

Chemical Residues and Contaminants in Foods of Animal Origin in Korea during the Past Decade

MeeKyung Kim,* Byung-Hoon Cho, Chae-mi Lim, Dong-Gyu Kim, So Young Yune, Jin Young Shin, Young Hoon Bong, JeongWoo Kang, Myeong-ae Kim, and Seong-Wan Son

Animal, Plant, Fisheries Quarantine and Inspection Agency, 175 Anyangro, Anyang, Gyeonggido 430-757, Republic of Korea

ABSTRACT: Residues of veterinary drugs, pesticides, and environmental contaminants in domestic and imported foods of animal origin were monitored by the National Residue Program and inspection service in Korea in the past decade. In all, 134 substances were analyzed in the monitoring plan; 35 substances were examined in the surveillance and enforcement testing program, and 27 substances were investigated in exploratory projects. The overall trend of violation rates gradually decreased over the past decade. Pesticides were not found in any domestic samples of animal origin. The violation rates of chlortetracycline and oxytetracycline decreased, but quinolone and penicillin detections increased in Korea. Several kinds of residue violations of veterinary drugs, endosulfan, or dioxins were found in the imported products each year. In an example event in 2008, the Korea monitoring plan contributed globally to investigate the dioxin contamination from Chilean pork. Continuous monitoring based on internationally harmonized standards and methods provides the essential scientific basis to manage and ensure food safety.

KEYWORDS: *veterinary drugs, pesticides, environmental contaminants, food of animal origin, Korean National Residue Program*

■ INTRODUCTION

Analyses of chemical residues, including veterinary drugs and contaminants such as pesticides and dioxins, are an important responsibility of the Animal, Plant and Fisheries Quarantine and Inspection Agency (QIA) to ensure food safety for domestic and imported products. Prior to June 2011, the QIA was known as the National Veterinary Research and Quarantine Service.

The National Residue Program (NRP) for domestic and import residue testing serves as a control system for chemical residues and contaminants in foods of animal origin in Korea.¹ The domestic NRP is for prevention of residue occurrence on the farm, and the import testing is for verification of residue control program of countries exporting to Korea. However, the sampling components are similar between domestic NRP and import testing. The NRP aims to ensure the safety of livestock products by providing guidance for effective implementation of testing and control of harmful chemical residues. Its focus is on public health protection to ensure safe food. Also, the purpose of the NRP is to assess human exposure, to restrain the slaughter and processing of adulterated animals, to identify violative product for removal from the food supply, and to verify hazard analysis critical control points (HACCP) in the slaughtering and processing of animal-derived foods. The obtained data are used for risk assessment, enforcement, educational activities, appropriate regulatory follow-up, and the planning of future activities. There are three components in both domestic NRP and import testing. First, the monitoring plan is conducted in accordance with a statistical random sampling of animals that have passed ante-mortem inspection at slaughterhouses. The monitoring data identify and evaluate compounds of health concern, and regulatory actions are taken when violations are found. Results are reported, which are taken into account to plan subsequent activities. Second,

surveillance and enforcement testing is conducted in accordance with targeted sampling plans, which are focused on individual animals or lots that do not appear healthy. The testing is targeted to animals from farms that previously had residue violations and residue-suspected animal samples taken from slaughterhouses. Third, exploratory projects entail studies to determine the occurrence of residues that do not have a safe limit. These results could reflect changes to the monitoring plan and surveillance and enforcement testing.

Chemicals that have maximum residue limits (MRLs) in Korea were selected for testing of veterinary drugs, pesticides, and environmental contaminants, which can develop antibiotic resistance, hypersensitivity, acute and chronic poisoning, endocrine disruption, or carcinogenesis to animals and/or humans. Currently, Korean MRLs have been established for 150 veterinary drugs, 83 pesticides, and 6 environmental contaminants.² There are 12 banned substances that should not be found in foods of animal origin, which consist of chloramphenicol, chlorpromazine, clenbuterol, colchicine, diethylstilbesterol, dimetridazole, malachite green and its metabolites, medroxyprogesterone acetate, pyrimethamine, thiouracil, vancomycin, and nitrofurans (furazolidone, furaltadone, nitrofurazone, nitrofurantoin, nitrovin, etc.) and their metabolites. Analytical processes were conducted at the QIA and the provincial veterinary services according to an annual sampling plan. The aim of this paper is to present the results obtained from the Korean NRP for domestic and inspection

