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Prioritization of chemical hazards in spices and herbs for European monitoring programs

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ABSTRACT

Monitoring programs are preferably risk-based, which allows focusing on the most relevant human health risks. In this study, a risk matrix was used to identify those chemical hazards that have the highest human health risk for the following spices and herbs: paprika/chilli powder, black pepper, nutmeg, basil, thyme, and parsley. Both the probability of occurrence and the severity of the hazard were assessed for 36 chemical compounds and classified into low, medium, high, and severe. Probability of occurrence was evaluated based on available monitoring data and RASFF notifications as well as possibilities for economic adulteration. Severity was assessed based on available toxicological reference values and classification of carcinogenicity. The results demonstrated that the mycotoxins aflatoxins and ochratoxin A, the pesticides chlorpyrifos and triazophos, and the dye Sudan I posed the highest human health risk for spices and herbs. These compounds should, therefore, have an increased monitoring frequency in these products.

1. Introduction

Spices and herbs are present in almost every processed food, including ready-to-eat products, and are often used by the consumer for flavouring purposes without further processing (Pilizota, 2014). In the European Union (EU), consumption of spices and herbs increased by 1.7% per year between 2010 and 2013, with a total consumption of 385,000 tonnes in 2012. Although there is a broad range of countries producing spices and herbs, China is currently the main supplier (CBI Market Intelligence, 2015). One of the reasons for the increased consumption is the growing demand for natural flavours and colourants. Colour and flavour are thus the main quality parameters of different spices (e.g., paprika powder, chilli powder or saffron) (Peter, 2006). However, other parameters, such as the presence of food safety hazards are as important to consider. Similar to other agricultural products, spices and herbs may be subjected to chemical contaminations within one or more stages of the supply chain. The European Rapid Alert System for Food and Feed (RASFF) has shown 1831 notifications for spices and herbs between 2004 and 2014. The chemical hazard most often notified is aflatoxins (Banach, Stratakou, van der Fels-Klerx, Besten, & Zwietering, 2016), which are toxic secondary metabolites produced by *Aspergillus* spp. Besides natural contaminants, spices and herbs may be subjected to deliberate contaminations. For example, dyes are added as colourants to make the spice look fresher. Sudan dyes have been added to chilli, curry and paprika powder to intensify and maintain the natural red colour of the spices (Everstine, Spink, & Kennedy, 2013). However, Sudan compounds are not authorised as food additives in the European Union due to their carcinogenic properties. The illegal presence of Sudan I was first reported in 2003. Since then, this dye as well as other azo-dyes, such as para red, have frequently been reported to RASFF between 2003 and 2005 (Everstine et al., 2013). Besides the addition of artificial dyes, chemical residues from pesticides may be present. Various pesticide residues have been reported, sometimes exceeding the maximum residue level (MRL) (Ferrer Amate, Unterluggauer, Fischer, Fernández-Alba, & Masselter, 2010). Organochloride pesticides (OCPs) like DDT (4,4′-Dichlorodiphenyltrichloroethane) have been banned from the European market since 1986 by Council Directive 79/117/EEC, but due to their persistent nature they are still present in the environment. Consequently, they can contaminate spices and herbs (Ferrer Amate et al., 2010). It must be noted that EU MRLs for pesticides are not always related to human health effects. Sometimes, MRLs are set at the LOD when a compound has not obtained a renewal of authorisation due to economic reasons or when there are environmental issues related

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to the compound. Exceedance of the MRL for pesticides, thus, may not automatically result in human health effects.

Control of chemical hazards along the food chain is important since consumers cannot substantially reduce them during food preparation. Therefore, chemical analysis of spices and herbs is relevant in terms of quality control and in order to protect human health. Food safety authorities are responsible for checking product compliance to the legal limits. As indicated above, a wide range of chemical hazards may be present in spices and herbs. Therefore, a monitoring program is preferably risk-based allowing for an efficient allocation of resources to the most relevant human health risks. Applying a prioritization method has been shown to increase the probability of detection and decrease surveillance budgets (Baptista, Alban, Olsen, Petersen, & Toft, 2012). Furthermore, an effective monitoring program along the supply chain allows the identification of critical points for the most relevant compounds. Then, mitigation actions can be implemented to reduce human health risks associated with the presence of these chemical hazards. Risk in this case is defined as the probability that a hazard may occur in a food product and the effect, or severity, of that hazard on human health. Probability of occurrence in this case is evaluated against the EU legal limits as the first priority is that spices and herbs sold on the EU market fulfil the legal requirements. Compounds may accidentally be present above legal limits, but also as a result of deliberate contaminations for economic reasons. Therefore, the potential for economic adulteration is also included in the assessment of probability analogous to Hanlon, Hlywka, and Scimeca (2015). Apart from probability of occurrence, the effect on human health is incorporated by establishing the toxic effect of the compound. Combining these two elements provides the human health risk of a chemical hazard in spices and herbs. Other than human health risks, there may be other reasons, such as political explanations or consumer concerns, for including chemical hazards in a priority list. These factors were, however, not included in the risk ranking performed in this study.

