we examined whether the application of those active substances could be justified based on the principles of integrated pest management and good practice in the application of pesticides. Considering the major pests and diseases of the selected crops as well as the need for the rotation of active substances and treatments with mixtures of pesticides to reduce the chance for the development of resistance, we concluded that the use of 23, 15, 12 and 11 different pesticides in apple, pepper, grape and strawberry, respectively, do not represent good plant protection practice in a normal growing season. On average, the application of three to four active substances within the four-week period before harvest of apples is well defensible. Similarly, three to four pesticide treatments of cherries and peaches and three to five in strawberries are reasonable. Even seven to nine active substances may be needed for effective protection under special circumstances (e.g., severe infestation of arthropod pests, serious and sustained infection of plant pathogens) and for resistance management.

When a high number of pesticide treatments is witnessed, even though there is no risk to the health of the consumers deriving from the exposure to pesticide residues, the farm owners should be informed and advised to seek the help of a plant protection specialist who would examine the actual growing conditions prevailed during the growing season and advise the farmers on the effective and economical use of pesticides.

Considering the results of our evaluation based on the selected crops, we can conclude that the national monitoring program conducted over the past 5-year period served its purpose and met the requirements of the European Commission specified in regulation 396/2005. Moreover, it provided well-supported information for the regulators on the appropriate level of plant protection practice in Hungary.

Nevertheless, the monitoring of pesticide residues should be continued to provide up-to-date information for exporters of agricultural products and regulators to take timely action assuring the safe and effective use of pesticides, if necessary.

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Abbreviations

AL	Action Limit
AS	Active Substance
EFSA	European Food Safety Authority
FAO	Food and Agriculture Organization
GAP	Good Agricultural Practices
JMPR	Joint Meeting on Pesticide Residues
LOD	Limit of Detection
LOQ	Limit of Quantification
MRL	Maximum Residue Limit
NFCSO	Hungarian Food Chain Safety Office
US FDA	United States Food and Drug Administration
WHO	World Health Organization

Appendix A

Table A1. Summary of number of samples and active substances tested.

Number of Tests	Apples	Cherries	Grapes	Green Peppers	Peaches and Nectarines	Active Substances [1]
No. of samples tested	803	122	783	588	468	349
No. of residues—matrix combinations tested	227,571	32,962	113,132	165,388	90,851	60,458
No. of ASs tested	459	441	459	459	445	447

Note: [1]: The residue components included in the residue definition defined by various European Commission regulations were measured separately or as their common derivative. The reported residue concentration was calculated from the measured residues.

Table A2. Number of samples in which the active substances were detected.

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
2,4-D	202	52	212	148	80	99
2,4-DB	48	12	42	19	13	32
2-Phenylphenol	588	81	499	438	367	153
3,5-Dichloroaniline	219	42	156	139	68	95
3-Chloroaniline	158	40	125	90	53	70
Abamectin (sum)	449	41	166	346	190	92
Acephate	759	99	386	573	302	204
Acetamiprid	765	108	394	586	321	212
Acetochlor	694	102	316	489	278	178
Aclonifen	163	10	63	92	61	32

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Acrinathrin	766	102	389	577	311	206
Alachlor	542	81	273	381	192	140
Aldicarb (sum)	572	92	343	471	248	165
Aldrin and Dieldrin (sum)	724	104	387	568	312	205
Alphamethrin	184	29	77	145	91	39
Ametoctradin	404	38	157	257	148	70
Ametryn	370	52	203	214	122	107
Amidosulfuron	140	18	36	97	75	35
Aminopyralid	14					
Amitraz (sum)	161	40	126	94	53	68
AMPA	14	1	11	15	4	2
Atraton	207	42	140	122	61	75
Atrazine	593	95	287	423	239	162
Azamethiphos	247	18	67	134	88	25
Azinphos-ethyl	700	103	320	494	287	178
Azinphos-methyl	769	103	393	581	318	206
Aziprotryne	370	52	203	214	122	107
Azoxystrobin	765	110	394	586	322	211
Beflubutamid	137	18	34	96	75	33
Benalaxyl (sum of isomers)	698	104	317	490	273	177
Bendiocarb	122	11	41	107	44	15
Benfluralin	379	71	210	289	131	108
Bentazone (sum)	218	41	104	207	103	48
Benthiavalicarb (Benthiavalicarb-isopropyl)	169	10	131	102	61	34
Benzovindiflupyr	151	13	320	83	31	8
Bifenazate	163	10	359	92	61	32
Bifenox	694	102	331	490	279	178
Bifenthrin (sum of isomers)	769	108	155	588	322	212
Biphenyl	565	79	316	417	210	151
Bitertanol (sum of isomers)	759	107	617	574	307	209
Bixafen	709	107	396	493	290	177
Boscalid	773	108	308	588	322	212
Bromfenvinfos	167	12	396	98	64	37
Bromide ion				14		
Bromophos-methyl	688	105	309	473	288	177
Bromophos-ethyl	691	105	170	477	287	177
Bromopropylate	771	108	395	587	321	212
Bromoxynil and its salts	382	65	356	246	132	106
Bromuconazole (sum of diasteroisomers)	696	108	63	499	290	184

