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*Published in:*  
Food Control

*Link to article, DOI:*  
[10.1016/j.foodcont.2016.11.022](https://doi.org/10.1016/j.foodcont.2016.11.022)

*Publication date:*  
2017

*Document Version*  
Peer reviewed version

[Link back to DTU Orbit](#)

*Citation (APA):*

Poulsen, M. E., Andersen, J. H., Petersen, A., & Jensen, B. H. (2017). Results from the Danish monitoring programme for pesticide residues from the period 2004–2011. *Food Control*, 74, 25-33. DOI: 10.1016/j.foodcont.2016.11.022

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# Accepted Manuscript

Results from the Danish monitoring programme for pesticide residues from the period 2004–2011

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PII: S0956-7135(16)30636-3

DOI: [10.1016/j.foodcont.2016.11.022](https://doi.org/10.1016/j.foodcont.2016.11.022)

Reference: JFCO 5352

To appear in: *Food Control*

Received Date: 1 September 2016

Revised Date: 14 November 2016

Accepted Date: 15 November 2016

Please cite this article as: Poulsen M.E., Andersen J.H., Petersen A. & Jensen B.H., Results from the Danish monitoring programme for pesticide residues from the period 2004–2011, *Food Control* (2016), doi: 10.1016/j.foodcont.2016.11.022.

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# 1 Results from the Danish monitoring programme for pesticide residues from the 2 period 2004 – 2011

## 3 Abstract

4 The Danish pesticide residue monitoring programme evaluates compliance with the maximum residue levels  
5 established by the EU and monitors the residue levels in foods to enable an evaluation of the exposure of the Danish  
6 population to pesticides. The latter part of the programme included 25 different fruits, vegetables and cereals and  
7 processed foods. The commodities were chosen based on their contribution to the intake of pesticides in the Danish  
8 population. A total of 17,309 samples were collected during 2004-2011.. The monitoring showed that the frequencies  
9 of pesticides were higher in samples of foreign origin than in samples of Danish origin both for samples with residues  
10 above or below the MRLs. Overall, pesticide residues were more frequently found in fruits and vegetables than the  
11 other groups of commodities; fruits had higher frequencies than vegetables. Residues above the MRLs were found in  
12 2.6% of the samples. In plant commodities, 163 different substances were found in measurable concentrations.  
13 Residues of more than one pesticide (multiple residues) were found in 27% of all samples. A comparison of the  
14 frequencies of pesticide residues in commodities from different countries showed that Danish commodities with a  
15 lower frequency of pesticides.

## 16 Keywords

18 Pesticide residues  
19 Monitoring programme  
20 Sampling plan  
21 Pesticide residue analysis  
22 Maximum Residues Levels

## 23 24 25 1. Introduction

26 Although food and health authorities worldwide ensure that the risk of exposure to pesticide residues in food is very  
27 limited for residues below the maximum residue limits, MRL (EU - Pesticides database, 2016), Danish consumers are in  
28 general very concerned about pesticide residues in foods. The focus of the Danish consumers is not explicitly on  
29 residues above the MRLs but on how often pesticides are found and how to avoid foods with pesticide residues.

30  
31 Since the beginning of the 1960s, Denmark has monitored fruit and vegetables for pesticide residues. The Danish  
32 monitoring programme for foods was established in 1983. The results are reported for periods of 5-8 years. This paper  
33 covers the fifth period, 2004-2011. The programme included commodities of fruit, vegetable, cereal and animal origin.

34  
35 The aim of the Danish monitoring programme is to evaluate compliance with the maximum residue levels established  
36 by the EU (The European Parliament, 2005) and to monitor the residue levels in foods to enable an evaluation of the  
37 dietary exposure of the Danish population to pesticides (Poulsen, Andersen, Petersen, & Hartkopp, 2005).

38  
39 This paper describes and compares in detail the 2004-2011 monitoring results to show the different factors that  
40 influence the exposure of the Danish population to pesticides. After describing the design of the monitoring  
41 programme, the sampling and analytical methods used are described, and a comparison of the frequency of samples  
42 with residues below and above MRLs between Danish, EU- and non-EU-produced commodities is shown, as well as the  
43 frequency of samples and commodity types with multiple residues. Additionally, the types of pesticides found in fruit,  
44 vegetables and cereals are listed. Finally, a detailed comparison of the frequencies of pesticide residues in  
45 commodities produced in different countries is presented. All residue data can be found in the Appendix.

46  
47 Although pesticide control programmes is implemented in many countries for many years, the amount of peer  
48 reviewed literature on results from pesticide residue monitoring in general is limited, since this type of data is usually  
49 published in reports and are often not in English language. Peer reviewed articles on food control results of pesticide

residues covers typically only one commodity like honey, tomatoes, wine, citrus fruits (Arias, Bojacá, Ahumada, & Schrevels, 2014; Bargańska, Ślebioda, & Namieśnik, 2013; Čuš, Česnik, Bolta, & Gregorčič, 2010; Juan-Borrás, Domenech, & Escriche, 2016; Ortelli, Edder, & Corvi, 2005; Rodríguez López, Ahumada, Díaz, & Guerrero, 2014; Uygun, Koksel, & Atli, 2005), limited number of commodities e.g. fruits (Oliva, Gemal, Nóbrega, & Araújo, 2003), limited number of samples (Knežević & Serdar, 2009), or limited number of pesticides (Chen et al., 2011). Thus comparison of the overall results to other published data is primarily done against results from EU (EFSA, 2013, 2014) and USD (U.S. FDA, 2010; U.S. Food and Drug Administration, 2011).

The residue data presented here have been combined with Danish consumption data for different consumer groups, and dietary exposure calculations have been estimated and published in another paper by Jensen et al., 2015.

## 2. Materials and methods

### 2.1. Design of sampling plan

The sampling plan for the period 2004 to 2006 was structured in the same manner as for the period 1998-2003. A detailed description can be seen in Poulsen et al. (2005). In 2006, the design of the sampling plan was changed, and it has remained the same since then. The sampling plan for fruit and vegetables was drafted in two parts. The first part was designed to enable estimation of the exposure to pesticides and included 25 different fruits, vegetables and cereal commodities as well as processed foods such as wine. The commodities were chosen based on their contribution to the intake of pesticides in the Danish population calculated using the monitoring results from the period 1998-2003 (Poulsen et al., 2005). For these commodities, a fixed number of 50 samples per year were collected. Additionally, 15 samples of commodities referenced in the EU Multiannual Pesticide Control Programme (EU Commission, 2008) were included. Part two included samples that contributed less to the intake of pesticides but was focussed specifically on the compliance with MRLs or labelling of the production method, e.g., organically grown, produced without growth regulators or surface treatment. Part one comprised 70% fruit and vegetable samples and 15% cereal samples. The remaining 15% of the samples were of animal origin, including milk, honey, baby food and organic commodities. The results from these samples are not included in the comparisons, but all results are listed in the Appendix.

