Trends of pesticide residues in foods imported to the United Kingdom from 2000 to 2020

Arzu Mert[ab]*, Aiming Qi[a], Aiden Bygrave[a], and Henrik Stotz[a]

[a]*School of Life and Medical Sciences, University of Hertfordshire, Hatfield, Hertfordshire, AL10 9AB, UK;*

[b]*Current address: Directorate of Plant Protection Central Research Institute, Republic of Turkey Ministry of Agriculture and Forestry, 06172, Yenimahalle, Ankara, Turkey.*

***Corresponding Author:**

E-mail: arzu.mert@tarimorman.gov.tr

Tel: (90)-312-344 59 93 / 1409

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Abstract

A total of 33,911 samples with determined pesticide residues were collated and analysed in the UK monitoring programme to determine trends in pesticide residue levels in imported foods during the period of 2000 to 2020. 17,027 of those samples (50.2%) contained detectable residues while 1,126 (3.3%) exceeded maximum residue levels (MRLs). An increased trend and a significant shift before and after 2010 in imported foods containing both detectable residues and exceeding MRLs were found. The main factors responsible for these changes were due to constant amendments in regulations and legal frameworks. With adoption of Regulation EC396/2005, there have been major changes that have affected the operations of the UK food monitoring programme including sampling methods, analysis methods, new MRLs, types of foods, and the accreditation system. The proportion of imported foods with residues and the amounts of residues in imported foods varied from country to country. Foods imported from non-European countries had more non-compliant rates than foods imported from EU. Levels of pesticide residues also varied between processed foods and unprocessed raw agricultural products and between plant-based and animal foods. Fruits and vegetables and cereals had higher occurrences of quantified residues as well as higher MRLs violation rates compared to animal products.

Keywords: food safety; pesticide residues; food monitoring programme; imported foods; the UK.

Abbreviations[1]

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[1] MRLs- maximum residue levels,
Defra- Department for Environment, Food & Rural Affairs,
FSA-Food Standards Agency,
EFSA- European Food Safety Authority,
EC- European Commission, EU- European Union,
GIS- Geographic Information System,
PRiF- The Expert Committee on Pesticide Residues in Food,
FSAI- The Food Safety Authority of Ireland, CAC- Codex Alimentarius Commission,
HSE- Health and Safety Executive,GLP- Good Laboratory Practice,
UKAS- United Kingdom Accreditation Service, RASFF- Rapid Alert System for Food and Feed,
WPPR- Working Party on Pesticide Residues and PRC- Pesticides Residues Committee,
AFBI- AgriFood and Biosciences Institute,
SASA-Science and Advice for Scottish Agriculture,
FERA-Food and Environment Research Agency.

1. Introduction

Protecting agricultural productivity is vital in meeting the rising food needs of the rapidly growing world population because worldwide losses of crop yields from pests and diseases can reach approximately 45% (Kolani et al., 2016). Pesticides are extensively used in agriculture to maintain crops yield and quality, and thus they constitute one of the main agricultural inputs (Bajwa & Sandhu, 2014). According to the Pesticide Manual 17th edition of the British Crop Production Council, more than 1,600 active substances have been developed and used as pesticides (Fussell, 2016). Many of them are restricted or banned due to adverse effects of pesticides on the environment, wildlife and human health.

Harmful pesticide residues in foods pose risks to human health (Blankson et al., 2016; Cieslik et al., 2011; López-Blanco et al., 2016). The presence of pesticide residues in foods even at low levels can cause many health problems (Fothergill & Abdelghani, 2013; Kiwango et al., 2018). Therefore, pesticide residues in foods are regulated by international organizations and national governments (Seo et al., 2013). Many countries have established their own monitoring programmes to measure pesticide residues in foods to control the allowable residue levels, also known as maximum residue levels (MRLs) (Seo et al., 2013). In the UK, monitoring of pesticide residues in both imported and domestic foods has been carried out since 1977 and results are regularly published in official reports (Reynolds, 1998).

Imported foods, accounted for approximately 50 percent of food consumed in the UK, increasingly constituting a large proportion of the UK food needs (Defra, 2017). Imported foods have been controlled and subjected to strict inspection and checks at entry points (FSA, 2013). However, the UK pesticide residue monitoring results have reported that imported foods, including from EU countries, have frequently exceed the MRLs (FSA, 2006). Food samples from abroad also more frequently contained detectable residues than those from the UK (FSA, 2006). Similarly, annual reports of the EU monitoring programme have indicated that MRLs of imported foods from outside the EU were often higher than foods from EU countries (EFSA, 2018a). Poulsen et al. (2017) analysed the results of the Danish monitoring programme between 2004 and 2011 and found that imported foods were higher than domestic foods in terms of the frequencies of samples with detectable residues and MRLs.

Countries exporting foods to the UK must comply with EU MRLs, but both UK and EU monitoring programmes have revealed that EU MRLs are often violated in imported foods. These findings have also increasingly concerned regulators, researchers and consumers (Galt, 2009).The objectives of this study were to assess pesticide residues in imported foods consumed in the UK and to investigate the trends of pesticide residues of imported foods from 2000 to 2020.In addition to determining changes in the frequencies of pesticide residues in imported foods, another objective was to determine differences between exporting countries by visualising their residue levels using Geographic Information System (GIS). The GIS technique was used for spatial assessment of the exporting countries by producing global patterns of pesticide residue levels in imported foods. The global and spatial patterns created in this study facilitated the understanding and interpretation of differences in pesticide residue levels of imported foods among exporting countries.