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Bupirimate	779	111	393	585	324	215
Buprofezin	759	108	203	583	314	211
Butocarboxim	146	18	70	103	77	35
Butralin	172	29	39	167	70	33
Butylate	370	52	39	214	122	107
Cadusafos	698	106	362	495	98	179
Captafol	221	32	291	200	298	55
Captan (sum)	692	99	386	540	304	211
Carbaryl	754	101	380	576	322	205
Carbendazim and benomyl (sum)	765	108	318	586	297	212
Carbofuran (sum)	697	103	336	556	231	349
Carboxin	696	108	39	499	16	184
Carfentrazone-ethyl (sum)	293	40	210	243	288	54
Chinomethionat	379	71	73	289	156	108
Chlorantraniliprole	766	108	7	578	228	202
Chlorbromuron	163	10	361	92	280	32
Chlordane (sum of cis- and trans-chlordane)	653	104	40	468	280	171
Chlorfenapyr	692	99	140	541	271	205
Chlorfenson	207	42	19	122	39	75
Chlorfenvinphos	690	102	58	487	61	177
Chlorfluazuron	134	15	36	73	75	11
Chloridazon	506	98	380	389	16	151
Chlorobenzilate	374	54	307	220	61	108
Chlorothalonil	772	108	390	587	283	212
Chlorotoluron	706	107	285	494	307	182
Chloroxuron	140	18	397	100	321	35
Chlorpropham	578	81	396	435	18	145
Chlorpyrifos	803	108	311	588	321	212
Chlorpyrifos-methyl	803	108	1	588	215	212
Chlorsulfuron	1	4	318	14	321	10
Chlozolinate	208	42	63	123	123	71
Cinidon-ethyl	370	52	398	214	122	107
Clethodim (sum)	184	30	318	167	314	33
Clofentezine	759	108	1	583	283	211
Clomazone	706	107	393	494	321	182
Clopyralid	1		248			
Clothianidin	722	99	206	535	60	189
Coumaphos	653	93	321	443	322	173
Cyanazine	113	10	140	58	32	10

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Cyanofenphos	207	42	3	122	61	75
Cyantraniliprole	47	2	328	7	8	
Cyazofamid	656	105	53	491	299	186
Cycloate	516	70	71	316	199	142
Cycloxydim (sum)	169	10	394	102	61	34
Cyflufenamid	273	45	186	254	102	37
Cyfluthrin (sum of isomers)	741	102	553	569	299	187
Cymoxanil	765	108	203	586	322	212
Cypermethrin (sum of isomers)	802	108	394	588	321	212
Cyproconazole	754	106	15	576	305	203
Cyprodinil	780	107	388	587	315	211
Cyprosulfamide				5		2
Cyromazine	21		307	70	8	14
Dazomet	58	17	322	17	14	40
DDT	701	108	322	501	290	184
Deltamethrin	803	108	204	588	322	212
Demeton-S-Methyl	367	54	82	219	124	108
Desethyl-Atrazine	552	86	275	385	196	144
Desisopropyl-Atrazine	552	86	214	385	196	144
Desmedipham	402	63	189	257	168	129
Dialifos	207	42	395	122	61	75
Diazinon	773	108	387	588	322	212
Dicamba	146	48	252	97	62	72
Dichlobenil	525	94	63	396	212	145
Dichlofenthion	535	96	320	401	227	145
Dichlofluanid	696	106	336	493	287	183
Dichlormid	338	59	146	276	121	94
Dichlorprop	210	52	74	139	74	82
Dichlorvos	771	108	176	586	322	212
Diclobutrazol	163	10	259	91	32	10
Dicloran	595	81	394	457	215	155
Dicofol (sum of p, p′ and o, p′ isomers)	712	99	615	519	340	213
Dicrotophos	372	62	1	232	135	114
Diethofencarb	765	108	318	586	322	212
Difenoconazole	766	108	141	586	322	212
Diflovidazin (Flufenzin)	204	44	394	127	63	76
Diflubenzuron	765	108	321	586	322	212
Diflufenican	696	108	120	499	290	184
Dimethachlor	700	107	203	491	275	181

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Dimethenamid (sum of isomers)	370	52	286	214	122	107
Dimethipin	379	71	394	289	131	108
Dimethoate	765	108	101	586	322	212
Dimethomorph (sum of isomers)	767	108	318	586	322	212
Dimoxystrobin	707	107	393	494	283	182
Diniconazole (sum of isomers)	759	108	140	583	314	211
Dioxacarb	1					
Dioxathion	207	42	273	122	61	75
Diphenylamine	591	81	394	439	215	153
Diquat			317		1	
Disulfoton (sum)	488	79	255	348	184	133
Ditalimfos	698	106	142	493	288	177
Dithianon	243	30	324	172	105	35
Dithiocarbamates	605	77	320	399	247	176
Diuron	690	108	202	496	282	183
Dodine	423	37	157	335	169	90
Emamectin B1a (free base)	421	52	535	337	171	84
Endosulfan (sum)	772	108	395	588	322	212
Endrin	701	108	322	501	290	184
Endrin Aldehyde	692	108	109	495	290	178
Endrin, Keto-	312	37	394	205	160	74
EPN	769	106	362	535	319	207
Epoxiconazole	766	108	29	586	322	212
epsilon-HCH	48	8	35	48	25	13
EPTC (ethyl dipropylthiocarbamate)	207	42	140	122	61	75
Ethephon	60		46	44	30	
Ethiofencarb	119	7	46	68	32	12
Ethiofencarb-Sulfone	119	7	46	68	32	12
Ethiofencarb-Sulfoxide	119	7	392	68	32	12
Ethion	767	103	394	579	318	206
Ethirimol	765	108	388	586	322	210
Ethofumesate	367	54	126	219	124	108
Ethoprophos	699	106	157	495	288	179
Ethoxyquin	222	55	143	164	116	93
Etofenprox	781	99	204	575	301	203
Etoxazole	404	38	148	257	148	74
Etridiazole	208	42	322	123	60	71

