Environmental Science and Pollution Research (2022) 29:65860–65872
https://doi.org/10.1007/s11356-022-20481-x

RESEARCH ARTICLE

Health risk associated with pesticide residues in vegetables from Incheon region of Korea

Byung Kyu Park[1,2] · Seung Hye Jung[1] · Sung Hee Kwon[1] · Sun Hoi Kim[1] · Eun Young Yeo[1] · Mi Sook Yeom[1] · Soon Jae Seo[1] · Kwang Sig Joo[1] · Myung Je Heo[1] · Geun Pyo Hong[2]

Received: 26 April 2021 / Accepted: 23 April 2022 / Published online: 30 April 2022

Abstract

This study was conducted to investigate the pesticide residue concentrations and assess potential human health risks from vegetable consumption in Incheon. A total of 960 samples were collected from the Incheon areas of Korea in 2019. The pesticide residues were analyzed by the multi-residue method of the Korean Food Code for 373 different pesticides using GC–MS/MS, LC–MS/MS, GC-ECD/NPD, and HPLC-UVD. Among the vegetable samples, 869 samples (90.5%) were free from detectable residues, while 91 samples (9.5%) contained residues, and 16 samples (1.7%) had residues exceeding the Korean maximum residue limit (MRLs). A total of 33 different pesticide residues were found, and 11 residues exceeded MRLs. The most frequently detected pesticide residues were chlorfenapyr, fludioxonil, pyridalyl, hexaconazole, and procymidone. Samples exceeding the MRLs were found in aster scaber, coastal hog fennel, lettuce (leaves), mustard green, mustard leaf, perilla leaves, *Pimpinella brachycarpa*, radish leaves, shepherd' purse, spinach, and winter-grown cabbage. The potential health risk assessment of pesticides was estimated by calculating the estimated daily intake (EDI) and the acceptable daily intake (ADI). The range of HQs was 0.002–90.621%, which was below 100%. Therefore, the results of this study show that the detected pesticide could not be considered a serious public health problem through the consumption of vegetables.

Keywords Pesticide residues · Vegetables · Risk assessment · Incheon · Korea

Introduction

Fruits and vegetables are important sources of carbohydrates, fiber, trace minerals, vitamins and antioxidants (Prodanov et al. 2004; Shashirekha et al. 2015). But, fruits and vegetables may have toxic residual pesticides by the use of pesticides in agricultural production. Pesticides are widely used to prevent and reduce diseases affecting agricultural products during the production cycle. The use of pesticides can improve the yield, quality, safety, and nutritional value of agricultural products (Cho et al. 2009). However, the presence of pesticide residues in agricultural products can reduce the quality of agricultural products and may be related to potential human health risks such as nausea, headaches, carcinogenesis, neurotoxicity, infertility, and immunological effects (Baldi et al. 2001; Mansour 2004; Nougadère et al. 2012; Alavanja et al. 2013).

International organizations and governments control the use of pesticides by establishing the maximum residue levels (MRLs) of pesticides in food commodities. The MRLs are used to determine the maximum amount of intake legally admitted by regulations (Darko and Akoto 2008). MRLs in Korea were first set up in 1988 by the Ministry of Food and Drug Safety. The number of pesticides increased to about 500 types in 2019. Korea's MRL is set in consideration of international standards and some agricultural products are set lower than those of other countries (Chun et al. 2002).

Because fruits and vegetables are eaten without processing, they are the important sources of pesticide residue intake for humans. The human intake of pesticide residues in food commodities is higher than the intake associated

Responsible Editor: Philippe Garrigues

✉ Byung Kyu Park
iampbk@korea.kr

[1] Samsan Agricultural Products Inspection Center, Incheon Institute of Public Health and Environment, 46, Yeongseongdong-ro, Bupyeong-gu, Incheon 21320, Republic of Korea

[2] Department of Food Science and Biotechnology, Sejong University, Seoul, Republic of Korea

with water intake and air inhalation. Therefore, it is very important to monitor pesticide residues in fruits and vegetables and assess the potential risks to human health (Juraske et al. 2007; Szpyrka et al. 2015).

Incheon metropolitan city is the third largest city in Korea and has about 3 million inhabitants. Agricultural products are supplied from all parts of Korea, and agricultural products exceeding the MRL are sometimes distributed. About 1% of agricultural products distributed in Incheon exceed the MRL every year. Therefore, monitoring of pesticide residues in agricultural products and removal of agricultural products distributed in excess of the MRL are very important for public safety.

This study was conducted with a residual pesticide monitoring program of the Agricultural Products Inspection Center, under the Incheon Metropolitan Government. The purpose of this study was to analyze pesticide residues of vegetables distributed in Incheon and we evaluated the human health risk assessment by calculating the estimated daily intake (EDI) and acceptable daily intake (ADI) with the pesticides that are detected in vegetables. The results can improve understanding of the importance of pesticide residues contained in agricultural products, and it is expected to help minimize human health risks for Korean consumers.

Materials and methods

Chemical and reagents

The 373 pesticide analytical standards (Table 1) were purchased for AccuStandard (New Haven, CT, USA). The purities of all pesticide standards and analytical reagents were 99% or more. Individual pesticide stock solutions (1000 mg/L) were prepared in acetone or methanol and stored at − 20 °C. The standard working solutions were prepared by diluting the stock solution with acetone/acetonitrile. HPLC grade acetone, acetonitrile, dichloromethane, methanol, and *n*-hexane purchased from Honeywell International Inc. (Muskegon, MI, USA). Anhydrous sodium chloride for residue analysis was purchased from Junsei (Tokyo, Japan). The solid phase extraction (SPE) cartridge for the purification of samples was purchased from Bekolut (Haupststuhl, Rhineland-Palatinate, Germany).

Samples

Sampling was performed by authorized personnel in accordance with the pesticide residue analysis manual of the Korean Food Code Guideline (KFC 2017). A total of 960 samples were collected from January to December 2019 at an agriculture product wholesale market, big retailers etc. in Incheon (Fig. 1). Generally, samples were selected from representative vegetables that Koreans eat a lot. All samples were analyzed within 24 h and 4 °C until the moment of extraction.

Sample extraction and clean up

Most samples were prepared from unwashed whole raw commodity. A representative portion of the sample (1 kg) was chopped and blended thoroughly using a food processor. The sample extraction procedures were conducted with the multi-residue method for pesticide residues, according to the Korean Food Code, which is based on the multi-residue method of the California Department of Food and Agriculture (Lee et al. 1991). After grinding, a ground sample (50 g) was homogenized with 100 mL of acetonitrile for 2 min in a high speed homogenizer. The homogenized mixture was filtered into a bottle with 15 g of anhydrous sodium chloride. The extraction was vortexed vigorously for 3 min. From the upper layer, an aliquot of 10 mL was transferred into a tube and evaporated to dryness on a 40 °C bath with a gentle stream of air. For gas chromatography analysis (GC–MS/MS, GC-ECD/NPD), the dried extracts were dissolved with 4 mL of *n*-hexane/acetone (80:20, v/v) and transferred to a florisil cartridge (1 g, 6 mL), which was activated and pre-conditioned with 5 mL of *n*-hexane/acetone (80:20, v/v) and 5 mL of *n*-hexane. After sample loading, the cartridge was eluted with 5 mL of *n*-hexane/acetone (80:20, v/v). This solvent was then evaporated slowly to dryness at 40°Cbath under a gentle stream of air. The dried residue was re-dissolved with 1 mL of *n*-hexane/acetone (80:20, v/v) for GC analysis. For liquid chromatography analysis (LC–MS/MS, LC-UVD), sample extracts were dissolved with 4 mL of dichloromethane/methanol (99:1, v/v) and transferred to amino-propyl cartridge (1 g, 6 mL) which was activated and pre-conditioned with 5 mL of dichloromethane. After sample loading, the cartridge was eluted with 7 mL of dichloromethane/methanol (99:1, v/v). This solvent was evaporated slowly to dryness at 40° bath under a gentle stream of air. The dried residue was re-dissolved with 1 mL of acetonitrile for LC analysis.