2. Material and methods

2.1.Data collection and extraction

The UK National Reference Laboratories, Food and Environment Research Agency (Fera Science), AgriFood and Biosciences Institute (AFBI) and Science and Advice for Scottish Agriculture (SASA), have conducted the analysis of pesticide residues for the UK monitoring programme (Fera, 2021; PRiF, 2016). The official laboratories are also accredited to the ISO/IEC 17025 standard, and so the analysis method applied must meet requirements of the current and valid laboratory criteria (EC, 2017; Fera, 2021). Collected samples have been analysed by the national laboratories mostly using multi-residue methods based on LC-MS and/or GC-MS instrumentation. However, polar analytes and other pesticides unsuitable for multi-residue methods have been also analysed using single residue methods that is more selective.

Every year, the results of the UK monitoring programme are published as quarterly and annual reports. Accessible and downloadable reports of the monitoring programme results are available on National Archives website (data.gov.uk). The data used in this study were collected from existing reports of UK monitoring programme results. The data of the 21years between 2000 and 2020 were collated and analysed after data collection was completed and organized.

According to structure of the UK monitoring programme, a year is divided into four periods of three months (i.e. per quarter of a year) and samples of foods are collected and analysed for each period. Each year, the results of each period are released in four quarterly reports and the results are published in a series of reports (e.g. 2017 Report on Quarter 1, 2017 Report on Quarter 2, 2017 Report on Quarter 3, 2017 Report on Quarter 4, and 2017 Annual Report). Quarterly reports were used to obtain data for this study, as they contained more detailed data. The databases provided by quarterly reports include records of residue analysis results for each type of foods imported from each country. The number of samples analysed for pesticide residues, the number of samples with detectable residues and the number of samples with residues above MRLs were provided for each record. The databases of quarterly reports also contained the countries of origin of samples analysed for each record.

Eighty-four quarterly reports between 2000 and 2020 were examined to extract the necessary data. The most significant issue regarding data extraction was the determination of imported foods and countries from where these foods were imported. Therefore, imported foods were separated from foods of UK origin and this process was carried out for each year from 2000 to 2020. Countries that exported their foods to UK were also determined. Then, the data of these foods and their origins of countries for all years were compiled and these databases were used to produce the annual data in Tables Supplemental S1 to S4.

2.2.Statistical analysis

The data was statistically analysed to determine whether the change in pesticide residues followed a changing trend over the years. First, proportion of samples with detectable pesticide residues was calculated. Then, the proportion of samples with pesticide residues above MRLs was calculated. Linear regression analysis was performed to test and determine whether there was a significant temporal trend for all annual samples analysed. The proportion of samples with detectable residues and proportion of samples above MRLs were also analysed. To detect whether there was a significant shift in proportion of samples with detectable residues and proportion of samples above MRLs before and after 2010, sums of annual samples, sums of samples with detectable residues and samples with residues above MRLs from 2000 to 2010 were determined and then analysed against those counterparts from 2011 to 2020 assuming a binomial distribution for proportion of samples with detectable residues and proportion of samples with residues above MRLs. All the above calculation and analyses were performed in Microsoft Excel.

2.3.Data processing for GIS maps

The study was also aimed to create global and spatial maps that allowed the visualisation of pesticide residue data from foods imported to the UK. GIS maps, which were created using the data on foods imported from each country, provided spatial patterns in detectable pesticide residues and MRLs in foods of these countries. To prepare the spatial data for map presentation, the total number of samples of each country was calculated from all imported samples analysed during 2000-2020. Samples with detectable residues of each country and samples exceeding MRLs of each country were each calculated as percentages. The data used in GIS maps were processed using Microsoft Excel. QGIS software was used to create maps for visualization of pesticide residue data.

3. Results and Discussion

3.1.Trends of pesticide residues

A total of 33,911 analysed samples from imported foods were reported in the UK pesticide residue-monitoring programme during the 21years from 2000 to 2020 to check for pesticide residues (Supplemental Table S1). Out of the total 33,911 analysed samples, 17,027 of those samples (50.2%) contained detectable residues and 1,126 of total analysed samples (3.3%) were above MRLs.

Figure 1 shows the number of imported food samples analysed annually for pesticide residues by the UK monitoring programme between 2000 and 2020. The number of samples analysed in 2000 was smaller than half of the annual total food samples in the following years. The numbers of annually analysed samples have stabilised after 2001. However, the number of total samples analysed in 2020 showed a sharp decline. This could be caused by the disruptions in activities both in imports and in staff restrictions during lockdown due to COVID-19 pandemic. There was no statistically significant change in annual samples analysed after 2000 ($p > 0.31$, Figure 1). The reason for this was the introduction of major changes and innovations in sampling methods since 2001, and the legalization of these changes and innovations with Directive 2002/63/EC adopted in 2002 (EC, 2017; EFSA, 2013). In fact, the biggest change in the sampling methods was made by this directive that introduced the representative number of samples, which took account of the population size for the number of products analysed in each country (FSAI, 2009). Before this directive, the UK monitoring programme implemented sampling methods according to Codex Alimentarius Commission (CAC) guidelines set in 1993 (EC, 1999).