### 2.2. Samples

Authorised personnel from the regional food control units under the Danish Veterinary and Food Administration performed the sampling and collected the samples randomly within each commodity. The sampling procedure conformed to the EU directive on sampling for the official control of pesticide residues (EU Commission, 2002). The samples were mainly taken at wholesalers, importers, slaughterhouses and at food processing companies.

The sampling of meat and other products of animal origin is regulated by Council Directive 96/23/EC (EU Commission, 1996). The aim of this directive is to ensure that the Member States monitor primarily their own production of commodities of animal origin for different substances, e.g., pesticides. However, imported samples from non-EU countries shall also be monitored. Depending on the animal species, the number of samples was between 0.03-0.15% of the production or import.

The aim has been to monitor the commodities representative of the Danish market, and for this reason more samples produced in EU Member States and non-EU countries were collected than samples of Danish origin. Thus, one-third of the plant origin samples were of Danish origin. For animal origin samples, more than 90% of the samples were of Danish origin as described by EU regulation 96/23 (EU Commission, 1996).

A total of 17,309 samples were collected over eight years. The number of fruit, vegetable and cereal samples increased, whereas the number of samples of animal origin decreased slightly during the period (see Figure 1). Most of the samples were conventionally grown fresh fruits and vegetables (70%), but conventionally grown cereals (10%) and samples of animal origin (11%) were also collected. In addition, 6% samples of organically grown crops (fresh,

103 frozen, processed) were collected, as well as processed foods (e.g., wine) and samples of baby food (see Table 1).  
104 Approximately 30% of the fruits, vegetables and cereals; 80% of the meat; and 100% of the milk were of Danish  
105 origin, whereas 99% of the wine was produced outside Denmark. Almost 175 different fruit, vegetable and cereal  
106 commodities were sampled; of these, 73 were also organically produced. Detailed results have been published each  
107 year in the period 2004-2011 (Christensen et al., 2007, 2008; Christensen, Petersen, Poulsen, Grossmann, & Holm,  
108 2006; Danish Veterinary and Food Administration, 2005; Jensen et al., 2010, 2011, 2012; Petersen et al., 2009).

109

110 [Insert Table 1 around her]

111

112 The number of samples was low compared directly with other EU countries. However, despite the low number of  
113 samples, Denmark was among the top four European Union (EU) Member States when comparing the number of  
114 samples taken related to the population size (EFSA, 2013). In 2010, Denmark collected 40 samples per 100,000  
115 inhabitants. Only Iceland (86), Cyprus (84) and Slovenia (60) collected more samples than Denmark (EFSA, 2013). In  
116 2010, the average number of monitoring samples per inhabitant in the EU was 14, and, for comparison, the number  
117 was 2.6 in the USA (U.S. FDA, 2010).

118

119

120 [Insert Figure 1 around here]

121

122

123

### 2.3. Analysis

124 The samples were mainly analysed at the Regional Food Laboratories. However, a few of the samples were analysed  
125 at the National Food Institute of the Technical University of Denmark. All laboratories involved in the monitoring  
126 were accredited to perform pesticide analysis in accordance to (International Standard Organization, 2005) by the  
127 Danish body of accreditation, DANAK.

128

129 Analytical methods were developed and documented at the National Food Institute of the Technical University of  
130 Denmark. Fruits and vegetables were analysed by up to five different analytical methods covering an increasing  
131 number of pesticides over the years, from 149 to 238 pesticides. Cereals were analysed by three different methods  
132 covering 105-166 pesticides, and meat was analysed by one method covering 30-44 pesticides. The multi-methods  
133 have changed over the period, but all methods were accredited. The GC-amendable pesticides for fruit and  
134 vegetables were based on an acetone/ethylacetate/cyclohexan extraction which in 2010 was changed to acetonitril  
135 extraction (QuEChERS). The LC-amendable pesticides for fruit and vegetables were based on a methanol extraction.  
136 In addition to the multi methods, three single-residue methods were used. One included carbendazim/thiabendazole  
137 until these pesticides were included in the LC-multimethod. Furthermore, two methods for dithiocarbamate and  
138 chlormequate/mepiquate were used. Cereals were analysed with a GC multi-method based on ethylacetate  
139 extraction and two single-residue methods covering chlormequate/mepiquate and glyphosate. Samples of animal  
140 origin were analysed by a GC multimethod based on acetone/ethylacetate/cyclohexan extraction.

141

## 3. Results and discussion

142

### 3.1. Pesticide residues

143

144 The results for the eight years of monitoring show that more residues were found in samples of foreign origin than in  
145 samples of Danish origin (see Figure 2) for both samples with residues above and below the MRLs. Overall, fruits and  
146 vegetables had higher frequencies of residues than the other groups of commodities; fruits had higher frequencies  
147 than vegetables. Overall, residues above the MRLs were found in 2.6% of the samples, most frequently in fruits.

148

149 The Danish results are in accordance with the results reported by the EU for 2010 and 2011, in which the exceedance  
150 rate for samples from the EU and EFTA was at 1.5%. For samples that originated from non-EU countries, the  
151 exceedance rates were 6.0% and 4.7% for 2010 and 2011, respectively (EFSA, 2013, 2014). The results were also in  
152 accordance with findings in the USA, where the exceedance rates for import samples were 4.9 and 7.1 (U.S. FDA,  
153 2010; U.S. Food and Drug Administration, 2011).

154

155 [Insert Figure 2 around here]

156

157 In plant commodities, 163 different substances were found in measurable concentrations. The residues exceeding the  
158 legal limits included 33 different pesticides. The pesticides which were detected in at least 1% of the samples of plant  
159 products are presented in Figure 3. Malathion was the pesticide analysed in the most samples (16,806). Imazalil was  
160 the pesticide most frequently found (14% of the samples), while dithiocarbamates and carbendazim were the  
161 pesticides that exceeded the MRLs most frequently—0.54% and 0.46% of the samples, respectively.

162

163 [Insert Figure 3 around here]

164

### 165 3.2. Multiple residues

166 Residues of more than one pesticide (multiple residues) were found in 27% of all samples and in 98 different  
167 commodities (see Figure 4). Correspondingly, the EU reported 27% of samples having multiple residues in both 2010  
168 and 2011 (EFSA, 2013, 2014).