Instrument analysis of pesticides

The analysis of residual pesticides was performed according to the multi-residue method of the Korea Food Code. When monitoring pesticide residues in agricultural products, analysis equipment that can be detected depending on the type of pesticide was used. GC–MS/MS and LC–MS/MS systems were mainly used for qualitative analysis. GC-ECD, GC-NPD, and HPLC were used for quantitative analysis of organochlorine and pyrethroid compounds, organophosphorus and nitrogen-containing compounds, and ultraviolet light-detected compounds, respectively.

Table 1 List of pesticides analyzed in this study

Instrument	Pesticide
GC–MS/MS GC-ECD GC-NPD	2,6-Diisoporpylnaphthalene, Acrinathrin, Aldrin, Allethrin, Allidochlor, Ametryn, Anilofos, Aspon, Atrazine, Azaconazole, Azinphos-ethyl, Benalaxyl, BHC, Benodanil, Benzoylprop-ethyl, Bifenox, Bifenthrin, Binapacryl, Bromacil, Bromobutide, Bromophos-methyl, Bromopropylate, Bupirimate, Butafenacil, Butralin, Butylate, Cadusafos, Captafol, Captan, Carbophenothion, Chinomethionat, Chlorbufam, Chlordane, Chlordane-trans, Chlorethoxyfos, Chlorfenapyr, Chlorfenson, Chlorfluazuron, Chlorfluenol-methyl, Chloridazon, Chlorobenzilate, Chloroneb, Chloropropylate, Chlorothalonil, Chlorpyrifos, Chlorpyrifos-methyl, Chlortal-dimethyl, Chlorthion, Chlorthiophos, Chlozolinate, Cinmethylin, Clomeprop, Cyanazine, Cyanophos, Cycloate, Cyflufenamid, Cyfluthrin, Cyhalofop-butyl, Cyhalothrin, Cypermethrin, Cyproconazole, Deltamethrin, Demeton-O, Demeton-S, Demeton-S-methyl-sulfone, Desmetryn, Diallate, Diazinon, Dichlofenthion, Dichlofluanid, Dichlormid, Dichlorvos, Dicloran, Dicofol, Dieldrin, Diethatyl-ethyl, Diethofencarb, Diflufenican, Dimepiperate, Dimethachlor, Dimetenamid, Dimethoate, Dimethylvinphos, Diniconazole, Dinitramine, Dioxathion, Diphenmaid, Diphenylamine, Dithiopyr, DDT, Edifenphos, Endosulfan, Endrin, EPN, Epoxiconazole, EPTC, Esprocarb, Etaconazole, Ethalfluralin, Etofenprox, Ethion, Ethofumestae, Ethoprophos, Etoxazole, Etridiazole, Etrimfos, Fenamidone, Fenarimol, Fenazaquin, Fenbuconazole, Fenchlorphos, Fenfuram, Fenitrothion, Fenobucarb, Fenoxanil, Fenoxycarb, Fenpropathrin, Fenson, Fenthion, Fenvalerate, Fipronil, Flamprop-isopropyl, Flonicamid, Fluchloralin, Fludioxonil, Flufenpyr-ethyl, Flumetralin, Flumiclorac-pentyl, Flumioxazine, Fluopyram, Fluorodifen, Flurochloridone, Flutamone, Flusilazole, Fluthiacet-methyl, Flutolanil, Flutriafol, Fluvalinate, Folpet, Fonofos, Fosthiazate, Fthalide, Furathiocarb, Halfenprox, Heptenophos, Heptachlor, Hexachlorbenzene, Hexaconazole, Imazali, Indanofan, Indoxacarb, Iprobenfos, Iprodione, Iprovalicarb, Isazofos, Isofenphos, Isofenphos-methyl, Isopropalin, Isoprothiolane, Isoxanthion, Kresoxim-methyl, Lactofen, Leptophos, Malathion, Mecarbam, Mefenacet, Mefenpyr-diethyl, Mepronil, Metconazole, Methidathion, Methoprotryne, Methoxychlor, Methyl pentachlorophenyl-sulfide, Methyl trithion, Metolachlor, Metrafenone, Metribuzin, MGK, Molinate, Myclobutanil, Napropamide, Nitrapyrin, Nitrothal-isopropyl, cis-Nonachlor, trans-Nonachlor, Nuarimol, Ofurace, Oxadixyl, Oxydemeton-methyl, Paclobutrazol, Parathion-ethyl, Parathion-methyl, Pebulate, Penconazole, Pendimethalin, Pentachloroaniline, Permethrin, Phenthoate, Phentoxazone, Phosalone, Phosmet, Phosphamidone, Picolinafen, Picoxystrobin, Piperophos, Pirimicarb, Pirimiphos-ethyl, Pirimiphos-methyl, Probenazole, Procymidone, Profenofos, Profluralin, Prometon, Pronamide, Propachlor, Propazine, Propetamphos, Propham, Propisochlor, Prothiofos, Pyracarbolid, Pyraclofos, Pyrazophos, Pyridaben, Pyridalyl, Pyrifenox, Pyrimidifen, Pyriminobac-methyl, Quinalphos, Quinoxyfen, Quintozene, Secbumeton, Simeconazole, Spiroxamine, Sulfotep, Sulprofos, TCMTB, Tebuconazole, Tebufenpyrd, Tebupirimfos, Tefluthrin, Terbacil, Terbufos, Terbumeton, Terbuthylazine, Tetrachlorvinphos, Tetraconazole, Tetradifon, Tetramethrin, Tetrasul, Thiazopyr, Thifluzamide, Thiometon, Tolclofos-methyl, Tolfenpyrad, Tolylfluanid, Tralomethrin, Triadimefon, Tridadimenol, Triazophos, Tribufos, Triflumizole, Triflumuron, Trifluralin, Uniconazole, Vernolate, Vinclozolin, Zoxamide

Table 1 (continued)

Instrument	Pesticide
LC–MS/MS LC-UVD	Aldicarb, Amisulbrom, Asulam, Azamethiphos, Azoxystrobin, Bendiocarb, Bensulide, Benzoximate, Bixafen, Boscalid, Butocarboxim, Carbaryl, Carbetamide, Chlorantraniliprole, Chlorimuron-ethyl, Chlorobenzuron, Chlorotoluron, Chromatenozide, Cinosulfuron, Cyazofamid, Cycloprothrin, Cymoxanil, Dicrotophos, Dimethomorph, Ethaboxam, Ethametsulfuron-methyl, Ethiofencarb, Fenhexamid, Fenpyroximate, Ferimzone, Fluacrypyrim, Fluazinam, Flubendiamide, Flufenacet, Flufenoxuron, Fluometuron, Fluquinconazole, Fluridone, Flusulfamide, Forchlorfenuron, Hexaflumuron, Imazamox, Imazapic, Imazaquin, Imazethapyr, Imibenconazole, Ipconazole, Isoprocarb, Isoproturon, Isoxaben, Lenacil, Lufenuron, Malaoxon, Mepanipyrim, Mesosulfuron-methyl, Metamifop, Metamitron, Methabenzthiazuron, Methiocarb, Methoxyfenozide, Metolcarb, Metominostrobin, Metosulam, Nitenpyram, Novaluron, Oxaziclomefon, Phenmedipham, Pinoxaden, Promecarb, Propaquizafop, Propoxur, Propyrisulfuron, Prosulfocarb, Prothioconazole, Pyrachlonil, Pyraclostrobin, Pyraflufen-ethyl, Pyrazolate, Pyributicarb, Pyridate, Pyrimethanil, Pyriproxyfen, Pyroquilon, Quinoclamine, Rimsulfuron, Spriodiclofen, Sulfentrazone-NH_4, Tebufenozide, Tebuthiuron, Teflubenzuron, Tepraloxydim, Thenylchlor, Thiodicarb, Tiadinil, Tralkoxydim, Triasulfuron, Tribenuron-methyl, Tricyclazole, Tridemorph, Trifloxystrobin, Trifloxysulfuron, 2,3,5-Trimethacarb, 3,4,5-Trimethacarb, Triticonazole, XMC