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Etrimfos	698	106	327	495	288	179
Famoxadone	592	90	393	458	229	171
Fenamidone	759	108	396	583	314	211
Fenamiphos (sum)	566	88	324	450	228	155
Fenarimol	760	102	393	574	303	205
Fenazaquin	759	108	394	583	314	211
Fenbuconazole	763	108	18	584	317	211
Fenbutatin oxide	47		394	27	19	9
Fenchlorphos (sum)	487	92	245	372	217	138
Fenhexamid	765	108	391	586	322	212
Fenitrothion	765	106	140	571	317	193
Fenoxycarb	765	108	1	586	322	212
Fenpicoxamid	15		151	3		
Fenpropathrin	771	106	392	582	319	207
Fenpropidin	763	108	392	585	320	212
Fenpropimorph (sum of isomers)	706	90	308	524	256	171
Fenpyrazamine	276	40	394	260	96	34
Fenpyroximate	765	108	396	586	322	212
Fenson (Fenison)	207	42	388	122	61	75
Fensulfothion	649	100	39	458	278	168
Fensulfothion-Oxon	113	7	39	58	32	10
Fensulfothion-Sulfone	113	7	388	58	32	10
Fenthion (sum)	515	83	242	399	221	145
Fenuron	1	4	398	14	18	10
Fenvalerate (sum)	801	108	140	588	322	212
Fipronil (sum)	733	103	390	570	313	204
Flazasulfuron	137	18	316	96	75	33
Flonicamid (sum)	447	78	255	353	176	110
Florasulam	385	80	138	302	151	116
Fluazifop-P	180	52	333	152	73	88
Fluazifop-P-butyl	22		270	46	9	18
Fluazinam	690	105	211	495	281	182
Flubendiamide	580	74	394	468	261	126
Flucythrinate (sum of isomers)	376	73	297	294	133	109
Fludioxonil	766	108	392	586	322	212
Flufenacet	646	105	3	465	261	162
Flufenoxuron	741	108	39	575	305	188
Flumethrin	113	7	243	58	32	10
Flumetralin	6		141	5		

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Flumioxazine	341	39	377	269	131	67
Fluometuron	513	72	351	322	201	143
Fluopicolide	764	105	268	573	321	210
Fluopyram	615	92	175	475	230	163
Fluoxastrobin	500	58	388	359	210	101
Flupyradifurone	29		3	3		
Fluquinconazole	755	104	15	578	305	206
Flurochloridone	597	88	1	376	230	153
Fluroxypyr (sum)	16	11	246	25	38	17
Flusilazole	761	104	354	580	313	207
Flutolanil	503	78	47	332	171	131
Flutriafol	771	104	182	578	313	206
Fluvalinate (sum of isomers)	768	106	783	581	410	249
Fluxapyroxad	421	49	361	296	179	88
Folpet (sum)	684	92	252	522	270	192
Fomesafen	163	10	39	92	61	32
Fonofos	521	77	297	327	210	143
Foramsulfuron	146	18	204	103	77	35
Forchlorfenuron	140	18	292	100	75	35
Formetanate	429	77	322	359	166	128
Formothion	624	92	8	428	243	161
Fosetyl-Al (efozit-Al)			330	24		
Fosthiazate	763	108	354	586	201	203
Fuberidazole	308	28	140	195	130	67
Furilazole	207	42	7	122	32	75
Glufosinate			27	11		
Glyphosate	82	2	36	27	75	17
Halosulfuron methyl	140	18	138	100	8	35
Haloxyfop	201	52	592	208	353	104
Heptachlor (sum)	653	104	307	471	280	177
Heptenophos	698	106	322	495	143	179
Hexachlorobenzene	701	108	205	501	315	184
Hexachlorocyclohexane, alpha-isomer	700	108	322	501	290	183
Hexachlorocyclohexane, beta-isomer	700	108	495	501	290	183
Hexachlorocyclohexane, delta-isomer	679	100	398	482	276	176
Hexaconazole	775	107	394	581	124	210
Hexaflumuron	394	60	391	245	290	110
Hexazinone	382	54	394	221	322	108

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Hexythiazox	765	108	207	586	322	212
Imazalil	765	108	128	586	143	212
Imazamox	353	78	7	278	53	109
Imazapyr	164	40	394	100	321	69
Imazethapyr	6		391	10		2
Imidacloprid	765	108	141	586	315	212
Indoxacarb	775	107	141	581	63	210
Iodosulfuron-methyl	184	27	94	164	288	33
Ioxynil	204	44	389	127	199	76
Ipconazole	516	70	394	317	313	142
Iprodione	765	104	38	578	322	206
Iprovalicarb	765	108	322	586	35	212
Isocarbophos	768	107	204	576	68	208
Isodrin	116	12	312	64	287	15
Isofenphos	698	106	390	495	270	178
Isofenphos-methyl	688	99	90	484	305	176
Isoprocarb	367	54	249	219	284	108
Isoprothiolane	693	108	34	508	178	197
Isoproturon	493	79	210	337	75	133
Isopyrazam	205	18	335	120	124	35
Isoxaben	137	18	56	96	143	33
Isoxadifen-ethyl	431	53	140	301	42	106
Isoxaflutole	376	60	393	239	175	109
Kresoxim-methyl	770	107	398	578	251	209
Lambda-cyhalothrin	802	108	322	588	290	213
Lenacil	696	108	394	499	290	184
Lindane	701	108	380	501	322	184
Linuron	765	108	371	586	276	212
Lufenuron	719	99	383	535	287	180
Malathion (sum)	721	100	383	564	322	199
Mandipropamid	765	108	319	586	70	212
MCPA and MCPB	180	52	138	121	73	81
Mecarbam	688	108	389	493	73	183
Mecoprop (sum)	180	52	169	121	305	81
Mefenpyr-diethyl	172	29	138	167	282	33
Mepanipyrim	764	104	204	575	124	205
Mepiquat			141			
Mepronil	367	54	36	219	63	108
Meptyldinocap	204	44	281	127	75	76
Mesosulfuron-methyl	146	18	3	103	62	35