The aim of this study was to prioritize the risks of chemical hazards in spices and herbs as input for setting up monitoring programs. Since there are more than 400 spices and herbs commercially available and there is a broad variability in supply chains, this study focused on a subset of spices and herbs. Paprika powder and pepper, primarily black pepper, are the most frequently consumed spices in Europe, whereas parsley and thyme are the most frequently consumed herbs (CBI Market Intelligence, 2011). Most RASFF notifications were for the spices chilli/paprika powder, curry, nutmeg and pepper and the herb basil. Monitoring data from Germany and the Netherlands and the WHO GEMS database showed that most chemical analyses were performed on the spices chilli/paprika powder, nutmeg and pepper and the herbs basil and parsley. Therefore, this study focused on three spices: paprika powder/chilli powder, black pepper and nutmeg and three herbs: basil, thyme and parsley based on available data from monitoring programs, notifications from RASFF, consumption data, and as recommended by experts.

2. Materials & methods

2.1. Selection of a prioritization method

Various methods are available for prioritizing chemical hazards ranging from quantitative to semi-quantitative to qualitative methods. An appropriate ranking scheme was selected based on the characteristics of the various prioritization methods and the data availability as described in van der Fels-Klerx et al. (2015). For this purpose, the following prerequisites were considered: the method selected should be as accurate as possible and should be easy to communicate to the risk manager; communication may be through graphs, lists or tables; stakeholder input and economic analysis were not required and time and resources available were limited (<6 man months). Besides the methods' characteristics, data availability is another criterion for selecting an appropriate prioritization method. For the selected spices and herbs, food consumption data, occurrence data and toxicological reference values of the chemical hazards were available. Based on these prerequisites and the available data, the risk matrix and expert synthesis method were deemed most appropriate (van der Fels-Klerx et al., 2015). Since the risk matrix method can include semi-quantitative as well as qualitative information, this method was selected for prioritization.

2.2. Scores attributed

Low, medium, high and severe scores were attributed to both the severity and the probability of occurrence of the chemical hazards. Adverse effects can range from acute to chronic. Toxicologists use the most sensitive adverse effect registered to derive the Acute Reference Dose (ARfD) and/or the Acceptable Daily Intake (ADI) for chronic effects of avoidable contaminants such as pesticides or the Tolerable Daily Intake (TDI) for chronic effects of unavoidable contaminants such as dioxins and mycotoxins. These general toxicological reference values were used to attribute a score to the severity of a hazard. Additionally, carcinogenicity was included in the analysis using the International Agency for Research on Cancer (IARC) or US Environmental Protection Agency (US EPA) grouping (Table 1). The classification in severe, high, medium and low was based on literature (Hanlon et al., 2015; van Asselt, van der Spiegel, Noordam, Pikkemaat, & van der Fels-Klerx, 2013) and derived such that a normal distribution of compounds was obtained over the four classes. For each compound included in the risk ranking, the highest score was selected from the acute, chronic and carcinogenicity criteria in order to classify the severity of a hazard.

The possible presence, or probability, of chemical hazards in the selected spices and herbs was based on the potential for economic adulteration (Hanlon et al., 2015) and historical data on compounds found in the selected spices and herbs (VRC, 2015). The classification is indicated in Table 2. The highest score from these two criteria was used to classify the probability of occurrence of the chemical hazard into low, medium, high or severe.

The chemical hazards were subsequently plotted in a 4×4 risk matrix based on their scores on severity and probability in the specified spices and herbs. The colouring scheme as presented by Hanlon et al. (2015) was used to indicate high priority compounds (red), medium priority compounds (yellow) and low priority compounds (green).

2.3. Data collection

Chemical hazards mentioned in literature, notified in the RASFF database, present in available monitoring data and indicated by experts were included in the prioritization of the selected spices and herbs: mycotoxins (aflatoxin B_1, B_2, G_1 and G_2, fumonisin B_1, ochratoxin A and sterigmatocystine), plant toxins (estragole, methyleugenol, safrole and trans-anethole), pesticides (bifenthrin, carbendazim, chlorpyrifos, cypermethrin, DDT, dimethoate, endosulfan, ethion, metalaxyl, profenofos, propamocarb, tefluthrin, triazophos and trifluralin), dioxins and dioxin-like polychlorinated biphenyls (dl-PCBs), polycyclic aromatic hydrocarbons (PAHs), heavy metals (arsenic, cadmium, lead and mercury) and dyes (Sudan I, III, IV, and Para Red).