Fig. 1 The location of Incheon Metropolitan City, where the samples were collected

GC–MS/MS analysis

GC–MS/MS analysis was performed using 7890B gas chromatograph coupled to a triple quadrupole mass spectrometer 7000D with electron impact ionization (EI) equipped with a 7693 autosampler (Agilent Technologies, Santa Clara, CA, USA). The analysis was conducted on DB-5 MS capillary columns (30 m × 0.25 mm × 0.25-μm film thickness, Agilent Technologies, Santa Clara, CA, USA). The temperature of injection port was 250 °C. The oven temperature was programmed from 70 °C (hold 3 min) to 180 °C by a rate of 20 °C/min and finally increased to 300 °C (hold 2.5 min) by a rate of 5 °C/min. The injection volume was 1 μL with splitless mode and helium carrier gas (99.999%) flowed at 1 mL/min. The mass spectrometry detector (MSD) was operated with electron impact ionization mode (ionization energy 70 eV), while the temperature of ion source and quadrupole were at 250 °C and 150 °C, respectively. The selected ion monitoring (SIM) mode with minimum three ions for each pesticide was used for detection and quantification of pesticides.

GC-ECD/NDP analysis

Gas chromatography analyses were performed with the GC-ECD system for organochlorine and pyrethroid compounds and the GC-NPD system for organophosphorus and nitrogen-containing compounds. An Agilent 6890 series

GC equipped with ^{63}Ni electron capture detector (ECD) and nitrogen phosphorous detector (NPD) were employed. Chromatographic separations were conducted on DB-5 capillary columns (30 m × 0.25 mm × 0.25-μm film thickness, Agilent Technologies, Santa Clara, CA, USA) for GC-ECD and GC-NPD. The operating conditions for GC-ECD were as follows: The oven temperature was programmed from 150 °C (hold 1 min) to 240 °C (hold 2 min) by a rate of 12 °C/min and finally increased to 280 °C (hold 13.5 min) by a rate of 10 °C/min. The injection volume was 1 μL with split mode (42.2:1), and nitrogen carrier gas flowed at 1.2 mL/min. The temperature of injector and detector were at 250 °C and 280 °C, respectively. The operating conditions for GC-NPD were as follows: The oven temperature was programmed from 120 °C (hold 1 min) to 240 °C (hold 2 min) by a rate of 12 °C/min, increased to 280 °C (hold 10 min) by a rate of 10 °C/min and finally increased to 300 °C (hold 1 min) by a rate of 10 °C/min. The injection volume was 1 μL with splitless mode and nitrogen carrier flowed at 1.2 mL/min. Hydrogen and air flowed at 3.0 mL/min, at 120.0 mL/min, respectively. The temperature of the injector and detector were at 270 °C and 300 °C, respectively.

LC–MS/MS analysis

LC–MS/MS analysis was performed using Vanquish UHPLC system coupled to a TSQ Altis triple quadrupole mass detector system (Thermo-Fisher Scientific, Waltham, Massachusetts, USA). Chromatographic separation was conducted on Accucore aQ (2.1 mm × 100 mm, 2.6-μm particle size, Thermo-Fisher Scientific, Waltham, Massachusetts, USA). UHPLC conditions consisted of mobile phase A (0.01% formic acid and 5-mM ammonium formate in water), mobile phase B (0.01% formic acid and 5-mM ammonium formate in MeOH), 2 μL injection volume, 0.3 mL/min flow rate and 40 °C oven temperature. The gradient programme was as follows: 0–0.5 min (80% A/20% B), 0.5–12 min (20–95% B), 12–12.1 min (5–80% A), and 12.1–15 min (80% A/20% B). For mass spectrometric analysis, LC-MSD was performed using an electrospray ionization source (ESI) in positive and negative modes, and data were acquired in SRM mode. Operating conditions were as follows: 350 °C vaporizer temperature, 325 °C ion transfer tube temperature, 3800 eV ion spray voltage, and 4.58 L/min sheath gas flow rate. Collision-induced dissociation was performed using argon as the collision gas pressure at a 1.5 mTorr in the collision tube.

HPLC analysis

The high performance liquid chromatography (HPLC) was carried out on Ultimate 3000 (Dionex, Sunnyvale, California, USA) with UV-VWD detector. Chromatographic separation was performed on a Capcell Core C18 column (4.6 mm × 100 mm, 2.7-μm particle size, Osaka Soda, Osaka, Japan). HPLC conditions consisted of mobile phase A (5% acetonitrile in water), mobile phase B (80% acetonitrile/ 20% methanol, v/v), 10 μL injection volume, 1.0 mL/min flow rate, and 40° oven temperature. UV absorbance was monitored at 220 nm and 250 nm. The gradient programme was as follows: initial (90% A/10% B), 0–13 min (10–80% B), 13–16 min (20% A), 16–16.1 min (20–90% A) and 16.1–20 min (90% A/10% B).

Method validation

The validation of the analytical methods was carried out in order to assess the performance of the method through the determination of precision, recovery, linearity, limit of detection (LOD), and limit of quantification (LOQ) for each pesticide, according to the Korea Food Code pesticide guidelines (KFC 2017). Assessment of recovery was performed using a mixture of the targeted pesticides at fortification levels of 0.1, 1.0 mg/kg using pesticide free sample extracts. The LOD and LOQ were calculated from the standard deviation of the five replicated analyses of spiked sample at low concentration level (LOD = 3.3 × SD and LOQ = 10 × SD). Precision was evaluated by performing recovery studies and is expressed as relative standard deviation (RSD, %). To assess linearity, the extracts from blank samples were fortified with standard solutions of 0.05, 0.1, 0.25, 1.0, 2.0 mg/kg and analyzed in triplicate at each concentration.

Risk assessment estimation

An exposure assessment was performed using the results of the pesticide residues in vegetables to determine the level of risk on human health. Risk assessment was performed using the estimated daily intake (EDI) and the established acceptable daily intake (ADI). EDI was calculated by multiplying the average pesticide concentration and the food consumption rate and dividing this by the body weight (Gad Alla et al. 2015). The average daily intake was referred to the intake of vegetables surveyed by Korea Disease Control and Prevention Agency (KDCA 2018). The criteria for ADI were in accordance with that of the pesticides and veterinary drug information of Korea Food and Drug Administration (KFDA 2020). The average body weight of Koreans (65.8 kg) was based on data provided by Korean Statistical Information Service (KSIS 2020). The hazard quotient (HQ) calculated the ratio of EDI and ADI to evaluate the risk of ingesting agricultural products with residual pesticides detected. The HQ was calculated using the formula: HQ = EDI/ADI × 100%. If the HQ value exceeds 100%, it is determined that it is hazardous for toxic effects. If the HQ

value is less than 100%, it is considered safe against toxic effects (Yu et al. 2016).

Statistical analysis

The detection rate of pesticides in agricultural products and the excess rate of MRL were calculated.

And, the lowest, highest, mean pesticide residue content and standard deviation were calculated for each pesticide. The LOD and LOQ values for method validation were calculated using the mean and standard deviation with the results of 5 replicates for the sample. In addition, EDI and HQ values for risk assessment were evaluated. The software for data analysis used the function of Microsoft Excel 2016 (Microsoft, Redmond, WA, USA).