169

170 In particular, citrus fruits contained multiple residues in more than 75% of the samples, which was a higher frequency  
171 than the 63% and 60% found in the EU in 2010 and 2011, respectively (EFSA, 2013, 2014). The samples with the  
172 highest number of pesticides were two chili peppers from Thailand, where 10 different pesticides were detected.  
173 Another chili sample from Thailand contained nine different pesticides, and this was also the case for two table grape  
174 samples from Italy and Chile. Samples with eight detected pesticides included three samples of chili peppers (Spain,  
175 Thailand and Vietnam), two samples of lettuce (Belgium and France), and one sample of apple (France), pear  
176 (Belgium) and peas with pods (Kenya). It should be emphasised that it is not necessarily an individual fruit or  
177 vegetable that contained all the detected pesticides because the analysed samples can comprise more than one fruit  
178 or vegetable, e.g., 10 individual fruits. The composite sample can also consist of commodities produced by different  
179 growers. Table 2 shows the commodities with multiple residues and for which more than 30 samples were analysed  
180 for the period 2004-2011.

181

182

183 [Insert Figure 4 around here]

184

184 [Insert Table 2 around here].

185

186

### 186 3.3. Detailed evaluation of pesticide residues in commodities produced in different countries.

187 The commodities consumed in Denmark are produced in many different countries, and the monitoring data were  
188 evaluated to determine any differences in the frequencies of samples with residues of Danish and foreign origin or  
189 between countries. Here, foreign origin signifies countries outside Denmark. For the commodities that contributed  
190 most to the exposure of pesticides for Danish consumers, the frequency of residues in samples has been compared  
191 between countries. Only commodities for which more than 10 samples from the same country were analysed are  
192 included in this evaluation. The results included in the evaluation below were tested by a chi-square test to if  
193 differences seen between countries were statistically significant. For bananas, grapefruit, lemon,  
194 mandarins/clementines and oranges no significant differences were seen, due to the low number of samples without  
195 pesticide residues. For the rest of the commodities a significant differences were seen with CHITEST values <0.05. The  
196 highest values were for apple (0.022) and strawberries (0.011).

197 The frequency of residues found in Denmark is also compared with the frequencies found by the EU monitoring  
198 programme. Because the EU monitoring programme is a rolling programme, all commodities are not analysed every  
199 year; therefore, a comparison has been performed for the years 2009, 2010 and 2011.

200 Bananas were mainly imported from Columbia, Costa Rica and Ecuador, and pesticides were found in 85% of the 425  
201 samples. The banana samples had residues of 14 different pesticides; the pesticides most frequently found were  
202 imazalil and thiabendazole. In 2009 in the EU, an average of 57% of the banana samples had residues of 35 different  
203 pesticides, most frequently imazalil, thiabendazole, chlorpyrifos and azoxystrobin (EFSA, 2011).

204 The main exporters to the Danish market of grapefruit were South Africa, Turkey, the USA and Israel, and pesticides  
205 were found in 99% of the 388 samples. The grapefruit samples had residues of 46 different pesticides, most frequently  
206 imazalil, orthophenylphenol and chlorpyrifos.

207 Most of the lemon and mandarin/clementine samples originated from Spain. Pesticide residues were found in 97% of  
208 the 388 lemon samples, 39 different pesticides were found and the most frequently found pesticides were imazalil  
209 and chlorpyrifos. Pesticide residues were found in 99% of the 411 mandarin samples, 55 different pesticides were  
210 found, and the most frequently found pesticides were imazalil and chlorpyrifos as for lemons. In 2011 in the EU, an  
211 average of 85% of the mandarin samples had residues of 59 different pesticides, most frequently imazalil, chlorpyrifos  
212 and thiabendazole (EFSA, 2014).

213 The samples of orange originated mainly from Spain, Greece, South Africa and Morocco. Pesticide residues were  
214 found in 98% of the orange samples. Forty-nine different pesticides were found, and here the most frequently found  
215 were also imazalil and chlorpyrifos. In 2011 in the EU, an average of 80% of the samples had residues of 73 different  
216 pesticides, the most frequently found being imazalil, bromide ion and chlorpyrifos (EFSA, 2014).

217 Figure 5 shows the frequencies of pesticide residues for 18 different commodities where we found significant  
218 differences in the frequencies between different countries. Below is a short summary of the results for each  
219 commodity.

220 [Insert figure 5 around here]

221 Apples are the commodity that contributes the most to the pesticide intake of the Danish consumer (Jensen et al.,  
222 2015). This is due to the high consumption of apples, the high frequency of pesticide residues in apples and main  
223 method of eating apples raw with the peel. For most pesticides a major part of the content can be found in the peel.  
224 Apples are therefore controlled frequently, and all together 585 samples were collected and analysed. Approximately  
225 35% of the samples originated from Denmark, and approximately 20% originated from Italy and France. The remaining  
226 25% originated from 7 other countries (see Figure 5). The frequencies of pesticide residues ranged from 46%  
227 (Denmark) to 100% (Chile and Brazil). The apple samples had residues of 54 different pesticides, most frequently  
228 chlorpyrifos, diphenylamine and carbendazim. In 2010 in the EU, an average of 68.4% of the samples had residues of  
229 94 different pesticides, most frequently found dithiocarbamates, captan/folpet (sum) and diphenylamine (EFSA,  
230 2013).

231 Approximately 30% of the collected pear samples in the period 2004-2011 were of Danish origin, and another 30%  
232 originated from the Netherlands. In total, 466 pear samples were collected. The frequencies of pesticide residues  
233 ranged from 55% (Denmark) to 100% (Belgium). The pear samples had residues of 48 different pesticides, most  
234 frequently pyraclostrobin, chlormequat and dithiocarbamates. In 2011 in the EU, an average of 70.8% of the samples  
235 had residues of 66 different pesticides, most frequently dithiocarbamates, boscalid and pyraclostrobin (EFSA, 2014).

236 Half of the peach and nectarine samples originated from Italy and Spain. Peaches and nectarines are not grown  
237 commercially in Denmark. In total, 678 samples of peaches and nectarine were collected, and the frequencies of  
238 pesticide residues ranged from 62% (Spain) to 92% (Chile). The peaches and nectarine samples had residues of 40  
239 different pesticides, most frequently tebuconazole, iprodione, carbendazim and chlorpyrifos. In 2010 in the EU, an  
240 average of 73.0% of the samples had residues of 79 different pesticides, most frequently tebuconazole followed by  
241 dithiocarbamates and iprodione (EFSA, 2013).