Special Issue: Florida Pesticide Residue Workshop 2012

Received: October 30, 2012

Revised: February 9, 2013

Accepted: February 12, 2013

Published: February 12, 2013

Table 1. Outlines of Residue Analysis for Veterinary Drugs, Pesticides, and Dioxins in Foods of Animal Origin

residue class	sample	sample extraction and cleanup	instrumentation	LOQ	ref
animal drugs					
penicillins	bovine, swine, poultry (muscle, kidney)	(1) Add 10 mL of 0.1 M sodium phosphate buffer (pH 4.5) to 2 g of sample and mix. (2) Add 2.5 mL of 0.17 M sulfuric acid and 2.5 mL of 5% sodium tungstate and mix thoroughly. (3) Centrifuge at 5000 rpm for 5 min and collect upper layer. (4) HLB SPE cartridge cleanup. (5) Concentrate and then filter through 0.2 μ m PVDF.	HPLC or LC-MS/MS equipped with C18 column (2.1 \times 150 mm, 3.5 μ m)	6–45 μ g/kg	3
					4
					5
tetracyclines	bovine, swine, poultry (muscle, kidney, liver)	(1) Add oxalic acid and EDTA to 0.1 g of sample and mix. (2) Extract with 5 mL of acetonitrile/ethyl acetate (2:1, v/v). (3) Centrifuge, collect upper layer, and concentrate. (4) Filter through 0.2 μ m filter.	HPLC or LC-MS/MS equipped with C18 column (2.1 \times 150 mm, 3.5 μ m)	6–80 μ g/kg	3
					4
					5
sulfonamides	bovine, swine, poultry (muscle)	(1) Add 1 mL of 5 mM potassium phosphate to 1 g of sample and mix. (2) Extract with 10 mL of acetonitrile and centrifuge. (3) Take upper layer, add 15 mL of hexane, and mix. (4) Centrifuge, collect lower layer, and concentrate. (5) Add 0.1% formic acid and deionized water. (6) Sonicate and centrifuge. (7) Collect upper layer and then filter through 0.2 μ m filter.	HPLC or LC-MS/MS equipped with C18 column (3.9 \times 150 mm, 3 μ m)	0.25–70 μ g/kg	3
					4
					5
quinolones	bovine, swine, poultry (muscle)	(1) Shake 2 g of sample with 2 mL of 2.5% trichloroacetic acid for 15 min. (2) Add 10 mL of acetonitrile/ethyl acetate (2:1, v/v) and shake. (3) Centrifuge for 15 min and collect the upper layer. (4) After triple extraction, add 1 mL of 0.1% formic acid. (5) Centrifuge and collect upper layer and then filter through a 0.2 μ m filter.	HPLC or LC-MS/MS equipped with C18 column (2.1 \times 100 mm, 3.5 μ m)	0.25–50 μ g/kg	3
					4
					5
					6
pesticides					
organochlorines, organophosphates, pyrethroids	bovine, swine, poultry (fat)	(1) Extract 2 g of fat sample with 50 mL of hexane. (2) Add hexane-saturated acetonitrile and shake. (3) Freeze acetonitrile layer at -70 $^{\circ}$ C. (4) Centrifuge, take upper layer, and concentrate to 1 mL.	GC-ECD equipped with DB-608 column (30 m \times 0.25 mm i.d., 0.25 μ m film thickness)	3–50 μ g/kg	3
					7
					8
					9
carbamate	bovine, swine, poultry (muscle)	(1) Extract 5 g of sample with 30 mL of acetonitrile. (2) Add hexane-saturated acetonitrile and shake. (3) Take acetonitrile layer and concentrate. (4) Clean up by Sep-Pak cartridge and make up to 1 mL.	HPLC-FLD (with postcolumn reaction system) equipped with carbamate analysis column (150 mm \times 3.9 mm)	<50 μ g/kg	3 10
dioxins					
PCDD/Fs	bovine, swine, poultry (muscle)	(1) Extract 5 g of sample with 150 mL of hexane/dichloromethane (1:1, v/v) using a Soxhlet extractor or melted fat of meat extract using a separatory funnel. (2) Clean up extracts by Power-Prep system (FMS, USA) using triphasic silica, basic alumina, and carbon columns. (3) Concentrate to 10 μ L.	HR-GC-MS equipped with DB5MS column (60 m \times 0.25 mm i.d., 0.25 μ m film thickness)	0.4–2.8 pg/g fat	11
					12
					13

service testing for imported products from 2002 to 2011 and to show monitoring trends.