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Mesotrione	194	44	391	113	315	76
Metaflumizone (sum of E- and Z- isomers)	548	90	2	395	265	148
Metalaxyl and metalaxyl-M (sum of isomers)	771	104	455	578	373	206
Metaldehyde	163	10	321	92	318	32
Metamitron	696	108	390	499	275	184
Metazachlor	700	107	395	491	302	181
Metconazole (sum of isomers)	688	105	345	498	287	177
Methabenzthiazuron	140	18	305	100	212	35
Methacrifos	616	100	388	461	1	167
Methamidophos	767	103	317	579	290	206
Methidathion	760	102	395	575	321	201
Methiocarb (sum)	598	90	327	455	316	162
Methomyl	766	108	394	586	322	212
Methoxychlor	701	108	356	501	32	184
Methoxyfenozide	765	108	39	586	290	212
Metobromuron	696	108	32	499	92	184
Metolachlor and S-metolachlor (sum of isomers)	705	98	678	496	468	171
Metoxuron	113	10	316	58	322	10
Metrafenone	764	108	113	547	273	211
Metribuzin	695	104	321	487	157	177
Metsulfuron-methyl	332	48	39	270	288	70
Mevinphos	698	106	133	493	77	179
Molinate	516	70	63	317	291	142
Monocrotophos	686	98	203	534	61	195
Monolinuron	163	10	39	92	122	32
Myclobutanil	775	107	362	581	199	210
N,N-Diethyl-m-toluamid (DEET)	652	105	302	462	266	174
Napropamide (sum of isomers)	370	52	243	214	77	107
Nicosulfuron	146	18	167	103	201	35
Nitenpyram	513	72	204	322	90	143
Nitrofen	300	49	203	165	124	84
Novaluron	367	54	294	219	122	108
Nuarimol	370	52	112	214	249	107
o.p′-DDD	631	92	322	434	251	164
o.p′-DDE	631	92	203	434	290	164

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Ofurace	370	52	73	214	309	107
Omethoate	728	108	393	575	105	205
Oxadiazon	198	19	394	130	314	53
Oxadixyl	759	108	39	583	322	211
Oxamyl	765	108	1	586	77	212
Oxasulfuron	146	18	247	103	168	35
Oxathiapiprolin	15		284	3		118
Oxycarboxin	163	10	322	92	249	172
Oxydemeton-methyl (sum)	551	83	374	442	216	161
Oxyfluorfen	492	78	294	347	61	32
Paclobutrazol	760	104	389	580	218	140
Paraoxon	526	77	362	324	318	206
Parathion	773	108	391	541	322	212
Parathion-methyl (sum)	768	106	391	580	318	211
Penconazole	772	104	110	578	269	175
Pencycuron	765	108	87	586	315	210
Pendimethalin	775	107	390	581	68	33
Penflufen (sum of isomers)	205	18	303	115	313	206
Penthiopyrad	282	42	203	202	321	212
perchlorate	1					107
Permethrin (sum of isomers)	772	108	242	587	122	142
Pethoxamid	370	52	320	214	199	182
Phenkapton	207	42	245	122	61	75
Phenmedipham	402	63	394	257	168	129
Phenthoate	516	70	5	317	200	143
Phorate (sum)	342	78	41	282	158	115
Phorate (sum)	700	106	8	495	288	179
Phosmet (sum)	595	81	394	438	216	151
Phosphamidon	700	106	358	495	288	179
Phosphane and phosphide salts	1		320	12		
Phoxim	513	72	102	323	270	143
Picolinafen	516	70	229	317	282	139
Picoxystrobin	690	108	63	496	244	32
Piperonyl butoxide	163	10	6	92	53	9
Pirimicarb	773	103	394	579	102	212
Pirimicarb, desmethyl-	333	63	140	219	107	110
Pirimiphos-ethyl	654	94	389	476	319	207
Pirimiphos-methyl	769	106	17	582	313	19
Prochloraz (sum)	507	87	54	412	42	18