Results and discussion

Method validation

From 373 pesticides, 14 pesticides were chosen based on their detection rate and violation rate of the MRLs. Table 2 presents linear correlation coefficients, limits of detection (LODs), limits of quantification (LOQs), and recoveries for the validation study. A linear correlation coefficient was detected in the range of 0.9947–0.9999. The LOD and LOQ values for the studied pesticides ranged from 0.002 to 0.030 mg/kg and from 0.006 to 0.090 mg/kg, respectively. The recovery rate was 83.5–101.3% for all pesticides, which is within the acceptable recovery range of 70–120% and the RSD of less than 10% also met requirement (Codex 1993). These results showed that the analytical method used in this work was appropriate for the analysis of targeted pesticide residues in vegetables.

Pesticide residues in vegetables.

In this survey, 960 samples of vegetables were analyzed for 373 pesticides contamination to assess health risk. After the screening for residual pesticides using GC–MS/MS and LC–MS/MS in vegetables, pesticides were analyzed for further validation and determination. The pesticide residues were analyzed by GC–MS/MS, GC-ECD/NPD, and LC–MS/MS, LC-UVD in consideration of their properties. In 869 of 960 analyzed vegetable samples (90.5%), no detectable residues were found, while 91 (9.5%) tested positive for pesticide residues. A number of 16 samples (1.7%) contained residues above MRLs established by the Korean Regulation for agricultural pesticides. Perilla leaves (21 samples, 19.8%), winter-grown cabbage (14 samples, 9.2%), lettuce leaves (10 samples, 6.7%) and spinach (10 samples, 10.0%) had the highest percentage of contaminated samples. Perilla leaves (3 samples, 2.8%), mustard leaf (2 samples, 12.5%), radish leaves (2 samples, 2.0%), and spinach (2 samples, 2.0%) showed the highest number of samples that violated MRLs (Table 3). Multi-residue pesticides were detected in perilla leaves, aster scaber, lettuce (leaves), mustard leaf, rape leaves, and spinach. The occurrence of multi-residues in vegetables is believed to be the result of the use of different types of pesticides to protect vegetables against various kinds of insects and diseases. Of all samples analyzed, 8.6% contained one pesticide residue, 0.8% contained two pesticide residues, and 0.1% contained three pesticide residues (Fig. 2).

Incidences and MRL violation

Table 4 shows the frequency and concentration of pesticide residues in the analyzed samples. Of the 373 pesticides tested, 33 were detected in the analyzed samples. The detection of pesticide by function was

Table 2 Validation parameters of the analytical method for pesticide residues detected in this study

Type	Pesticide	R^2	LOD (mg/kg)	LOQ (mg/kg)	Recovery (%)	RSD (±, %)
Insecticides	Chlorfenapyr	0.9947	0.012	0.035	98.3	3.5
	Chlorpyrifos	0.9975	0.017	0.068	88.7	5.5
	Diazinon	0.9998	0.002	0.006	94.7	0.6
	Flubendiamide	0.9965	0.013	0.039	91.3	2.9
	Pyridalyl	0.9988	0.012	0.037	87.7	2.8
	Tebupirimfos	0.9958	0.006	0.017	93.7	1.5
Fungicides	Fludioxonil	0.9956	0.013	0.038	86.5	3.0
	Fluquinconazole	0.9991	0.022	0.066	94.3	6.3
	Fthalide	0.9992	0.009	0.028	91.1	3.2
	Hexaconazole	0.9961	0.030	0.090	85.7	5.0
	Prochloraz	0.9991	0.010	0.029	88.7	4.6
	Procymidone	0.9993	0.015	0.045	83.5	5.9
	Uniconazole	0.9999	0.005	0.016	88.3	1.8
Herbicides	Pendimethalin	0.9969	0.005	0.015	101.3	1.3

Table 3 Occurrence of pesticide residues in vegetables

Vegetable	Samples	*N* < LOD		*N* > LOD		*N* > MRL	
Amaranth leaves	8	8	100%	0	0.0%	0	0.0%
Aster scaber	33	28	84.8%	4	12.1%	1	3.0%
Butterbur	9	6	66.7%	3	33.3%	0	0.0%
Chard	25	22	88.0%	3	12.0%	0	0.0%
Chicory	29	29	100%	0	0.0%	0	0.0%
Chili pepper leaves	4	4	100%	0	0.0%	0	0.0%
Coastal hog fennel	14	13	92.8%	0	0.0%	1	7.1%
Crown daisy	65	61	93.8%	4	6.2%	0	0.0%
Curled mallow	25	24	96.0%	1	4.0%	0	0.0%
Dandelion	5	5	100%	0	0.0%	0	0.0%
Lettuce(head)	29	28	96.6%	1	3.4%	0	0.0%
Lettuce(leaves)	149	139	93.3%	9	6.0%	1	0.7%
Mustard green	1	0	0.0%	0	0.0%	1	100%
Mustard leaf	16	12	75.0%	2	12.5%	2	12.5%
Pak choi	25	24	96.0%	1	4.0%	0	0.0%
Perilla leaves	106	85	80.2%	18	17.0%	3	2.8%
Pimpinella brachycarpa	20	18	90.0%	1	5.0%	1	5.0%
Pumpkin (leaves)	4	4	100%	0	0.0%	0	0.0%
Rape leaves	3	2	66.7%	1	33.3%	0	0.0%
Radish leaves	102	97	95.1%	3	2.9%	2	2.0%
Shepherd's purse	21	18	85.7%	2	9.5%	1	4.8%
Spinach	100	90	90.0%	8	8.0%	2	2.0%
Winter-grown cabbage	152	138	90.8%	13	8.6%	1	0.7%
Others	15	14	93.3%	1	6.7%	0	0.0%
Total	960	869	90.5%	75	7.8%	16	1.7%

fungicides 14 (42.4%), insecticides 13 (39.4%), herbicides 3 (9.1%), acaricides 2 (6.1%), and growth regulator 1 (3.0%). The most frequently detected pesticides in vegetables were chlorfenapyr (13 samples, 0.06–3.11 mg/kg), fludioxonil (7 samples, 0.31–3.30 mg/kg), pyridalyl (7 samples, 0.26–3.02 mg/kg), hexaconazole (6 samples, 0.14–1.11 mg/kg), procymidone (6 samples, 0.028–0.185 mg/kg), azoxystrobin (4 samples, 0.31–7.40 mg/kg), bifenthrin (4 samples, 0.06–0.62 mg/kg), chlorpyrifos (4 samples, 0.014–0.106 mg/kg), etofenprox (4 samples, 0.04–0.32 mg/kg), and flubendiamide (4 samples, 0.120–4.870 mg/kg). Similar results were reported in Korea that the detection rates of procymidone, azoxystrobin, and chlorfenapyr were higher than those of other pesticides. Park et al. (2016) found that procymidone (0.1–4.7 mg/kg), azoxystrobin (0.01–7.0 mg/kg), dimethomorph (0.01–6.8 mg/kg), and lufenuron (0.03–-1.3 mg/kg) were frequently detected pesticides in leafy vegetables, and stalk and stem vegetables from South Korea. Yi et al. (2020) reported the detection of azoxystrobin (0.033–40.547 mg/kg), cypermethrin (0.024–9.157 mg/kg), procymidone (0.008–56.049 mg/kg), and tebufenpyrad (0.023–2.8 mg/kg) in vegetables during 2010–2014. In addition, Kim et al. (2014) reported that chlorfenapyr (0.006–2.062 mg/kg), fludioxonil (0.037–1.802 mg/kg), pyridalyl (0.079–1.820 mg/kg), procymidone (0.013–29.521 mg/kg), azoxystrobin (0.536–2.871 mg/kg), bifenthrin (0.012–0.657 mg/kg), and chlorpyrifos (0.010–0.351 mg/kg) were detected in distributed agricultural products. Figure 3 shows the number of pesticide residues detected in vegetables and the number of excess MRLs. It was detected mainly in perilla leaves, winter-grown cabbage, mustard leaf, chard, and aster scaber.