MATERIALS AND METHODS

Sample Collection. Monitoring Plan. Veterinarians in provincial veterinary service laboratories collect samples of urine or blood from livestock animals and screen for antimicrobial residues on the basis of requests by farmers who want to make a shipment for slaughter. Inspectors collect random samples of tissue (muscle, fat, kidney, or liver) at the slaughterhouse and send the samples to provincial veterinary service laboratories to test for antibiotics, synthetic antimicrobials, hormones, and pesticides. Imported samples were randomly selected by the Korean automated import information system (AIIS) based on the annual residue testing plan.

Surveillance and Enforcement Testing. Inspectors take samples from individual suspect animals and suspect population animals at the slaughterhouse and send the samples to provincial veterinary service laboratories to test for penicillin, tetracycline, and quinolone antibiotics, synthetic antimicrobials, hormones, and pesticides.

Exploratory Projects. Inspectors take random samples at the slaughterhouse and ship the samples to the veterinary service laboratory headquarters to investigate certain chemical residues of interest including environmental contaminants such as dioxins.

Chemicals and Reagents. Reference standards were purchased from Sigma-Aldrich (St. Louis, MO, USA) for veterinary drugs and from AccuStandard (New Haven, CT, USA) for pesticides. Dioxins (PCDD/Fs) were purchased from Cambridge Isotope Laboratories (Andover, MA, USA) and Wellington Laboratory (Guelph, ON, Canada). All solvents used for the analysis were of HPLC grade, purchased from J. T. Baker (Phillipburg, NJ, USA). Silica, alumina, and carbon columns for dioxin analysis were obtained from FMS Inc. (Waltham, MA, USA).

Chemical Analysis. Multiresidue analyses were applied to residues of veterinary drugs, pesticides, and persistent organic pollutants (POPs), respectively. International or local interlaboratory proficiency tests were regularly conducted for analytical quality assurance purposes. Table 1 describes the outlines of residue analysis for veterinary drugs, pesticides, and dioxins in products of bovine, swine, and poultry. Residues of veterinary drugs were first analyzed using a

bioassay for screening purposes and by HPLC and LC-MS/MS for confirmation and quantification.^{3–6} Residues of pesticides were analyzed by HPLC, LC-MS, GC, and GC-MS.^{3,7–10} Residues of dioxins were analyzed using isotope dilution high-resolution gas chromatography–mass spectrometry (HR-GC-MS).^{3,11–13} Quality controls of the analyses were conducted, and the proficiency tests were conducted on a regular basis for samples obtained from FAPAS in the United Kingdom. The laboratories were accredited (ISO/IEC 17025) for the veterinary drugs, pesticides, and dioxin analyses.

RESULTS AND DISCUSSION

The sampling plans were varied every year depending on the violation rates in the previous year. For example, Table 2 shows

Table 2. Summary of Substances Tested by Domestic Sampling Plan in 2011

program	species	substances tested	no. of samples (head)	rate of violation (%)
monitoring	cattle, pig, chicken, duck, sheep, goat, horse	134 ^a	100820	0.09
surveillance and enforcement	cattle, pig, chicken, duck, sheep, goat, horse	35 ^b + α	23000	0.26
exploratory	cattle, pig, chicken, milk, egg	27 ^c	1000	

^aAntibiotics (47), synthetic antimicrobials (57), hormones (2), and pesticides (28). ^bPenicillins (6), tetracyclines (4), sulfonamides (14), quinolones (11), and additional violative compound by former monitoring program. ^cSubstances established for MRLs of veterinary drugs in animal products or issued internationally on food safety in 2010 (dexamethasone, prednisolone, flunixin, etc).