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Procymidone	750	99	388	569	199	205
Profenofos	759	102	348	574	131	178
Profluralin	379	71	255	289	273	133
Promecarb	326	48	245	267	215	138
Prometryn	571	86	394	398	201	212
Propachlor	516	73	337	316	298	184
Propamocarb	715	108	359	586	252	178
Propaquizafop	467	68	203	290	279	107
Propargite	650	96	102	518	122	67
Propazine	370	52	390	214	138	209
Propetamphos	309	28	394	195	307	212
Propham	516	70	210	317	303	108
Propiconazole (sum of isomers)	768	107	276	578	322	141
Propisochlor	547	81	321	387	282	162
Propoxur	690	108	271	496	242	137
Propyzamide	765	108	320	586	192	183
Proquinazid	618	89	63	474	274	32
Prosulfocarb	605	74	309	483	61	180
Prosulfuron	163	10	178	92	291	80
Prothioconazole: prothioconazole-desthio (sum of isomers)	542	90	333	450	296	184
Prothiofos	693	107	102	481	136	67
Pymetrozine	540	74	43	482	61	20
Pyraclostrobin	765	108	164	586	288	78
Pyraflufen-ethyl	135	19	394	111	322	211
Pyrazophos	698	106	393	495	85	211
Pyrethrins	271	47	320	152	313	178
Pyridaben	759	108	100	583	286	6
Pyridalyl	151	13	204	134	143	108
Pyridaphenthion	696	102	207	486	31	109
Pyridate	353	78	391	278	124	210
Pyrifenox	367	54	285	219	315	164
Pyrimethanil	776	107	395	581	277	207
Pyriofenone	273	36	70	201	322	33
Pyriproxyfen	765	108	383	586	78	204
Pyroxsulam	184	27	242	161	301	142
Quinalphos	700	106	117	495	131	179
Quinmerac	503	98	393	388	68	211
Quinoclamine	190	18	300	112	314	170

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Quinoxyfen	759	108	304	583	262	175
Quintozene (sum)	658	102	204	466	123	25
Resmethrin (sum of isomers)	366	54	243	216	63	143
Rimsulfuron	69	15	203	75	201	107
Rotenone	513	72	63	322	122	32
Secbumeton	309	28	102	196	138	177
Sedaxane	26		4	3		67
Silthiofam	309		65	196		168
Simazine	370	52	104	214	32	10
Simetryn	113	10	394	91	31	212
Spinetoram (XDE-175)	151	13	394	139	322	212
Spinosad (sum)	765	108	394	586	322	212
Spirodiclofen	765	108	187	586	317	78
Spiromesifen	770	106	38	582	175	39
Spirotetramat (sum)	540	67	344	406	324	142
Spiroxamine (sum of isomers)	775	107	102	581	138	2
Sulfotep	690	102	346	487	31	211
Sulfoxaflor (sum of isomers)	151	13	393	83	224	212
Tau-Fluvalinate	613	81	394	468	314	206
Tebuconazole	774	104	393	580	226	205
Tebufenozide	759	108	391	583	322	147
Tebufenpyrad	766	108	260	586	313	211
Tecnazene	535	96	388	403	314	33
Teflubenzuron	759	108	34	583	303	33
Tefluthrin	760	102	34	574	75	144
Tepraloxydim	137	18	322	96	201	32
Terbacil	516	73	63	320	288	10
Terbufos	698	106	39	494	61	10
Terbufos-sulfone	163	10	39	92	32	182
Terbufos-sulfoxide	113	7	361	1	61	162
Terbumeton	113	10	285	58	273	151
Terbuthylazine	651	98	396	516	237	146
Terbutryn	583	93	291	422	321	210
Tetrachlorvinphos	526	77	316	328	315	212
Tetraconazole	775	107	392	581	279	212
Tetradifon	772	108	252	587	201	178
Tetramethrin	695	102	394	490	321	32
Thiabendazole	765	105	394	585	321	70
Thiacloprid	766	108	40	586	321	212

Table A2. *Cont.*

Active Substances [1]	Apples	Cherries	Grapes, Table	Green Peppers	Peaches and Nectarines	Strawberries
Thiamethoxam	766	108	113	586	32	180
Thiencarbazone-methyl	128	10	394	61	157	69
Thifensulfuron-methyl	332	48	361	270	322	117
Thiodicarb	765	108	112	586	271	184
Thiofanox	326	48	340	267	180	205
Thiometon	470	74	254	287	270	35
Thiophanate-methyl	684	97	227	501	149	112
Tolclofos-methyl	759	102	307	574	75	48
Tolylfluanid (sum)	445	76	210	345	303	210
Tralkoxydim	140	18	391	100	280	210
Triadimefon	775	107	391	581	85	36
Triadimenol	775	107	39	581	124	35
Tri-allate	367	54	395	219	78	171
Triasulfuron	146	18	39	103	319	212
Triazophos	770	106	306	582	77	182
Tribenuron-methyl	146	18	394	103	267	108
Trichlorfon	518	70	313	319	149	12
Triclopyr	7		112	10		25
Tricyclazole	600	105	318	457	322	211
Trifloxystrobin	765	108	200	586	281	178
Triflumizole	690	105	394	495	132	101
Triflumuron	765	108	191	586	279	143
Trifluralin	694	102	7	506	114	69
Triflusulfuron	15		46	3		42
Triforine	327	50	243	192	200	177
Trimethacarb	326	48	1	267	272	151
Triticonazole	685	101	38	490	76	206
Uniconazole	144	18	279	95	32	182
Valifenalate	119	7	389	68	269	
Vamidothion	643	96	33	477	311	
Vinclozolin	765	102	319	577	13	
Zoxamide	690	122		495		

Note: [1]: The residue components included in the residue definition defined by various European Commission regulations were measured separately or as their common derivative. The reported residue concentration was calculated from the measured residues.

References

1. Research Institute of Organic Agriculture (FIBL). European and Global Organic Farming Statistics. 2023. Available online: https://www.organic-world.net/statistics.html (accessed on 25 May 2023).
2. European Council Regulation No 1107/2009 of the European Parliament and of the Council of 21 October 2009 Concerning the Placing of Plant Protection Products on the Market and Repealing Council Directives 79/117/EEC and 91/414/EEC. Available online: https://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri=CELEX:32009R1107 (accessed on 25 May 2023).