Figure 2 and 3 show changes in the proportion of imported samples containing detectable residues and samples with residues above MRLs from 2000 to 2020. It was found that there were apparent increases in the annual proportion of samples with detectable residues and exceeding MRLs and the general trends were statistically significant ($p < 0.01$ in both cases). Of note, MRLs did not change much after 2010, suggesting that a change in legislation brought about a stabilisation of contaminated food imports. Annual EU reports stated that there was an increased trend in the annual number of imported and domestic foods with measurable pesticide residues and MRLs exceedances (EFSA, 2018a). Similarly, Nordic Projects performed by Hjorth et al. (2011) and Skretteberg et al. (2015) reported the increased trends in pesticide residues in imported foods.

To detect whether there was a significant shift in proportion of samples with detectable residues and proportion of samples with residues above MRLs before and after 2010, total annual samples with detectable residues and samples with residues above MRLs from 2000 to 2010 were analysed against those counterparts from 2011 to 2019 (i.e. the abnormal data point in 2020 was excluded) assuming a binomial distribution. Binomial distribution analysis was assessed approximately using the rejection region (z value of 1.96 at p=0.05) in a normal distribution because of many samples used. As a result, there was a significant shift in the proportion of total detectable samples before and after 2010 (z=27.2491) in samples with detectable residues. The calculated proportion of samples with detectable residues was 45.0% before 2010 while the calculated proportion of samples with detectable residues was 56.7% after 2010. The proportion of samples exceeding MRLs has also significantly changed before and after 2010 (z=10.7692) in samples with residues above MRLs. The calculated proportion of samples with residues above MRLs was 2.5% before 2010 while the calculated proportion of samples with residues above MRLs was 4.3% after 2010. Importantly, however, no significant increases in detectable pesticide residues and MRLs were observed after 2010.

It is important to point out that the changes in pesticide residues can be affected by several factors such as legal frameworks, structure of the monitoring programme and introduction of new pesticide use patterns (EFSA, 2013). However, it can be argued that the main reasons that affected the changes are regulations that have been substantially amended several times.

Until Regulation EC 396/2005 adopted in 2005 and fully harmonized in September 2008 throughout the EU, the UK like all other EU Member States, had its own pesticide regulations and national MRLs (FSAI, 2009). With the introduction of this pesticide residue regulation, substantial changes and developments occurred in pesticide residue regulations, directives and decisions (Fussell, 2006). This new legislative framework consolidated and replaced all existing pesticide residues directives, such as 90/642/EEC (plant products), 86/363/EEC (foodstuffs of animal origin), 86/362/EEC (cereals), and 76/895/EEC (fruits and vegetables) (Fussell, 2006; FSAI, 2009). This new regulation also replaced all old national MRLs and provided for full harmonisation of MRLs in the EU (Delcour et al., 2015; EFSA, 2013). Additionally, with the introduction of EC 396/2005, UK pesticide regulations, The Pesticides (Maximum Residue Levels in Crops, Food and Feeding Stuffs) Regulations 1997, which was amended in 1999 and 2001, was renewed as The Pesticides (Maximum Residue Levels) (England and Wales) Regulations 2008 (legislation.gov.uk, 2008). These changes in both EU and UK pesticide residue regulations has led to major structural changes and improvements in the UK monitoring programme.

Due to Regulation EC 396/2005, there have been significant innovations and changes in the design of monitoring programmes, such as method of sampling, method of analysis, pesticides sought, and the MRLs (HSE, 2015). Article 27 of EC 396/2005 set out the sampling method of monitoring programmes, such as product type and number of samples analysed (EC, 2005). According to sampling methods set by this regulation, sufficient number of domestic and imported products should be analysed every year, and the number of products analysed in each country is expected to reflect population size, and thus the number of samples analysed each year is expected to be updated (EC, 2005). Product type should also be determined according to dietary consumption patterns of each country provided that most of them are fresh agricultural products (FSAI, 2009). Allocation of the majority of analysed product types into fresh fruits and vegetables may directly affect changes in numbers and proportions of foods containing pesticide residues because fresh produce contains higher frequencies and quantities of pesticide residues than processed, animal or cereal food samples (EFSA, 2018a; FSA, 2006). Galt (2010) also found linear relationships between volumes of analysed samples and MRLs violation increases.

Article 28 in Regulation EC 396/2005 sets out analysis methods for pesticide residues in monitoring programmes, and the analysis method applied must meet requirements of the current and valid laboratory criteria (EC, 2005). Directive 882/2004/EC adopted in EU to provide the current and valid laboratory criteria with the ISO/IEC 17025:2005 accreditation that imposes all official control laboratories used in monitoring programmes (EC, 2017). Guidelines, which determine laboratory criteria, are constantly changed and updated with technological advances (EFSA, 2013). Until the accreditation of ISO/IEC 17025, the official control laboratories used in the UK monitoring programme were accredited with Good Laboratory Practice (GLP) or United

Kingdom Accreditation Service (UKAS) (EC, 1999). The ISO/IEC 17025 accreditation incorporates analysis methods and methodology that are constantly improved and renewed through technological developments as well as keeping the analysis methods up-to-date, which increases the quality and accuracy of the results in the control laboratories (Fussell, 2016). This means that the latest analytical methods allowed measurement of residues at low levels and ensured that the residue results were more accurate and reliable (PRiF, 2016).