the domestic sampling plan in 2011. The monitoring plan was conducted for 134 substances including 47 antibiotics, 57 synthetic antimicrobials, 2 hormones, and 28 pesticides for 100,820 samples from 6 major species. Surveillance and enforcement testing was conducted for 35 substances including 6 penicillins, 4 tetracyclines, 14 sulfonamides, 11 quinolones, and additional violative substances (from the former monitoring program) for 23,000 samples. Exploratory projects were conducted for 27 substances that have no MRL established in animal products or issued internationally in the previous year. The average violation rate in 2011 was 0.09% among 100,820 samples and 0.26% among 23,000 samples in the monitoring plan and surveillance and enforcement testing, respectively. The total violation rate was 0.12% from 124,820 samples tested in 2011.

Violation rates of veterinary drugs in the past decade in Korea are presented in Figure 1. The violation rate measured from the surveillance and enforcement efforts have significantly decreased over time, which demonstrates the effectiveness of the Korean NRP. The samples for surveillance and enforcement testing were collected from animals of residue-violative farms, animals for immediate slaughtering, and animals or carcasses having abscesses, remarkable injection site lesions, or high potential for residue violations. One of the main reasons for the drop in violation rate was due to education provided by provincial veterinarians to farmers. However, violation rates in the monitoring plan did not show a significant variation because of the very low violation rates in the first place. Pesticides were not found in any samples of domestic products.

Figure 2 shows violation rates of veterinary drugs in animal species in Korea. Violation rates in cattle decreased slightly and in pig did not show a significant trend. Violation rates in chickens were very low during 2002–2004, increased to 2006,

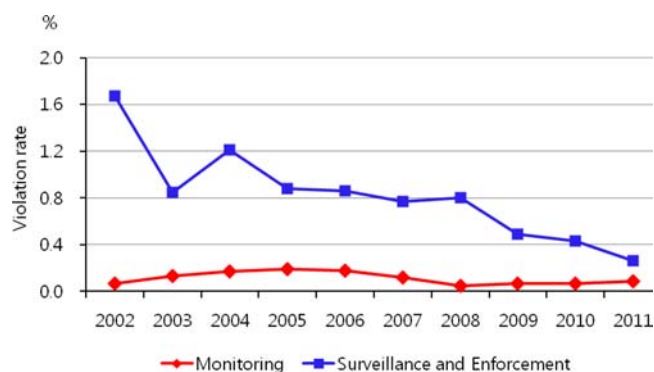


Figure 1. Violation rates for veterinary drugs from monitoring and surveillance and enforcement testing of domestic meat in Korea. No violations for pesticides and environmental contaminants such as dioxins were from 2002 to 2011.

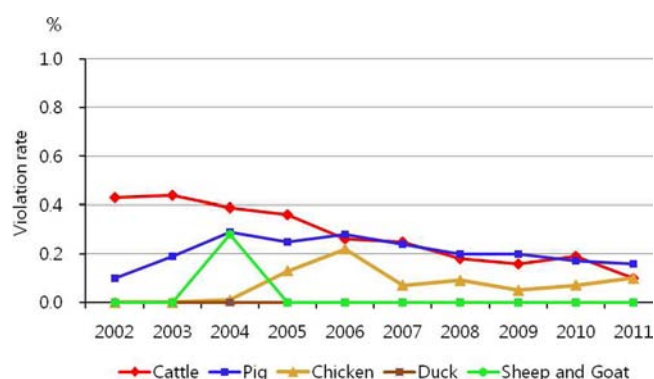


Figure 2. Violation rates of veterinary drugs in different animal species.

and then decreased, but did not reach the lower levels found in 2004. The relatively high violation rate in 2006 was probably associated with the establishment of an MRL and subsequent testing for quinolone residues in chicken. It is also relevant to point out that avian influenza (AI) outbreaks occurred in Korea from December 2003 to March 2004, from November 2006 to March 2007, from April 2008 to May 2008, and from December 2010 to May 2011. The use of animal drugs probably increased during or after the outbreaks of AI. However, the overall trend toward decreasing violative residues seems to be a result of decreased use of veterinary drugs, a reflection of increased on-site education for farmers and public instruction on the establishment of legal limits of residues in food animals. In addition, the HACCP system, which was implemented in slaughter and processing establishments, contributed to ensuring food safety.