3. European Council. Commission Regulation No 2022/1438, Amending Annex II to Regulation (EC) No 1107/2009 as Regards Specific Criteria for the Approval of Active Substances that are Micro-Organisms. Available online: http://data.europa.eu/eli/reg/2022/1441/oj (accessed on 25 May 2023).
4. European Commission Regulation No 283/2013 of 1 March 2013 Setting Out the Data Requirements for Active Substances, in Accordance with Regulation (EC) No 1107/2009 of the European Parliament and of the Council Concerning the Placing of Plant Protection Products on the Market. Available online: https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2013:093:0001:0084:EN:PDF (accessed on 25 May 2023).
5. European Commission Regulation No 546/2011 of 10 June 2011 Implementing Regulation (EC) No 1107/2009 of the European Parliament and of the Council as Regards Uniform Principles for Evaluation and Authorisation of Plant Protection Products. Available online: http://data.europa.eu/eli/reg/2009/1107/2022-11-21 (accessed on 25 May 2023).
6. European Council Regulation No 396/2005 of the European Parliament and of the Council of 23 February 2005 on Maximum Residue Levels of Pesticides in or on Food and Feed of Plant and Animal Origin and Amending Council Directive 91/414/EEC. Available online: http://data.europa.eu/eli/reg/2005/396/2023-02-28 (accessed on 25 May 2023).
7. Austrian Agency for Health and Food Safety. National Pesticide Residue Control Programmes. Available online: https://www.ages.at/en/plant/pesticides/pesticide-residues#c5252 (accessed on 25 May 2023).
8. Australian Government Department of Agriculture, Fisheries and Forestry. National Residue Survey. Available online: https://www.agriculture.gov.au/agriculture-land/farm-food-drought/food/nrs (accessed on 25 May 2023).
9. German Federal Office of Consumer Protection. Tables for the National Reporting of Pesticide Residues in food 2020. Available online: www.bvl.bund.de/DE/Arbeitsbereiche/01_Lebensmittel/01_Aufgaben/02_AmtlicheLebensmittelueberwachung/07_PSMRueckstaende/01_nb_psm_2020_tabellen/nbpsm_2020_tabellen_node.html (accessed on 25 May 2023).
10. Japanese Ministry of Health, Labour and Welfare. Agricultural Chemical Residues in Foods. Available online: https://www.mhlw.go.jp/stf/seisakunitsuite/bunya/kenkou_iryou/shokuhin/zanryu/index_00016.htm (accessed on 25 May 2023).
11. USDA. PDP Databases and Annual Summaries (1992–2020). Available online: www.ams.usda.gov/datasets/pdp/pdpdata (accessed on 25 May 2023).
12. Zhang, M.; Zeiss, M.R.; Geng, S. Agricultural pesticide use and food safety: California's model. *J. Integr. Agric.* **2015**, *14*, 2340–2357. [CrossRef]
13. Jardim, A.N.O.; Caldas, E.D. Brazilian monitoring programs for pesticide residues in food-results from 2001 to 2010. *Food Control* **2012**, *25*, 607–616. [CrossRef]
14. Lei, Y.; Luo, Y.; Fang, N.; Li, Y.; Wang, X.; He, H.; Jiang, J.; Yu, J.; Zhang, C.; Zhao, X. Residues and dietary risk assessment of imidacloprid in bamboo shoot (*Phyllostachys praecox*), winter jujube (*Ziziphus jujuba* Mill. cv. Dongzao), *Dendrobium officinale* Kimura et Migo, and *Fritillaria*. *Agronomy* **2023**, *13*, 1076. [CrossRef]
15. Wang, X.; Fang, N.; Wang, X.; Zhao, X.; Zhang, C.; Wang, Q. Residue analysis and dietary risk assessment of metalaxyl in Chinese bayberry and *Dendrobium officinale*. *Agronomy* **2023**, *13*, 186. [CrossRef]
16. Meftaul, M.; Venkateswarlu, K.; Parven, A.; Annamalai, P.; Megharaj, M. Human health risk assessment of pesticides in lettuce and spinach grown in urban backyard garden soils. *J. Food Compos.* **2023**, *115*, 104977. [CrossRef]
17. Mandal, K.; Singh, R.; Sharma, S.; Kataria, D. Dissipation and kinetic studies of fluopyram and trifloxystrobin in chilli. *J. Food Compos.* **2023**, *115*, 105008. [CrossRef]
18. Li, Q.; Zhang, J.; Lin, T.; Fan, C.; Li, Y.; Zhang, Z.; Li, J. Migration behavior and dietary exposure risk assessment of pesticides residues in honeysuckle (*Lonicera japonica* Thunb.) based on modified QuEChERS method coupled with tandem mass spectrometry. *Food Res. Int.* **2023**, *166*, 112572. [CrossRef]
19. Penagos-Tabares, F.; Sulyok, M.; Faas, J.; Krska, R.; Khiaosa-ard, R.; Zebeli, Q. Residues of pesticides and veterinary drugs in diets of dairy cattle from conventional and organic farms in Austria. *Environ. Pollut.* **2023**, *316*, 120626. [CrossRef]
20. Lajmanovich, R.; Repetti, M.R.; Cuzziol Boccioni, A.P.; Michlig, M.P.; Demonte, L.; Attademo, A.M.; Peltzer, P.M. Cocktails of pesticide residues in *Prochilodus lineatus* fish of the Salado River (South America): First record of high concentrations of polar herbicides. *Sci. Total Environ.* **2023**, *870*, 162019. [CrossRef]
21. De Cock, A.; Forio, M.A.E.; Croubels, S.; Dominguez-Granda, L.; Jacxsens, L.; Lachat, C.; Roa-López, H.; Ruales, J.; Scheyvaerts, V.; Hidalgo, M.C.S.; et al. Health risk-benefit assessment of the commercial red mangrove crab: Implications for a cultural delicacy. *Sci. Total Environ.* **2023**, *862*, 160737. [CrossRef]
22. Environmental Working Group. Dirty Dozen Clean Fifteen. Available online: https://www.ewg.org/https://www.ewg.org/foodnews/clean-fifteen.php (accessed on 25 May 2023).
23. European Commission Implementing Regulation (EU) 2021/601 of 13 April 2021 Concerning a Coordinated Multiannual Control Programme of the Union for 2022, 2023 and 2024 to Ensure Compliance with Maximum Residue Levels of Pesticides and to Assess the Consumer Exposure to Pesticide Residues in and on Food of Plant and Animal Origin. Available online: http://data.europa.eu/eli/reg_impl/2021/601/oj (accessed on 25 May 2023).
24. EFSA. National summary reports on pesticide residue analysis performed in 2020. *EFSA Support. Publ.* **2022**, *19*, 7216E. [CrossRef]
25. EFSA; Carrasco Cabrera, L.; Medina Pastor, P. The 2019 European Union report on pesticide residues in food. *EFSA J.* **2021**, *19*, e06491. [CrossRef]
26. EFSA. Eurobarometer 2022. Available online: www.efsa.europa.eu/sites/default/files/2022-09/EB97.2-food-safety-in-the-EU_report.pdf (accessed on 25 May 2023).