Official laboratories accredited with ISO/IEC 17025 started to be established in 2006 and most of EU Member States, including the UK and accredited their control laboratories as of 2010 (EFSA, 2013, Fussell, 2016). We found significant increases in the proportions of samples with detected pesticide residues ($z = 27.2491$) and samples exceeding MRLs ($z = 10.7692$) after 2010. This substantial change may be attributed to new analytical methods and improvements provided ISO/IEC 17025 accreditation to official control laboratories (EFSA, 2013; Fussell, 2016). Furthermore, the annual number of samples with pesticide residues and MRL violations increased over time. PRiF (2016) has reported that UK monitoring programme uses state-of-the-art methods for residue analysis, which means that it was expected to see an increase in the number of active substances detected and samples containing pesticide residues.

Up to EU-harmonized MRLs, pesticide residues were evaluated in the UK monitoring programme according to UK, EU and Codex MRLs (HSE, 2015). Since the fully harmonized EU MRLs were provided by EC 396/2005 in September 2008, it can be considered that a major factor affecting the results of the monitoring programme was the MRLs which were amended by this regulation. The EU MRLs for more than 500 active substances and about 370 foods are defined and the default MRLs (0.01 mg/kg) was used for the unspecified active substances in this regulation (EFSA, 2018a). Regulation EC 396/2005 not only modified the MRLs, but also led to an increase in the number of MRL-defined active substances, which result in an increase in the number of pesticides sought in monitoring programmes (Fussell, 2006). With this regulation, new types of foods, such as cocoa, coffee and sugar beet, were also included, and thus there was an increase in the number of products exceeding MRLs, which meant that greater numbers and types of products were analysed in monitoring programmes (Fussell, 2006). In short, more numbers of foods containing pesticide residues were determined in the monitoring programmes due to expanded analytical scopes, greater numbers of foods analysed, and more active substances sought (EFSA, 2013).

MRL values are the levels determined for safe exposure of people to pesticide residues in foods (Winter & Jara, 2015). Under Regulation EC 396/2005, proposals are made on a regular basis to improve, change or delete MRLs (EC, 2005). The Food Standards Agency (FSA) established in 2000 and European FSA (EFSA) established in 2002 are considered two important agents for proposals made for potential changes to MRLs (Fussell, 2006). FSA principally focuses on consumer protection and thus notices Rapid Alert System for Food and Feed (RASFF) for risky foods analysed in monitoring programmes (PRiF, 2016). EFSA has performed risk assessment and communication, but it began to perform the risk assessment needed for all MRLs with the adoption of Regulation EC 396/2005 in 2008 (Fussell, 2006). The proposals for MRLs arrangements are based on EFSA opinions under Regulation EC 396/2005. In short, the establishment of FSA and EFSA and their activities on MRLs can consider two important political issues because setting, modifying or deleting of MRLs directly affected the results of pesticide residue analysis.

Studies on toxicological reference values for pesticide residues are continuously carried out and thus some MRLs values are lowered (EFSA, 2018a). Lowered MRLs directly affected the results of pesticide residue analysis and may cause an increase in the number of samples with residues. Furthermore, MRLs values have been changed and regulated for many reasons. To date, changes and amendments in commodities and pesticides for which MRLs are set have been made with many MRLs and pesticide residues regulations. Although new laws and regulations on MRLs have been introduced from time to time, the biggest change may be from Regulation EC396/2005 because a large number of new active substances and products have been introduced in which the MRLs have been set for the first time (EC, n.d.; HSE, 2012). Since 2009 and 2010, this regulation and its new schemes have been implemented by all EU Member States, including the UK (EFSA, 2013). The UK has included these new products and active substances in its monitoring programme and can be considered one of the major reasons for the significant change after 2010.

Under Regulation EC396/2005, each member state is obliged to update its national monitoring programmes in accordance with the existing regulations and to fulfil their requirements (EC, 2005). Therefore, the UK monitoring programme had then been subjected to an indirect but effective change with amended and renewed regulations and directives mentioned above. However, it can be claimed that it was influenced by the changes in its own managerial structure. The HSE is responsible for both monitoring of pesticide residues and implementation of EU MRLs in the UK (HSE, 2015). PRiF has carried out the UK monitoring programme on behalf of the HSE (PRiF, 2016). However, official bodies responsible for conducting the UK monitoring programme before PRiF were Working Party on Pesticide Residues (WPPR) and Pesticides Residues Committee (PRC) (Defra, 2018). Since each management period has been designed to expand with regard to number of samples and product types analysed and active substances sought and to ensure more independent and reliable works, changes in the official bodies may affect the structure of monitoring programme and thus resulting in changes in pesticide residues (PRC, 2000; PRiF, 2016).