Residue violation rates of veterinary drugs in domestic use have changed during the past decade (Figure 3). The violation rates of chlortetracycline and oxytetracycline decreased and rates for quinolone and penicillin classes increased. The maximum 46% relative violation rate of chlortetracycline in 2004 was down to 3.8% in 2011. Similarly, the maximum 38% violation rate of oxytetracycline in 2002 was down to 3.3% in 2011. Violations of tetracycline had the generally lower rate. It is likely that this decrease in violation rate is related to a regulatory change from the individual MRLs for chlortetracycline, oxytetracycline, and tetracycline to the combined MRL that occurred in 2008. Sulfamethazine was the major violation drug in sulfonamides. It was more than half of the violation rates of total sulfonamides which varied only marginally year-to-year,

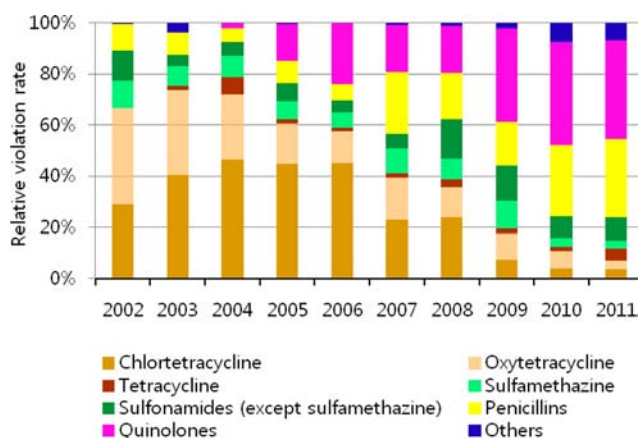


Figure 3. Relative violation rates of veterinary drugs used in domestic meat.

and there was no significant trend in the violation rates. The increasing violation rate associated with penicillin antibiotics was probably related to increased penicillin use. Improvements in analytical capabilities very likely contributed to the increased rate of quinolone antibiotic violations. For example, the use of LC-MS/MS lowered the detection limit of quinolones to 0.25–5.0 $\mu\text{g}/\text{kg}$ compared to 5.0–50 $\mu\text{g}/\text{kg}$ for HPLC, the method that was previously employed.

Residues found over the maximum legal limits were mostly veterinary drugs, especially tetracyclines, penicillins, aminoglycosides, macrolides, amphenicols, sulfonamides, quinolones, and nitrofurans. The history of residue violations from the imported products is presented in Table 3. MRL exceedances of chlortetracycline, nitrofurans metabolites (3-amino-2-oxazolidinone, AOZ, and 3-amino-5-morpholinomethyl-2-oxazolidinone, AMOZ), enrofloxacin, ciprofloxacin, doxycycline, and sulfamethazine were found in pork. AOZ, chloramphenicol, enrofloxacin, and/or ciprofloxacin were found in chicken or egg products. Clenbuterol was found in a beef product imported from China. Endosulfan was found in beef produced from cattle raised near a cotton field sprayed with endosulfan in New Zealand. As an environmental contaminant, excessive dioxin concentrations were found in pork samples imported from Chile and in beef samples from the United States. The number of violations was higher in pork because the amounts of imports were higher than those of other products. The violation rates of imported products on a yearly basis are presented in Figure 4. It is difficult to explain the variable annual violation rates because no information could be gathered from the countries involved in the violations. The countries that exported the most beef to Korea during the past decade were Australia, Canada, Mexico, New Zealand, and the United States. Pork was imported from Australia, Austria, Belgium, Canada, Chile, Denmark, France, Finland, Germany, Hungary, Mexico, The Netherlands, Poland, Spain, Sweden, and the United States. Chicken imports originated from Belgium, Brazil, Canada, China, Denmark, France, Thailand, the United Kingdom, and the United States.