27. FAO. *Submission and Evaluation of Pesticide Residues Data for the Estimation of Maximum Residue Levels in Food and Feed*, 3rd ed.; FAO Plant Production and Protection Paper 225; FAO: Rome, Italy, 1996; Available online: https://www.fao.org/3/i5452e/i5452e.pdf (accessed on 25 May 2023).
28. Szenczi-Cseh, J.; Ambrus, Á. Uncertainty of exposure assessment of consumers to pesticide residues derived from food consumed. *J. Environ Sci. Health B* **2017**, *52*, 658–670. [CrossRef]
29. FAO. Portion of Commodities to Which Codex MRLs Apply and Which Is Analyzed (CAC/GL 41-1993). Available online: http://www.fao.org/fao-who-codexalimentarius/sh-proxy/en/?lnk=1&url=https%253A%252F%252Fworkspace.fao.org%252Fsites%252Fcodex%252FStandards%252FCXG%2B41-1993%252FCXG_041e.pdf (accessed on 25 May 2023).
30. FAO. Recommended Methods of Sampling for the Determination of Pesticide Residues for Compliance with MRLs CXG33-1999. Available online: www.fao.org/fao-who-codexalimentarius/sh-proxy/en/?lnk=1&url=https%253A%252F%252Fworkspace.fao.org%252Fsites%252Fcodex%252FStandards%252FCXG%2B33-1999%252FCXG_033e.pdf (accessed on 25 May 2023).
31. European Commission Directive 2002/63/EK Establishing Community Methods of Sampling for the Official Control of Pesticide Residues in and on Products of Plant and Animal Origin and Repealing Directive 79/700/EEC. Available online: https://eur-lex.europa.eu/legal-content/EN/ALL/?uri=celex%3A32002L0063 (accessed on 25 May 2023).
32. ISO. ISO/IEC 17025:2017 General Requirements for the Competence of Testing and Calibration Laboratories. Available online: www.iso.org/ISO-IEC-17025-testing-and-calibration-laboratories.html (accessed on 25 May 2023).
33. FAO. CAC/GL 70-2009 Guidelines for Settling Disputes on Analytical (Test) Results. Available online: https://www.fao.org/fao-who-codexalimentarius/sh-proxy/en/?lnk=1&url=https%253A%252F%252Fworkspace.fao.org%252Fsites%252Fcodex%252FStandards%252FCXG%2B70-2009%252FCxg_070e.pdf (accessed on 25 June 2023).
34. Ambrus, Á.; Doan, V.V.N.; Szenczi-Cseh, J.; Szemánné-Dobrik, H.; Vásárhelyi, A. Quality control of pesticide residue measurements and evaluation of their results. *Molecules* **2023**, *28*, 954. [CrossRef]
35. European Commission. Analytical Quality Control and Method Validation Procedures for Pesticide Residues Analysis in Food and Feed SANTE 11312/2021. Available online: https://food.ec.europa.eu/system/files/2022-02/pesticides_mrl_guidelines_wrkdoc_2021-11312.pdf (accessed on 25 May 2023).
36. Ambrus, Á.; Szenczi-Cseh, J.; Doan, V.V.N.; Vásárhelyi, A. Evaluation of monitoring data in foods. *Agrochemicals* **2023**, *2*, 69–95. [CrossRef]
37. Farkas, Z.; Slate, A.; Whitaker, T.; Kötelesné Suszter, G.; Ambrus, Á. Use of combined uncertainty information for testing compliance with MRLs. *J. Agric. Food Chem.* **2015**, *63*, 4418–4428. [CrossRef]
38. Farkas, Z.; Cook, J.M.; Ambrus, Á. Estimation of uncertainty of measured residues and testing compliance with MRLs. In *Food Safety Assessment of Pesticide Residues*; Ambrus, Á., Hamilton, D., Eds.; World Scientific: Hackensack, NJ, USA, 2017; pp. 404–466.
39. Sharma, G.M.; Pereira, M.; Wang, S.S.; Chirtel, S.J.; Whitaker, T.B.; Wehling, P.; Arlinghaus, M.; Canida, T.; Jackson, L.S.; Williams, K.M. Evaluation of sampling plans for measurement of gluten in oat groats. *Food Control* **2020**, *114*, 107241. [CrossRef]
40. Farkas, Z.; Horváth, Z.; Szabó, I.J.; Ambrus, Á. Estimation of sampling uncertainty of pesticide residues based on supervised residue trial data. *J. Agric. Food Chem.* **2015**, *63*, 4409–4417. [CrossRef]