242 Plums are grown in Denmark, and approximately 20% of the collected samples were of Danish origin. Most of the  
243 foreign samples originated from Spain, South Africa and Chile, covering approximately one-third of the samples taken.  
244 In total, 391 samples of plums were collected, and the frequencies of pesticide residues ranged from 15% (Argentina)  
245 to 85% (Chile). The plum samples had residues of 31 different pesticides. The most frequently found pesticides were  
246 iprodione, tebuconazole and chlorpyrifos.

247 Strawberries are grown in Denmark, and approximately 40% of the samples were of Danish origin in the period 2004-  
248 2011. The main part of the samples (45%) was produced in Spain, Poland, Germany and Belgium. In total, 429  
249 strawberries samples were collected, and the frequencies of pesticide residues ranged from 50% (China and Poland)  
250 to 85% (Belgium). The strawberry samples had residues of 45 different pesticides, most frequently cyprodinil,  
251 fenhexamid and pyraclostrobin. In 2010 in the EU, an average of 67.9% of the samples had residues of 82 different  
252 pesticides, most frequently cyprodinil followed by fludioxonil and boscalid (EFSA, 2013).

253 Most of the table grapes samples originated from South Africa, Italy, Chile, India and Spain (77%). The commodity  
254 table grape is not grown commercially in Denmark. In total, 460 samples of grapes were collected, and the frequencies  
255 of pesticide residues ranged from 55% (Brazil) to 91% (Chile). The grape samples had residues of 54 different  
256 pesticides, most frequently fenhexamid, iprodione and cyprodinil. In 2009 in the EU, an average of 73.3% of the  
257 samples had residues of 76 different pesticides, most frequently fenhexamid, followed by cyprodinil and boscalid  
258 (EFSA, 2011).

259 Cucumbers are grown in Denmark, and approximately 50% of the collected samples were Danish cucumbers. Most of  
260 the foreign-produced samples originated from the Netherlands and Spain (50%). In total, 417 cucumber samples were  
261 collected, and the frequencies of pesticide residues ranged from 38% (Denmark) to 85% (Spain). The cucumber  
262 samples had residues of 36 different pesticides, most frequently azoxystrobin and propamocarb. In 2011 in the EU, an  
263 average of 47.0% of the samples had residues of 67 different pesticides, most frequently bromide ion and  
264 propamocarb (EFSA, 2014).

265 Tomatoes are grown in Denmark, and approximately 45% of the collected samples were of Danish origin. Most of the  
266 foreign-produced samples originated from Spain and the Netherlands (45%). In total, 462 samples of tomato were  
267 collected and analysed. The Danish tomatoes had residues of six different pesticides in only 5% of the samples, with  
268 none above the MRLs. The frequencies of pesticide residues in the samples of foreign origin ranged from 33% (the  
269 Netherlands) to 82% (Spain). The tomato samples had residues of 48 different pesticides, most frequently  
270 procymidone, cyprodinil and iprodione. In 2010 in the EU, an average of 48.9% of the samples had residues of 84  
271 different pesticides, most frequently bromide ion, followed by dithiocarbamates and cyprodinil (EFSA, 2013)

272 Sweet peppers are hardly grown commercially in Denmark. Only two samples were analysed, and both contained  
273 fenhexamid. Most of the samples originated from the Netherlands and Spain. In total, 387 sweet pepper samples  
274 were collected, 72% of them being Dutch or Spanish in origin. The frequencies of pesticide residues ranged from 11%  
275 (Netherlands) to 81% (Turkey). The sweet pepper samples had residues of 52 different pesticides where the most  
276 frequently found were flutriafol, procymidone and azoxystrobin. In 2009 in the EU, an average 34.5% of the samples  
277 had residues of 79 different pesticides, most frequently imidacloprid, flutriafol, and triadimefon (EFSA, 2011)

278 Lettuce is grown in Denmark, and approximately 50% of the collected samples in the period 2004-2011 were covered  
279 by Danish lettuce. One-third of the samples originated from Spain and Germany. In total, 371 lettuce samples were  
280 collected, and the frequencies of pesticide residues ranged from 13% (Denmark) to 65% (the Netherlands). The lettuce  
281 samples had residues of 36 different pesticides, most frequently dithiocarbamates and cyprodinil. In 2010 in the EU,  
282 an average of 57.6% of the samples had residues of 68 different pesticides, most frequently bromide ion,  
283 dithiocarbamates and iprodione (EFSA, 2013)



284 Spinach is grown in Denmark, and approximately 30% of the collected samples in the period 2004-2011 were covered  
285 by Danish spinach. Half of the samples originated from Italy and Germany. In total, 244 spinach samples were  
286 collected, and the frequencies of pesticide residues ranged from 17% (Belgium) to 69% (Spain). The spinach samples  
287 had residues of 28 different pesticides, most frequently bromide ion, lambda-cyhalothrin and deltamethrin. In 2011 in  
288 the EU, an average of 46.7% of the samples had residues of 50 different pesticides, most frequently bromide ion and  
289 dithiocarbamates (EFSA, 2014).

290 Most of the beans with pod samples originated from Kenya, Germany, Egypt, the Netherlands and Morocco. In total,  
291 311 bean samples were collected, and the frequencies of pesticide residues ranged from 29% (Egypt) to 75%  
292 (Netherlands). The bean samples had residues of 43 different pesticides, most frequently cyprodinil and azoxystrobin.  
293 In 2011 in the EU, an average of 46.1% of the samples had residues of 64 different pesticides, most frequently  
294 bromide ion, boscalid and iprodione (EFSA, 2014).

295 Carrots are grown in Denmark, and approximately 70% of the collected samples were covered by Danish carrots. Most  
296 of the foreign samples originated from Italy and Germany, covering 20% of the samples taken. In total, 499 carrot  
297 samples were collected and analysed, and the frequencies of pesticide residues ranged from 0% (Belgium) to 43%  
298 (Italy). The carrot samples had residues of 19 different pesticides, most frequently linuron. In 2011 in the EU, an  
299 average of 42.7% of the samples had residues of 51 different pesticides, frequently bromide ion, boscalid and linuron  
300 (EFSA, 2014).