Finally, it should be pointed out that the results of the 21-year pesticide residues were influenced by the legal regulations and directives set for product groups, such as baby foods. Since 2009, all member states have also been required to analyse at least 10 products from infant formula and foods (EFSA, 2013). As infants and young children are the most affected groups of pesticide residues, more restrictive regulations are being introduced with ongoing legal arrangements (EFSA, 2018a). The compulsory inclusion of baby foods in monitoring programmes may lead to changes in the trends of food samples analysed and residue levels.

3.2.Differences between exporting countries

The distribution of imported foods by country and by country groups separated as European and non-European countries is shown is Supplemental Table S2. The total of 33,911 food samples was distributed to125 different countries during the period 2000-2020. 16,440 samples imported from non-European countries had more non-compliant rates (detectable rate 53.7%; MRL exceeding rate 5.6%) than foods (17,471 samples) imported from European countries (detectable rate47.0% and MRLs exceeding rate 1.2%).

From Supplemental Table S2, five points can be highlighted. First, the majority of food samples analysed came from Spain (20.2%). The other important countries with the highest number of analysed samples were South Africa (7.5%), the Netherlands (7.0%), Italy (6.8%), France (5.0%), India (3.6%), the United States (3.4%), Belgium (3.2%), Israel (2.8%), Chile (2.8%), New Zealand (2.6%), Brazil (2.5%), Egypt (2.5%) and Kenya (2.1%). Samples from these countries with detectable residues included Chile (77%), South Africa (74.8%), Brazil (68.3%), Belgium (57.6%), Spain (56.2%), India (55.1%), Egypt (51.3%), the United States (46.1%), the Netherlands (46.0%), Kenya (45.7%), and France (43.6%). Moreover, some of these countries with the highest rates of violated MRLs were India (18.1%), Kenya (11.4%), Brazil (7.8%), Egypt 5.1%), Chile (3.2%), the United States (2.0%), Israel (1.6%), Spain (1.4%), Italy (1.3%), France (1.1%) and South Africa (1.1%).

Second, despite a small number of analysed samples, the following countries exceeded MRLs at very high percentages: Ukraine (MRLs violation was 50% in 6 samples), Romania (MRLs violation was 20% in 10 samples), Cambodia (MRLs violation was 12.5% in 8 samples), Iran (MRLs violation was 25% in 8 samples), Lebanon (MRLs violation was 20% in 5 samples), Saudi Arabia (MRLs violation was 50% in 2 samples) and Tunisia (MRLs violation was 11.1% in 9samples).

Third, MRL violations for samples imported from EU countries were remarkable because high residue rates were determined in food samples imported from certain EU countries. For example, Spain had a large number of samples with detectable residues (56.2%) and above MRLs (1.4%) especially in fruits and vegetables. In samples imported from Cyprus, rate of detectable residues was 66.0% and MRLs violation rate was 9.4%. France had samples containing measurable residues at 43.6% and breaching MRLs at 1.1%. Samples from Poland were also found to have serious residue rates (detectable 36.8%; MRL violation 4.8%). Another example of high residue levels was Italian samples with 37.3% for detectable residues and 1.3% for exceeding MRLs. Samples from Greece, the Netherlands, Germany, Ireland and Bulgaria had detectable residue rates at 52.8%, 46.0%, 19.1%,17.4% and 21.7%, and violating MRLs rates at 0.9%, 0.7%, 1.3%,0.8% and 4.3% respectively.

Fourth, high residue rates in foods imported from developed countries were of great concerns. There is a common belief that food commodities of developed countries are not alarming in terms of pesticide residues (Galt, 2010) but this study found contradictory results. For example, detectable residues (36.3%) and MRLs violation (5.3%) were determined in 171 Canadian samples analysed. The United States had samples with detectable residues (46.1%) and with residues above MRLs (2.0%), as well. Foods imported from Norway did not violate MRLs but residues were detected in 46.1% of the 152 samples. On the other hand, none of the samples analysed from the products of Finland, Iceland and Sweden had MRL violations.

Fifth, the highest non-compliance rates were determined in foods imported from outside European countries. Samples from Bangladesh had the most samples with residues above MRLs (50.0%) and with detectable residues (64.6%). Samples from Cameroon had residues in 29 of 32 samples (90.6%) and samples from Uruguay had residues in 48 of 56 samples (85.7%), but no MRL violations were found in any of their samples. Seven countries with the highest level of MRLs violations were Malaysia (38.8%), Jordan (20.8%), Jamaica (18.6%), Pakistan (12.2%), Ghana (11.4%), Thailand (11.5%) and Colombia (10.0%). Moreover, food samples from Dominican Republic (8.7%), Venezuela (9.1%), Serbia (3.7%), Uganda (4.3%), Puerto Rico (4.8%), Costa Rica (3.2%), and Turkey (3.6%) had residues above MRLs. Windward Islands (detectable 75.8%; MRLs exceedance 6.1%) and Namibia (detectable 75.0%; MRLs exceedance 4.2%) had also other highest non-compliant rates. However, 41 samples from Indonesia were analysed and no residue was found in all samples.