Table 4 shows the levels of PCDD/Fs in domestic and imported beef, pork, and chicken for distribution in Korea from 2002 to 2011. The average concentrations in samples were 0.16, 0.05, and 0.17 pg TEQ/g fat in 761 beef samples from 6 countries, 1067 pork samples from 17 countries, and 290 chicken samples from 10 countries, respectively. The concentrations were calculated using zero for nondetects. Toxic equivalent (TEQ) values were calculated with the 1998

Table 3. Residues Found in Imported Products from Various Countries

year	residue in product	country where produced
2002	chlortetracycline in pork	USA
	sulfamethazine in pork	Belgium
2003	dioxins in pork	Chile
	nitrofurans metabolites (AOZ) in egg product	India
	sulfamethazine in pork	France
2004	chlortetracycline in pork	USA
	sulfamethazine in pork	Austria
2005	endosulfan in beef	New Zealand
	enrofloxacin in pork	Spain
	nitrofurans metabolites (AOZ/AMOZ) in pork	Mexico
2006	dioxins in beef	USA
2007	sulfamethazine in pork	Spain, USA
2008	chloramphenicol in egg product	China
	dioxins in pork	Chile
	endosulfan in beef	New Zealand
2009	clenbuterol in beef product	China
	chloramphenicol in chicken	Brazil
	enrofloxacin and sulfamethazine in pork	Spain
2010	doxycycline in pork	France
	enro/ciprofloxacin in chicken	Brazil
	enro/ciprofloxacin in pork	Spain
2011	sulfamethazine in pork	USA

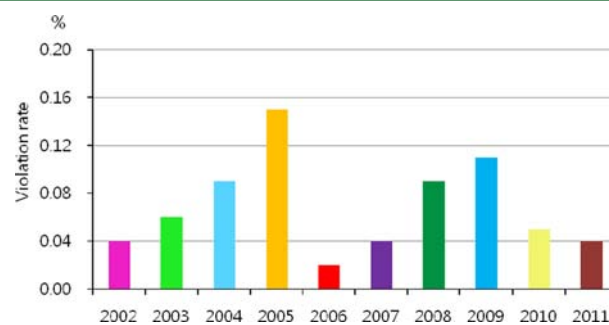


Figure 4. Total violation rates for all countries and all classes of veterinary drugs in imported meat products from 2002 to 2011.

WHO-TEF (World Health Organization toxic equivalency factor). The average concentrations showed differences between countries and types of food. For example, the measured levels of dioxins in chicken were 5.7 times higher than in the case of beef from Canada. However, the levels of dioxins in beef were 4.6 and 1.6 times higher than in chicken from Korea and the United States, respectively. The levels in pork were higher than in chicken for the samples of most countries except Korea. Samples of Chilean pork were particularly targeted because of dioxins found (3.9–8.3 pg TEQ/g fat) over the Korean MRL of 2.0 pg TEQ/g fat in 2003 and 2008. The products related to this incident were not

Table 4. Levels of PCDD/Fs in Domestic and Imported Beef, Pork, and Chicken from 2002 to 2011

country	beef (pg TEQ/g fat)	pork (pg TEQ/g fat)	chicken (pg TEQ/g fat)
Australia	0.07 (287) ^a	0.19 (9)	
Austria		0.04 (28)	
Belgium		0.04 (58)	0.24 (1)
Brazil			0.54 (33)
Canada	0.17 (7)	0.04 (56)	0.96 (2)
Chile		0.06 (438)	
China			1.26 (1)
Denmark		0.04 (32)	0.09 (19)
France		0.02 (41)	0.04 (6)
Finland		nd ^b (3)	
Germany		nd (1)	
Hungary		0.03 (28)	
Korea	0.30 (219)	0.07 (161)	0.07 (85)
Mexico	0.37 (8)	0.02 (6)	
Poland		0.09 (22)	
The Netherlands		0.01 (25)	
New Zealand	0.10 (125)		
Spain		0.01 (17)	
Sweden		nd (2)	
Thailand			0.23 (39)
UK			0.01 (2)
USA	0.16 (115)	0.04 (140)	0.10 (102)
av of total	0.16 (761)	0.05 (1067) ^c	0.17 (290)

^aThe number of samples is given in parentheses. ^bnd, not detected.