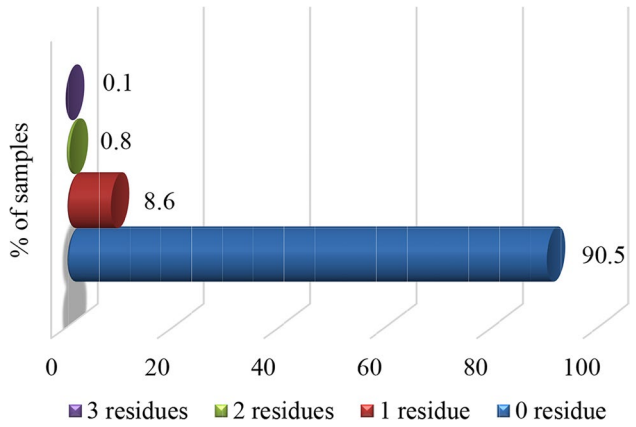

Fig. 2 Incidence percentage of multiple pesticide residues

Table 4 Pesticide residues detected in vegetables

Pesticide	Category	N > LOD (%)	N > MRL (%)	Range (mg/kg)	Mean(mg/kg)	Standard deviation (mg/kg)	MRL (mg/kg)
Bifenthrin	I	4 (0.4%)	0 (0.0%)	0.06–0.62	0.313	0.22	1.0–7.0
Chlorantraniliprole	I	2 (0.2%)	0 (0.0%)	0.41–0.80	0.605	0.28	7.0
Chlorfenapyr	I	13 (1.4%)	0 (0.0%)	0.06–3.11	0.678	0.58	3.0–9.0
Chlorpyrifos	I	4 (0.4%)	1 (0.1%)	0.014–0.106	0.064	0.06	0.05–0.2
Cyhalothrin	I	2 (0.2%)	0 (0.0%)	0.32–0.48	0.400	0.11	3.0
Cypermethrin	I	1 (0.1%)	0 (0.0%)	3.05	3.050	0.00	5.0
Diazinon	I	3 (0.3%)	3 (0.3%)	0.030–0.999	0.425	0.56	0.01
Etofenprox	I	4 (0.4%)	0 (0.0%)	0.04–0.32	0.142	0.11	0.5–15
Fenitrothion	I	1 (0.1%)	0 (0.0%)	0.011	0.011	0.00	0.05
Flubendiamide	I	4 (0.4%)	2 (0.2%)	0.120–4.870	2.109	2.224	0.02–15
Lufenuron	I	3 (0.3%)	0 (0.0%)	0.22–0.31	0.255	0.05	5.0–7.0
Pyridalyl	I	7 (0.7%)	0 (0.0%)	0.26–3.02	1.145	1.09	5.0–15
Tebupirimfos	I	1 (0.1%)	1 (0.1%)	0.033	0.033	0.00	0.01
Azoxystrobin	F	4 (0.4%)	0 (0.0%)	0.31–7.40	3.253	3.20	3.0–30
Dimethomorph	F	1 (0.1%)	0 (0.0%)	1.31	1.310	0.00	30
Diniconazole	F	3 (0.3%)	0 (0.0%)	0.03–0.10	0.075	0.035	0.3
Fludioxonil	F	7 (0.7%)	0 (0.0%)	0.31–3.30	2.380	0.552	15–40
Fluopyram	F	3 (0.3%)	0 (0.0%)	0.08–0.21	0.127	0.06	0.5
Fluquinconazole	F	2 (0.2%)	2 (0.2%)	0.108–0.138	0.123	0.02	0.01
Fthalide	F	1 (0.1%)	1 (0.1%)	0.075	0.075	0.00	0.01
Hexaconazole	F	6 (0.6%)	1 (0.1%)	0.14–1.11	0.246	0.17	0.7
Iprodione	F	2 (0.2%)	0 (0.0%)	1.56–4.03	2.795	1.75	20
Prochloraz	F	1 (0.1%)	1 (0.1%)	0.039	0.039	0.00	0.01
Procymidone	F	6 (0.6%)	2 (0.2%)	0.028–0.185	0.053	0.04	0.05
Tebuconazole	F	3 (0.3%)	0 (0.0%)	0.042–3.985	1.377	2.27	0.05–15
Thifluzamide	F	2 (0.2%)	0 (0.0%)	0.013–0.080	0.047	0.05	0.05–5.0
Uniconazole	F	1 (0.1%)	1 (0.1%)	0.025	0.025	0.00	0.01
Napropamide	H	1 (0.1%)	0 (0.0%)	0.018	0.018	0.00	0.1
Pendimethalin	H	3 (0.3%)	1 (0.1%)	0.037–2.204	0.769	1.239	0.05–0.2
Trifluralin	H	1 (0.1%)	0 (0.0%)	0.013	0.013	0.00	0.05
Fenpyroximate	A	1 (0.1%)	0 (0.0%)	1.33	1.330	0.00	7.0
Tebufenpyrad	A	3 (0.3%)	0 (0.0%)	0.18–0.42	0.260	0.12	5.0
Paclobutrazol	G	1 (0.1%)	0(0.0%)	0.13	0.130	0.00	5.0

A acaricide, *F* fungicide, *G* growth regulator, *H* herbicide, I insecticide

Chlorfenapyr is a pyrrole insecticide used for control of various insects and mites on vegetable crops. It has been accepted for use in several countries including the Republic of Korea, USA, Australia, Brazil, Japan, and Mexico. Fludioxonil is a phenylpyrrole fungicide that is widely used in the agriculture sector. In many toxicological studies, fludioxonil toxicity is low or negligible for mammals, but is highly toxic to some aquatic organisms (Buschart et al. 2012).

Of the 960 samples analyzed, 16 (1.7%) samples exceeded the MRLs. Agricultural products exceeding the MRLs are mostly leafy vegetables with wide leaves. Leafy vegetables have a large surface area, so they are more likely to be attached when pesticides are used. In addition, pesticide residues on the surface of leafy vegetables are less likely to be decomposed by rain, wind, and photolysis due to the uneven surface properties. The types of pesticides exceeding MRLs in agricultural products are fungicides 6 (54.5%), insecticide 4 (36.4), and herbicide 1 (9.1%). Chlorpyrifos, diazinon, flubendiamide, fluquinconazole, fthalide, hexaconazole, pendimethalin, prochloraz, procymidone, tebupirimfos, and uniconazole had residues that violated MRLs. Perilla leaves, mustard leaf, radish leaves, and spinach were the vegetables with the highest number of samples with residues above MRLs. Mustard green, mustard leaf, and coastal hog