301 Potatoes are grown in Denmark, and approximately 85% of the collected samples were covered by Danish potatoes.  
302 Only 9% originated from the United Kingdom, France and Spain. In total, 669 potato samples were collected, and 424  
303 were from Denmark. In the potato samples of Danish origin four different pesticides were found in only 2% of the  
304 samples, none above the MRLs. One of the pesticides was quintozone, which is a pollutant in the soil from earlier uses.  
305 The frequencies of pesticide residues in samples of foreign origin ranged from 14% (UK) to 54% (France). The potato  
306 samples had residues of 7 different pesticides, most frequently chlorpropham, propamocarb and metalaxyl. In 2011 in  
307 the EU, an average of 23.8% of the samples had residues of 30 different pesticides, most frequently bromide ion,  
308 chlorpropham and propamocarb (EFSA, 2014).

309 The volume of red wine produced in Denmark is very small. Only two samples were analysed, and no pesticide  
310 residues were found. Most of the samples originated from Argentina, Chile, France, Italy and Spain. In total, 268  
311 samples of red wine were collected. The frequencies of pesticide residues ranged from 15% (USA) to 87% (Argentina).  
312 The wine samples had residues of 16 different pesticides, most frequently carbendazim, dimethomorph and  
313 fenhexamid. Wine was not included in the EU multi-annual pesticide control programme before 2013. However, in  
314 2013 an average of 45.0% of the white and red wine samples had residues of 37 different pesticides, most frequently  
315 boscalid, followed by dimethomorph and fenhexamid (EFSA, 2015)

316 Wheat is grown in Denmark, and approximately 40% of the wheat flour samples were of Danish origin. For  
317 approximately 38% of the analysed samples the origin was unknown. In total, 143 samples of wheat flour were  
318 collected, and the frequencies of pesticide residues ranged from 22% (Denmark) to 84% (Germany). The wheat flour  
319 samples had residues of 7 different pesticides, most frequently chlormequat. In 2011 in the EU, an average of 52.1% of  
320 the samples had residues of 18 different pesticides, most frequently found chlormequat and pirimiphos-methyl (EFSA,  
321 2014).

322 Samples of wheat kernels were also collected, and approximately 50% of the wheat kernel samples were of Danish  
323 origin. In total, 442 samples were collected, and the frequencies of pesticide residues ranged from 22% (Denmark) to  
324 41% (Germany). The wheat kernel samples had residues of 9 different pesticides, most frequently chlormequat and  
325 pirimiphos-methyl. In 2009 in the EU, an average of 32.2% of the samples had residues of 38 different pesticides, most  
326 frequently chlormequat and pirimiphos-methyl (EFSA, 2011).

327 Oat is grown in Denmark, and approximately 40% of the samples of rolled oats were of Danish origin. For  
328 approximately 30% of the analysed samples the origin was unknown. Samples from Germany, Sweden, and the United  
329 Kingdom were collected and analysed. In total, 184 samples of rolled oats were collected, and the frequencies of  
330 pesticide residues ranged from 4% (Denmark) to 84% (Germany). The oat samples had residues of 6 different  
331 pesticides, most frequently chlormequat. In 2010 in the EU, an average of 54.4% of the samples had residues of 20  
332 different pesticides, most frequently chlormequat, glyphosate and pirimiphos-methyl (EFSA, 2013).

333 The differences in the pesticide residue frequencies of samples originating from different countries indicate that if  
334 Danish consumers chose commodities of Danish origin, they would have had a lower exposure to pesticides. This is  
335 especially the case for apples, pears, cucumbers, tomatoes, lettuce, spinach, potatoes, wheat flour and rolled oats.  
336 The reasons for the lower frequencies of pesticide residues in commodities of Danish origin are due to several factors.  
337 Denmark has for many years had a stricter regulation on pesticide use, and the pest pressure in Denmark is lesser than  
338 in countries with a warmer climate. In addition, the use of biological pest control is common for commodities grown in  
339 greenhouses. Dietary exposure calculations have shown that choosing Danish-produced commodities whenever  
340 possible could reduce the exposure and the Hazard Index by a factor of approximately 2 (Jensen et al., 2015).

341 When looking at the frequency profile for the different commodities shown in Figure 5, more or less same profile was  
342 found for apple, pear, peaches and nectarines, strawberries, table grapes in the pesticide residue with frequencies  
343 between 50-100%. All these commodities are grown in open fields. The rest of the commodities pesticide residues  
344 were found 2-7 times more frequently in samples from the country with the highest frequency compared with the  
345 country with the lowest frequency. For cucumber, tomatoes, sweet peppers, lettuce, spinach the larger differences  
346 could result from that the commodities in some of the countries (with low frequencies) were grown in greenhouses,  
347 where pest can be controlled by biological treatment instead of by pesticides. However, plums, beans with pods,  
348 carrots, potatoes, the grape used for producing red wine and the cereals are grown in open fields in all countries. The  
349 explanation could be different pests, different treatment practices or different approval of pesticides to use in the  
350 production.

351 [Insert Table 3 around here]

#### 352 3.4. General remarks on the pesticide residue findings and human health.

353 Regulation (EC) 1107/2009 (EU Commission, 2009) concerning the placing of plant protection products on the market  
354 states that pesticide residues detected after application consistent with Good Agricultural Practice (GAP) (FAO, 2008)  
355 shall not have any harmful effects on human health. As described above pesticide residues were found in many of the  
356 samples. In about 2-3% of the samples, residues above the MRLs were found and often more than one pesticide  
357 residue per sample was found. An exceedance of an MRL will not necessarily causes health problems, because the  
358 MRLs are set based on GAP which may result in MRLs giving exposures well below any toxicological based guidance  
359 values, e.g. acceptable Daily Intake (ADI) and Acute Reference Dose (ARfD). If residues from a specific pesticide would  
360 cause health problems, the pesticide will not be approved for use according to Regulation (EC) 1107/2009 (EU  
361 Commission, 2009). When setting the MRLs, the evaluation of the pesticides is done on the individual pesticides and  
362 this do not cover effects from multiple residues. However, the pesticide residue monitoring results described above  
363 have been used to calculate the exposure for pesticides in the Danish population (Jensen et al., 2015) as mentioned in  
364 the introduction. In this paper the cumulative exposure was calculated using the Hazard Index method and as all  
365 pesticides had the same effect. The highest HI was calculated for children 4-6 years of age and amounted to 0.44  
366 which is well below 1. It was concluded that there is no risk of adverse health effects following chronic cumulative  
367 exposure to the pesticides found in fruit, vegetables and cereals on the Danish market. As it is shown in this paper,  
368 samples of Danish have general have lower contents than samples of foreign origin so eating "Danish" whenever  
369 possible will reduce the exposure by a factor of 2 (Jensen et al., 2015).