The results mentioned above were comparable with EU-coordinated control programme (EUCP), which was conducted with the same scheme across Europe, and can be concluded to be within the range of those found for residue results here within. EFSA (2013) reported in the 2010 EUCP that foods imported from outside European countries (7.9%) had a higher average rate of MRLs violations than foods from EU countries (1.5%). Similarly, according to 2016 EUCP results, foods of European origin were found to have less measurable residue rates (43.9%) and less MRL violations (2.4%), compared to foods from outside European countries (measurable 52.1%; MRL violation 7.2%) (EFSA, 2018a). In this study, average MRL exceedance rate was found to be relatively lower but closer to 5.6% for samples from outside European countries and 1.2% for samples from EU countries.

Among the European countries, the highest rate of MRLs violations were found for samples from Slovenia, Portugal, Cyprus and Slovakia in the 2010 EUCP (more than 3%) (EFSA, 2013) as well as for samples from Poland, France, Norway, Cyprus and Iceland in the 2016 EUCP (above 4%) (EFSA, 2018a). In this study, the highest MRLs violation rates for EEA countries were found in samples from Cyprus (9.4%), Poland (4.8%), Spain (1.4%), Italy (1.3%) and France (1.1). Among the outside European countries, the highest rate of MRL violations were found for samples from Mauritius (20.0%), Thailand (20.9%), Iran (21.4%), Jordan (21.7%), Burundi (22.2%), Uganda (23.6%), India (28.3%), Bolivia (33.3%), Bangladesh (44.4%), Cambodia (50.0%) in 2010 EUCP (EFSA, 2013). Similar results were also found for outside European countries in 2016 EUCP and accordingly, samples from India, Brazil, Kenya, Tunisia, Dominican Republic, Egypt, Colombia and Israel had the highest MRLs breaches rates (7.2%) and other countries with the high non-compliance rates were Suriname, Cambodia, Pakistan, Thailand, Sri Lanka, Uganda, China, Vietnam and Laos (EFSA, 2018a).

In addition to the EUCP results, two Nordic project studies can be compared with the results of this study since the monitoring programmes were carried out under the same law in all European countries. Hjorth et al. (2011) examined the pesticide residues in fruits and vegetables exported from South American countries including Uruguay, Suriname, Peru, Ecuador, Columbia, Chile, Brazil and Argentina to Nordic countries such as Sweden, Finland, Estonia, Denmark and Norway and concluded that 72% of the samples analysed had detectable residues and more than 8% of them exceeded MRLs. Hjorth et al. (2011) have also found that foods from Brazil (13%), Uruguay and Columbia (both 10%) were the samples with the highest MRLs violations. Similarly, as in another Nordic project, Skretteberg et al. (2015) analysed the Southeast Asian fruits and vegetables imported to Sweden, Norway, Finland, Denmark to examine the adherence to EU MRLs and legislation but reported the requirement for effective and continuous monitoring of foods from Southeast Asia due to high rates of samples containing detectable residues (28%) and breaching EU MRLs (12%). Skretteberg et al. (2015) have also concluded that Vietnam (33%), Malaysia (11%) and Thailand (9%) had the highest samples exceeding MRLs.

We found similar results with both EUCP and Nordic Project results for these same outside European countries. Bangladesh (50.0%), Malaysia (38.8%), Burma (7.7%), Cambodia 12.5%), Colombia (10.0%), Ghana (11.4%), India (18.1%), Jamaica (18.6%), Jordan (20.8%), Kenya (11.4%), Pakistan (12.2%) and Thailand (11.5%) had the highest MRLs exceeding samples. The highest detectable residue rates were found from Belize (100%), Bolivia (95%), Cameroon (90.6%), Uruguay (85.7%), Bangladesh (64.6%), Brazil (68.3%), Chile (77%), Costa Rica (76.4%), Dominican Republic (54.7%), Guatemala (62.5%), India (55.1%), Namibia (75.0%), South Africa (74.8%) and Windward Islands (75.8%). About half of the food samples had detectable residues in Argentina (53.7%), Colombia (58.9%), Egypt (51.3%), Honduras (43.7%), Jordan (50.2%), Kenya (45.7%), Malaysia (45.9%), Morocco (59.6%), Turkey (55.6%) and the United States (46.1%).

All of these results revealed the high incidences of pesticide residues in imported foods consumed in the UK. Regulation 669/2009 was thus adopted for certain countries outside European to perform import controls for specific pesticide residues in their specific foods (EFSA, 2018a). Although this law has been implemented and regularly modified since 2010 (Fussell, 2016), this study demonstrated that the high levels of pesticide residues in foods imported from non-European countries still continue to exist.

3.3.Problematic pesticides

Over the 21 years from 2000 to 2020, there were in total 141 different active substances that were quantified with above MRLs (Supplemental Table S3), the most occurrence of active substance in violation of MRLs was dimethoate followed by dithiocarbamates and carbendazim, 98 different products were analysed with active substance(s) above MRLs (Supplemental Table S4), the product with the most frequent violation of MRLs was beans followed by okra and grapes, and 64 countries that exported foods with active substance(s) above MRLs.