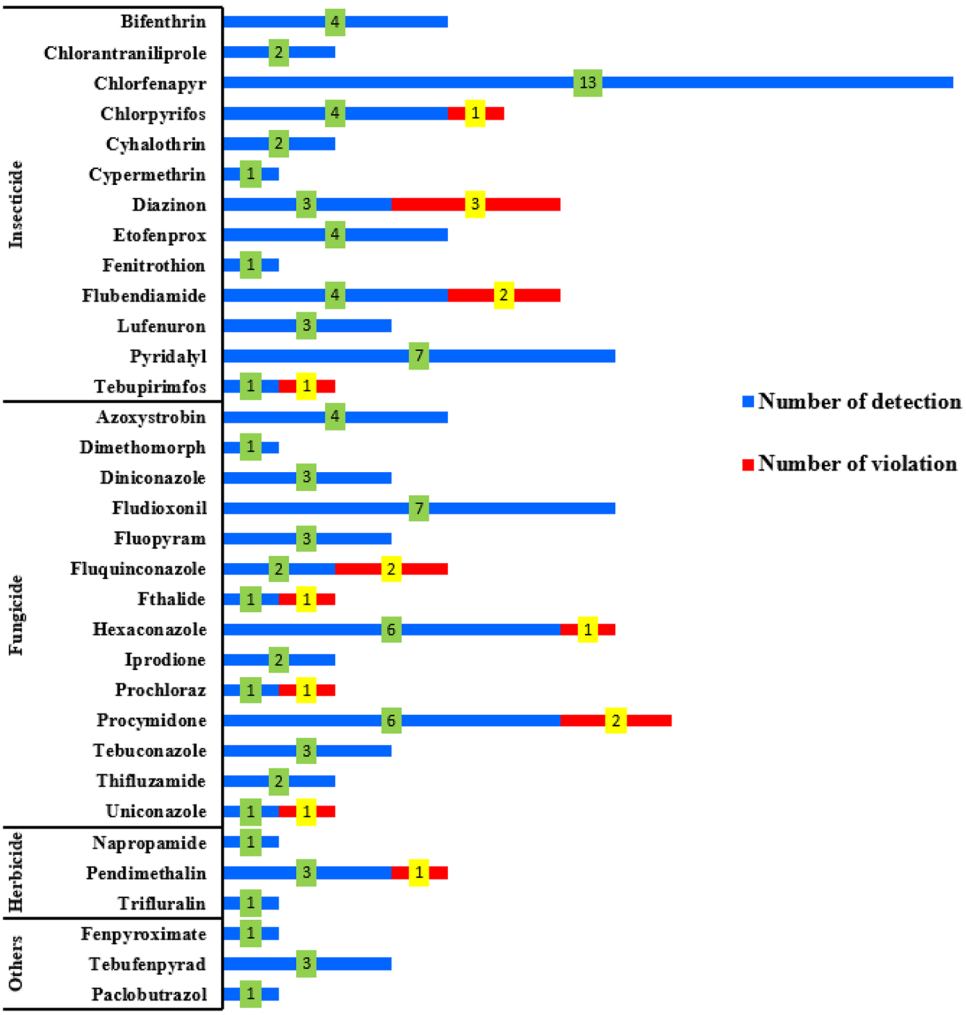

Fig. 3 Number of pesticide residues detected in vegetables and number of excess MRLs

fennel had the highest violations rate of 100%, 12.5%, and 7.1%, respectively. The most frequently violated pesticides were diazinon, flubendiamide, fluquinconazole, and procymidone. In a report by the Gyeonggido Institute of Health and Environment (GIHE 2012), procymidone, diazinon, and chlorpyrifos were shown to have the highest percentage of residues exceeding MRLs. In this study, the violations of diazinon were the highest. Diazinon was detected in *Pimpinella brachycarpa* and radish leaves. It is a widely used pesticide in agriculture to kill insects on fruit, vegetable, nut, and filed crops. Diazinon is an insecticide that belongs to the organophosphorus group and is able to inhibit the function of enzyme acetylcholinesterase and affect the central nervous system (Dhull et al. 2013).

The types of pesticides that exceed the MRLs are generally similar to other reported results in Korea (Yi et al. 2020; Park et al. 2016). According to other research papers in foreign countries, Szpyrka et al. (2015) reported 1.8% exceedance of MRLs in fruits and vegetables analyzed in Poland. The United States Food and Drug Administration reported that violation was detected in 2% of the domestic vegetables and 7% of the imported vegetables. Algharibeh and Alfararjeh (2018) found that 22% of fruits and vegetables exceeded MRLs in Jordan. Each country has different growing environments and eating habits of agricultural products, so even the same crops have different MRLs. Korea's MRLs were established by harmonizing with the CODEX standard. However, some agricultural products have recently been set lower than the limits of other countries and CODEX. In the EU, Japan, and Taiwan, Positive List System (PLS) is implemented for pesticides for which residue standards have not been established. The United States and Australia have applied the "zero tolerance" principle for pesticides for which standards have not been established. Table 5 shows the comparison of pesticides exceeding MRL in Korea and MRL in other countries. Differences in maximum residual levels were found in several countries, and the EU's MRLs were considered to be the most stringent. Although Korea's MRL was exceeded, it was found that some agricultural products did not exceed the MRL of other countries.

Table 5 Comparison with other countries for pesticides detected exceeding MRL in vegetables

Pesticide	Vegetable	Maximum (mg/kg)	Korea (mg/kg)	EU (mg/kg)	Japan (mg/kg)	Taiwan (mg/kg)	Australia (mg/kg)
Chlorpyrifos	Coastal hog fennel	0.106	0.05	0.01	0.01	1.0	0.01
Diazinon	*Pimpinella brachycarpa*	0.030	0.01	0.01	0.1	0.5	0.7
	Radish leaves	0.999	0.01	0.01	0.03	0.5	0.7
Flubendiamide	Mustard green	0.507	0.02	0.01	5.0	NA[a]	10
	Mustard leaf	0.120	0.02	0.01	5.0	NA	10
Fluquinconazole	Spinach	0.138	0.01	0.01	0.01	UP[b]	NA
Fthalide	Aster scaber	0.075	0.01	UP[b]	0.01	0.01	UP
Hexaconazole	Mustard leaf	1.110	0.7	0.01	0.01	0.5	NA
Pendimethalin	Shepherd's purse	2.204	0.05	0.05	0.1	0.1	0.05
Prochloraz	Lettuce(leaves)	0.039	0.01	0.03	0.01	0.01	NA
Procymidone	Perilla leaves	0.185	0.05	0.03	2.0	5.0	NA
Tebupirimfos	Winter-grown cabbage	0.033	0.01	UP	UP	UP	UP
Uniconazole	Perilla leaves	0.025	0.01	UP	0.01	UP	NA

[a]NA not applicable, because of no existing MRLs

[b]UP unregistered pesticide

Risk assessment

The risk from pesticide residues in vegetables was assessed for all detected pesticide in the samples. The results of the risk assessment to pesticides in consideration of vegetable consumption are shown in Table 6. The EDIs of pesticide range from 3.00×10^{-7} to 1.57×1^{-2} mg/kg bw/day, and the range of HQs (percent EDI to ADI ration) was 0.002–90.621%. The HQs for pyridalyl and flubendiamide were 0.018–56.143% and 0.007–90.621%, respectively, and were higher than for other pesticides. However, it was less than 100%. The results show that potential health risks are mainly associated with flubendiamide and pyridalyl detected in spinach and perilla leaves. Park et al. (2016) reported health risk for consumer in vegetables. They found HQ values for procymidone (0.124–0.446%), diazinon (0.240–2.410%), azoxystrobin (0.042–0.150%), EPN (0.272–30.389%), and chlorfenapyr (0.213–1.707%). Chen et al. (2011) reported pesticide residues in fruit and vegetables including pakchoi cabbage and lettuces. They found HQ values for omethoate (2.6%), methamidophos (2.2%), and chlorpyrifos (0.24%). Kim et al. (2014) reported that the HQs for cypermethrin, dimethomorph, procymidone, chlorpyrifos, and chlorothalonil in vegetables were 28.6%, 16.2%, 11.7%, 11.1%, and 8.7%, respectively. The HQ value of the results of this study was similar to or slightly higher than that of other papers. In a recent study in Chile, Elgueta et al. (2017) revealed that the HQs ranges from 0.0 to 73.9% in the leafy vegetables. The HQs for pesticides decreased in the order of methamidophos (73.9) > cypermethrin (30.4) > mancozeb (11.5) > cyfluthrin (4.5) > lambda-cyhalothrin (3.0). The results showed that methamidophos and cypermethrin were potentially associated with health risks in leafy vegetables. The HQ value above 100% indicates a potential risk to consumers (Yu et al. 2016; Chun and Kang 2003). Therefore, the results indicate that the detected pesticides in this study are not harmful to human health. Also, vegetables are usually consumed after washing. Since the pesticide residues are reduced by 8–68% with flowing water at home depending on the type of pesticide residues and characteristics of vegetables, the risk of pesticide residues can be lowered (Kwon et al. 2013). Additionally, agricultural products exceeding MRLs are seized by the authorities and discarded immediately in Korea, which make the risk of pesticides for consumer's intake of vegetables not serious. However, it is necessary to continuously monitor and manage agricultural products for food safety because the amount of intake can vary depending on the actual consumer's preference, season, and region (Park et al. 2020).