370 **4. Conclusion**

371 A total of 17,309 samples were collected in the Danish monitoring programme during 2004-2011. Most of the  
 372 samples were conventionally grown fresh fruits and vegetables (70%), but conventionally grown cereals (10%) and  
 373 samples of animal origin (11%) were also collected. Denmark is one of the EU member states that include the most  
 374 samples in the pesticide monitoring programme in relation to population size. The results for the eight-year  
 375 monitoring show that residues were more frequently found in samples of foreign origin than in samples of Danish  
 376 origin (see Figure 2) both for samples with residues above and below the MRLs. In general, fruits and vegetables had  
 377 higher frequencies of residues than the other groups of commodities; fruits had higher frequencies than vegetables.  
 378 Overall, residues above the MRLs were found in 2.6% of the samples, most frequently in fruit. In plant commodities,  
 379 163 different substances were found in measurable concentrations. Residues exceeding the legal limits included 33  
 380 different pesticides. Residues of more than one pesticide (multiple residues) were found in 27% of all samples and in  
 381 98 different commodities. The results indicate that if Danish consumers choose commodities of Danish origin  
 382 whenever possible, they will have a lower exposure to pesticides. This is especially the case for apples, pears,  
 383 cucumbers, tomatoes, lettuce, spinach, potatoes, wheat flour and rolled oats. This is confirmed in the paper by Jensen  
 384 et. al 2015, whose exposure calculations showed that choosing Danish commodities whenever possible reduced the  
 385 exposure and the hazard by a factor of two.  
 386

387 **Acknowledgement**

388 Current publication was performed in the framework of the National Reference Laboratories for pesticide residues in  
 389 cereals and feeding stuff, financed by the Danish Veterinary and Food Administration.

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1 Table 1. Number of samples analysed for the period 2004-2011 of Danish and foreign origin.

<b>Foods</b>	<b>Danish</b>	<b>Foreign</b>	<b>Total</b>
Fruit and vegetables (fresh, frozen, processed)	2844	9182	12026
Cereals (including processed)	717	1060	1777
Wine	2	273	275
Meat	1589	358	1947
Milk and honey	146	0	146
Baby food	28	38	66
Organically grown fruit, vegetables and cereals (fresh, frozen, processed)	358	714	1072
<b>Total</b>	<b>5684</b>	<b>11625</b>	<b>17309</b>

2

1 Table 2. Percentage of samples with multiple residues. Only commodities where more than 30 samples were analysed  
 2 during the period 2004-2011 are included.

<b>Commodities</b>	<b>Samples, multiple residues</b>	<b>Commodities</b>	<b>Samples, multiple residues</b>	<b>Commodities</b>	<b>Samples, multiple residues</b>
Apricots	52%	Lettuce	15%	Potatoes	1%
Apples	39%	Limes	50%	Rambutan	26%
Aubergines	11%	Mandarin, clementine	93%	Raspberries	31%
Bananas	50%	Mangoes	17%	Red currants	51%
Beans with pods	18%	Melons	1%	Rice	2%
Blackberry	9%	Mushrooms, cult.	8%	Rye flour	6%
Blue berries	20%	Oat kernels	7%	Rye kernels	2%
Carrots	4%	Oranges	82%	Spelt	16%
Celery	19%	Papaya	61%	Spelt flour	17%
Chilies	48%	Parsley root	5%	Spinach	4%
Courgettes	5%	Parsnip	3%	Spring onions	17%
Cucumbers	17%	Passionsfruits	37%	Star fruit	32%
Grapefruits	86%	Peaches, nectarines	38%	Strawberries	37%
Grapes	49%	Pears	44%	Sweet peppers	17%
Rolled oats	3%	Peas with pods	61%	Tea	13%
Kakis	7%	Peas without pods	6%	Tomatoes	19%
Kiwis	10%	Pineapples	23%	Water melons	8%
Leeks	2%	Plums	11%	Wheat flour	7%
Lemons	79%	Pomelos	61%	Wheat kernels	6%

3

1 **Table 3.** The ISO 3166 Codes (Countries) used in Figure 5. The codes can be found on  
2 <https://www.iso.org/obp/ui/#search> (Accessed on 7 July 2016)

Country	Code	Country	Code
Argentina	AR	Italy	IT
Australia	AU	Kenya	KE
Belgium	BE	Morocco	MA
Brazil	BR	Netherlands	NL
Chile	CL	Poland	PL
China	CN	South Africa	ZA
Denmark	DK	Spain	ES
Egypt	EG	Sweden	SE
France	FR	Turkey	TR
Germany	DE	United Kingdom	GB
Greece	GR	USA	US
Israel	IL	Unknown origin	UO

3

4



1

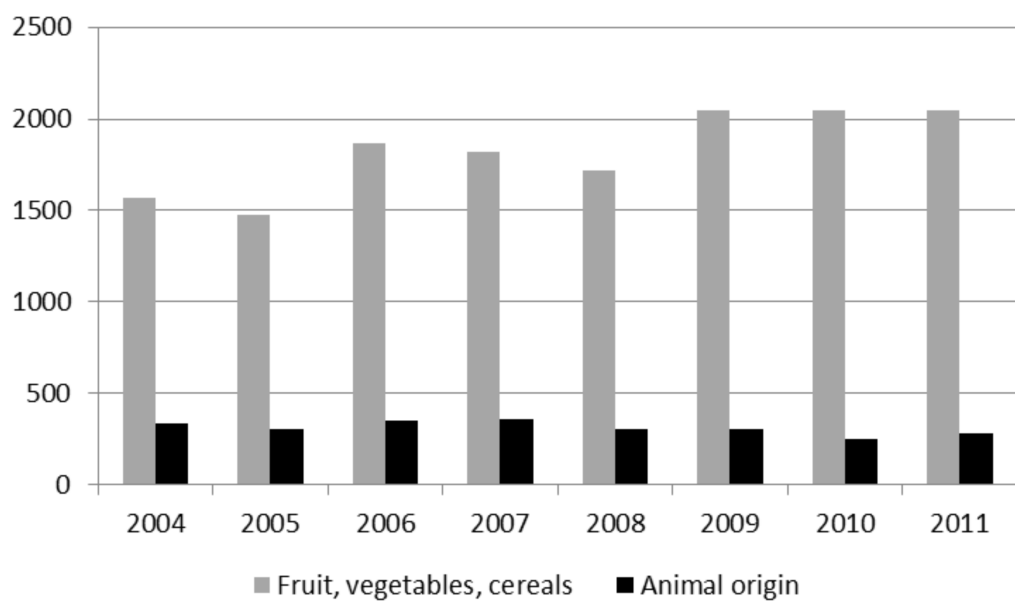
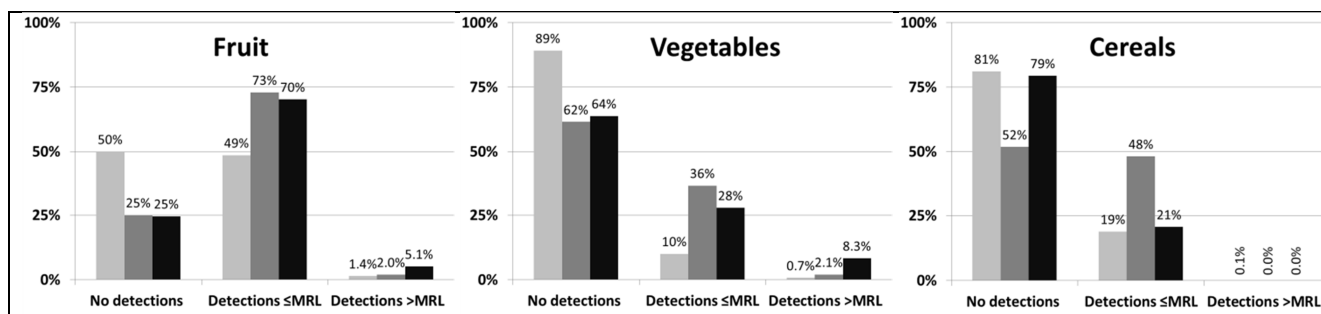
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3  
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Figure 1. Numbers of fruit, vegetables and cereal samples as well as samples of animal origin analysed during the period 2004-2011.