There were not any trends in the kinds of active substances that were above MRLs through time. However, the products with above MRLs were mostly grocery vegetables and fresh fruits (Supplemental Table S4). In particular, beans, okra, grapes and yam had the highest violating MRLs rates at 26.6%, 13.7%, 6.9%, 5.2%, respectively. Beans were mainly imported from Kenya, Thailand, Egypt, Spain, Cyprus, Bangladesh, China, India, Dominican Republic and Malaysia. Okra was mostly imported from India, Jordan, Colombia, Dominican Republic, Spain and Cyprus. Grapes were mainly imported from Chile, Greece, India, Egypt, Spain, Turkey, USA and Brazil. Yam was mostly imported from Brazil, Ghana, Jamaica, South Africa, China and Jordan. EFSA (2021) reported in the 2019 EUCP that fresh vegetables and fruits (beans, peppers, teas, vine leaves, herbs, celery leaves, water cresses, dragon fruit, prickly pears, passion fruits, pomegranates) had the highest MRL exceedance rates.

Although it was the product group with the lowest violation rate (0.7% in Table 1), pesticides which were banned years ago such as organochlorine pesticides were detected in animal foods. It is not surprising that residues of these pesticides were found in animal products, as DDT and its metabolites have a high potential to bioaccumulate in foods high in fat and have a long-term persistence in the environment, resulting in exposure of animal foods. For example, low levels of DDT were detected in oily fish (samples imported from Ireland, Norway, Peru, and Portugal), salmon (imported from USA, Ireland and France), lamb's liver, lamb’s mince, venison and butter (imported from New Zealand). Moreover, DDT and other organochlorine pesticides were detected in sea fish farming at the Pacific and South West Atlantic. All these findings prove that these pesticides still exist in aquatic and terrestrial ecosystems and continue to exist in the food chain. However, it has been emphasized in the annual reports of UK monitoring programme that these banned pesticides were at low levels and did not pose any risk to human health. It has been found the similar results with the EUCP that banned pesticides, DDT, were detected in fish and in animal foods (EFSA, 2021).

Dimethoate, dithiocarbamates, carbendazim, metomyl, and chlorpyrifos in imported foods from 2000 to 2020 were the most common types of violating active substances, with the highest MRLs exceedance rates of 8.24%, 7.66%, 6.44%, 3.35% and 3.15%, respectively. Poulsen et al. (2017) stated in their research that active substances that violate MRLs most frequently were dithiocarbamates and carbendazim. Other pesticides with high MRLs exceedance rates were omethoat (2.77%), acephate (2.7%), profenofos (2.7%), and BAC (2.64%).

3.4.GIS maps

To visualise differences between countries which exported foods to UK from 2000 to 2020, analysed and processed data were integrated into QGIS software and three different maps were created. Figure 4 shows the distribution of total number of samples analysed for pesticide residues in imported foods for each country. Figure 5 shows the distribution of analysed samples for each country according to the percentages of food samples with detectable residues. Figure 6 shows the distribution of analysed samples for each country according to the percentages of food samples with residues above MRLs.

These maps showed that the proportion of foods with residues greatly varied with country. Although the fluctuation might result from several factors mentioned above, such as sample numbers and types of foods analysed, sampling and analysis methods used, and even analytical instruments development by technological improvements, imported foods could be a major factor responsible for the variation from country to country (FSA, 2006).

Pesticides, pesticide regulations or authorizations and pest problems differed in each exporting country because of different environmental conditions and types of products (Galt, 2009). Environmental conditions, such as warm and humid climates, provide more favourable environments for different weeds, diseases and pests (Fussell, 2016). Countries with conducive environmental conditions tend to apply more pesticides to protect their products from intense diseases and pests, and thus pesticides and regulatory regimes vary appreciably from country to country (Hjorth et al., 2011). Additionally, chemicals show different degradation in different foods and environmental conditions (Galt, 2010). Levels of pesticide residues are influenced by the intensive use of pesticides, applying different active substances and degradation rates of pesticides, resulting in residue differences between countries (Vannoort, 2001).

Product types, such as processed and unprocessed foods, cause significant differences in residue levels because pesticide residues in foods are reduced and removed during commercial processing of foods, including preparation, transport and storage and post-harvest treatments (FSAI, 2009). Several studies (Al-Taher et al., 2013; Bajwa & Sandhu, 2014; HSE, 2015) have reported that foods are exposed to many stages from raw material to final product and this leads to a reduction in levels of pesticide residues. Preparation stages (e.g. trimming and peeling), which directly move away pesticide residues in shell and surface of foods; thermal processing stages (e.g. scrambling, canning, steaming, cooking, boiling, blanching and pasteurization), which are effective in degradation of chemicals and in lowering of residue levels; and other stages (e.g. brewing, malting, curing, wine making, baking, milling, fermentation and refining), which result in reduction of residue levels in final products. However, later processing stages like concentration and dehydration or drying, lead to an increase in concentration of residues and hence increase in residue levels (HSE, 2015).