Conclusion

A total of 960 vegetables obtained from different markets in Incheon during 2019 were analyzed for pesticide residues in order to evaluate a health risk assessment to pesticides. A multi-residue method was applied to extract pesticides for vegetables. GC–MS/MS, GC-ECD/NPD, LC–MS/MS, and LC-UVD were used to identify and quantify the compounds. According to the results, pesticide residues were found in 9.5% of the samples and exceeded the MRLs in 1.7% of the

Table 6 Risk assessment for detected pesticides in vegetables

Pesticide	Vegetable	Average of detection value (mg/kg)	ADI[a] (mg/kg bw/day)	EDI[b] (mg/kg bw/day)	HQ[c] (%)
Azoxystrobin	Aster scaber	0.31	0.2	0.0003906	0.195
	Crown daisy	7.40	0.2	0.0038480	1.924
	Lettuce(leaves)	1.20	0.2	0.0073800	3.690
	Perilla leaves	4.10	0.2	0.0113160	5.658
Bifenthrin	Butterbur	0.47	0.01	0.0001645	1.645
	Spinach	0.41	0.01	0.0021484	21.484
	Wintergrown cabbage	0.06	0.01	0.0000006	0.006
Chlorantraniliprole	Lettuce(leaves)	0.601	2.0	0.0036962	0.185
Chlorfenapyr	Aster scaber	0.665	0.026	0.0008379	3.223
	Butterbur	0.905	0.026	0.0003168	1.218
	Chard	1.805	0.026	0.0003430	1.319
	Lettuce(leaves)	0.18	0.026	0.0011070	4.258
	Perilla leaves	0.45	0.026	0.0012420	4.777
	Shepherd's purse	0.06	0.026	0.0000210	0.081
	Wintergrown cabbage	0.683	0.026	0.0000068	0.026
Chlorpyrifos	Coastal hog fennel	0.106	0.01	0.0000085	0.085
	Curled mallow	0.017	0.01	0.0000078	0.078
	Radish leaves	0.014	0.01	0.0000003	0.003
	Wintergrown cabbage	0.12	0.01	0.0000012	0.012
Cyhalothrin	Perilla leaves	0.40	0.02	0.0011040	5.520
Cypermethrin	Rape leaves	3.05	0.02	0.0000305	0.153
Diazinon	*Pimpinella brachycarpa*	0.030	0.0002	0.0000111	5.550
	Radish leaves	0.819	0.0002	0.0000164	8.190
Dimethomorph	Rape leaves	1.30	0.2	0.0000130	0.007
Diniconazole	Rape leaves	0.10	0.0023	0.0000010	0.043
	Wintergrown cabbage	0.05	0.0023	0.0000005	0.022
Etofenprox	Shepherd's purse	0.01	0.03	0.0000035	0.012
	Spinach	0.20	0.03	0.0010480	3.493
	Wintergrown cabbage	0.215	0.03	0.0000022	0.007
Fenitrothion	Lettuce(leaves)	0.011	0.005	0.0000677	1.353
Fenpyroximate	Perilla leaves	1.30	0.01	0.0035880	35.880
Flubendiamide	Mustard green	0.507	0.017	0.0000101	0.060
	Mustard leaf	0.12	0.017	0.0000012	0.007
	Perilla leaves	4.87	0.017	0.0134412	79.066
	Spinach	2.94	0.017	0.0154056	90.621
Fludioxonil	Chard	3.00	0.4	0.0005700	0.143
	Lettuce(head)	2.20	0.4	0.0025080	0.627
	Perilla leaves	1.94	0.4	0.0053599	1.340
Fluopyram	Lettuce(leaves)	0.127	0.01	0.0007811	7.811
Fluquinconazole	Spinach	0.123	0.002	0.0006445	32.226
Fthalide	Aster scaber	0.075	0.04	0.0000945	0.236
Hexaconazole	Crown daisy	0.140	0.005	0.0000728	1.456
	Mustard leaf	0.438	0.005	0.0000044	0.088
	Perilla leaves	0.16	0.005	0.0004416	8.832
Iprodione	Perilla leaves	4.0	0.06	0.0110400	18.400
	Spinach	1.5	0.06	0.0078600	13.100
Lufenuron	Perilla leaves	0.29	0.015	0.0008004	5.336
	Spinach	0.22	0.015	0.0011528	7.685
Napropamide	Crown daisy	0.018	0.3	0.0000094	0.003

Table 6 (continued)

Pesticide	Vegetable	Average of detection value (mg/kg)	ADI[a] (mg/kg bw/day)	EDI[b] (mg/kg bw/day)	HQ[c] (%)
Paclobutrazol	Perilla leaves	0.10	0.022	0.0002760	1.255
Pendimethalin	Shepherd's purse	2.20	0.13	0.0007714	0.593
	Spinach	0.07	0.13	0.0003668	0.282
	Wintergrown cabbage	0.037	0.13	0.0000004	0.0003
Prochloraz	Lettuce(leaves)	0.039	0.01	0.0002399	2.399
Procymidone	Crown daisy	0.028	0.1	0.0000146	0.015
	Perilla leaves	0.114	0.1	0.0003146	0.315
	Pimpinella brachycarpa	0.037	0.1	0.0000137	0.014
	Spinach	0.034	0.1	0.0001782	0.178
Pyridalyl	Aster scaber	0.87	0.028	0.0010962	3.915
	Pak choi	1.1	0.028	0.0000550	0.196
	Radish leaves	0.26	0.028	0.0000052	0.019
	Spinach	3.0	0.028	0.0157200	56.143
	Wintergrown cabbage	0.494	0.028	0.0000049	0.018
Tebuconazole	Aster scaber	0.042	0.03	0.0000529	0.176
	Godeulppaegi	0.09	0.03	0.0000009	0.003
	Perilla leaves	4.0	0.03	0.0110400	36.800
Tebufenpyrad	Perilla leaves	0.26	0.01	0.0007176	7.176
Tebupirimfos	Wintergrown cabbage	0.033	0.0002	0.0000003	0.165
Thifluzamide	Lettuce(leaves)	0.013	0.014	0.0000800	0.571
	Perilla leaves	0.08	0.014	0.0002208	1.577
Trifluralin	Radish leaves	0.013	0.015	0.0000003	0.002
Uniconazole	Perilla leaves	0.025	0.016	0.0000690	0.431

[a]ADI: acceptable daily intake

[b]EDI: estimated daily intake, average of detection value (mg/kg) × daily food intake (g/day)/1000

[c]HQ: hazard quotient (EDI/ADI × 100)

total vegetables. Risk assessment through intake of vegetables was done for all detected pesticides. The most frequently detected pesticides were chlorfenapyr, fludioxonil, hexaconazole, procymidone, and pyridalyl, while the highest rate of violations were diazinon, flubendiamide, fluquinconazole, and procymidone. Based on the findings, the range of HQs was 0.002–90.621%, which was below 100%. Therefore, pesticides detected in this study were not exposed to health risks with the consumption of vegetables. The results can improve understanding of the importance of pesticide residues in agricultural products. Also, monitoring and health risk assessment of pesticide residues in agricultural products are expected to improve the food safety for the Korean consumer.