^cThe average concentration was 0.05 pg TEQ/g fat when the intended sampling of Chilean pork was excluded.

approved for import. Therefore, the concentrations of these unapproved samples were excluded from the calculation of the average for retail pork in Korea. A high concentration (6.26 pg TEQ/g fat) in beef over the MRL (4.0 pg TEQ/g fat) from the United States in 2006 was also excluded. High concentrations of dioxin in Chilean pork in 2008 were due to contamination from zinc oxide, which was collected from a metal refinery process to used premix ingredients of animal feed.¹⁴ However, the contaminating sources remain unknown for dioxins in Chilean pork in 2003 and in U.S. beef in 2006. Relatively higher dioxin levels were found in chicken from China followed by Canada, but the number of samples was limited. Figure 5

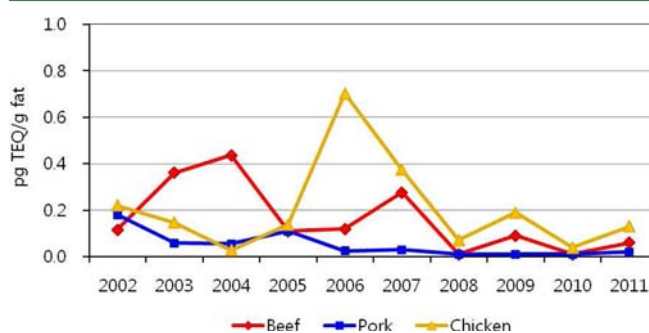


Figure 5. Trends of PCDD/Fs in beef, pork, and chicken including domestic and imported products from 2002 to 2011.

presents the yearly trends of PCDD/Fs in beef, pork, and chicken. The levels of dioxins in beef appear to be gradually decreasing overall, although the years 2003, 2004, and 2007 had

relatively higher levels than the remaining years. Pork showed a decreasing trend to <0.1 pg TEQ/g fat, except 0.113 pg TEQ/g fat in 2005. Samples of Brazilian chicken contributed relatively high levels of dioxins in 2006 and 2007. These levels ranged from 0.274 to 3.435 pg TEQ/g fat in 2006 and from not detected to 3.210 pg TEQ/g fat in 2007. Data on PCDD/Fs in chickens were collected from only two countries in 2006 and 2007; therefore, the results largely reflect the highly contaminated Brazilian samples. Studies have shown that the concentrations of dioxins in beef were relatively higher than in pork or chicken.^{15,16} This result is expected considering that the lifetime for accumulation in cattle is longer than in pig and chicken. However, the average concentration did not show those trends in this sample set. This may be because the environment in which animals are raised and their feedstuffs are different in each country. The HACCP inspection system at the farm level including feedingstuffs and processing establishments will decrease the risk of chemical residues in foods of animal origin. Nevertheless, continuous monitoring is one of the key ways to ensure food safety in a food chain.

AUTHOR INFORMATION

Corresponding Author

*Phone: +82 31 467 1982. Fax: +82 31 467 1833. E-mail: mkim@korea.kr.

Notes

The authors declare no competing financial interest.

ABBREVIATIONS USED

QIA, Animal, Plant and Fisheries Quarantine and Inspection Agency; NRP, National Residue Program; HACCP, hazard analysis critical control points; MRL, maximum residue limit; PCDD/Fs, polychlorinated dibenzo-*p*-dioxins and dibenzofurans; HPLC, high-performance liquid chromatography; POPs, persistent organic pollutants; LC-MS/MS, liquid chromatography–tandem mass spectrometry; HR-GC-MS, high-resolution gas chromatography–mass spectrometry; FAPAS, Food Analysis Performance Assessment Scheme; ISO/IEC, International Organization for Standardization/International Electrotechnical Commission; AI, avian influenza; AOZ, 3-amino-2-oxazolidinone; AMOZ, 3-amino-5-morpholinomethyl-2-oxazolidinone; WHO, World Health Organization; TEF, toxic equivalency factor; TEQ, toxic equivalent