1



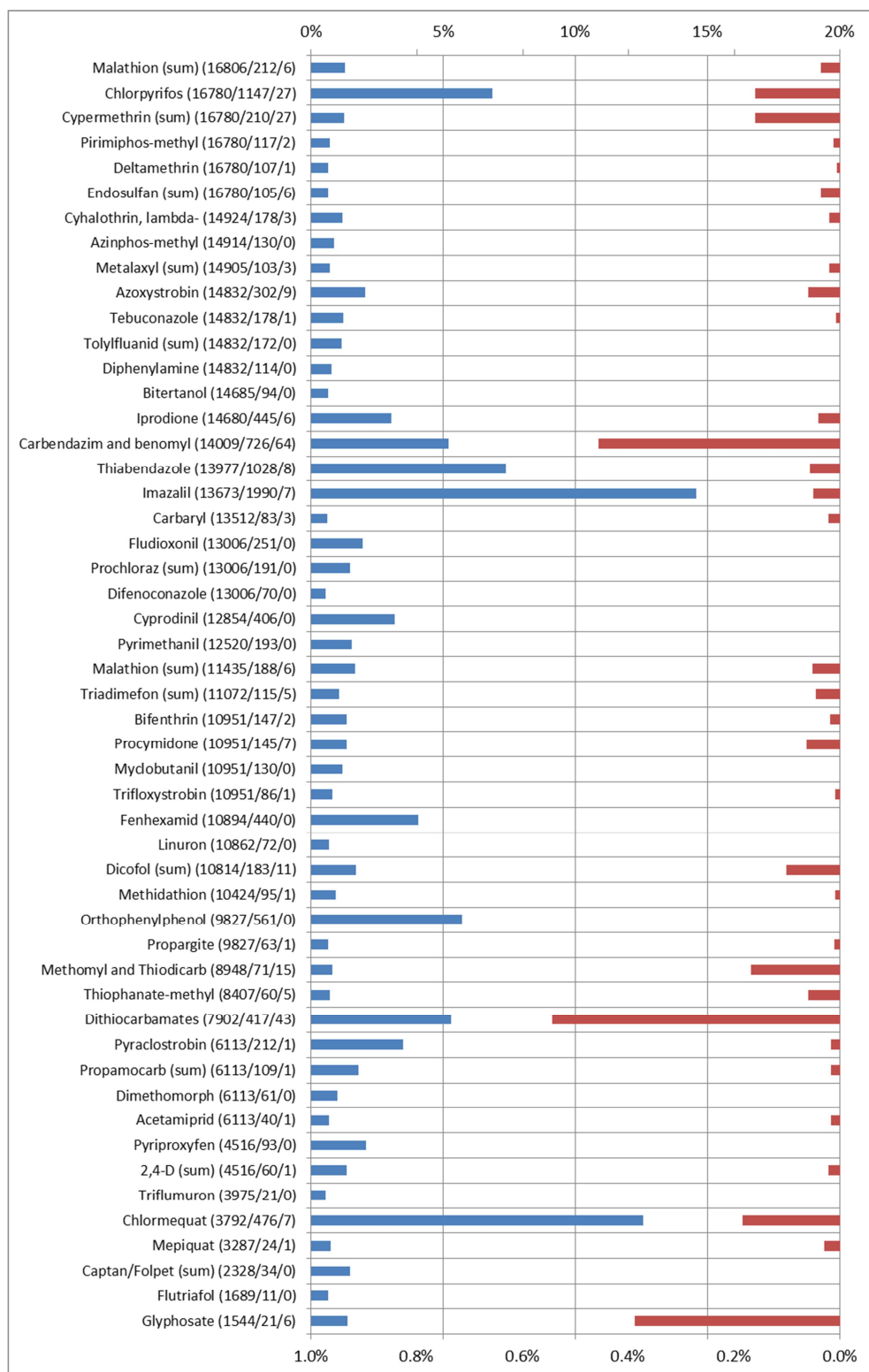
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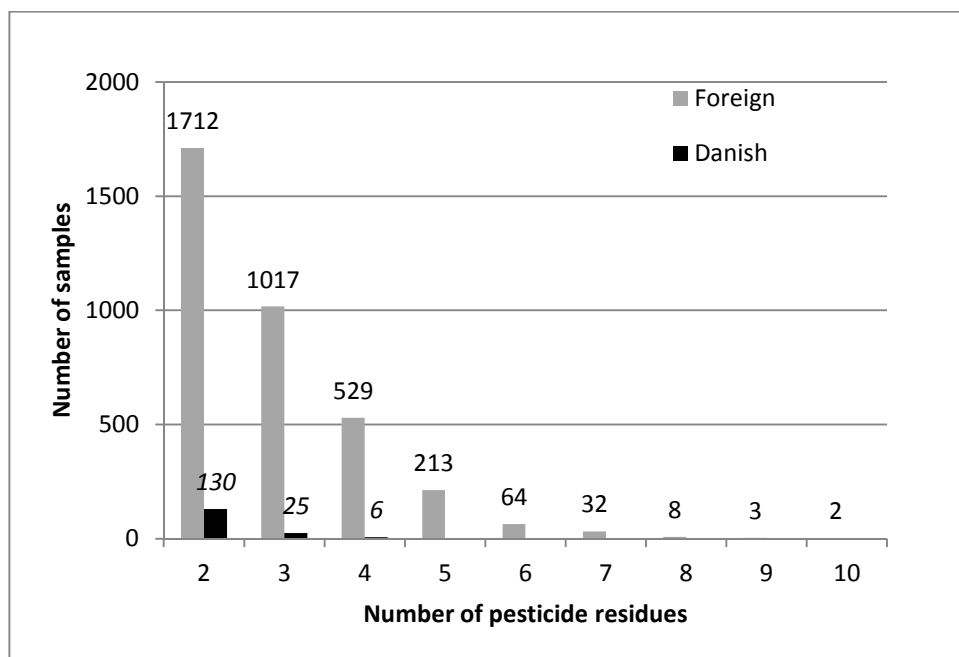
Figure 2. Pesticide residues detected in fruit, vegetables and cereals produced in Denmark (light grey bars), the EU (dark grey bars), and outside the EU (black bars).