Acknowledgements This work was supported by a project of Samsan Agricultural Products Inspection Center, Incheon Institute of Public Health and Environment, Republic of Korea. Thanks to the members of Samsan Agricultural Products Inspection Center for their help and advice in analyzing pesticide residues.

Author contribution All the authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by Byung Kyu Park, Seung Hye Jung, Sung Hee Kwon, Sun Hoi Kim, Eun Young Ye, Mi Sook Yeom, Soon Jae Seo, Kwang Sig Joo, Myung Je Heo, and Geun Pyo Hong. The first draft of the manuscript was written by Byung Kyu Park, and all the authors commented on the previous versions of the manuscript. All the authors read and approved the final manuscript.

Data availability Not applicable.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

References

Alavanja MCR, Ross MK, Bonner MR (2013) Increased cancer burden among pesticide applicators and others due to pesticide exposure. Cancer J Clin 63(2):120–142

Algharibeh GR, Alfararjeh MS (2018) Pesticide residues in fruits and vegetables in Jordan using liquid chromatography/tandem mass spectrometry. Food Addit Contam Part B 12(1):65–73

Baldi I, Filleul L, Mohammed-Brahim B, Fabrigoule C, Dartigues JF, Schwall S, Drevet JP, Salamon R, Brochard P (2001) Neuropsychologic effects of long-term exposure to pesticides: results from the French Phytoner study. Environ Health Perspect 109(8):839–844

Buschart A, Burakowska A, Bilitewski U (2012) The fungicide fludioxonil antagonizes fluconazole activity in the human fungal pathogen candida albicans. J Medi Micro 61(12):1696–1703

Chen C, Qian Y, Tao C, Li Y (2011) Evaluation of pesticide residues in fruits and vegetables from Xiament. China Food Control 22(7):1114–1120

Cho TH, Kim BS, Jo SJ, Kang HG, Choi BY, Kim MY (2009) Pesticide residue monitoring in Korean agricultural products, 2003–05. Food Addit Contam Part B 2(1):27–37

Chun OK, Kang HG (2003) Estimation of risks of pesticide exposure by food intake to Koreans. Food Chem Toxicol 41(8):1063–1076

Chun OK, Shin KY, Lee JH, Bak JS, Cho TH, Kim TR, Kang HG (2002) A study on current status of pesticide residues in commercial agricultural products, 2001. Food Sci Biotech 11(6):602–607

Codex (1993) Guidelines on good laboratory practice in residue analysis CAC/GL 40–1993. CODEX Alimentarius Committee, Italy

Darko G, Akoto O (2008) Dietary intake of organophosphorus pesticide residues through vegetables from Kumasi. Ghana Food Chem Toxicol 46(12):3703–3706

Dhull V, Gahlaut A, Dilbaghi N, Hooda V (2013) Acetylcholinesterase biosensors for electrochemical detection of organophosphorus compounds: a review. Biochem Res Int 2013:18

Elgueta S, Moyano S, Sepúlveda P, Quiroz C, Correa A (2017) Pesticide residues in leafy vegetables and human health risk assessment in north central agricultural areas of Chile. Food Addit Contam Part B 10(2):105–112

Gad Alla SA, Loutfy NM, Shendy AH, Ahmed MT (2015) Hazard index, a tool for a long term risk assessment of pesticide residues in some commodities, a pilot study. Regul Toxicol Pharmacol 73(3):985–991. https://doi.org/10.1016/j.yrtph.2015.09.016

GIHE (2012) Risk assessment for pesticide residues of agricultural products in Gyeonggido. Gyeonggido Institute of Health and Environment, Korea

Juraske R, Antón A, Castells F, Huijbregts MAJ (2007) Human intake fractions of pesticides via greenhouse tomato consumption: comparing model estimates with measurements for captan. Chemosphere 67(6):1102–1107

KDCA (2018) Korea Disease Control and Prevention Agency. Korean national health and nutrition examination survey 2018. https://knhanes.cdc.go.kr/knhanes/main.do. Accessed 01 Oct 2020

KFC (2017) Korea Food Code 2017. Analytical manual for pesticide residues in food, 5rd edn. Korea Food and Drug Administration, Korea

KFDA (2020) Korea Food and Drug Administration. http://foodsafetykorea.go.kr/residue/prd/mrls/list.do?menuKey=1&subMenuKey=161. Accessed 19 Sep 2020

Kim NH, Lee JS, Kim OH, Choi YH, Han SH, Kim YH, Kim HS, Lee SR, Lee JM, Yu IS, Jung K (2014) Monitoring of pesticide residues and risk assessment on agricultural products marketed in the northern area of Seoul in 2013. J Food Hyg Saf 29(3):170–180

KSIS (2020) Korean Statistical Information Service. Status of the average weight distribution by gender and age by city. http://kosis.kr/statHtml/statHtml.do?orgId=350&tbIId=DT_35007_N132&vw_cd=MT_ZTITLE&list_id=350_35007_A007&seqNo=&lang_mode=ko&language=kor&obj_var_id=&itm_id=&conn_path=MT_ZYTTLE. Accessed 01 Oct 2020

Kwon HY, Kim TK, Hong SM, Kim CS, Baeck MK, Kim DH, Son KA (2013) Removal of pesticide residues in field sprayed leafy vegetables by different washing method. Korean Soc Pesti Sci 17(4):237–243. https://doi.org/10.7585/kjps.2013.17.4.237

Lee SM, Papathakis ML, Feng HMC, Hunter GF, Carr JC (1991) Multipesticide residue method for fruits and vegetables: California department of food and agriculture. Fresenius J Anal Chem 339(6):376–383

Mansour SA (2004) Pesticide exposure – Egyptian scene. Toxicology 198(1):91–115

Nougadère A, Sirot V, Kadar A, Fastier A, Truchot E, Vergnet C, Hommet F, Baylé J, Gros P, Leblanc J-C (2012) Total diet study on pesticide residues in France: levels in food as consumed and chronic dietary risk to consumers. Environ Int 45(15):135–150

Park DW, Kim KG, Choi EA, Kang GR, Kim TS, Yang YS, Moon SJ, Ha DR, Kim ES, Cho BS (2016) Pesticide residues in leafy vegetables, stalk and stem vegetables from South Korea: a long-term study on safety and health risk assessment. Food Addit Contam Part A 33(1):105–118

Park BK, Jung SH, Kwon SH, Ye EY, Lee HJ, Seo SJ, Joo KS, Heo MJ (2020) Monitoring and risk assessment of pesticide residues on stalk and stem vegetables marketed in Incheon metropolitan area. J Food Hyg Saf 35(4):365–374. https://doi.org/10.13103/JFHS.2020.35.4.365

Prodanov M, Sierra I, Vidal-Valverde C (2004) Influence of soaking and cooking on the thiamin, riboflavin and niacin contents of legumes. Food Chem 84(2):271–277

Shashirekha MN, Mallikarjuna SE, Rajarathnam S (2015) Status of bioactive compounds in foods with focus on fruits and vegetables. Crit Rev Food Sci Nutr 55(10):1324–1339

Szpyrka E, Kurdziel A, Matyaszek A, Podbielska M, Rupar J, Slowik-Borowiec M (2015) Evaluation of pesticide residues in fruits and vegetables from the region of south-eastern Poland. Food Control 48:137–142

Yi YJ, Joung HJ, Kum JY, Hwang IS, Kim MS (2020) Pesticide residues in vegetables and risk assessment for consumers in Korea during 2010–2014. Food Addit Contam Part A 37(8):1300–1313

Yu R, Liu Q, Liu J, Wang Q, Wang Y (2016) Concentrations of organophosphorus pesticides in fresh vegetables and related human health risk assessment in Changchun, Northeast China. Food Control 60:353–360. https://doi.org/10.1016/j.foodcont.2015.08.013