For these reasons, levels of pesticide residues vary between processed foods and unprocessed raw agricultural products. EFSA (2018a) reported that in 2016 EUCP, unprocessed foods had more detectable residues (47.9%) than processed products (33.4%) and had more MRL violations (3.9%) than processed foods (2.8%). It was also calculated in 2006 EUCP that, about 3% of fresh produce had samples with residues above MRLs while MRLs exceedances was not found in processed foods (FSAI, 2009). Moreover, there are also residue differences between food of origin (e.g. cereals, fruits and vegetables) and animal foods. EFSA (2018b) reported by 2013, 2014 and 2015 EUCP that plant-based foods had a higher rate of samples with measurable residues and above MRLs than animal foods. Our results were consistent with these studies that fruits and vegetables and cereals had a higher occurrence of quantified residues (59.8% and 34.2%) as well as higher MRLs violation rate (4.0% and 2.1%), compared to animal products (14.0% and 0.7%) (Table 1). Given that countries exported different foodstuffs, it was expected that there were differences between the residue levels of exporting countries.

There are different MRLs being set throughout world and countries regulate pesticide residues according to their national MRLs (e.g. EPA tolerances used in the USA; EU MRLs used in EU countries; Canadian MRLs) or according to Codex MRLs employed in many countries, which result in creation of dissimilar pesticide / commodity MRLs combinations. Thus, MRLs differences in imported produce will affect residue assessment results (Winter & Jara, 2015).

Most developing countries may be less aware of the importance of pesticide residues in foods and do not have monitoring programmes or may not effectively administer them due to deficiency of the strict enforcement of regulations and enough infrastructure (Hjorth et al., 2011; Kolani et al., 2016; Mutengwe et al., 2016). Since the Circle of Poison, published by Weir and Schapiro in 1981, specified that developed countries are at risk with pesticide residues prohibited or restricted on imported foods, imported foods have been of great concern (Galt, 2009). Similarly, Ecobichon (2001) has asserted that developing countries intensively use inexpensive, more toxic and non-authorized pesticides without the concern of their pesticide residues in local foods and even in foods they export. While farmers and growers of developing countries use chemicals that are relatively more effective and cheaper in combating pests due to economic concerns, biological control and integrated pest management are widely applied in many developed countries (Ferro et al., 2015; Fussell, 2016; Poulsen et al, 2017). These methods lead to less use of pesticides and thus reducedlevels of residues, but many developing countries are even lack of GoodAgricultural Practice (GAP) (Ecobichon, 2001). Samples from developing countries which applied pesticides “that are approved or no longer approved in the EU on crops for which no import tolerances have been requested by the importers” (EFSA, 2018a, p.80). For these reasons, given the increasing trade in food commodities, governments have set and imposed obligatory regulations and legislation to reduce consumer concerns for imported foods as well as particularly monitored imported foods through their pesticide residue monitoring programmes (Wilson & Otsuki, 2004).

4. Conclusions

33,911 sample residues records from UK monitoring programme in 2000-2020 were analysed to reveal changes in the frequency of detectable pesticide residues and breaching MLRs in foods imported into the UK. Imported foods with both detectable pesticide residues and MRLs breaches increased through time and this increased trend was statistically significant. Several factors, such as structure of the monitoring programme, introduction of new pesticide patterns and changes in MRLs and MRL-defined foods, were jointly responsible for this trend. However, among the main factors were likely to be regulations and legal frameworks that have been constantly amended and changed. The proportion of imported foods with residues or the amounts of residues in imported foods were found to vary with year and between countries because of differences in production conditions, product types and MRLs. Thanks to the monitoring programmes, active substances exceeding MRLs and banned pesticides have been detected and monitored. GIS application was found to be a useful tool to display the differences in pesticide residues among exporting countries**.**

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Table and Figure Captions

Table 1. The total number of samples in imported foods analysed for pesticide residues, the number of samples with detectable residues, the number of samples with residues above maximum residue levels (MRLs), samples with detectable residue calculated as a percentage of total analysed samples and samples exceeding MRLs calculated as a percentage of total analysed samples in food product groups during the period in 2000-2020.

Figure 1. Trends in the total number of samples analysed for imported foods to the UK from 2000 to 2020. The dotted line is the fitted regression line to the data without 2000 and 2020. The fitted equation is y=14325.0 -6.286*x with the coefficient of determination (R^2) of 0.058 which measured the proportion of variance accounted for by the year and was not statistically significant ($p>0.31$).

Figure 2. Trends in the percentage of total samples with detectable residues in imported foods to the UK. The dotted line is the fitted regression line to the data without 2020. The fitted equation is y=-2368.0 + 1.2034*x with the coefficient of determination (R^2) of 0.6175 which measured the proportion of variance accounted for by the year and was statistically significant ($p<0.01$).

Figure 3. Trends in the percentage of total samples with residues above MRLs in imported foods to the UK. The dotted line is the fitted regression line to the data without 2020. The fitted equation is y=-363.5 + 0.1825*x with the coefficient of determination (R^2) of 0.6547 which measured the proportion of variance accounted for by the year and was statistically significant ($p<0.01$).

Figure 4. Global map showing total number of samples analysed out of the total samples in imported foods to the UK from each country in the period of 2000-2020.Country outlines © 2018 GADM.

Figure 5. Global map showing percentage of samples with detectable residues out of the total samples in imported foods to the UK from each country in the period of 2000-2020.Country outlines © 2018 GADM.

Figure 6. Global map showing percentage of samples exceeding maximum residue levels (MRLs) out of the total samples in imported foods to the UK from each country in the period of 2000-2020.Country outlines © 2018 GADM.