41. Horváth, Z.; Ambrus, Á. Principles of control of small-scale production of fruits and vegetables and planning risk-based monitoring programmes. In *Food Safety Assessment of Pesticide Residues*, 1st ed.; Ambrus, Á., Hamilton, D., Eds.; World Scientific: Hackensack, NJ, USA, 2017; pp. 467–506.
42. European Commission Implementing Regulation (EU) 2019/533 of 28 March 2019 Concerning a Coordinated Multiannual Control Programme of the Union for 2020, 2021 and 2022 to Ensure Compliance with Maximum Residue Levels of Pesticides and to Assess the Consumer Exposure to Pesticide Residues in and on Food of Plant and Animal Origin. Available online: https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32019R0533 (accessed on 25 May 2023).
43. Anastassiades, M.; Lehotay, S.J.; Štajnbaher, D.; Schenck, F.J. Fast and easy multiresidue method employing acetonitrile extraction/partitioning and 'dispersive solid-phase extraction' for the determination of pesticide residues in produce. *J. AOAC Int.* **2003**, *86*, 412–431. [CrossRef]
44. Anastassiades, M.; Scherbaum, E.; Tasdelen, B.; Stajnbaher, D. Recent developments in QuEChERS methodology for pesticide multiresidue analysis. In *Pesticide Chemistry. Crop Protection, Public Health, Environmental Safety*; Ohkawa, H., Miyagawa, H., Lee, P.W., Eds.; Wiley-VCH Verlag GmbH & Co.: Weinheim, Germany, 2007; pp. 439–458. [CrossRef]
45. Anastassiades, M.; Wachtler, A.K.; Kolberg, D.I.; Eichhorn, E.; Marks, H.; Benkenstein, A.; Zechmann, S.; Mack, D.; Wildgrube, C.; Barth, A.; et al. Quick Method for the Analysis of Highly Polar Pesticides in Food Involving Extraction with Acidified Methanol and LC-or IC-MS/MS Measurement I. Food of Plant Origin (QuPPe-PO-Method). Available online: https://www.eurl-pesticides.eu/userfiles/file/EurlSRM/EurlSrm_meth_QuPPe_PO_V11_1.pdf (accessed on 27 June 2023).
46. Ambrus, Á.; Buczkó, J.; Hamow, K.Á.; Juhász, V.; Solymosné Majzik, E.; Szemánné Dobrik, H.; Szitás, R. Contribution of sample processing to variability and accuracy of the results of pesticide residue analysis in plant commodities. *J. Agric. Food Chem.* **2016**, *64*, 6071–6081. [CrossRef]
47. Ambrus, Á.; Vásárhelyi, A. Pesticide residue analysis in Hungary between 1967 and 2015. *J. Food Investig.* **2016**, *62*, 919–942. Available online: http://real-j.mtak.hu/18410/1/EVIK2016-1.pdf (accessed on 20 June 2023).
48. Ambrus, Á.; Szenczi-Cseh, J.; Griff, T.; Kerekes, K.; Miklós, G.; Szigeti, T.; Vásárhelyi, A. Food safety assessment of the mycotoxin and pesticide residue contamination of our foods, Part 1. Pesticide residues. *J. Food Investig.* **2020**, *66*, 2773–2801. Available online: https://eviko.hu/Portals/0/ujsagok/Arcivum/2020/1_szam/EVIK2020-1.pdf (accessed on 20 June 2023).

49. Ambrus, Á.; Szenczi-Cseh, J.; Griff, T.; Kerekes, K.; Miklós, G.; Szigeti, T.J.; Vásárhelyi, A. Food safety assessment of the mycotoxin and pesticide residue contamination of our foods, Part 2. Mycotoxins. *J. Food Investig.* **2020**, *66*, 2923–2949.
50. Ambrus, Á. Variability of pesticide residues in crop units. *Pest Manag. Sci.* **2006**, *62*, 693–714. [CrossRef]
51. Horváth, Z.; Ambrus, Á.; Mészáros, L.; Braun, S. Characterization of distribution of pesticide residues in crop units. *J. Environ. Sci. Health—B Pestic. Food Contam. Agric. Wastes* **2013**, *48*, 615–625. [CrossRef]
52. Commission Regulation No 149/2008 of 29 January 2008 Amending Regulation No 396/2005 of the European Parliament and of the Council by Establishing Annexes II, III and IV Setting Maximum Residue Levels for Products Covered by Annex I Thereto. Available online: https://eur-lex.europa.eu/legal-content/EN/ALL/?uri=celex%3A32008R0149 (accessed on 20 June 2023).