REFERENCES

- (1) MIFAFF. National Residue Program. *Ministry for Food, Agriculture, Forestry and Fisheries Notification No. 2011-39*, 2011.
- (2) KFDA. Food Code. *Korea Food and Drug Administration*, 2011.
- (3) QIA. Method of residue analysis. *Animal Plant and Fisheries Quarantine and Inspection Agency*, 2011.
- (4) Goto, T.; Ito, Y.; Yamada, S.; Matsumoto, H.; Oka, H. High-throughput analysis of tetracycline and penicillin antibiotics in animal tissues using electrospray tandem mass spectrometry with selected reaction monitoring transition. *J. Chromatogr. A* **2005**, *1100*, 193–199.
- (5) Granelli, K.; Branzell, C. Rapid multi-residue screening of antibiotics in muscle and kidney by liquid chromatography–electrospray ionization–tandem mass spectrometry. *Anal. Chim. Acta* **2007**, *586*, 289–295.
- (6) Lim, C. M.; Kwon, H. J.; Cho, B. H.; Park, S. J.; Chung, G. S.; Son, S. W. Multi-residues determination of 13 quinolones in pork by liquid chromatography with tandem mass spectrometry. *Korea J. Publ. Health* **2010**, *34*, 331–340.
- (7) Pang, G. F.; Cao, Y. Z.; Fan, C. L.; Zhang, J. J.; Li, X. M. Multiresidue GC method for determination of synthetic pyrethroids in

agricultural products: collaborative study. *J. AOAC Int.* **1999**, *82*, 186–212.

(8) Park, J. W.; Abd El-Aty, A. M.; Lee, M. H.; Song, S. O.; Shim, J. H. Residue analysis of organophosphorus and organochlorine pesticides in fatty matrices by gas chromatography coupled with electron-capture detection. *Z. Naturforsch. C* **2006**, *61*, 341–346.

(9) Pang, G.-F.; Cao, Y.-Z.; Zhang, J.-J.; Fan, C.-L.; Liu, Y.-M.; Li, X.-M.; Jia, G.-Q.; Li, Z.-Y.; Shi, Y.-Q.; Wu, Y.-P.; Guo, T.-T. Validation study on 660 pesticide residues in animal tissues by gel permeation chromatography cleanup/gas chromatography-mass spectrometry and liquid chromatography-tandem mass spectrometry. *J. Chromatogr., A* **2006**, *1125*, 1–30.

(10) Schenck, F. R.; Podhorniak, L. V.; Hobbs, J.; Casanova, J.; Donoghue, D. Liquid chromatographic determination of *N*-methyl carbamate pesticide residues at low parts-per-billion levels in eggs. *J. AOAC Int.* **2006**, *89*, 196–200.

(11) U.S. EPA. Method 1613, Revision B. Tetra-through octachlorinated dioxins and furans by isotope dilution HRGC/HRMS. U.S. Environmental Protection Agency, Office of Water Engineering and Analysis Division, 1994.

(12) Huwe, J. K.; Larsen, G. Polychlorinated dioxins, furans, and biphenyls, and polybrominated diphenyl ethers in a U.S. neat market basket and estimates of dietary intake. *Environ. Sci. Technol.* **2005**, *39*, 5606–5611.

(13) Kim, M.; Kim, S.; Yun, S. J.; Kim, D.-G.; Chung, G.-S. Background levels and dietary intake of PCDD/Fs in domestic and imported meat in South Korea. *Chemosphere* **2007**, *69*, 479–484.

(14) Kim, M.; Kim, D.-G.; Choi, S.-W.; Guerrero, P.; Norambuena, J.; Chung, G.-S. Formation of polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) from a refinery process for zinc oxide used in feed additives: a source of dioxin contamination in Chilean pork. *Chemosphere* **2011**, *82*, 1225–1229.

(15) Tard, A.; Gallotti, S.; Leblanc, J.-C.; Volatier, J.-L. Dioxins, furans and dioxin-like PCBs: occurrence in food and dietary intake in France. *Food Addit. Contam.* **2007**, *24*, 1007–1017.

(16) Pirard, C.; Focant, J.-F.; De Pauw, E. An improved clean-up strategy for imultaneous analysis of polychlorinated dibenzo-*p*-dioxins (PCDD), polychlorinated dibenzofurans (PCDF), and polychlorinated biphenyls (PCB) in fatty food samples. *Anal. Bioanal. Chem.* **2002**, *372*, 373–381.