1

2 Figure 3. Detected pesticides. The pesticides that were detected in at least 1% of the plant product samples are  
 3 ordered by the total number of samples analysed for the pesticide. The figures in brackets next to the name of the  
 4 pesticide refer to the total number of samples analysed for this pesticide, the number of samples with residues within the  
 5 legally permitted concentrations and the number of samples exceeding the MRLs. The blue bars represent the  
 6 percentage of samples within the legally permitted concentration. The axis for these results is shown at the top (0% -  
 7 20%). The red bars represent the percentage of samples with residues above the MRL. The axis for these results is  
 8 shown at the bottom (0.0% - 1.0%).

1



2

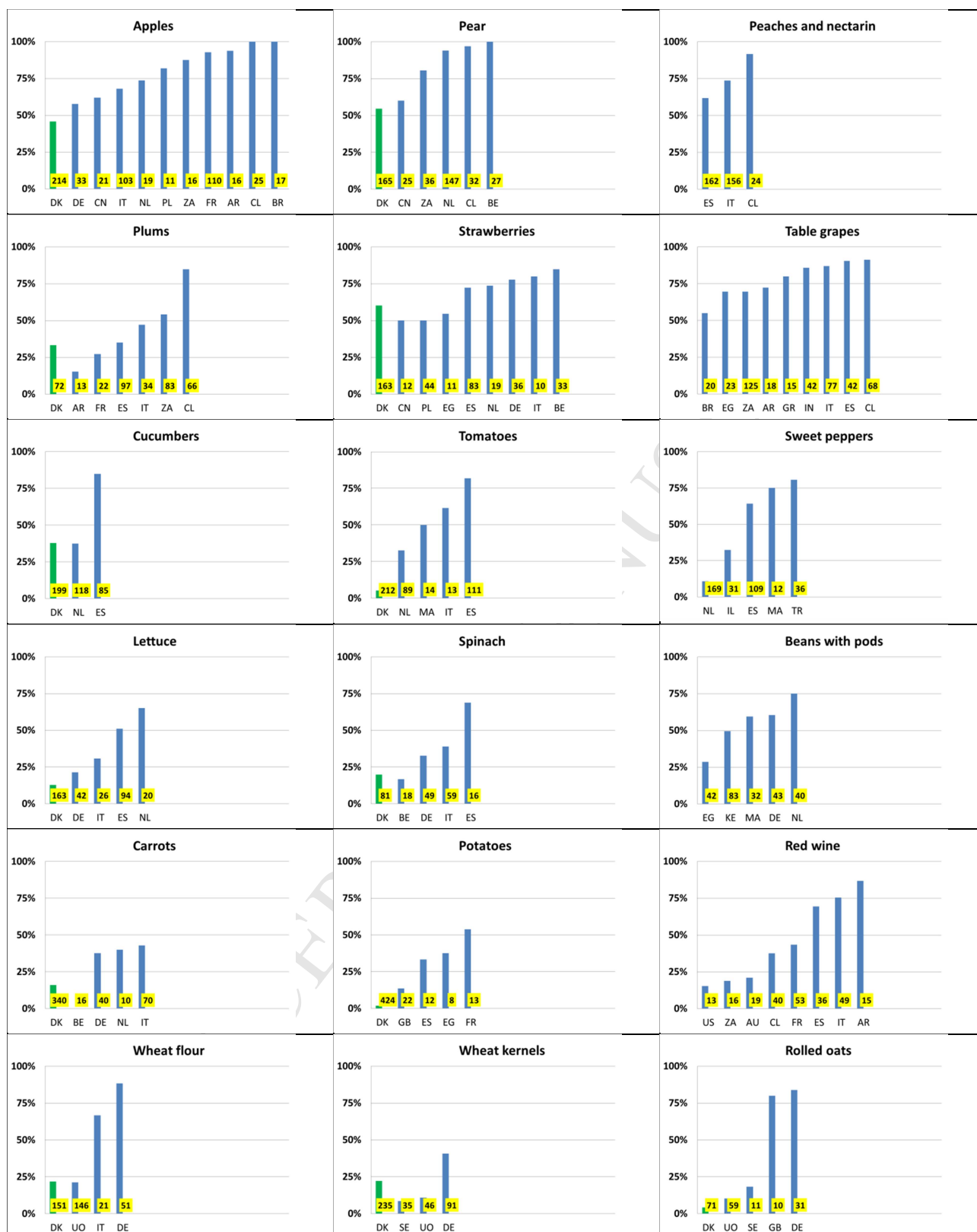
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5

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Figure 4. Number of samples with 2-10 residues per sample for the period 2004-2011.



1

2 Figure 5. Frequencies of samples with pesticide residues. The figures marked in yellow are the number of samples  
 3 originating from the countries listed. The country codes are ISO 3166 codes (see table 3).

## Highlights

- Results of 17,309 samples from the Danish Pesticide Monitoring Programme 2004-2011
- Fruits and vegetables had higher frequencies of residues than the other commodities
- Residues were more frequently found in samples of foreign origin than in samples of Danish origin
- 163 different substances were found in measurable
- Multiple residues were found in 27% of all samples