In order to classify the severity of the selected hazards, general toxicological reference values were obtained from the EU pesticide

Table 1
Scores for severity of a compound (Hanlon et al., 2015; van Asselt et al., 2013).

	Severe	High	Medium	Low
ARfD (μg/kg bw/day)	<10	10–50	50–200	≥200
ADI/TDI (μg/kg bw/day)	<1	1–10	10–30	≥30
Carcinogenicity	Carcinogenic to humans (IARC group 1 or US EPA Group A)	Probably carcinogenic (IARC Group 2A or US EPA Group B1 and B2) OR no evidence base on which to make the assessment	Possibly carcinogenic (IARC Group 2B or US EPA Group C)	Not classifiable as to its carcinogenic to humans (IARC group 3 or USEPA group D) OR probably not carcinogenic to humans (IARC Group 4 or US EPA Group E)

Table 2
Scores for probability of occurrence of a compound (Hanlon et al., 2015).

	Severe	High	Medium	Low
Potential for economic adulteration	**Known examples** of economic adulteration in spices and herbs	**Potential** for economic adulteration (e.g. examples of compounds adulterated in other foods,)	**Low potential** for economic adulteration (e.g. compound is not approved for use in the EU)	**Unlikely potential** for economic adulteration (e.g. compounds approved for use within the EU and/or compounds not demonstrated to be fraudulent in the past)
Historical data demonstrating presence of a contaminant in spices or herbs	Residues detected at MRL/ML or above in >1% of samples **OR** multiple RASFF notifications (>2) in last 10 years	Residues detected at MRL/ML or above in ≤1% of samples **OR** ≤2 RASFF notifications in the last 10 years **OR** no evidence base on which to make assessment (no MRL/ML specified or little to no testing)	Residues detected during last 10 years at concentrations below MRL/ML **AND** no RASFF notifications in the last 10 years	No evidence for residues where >100 samples have been included in an EU state monitoring programs in the last 10 years **AND** no RASFF notifications in the last 10 years

database, European Food Safety Authority (EFSA), World Health Organization (WHO), Joint Meeting on Pesticide Residues (JMPR), IARC and US EPA. Data for classifying the hazards based on economic adulteration were obtained from google and literature search for fraud incidences with spices and other food products. Data for classifying the hazards based on historical data were obtained from national monitoring data from two EU Member States (Germany and The Netherlands), monitoring data from WHO selecting the European region (https://extranet.who.int/gemsfood/) and RASFF notifications (https://webgate.ec.europa.eu/rasff-window/portal/) for the years 2004 up to 2014. Monitoring data were compared to the EU legal limits as indicated in Table 3. Data from the various monitoring programs included 6614 samples for chilli/paprika powder, 6740 samples for black pepper, 5679 for nutmeg, 4841 for basil, 153 for thyme and 6121 for parsley. RASFF notifications included 461 notifications for chilli/paprika powder, 50 for black pepper, 72 for nutmeg and 30 for basil.

3. Results

Various chemical hazards may be present in spices and herbs including natural contaminants such as mycotoxins and plant toxins, residues of agrochemicals, such as pesticides, environmental contaminants such as dioxins, heavy metals and PAHs and deliberate contaminants such as dyes. For each selected chemical hazard, both the severity and the probability were assessed based on the available data. Results for the severity of the hazards are indicated in Table 4. Toxicological reference values for acute and chronic effects were established using general ARfDs and ADI or TDI values as derived by EFSA, WHO and JMPR. Results for the probability of the hazards are indicated in Table 5.

Based on the outcome of the evaluation, the chemical hazards were plotted in a 4 × 4 risk matrix analogous to Hanlon et al. (2015) (Fig. 1). Chemical hazards with a low priority for monitoring are indicated in green, hazards with a medium priority are indicated in yellow and hazards with a high priority for monitoring are indicated in red in this figure. For natural contaminations, aflatoxins and ochratoxin A were determined to be high priority compounds in the selected spices and herbs. These mycotoxins have severe toxicity (carcinogenic characteristics and/or severe chronic toxic effects) and had multiple RASFF notifications in the last 10 years and/or more than 1% of the samples contained levels above the maximum limits (MLs) within the available national monitoring programs or the WHO database. With respect to the agrochemicals, the pesticides chlorpyrifos and triazophos were classified as high priority compounds in the selected spices and herbs. They may have a severe acute effect on human health (low ARfD) and showed multiple RASFF notifications. Furthermore, they were detected at levels above the MRL in more than 1% of the samples (Fig. 1). For deliberate contaminations, Sudan I was determined to be of high priority in the selected spices and herbs. In 1987, the IARC classified the Sudan dyes in group 3 'Not classifiable as to its carcinogenicity to humans' (IARC, 2015). A more recent study by EFSA, however, concluded that Sudan I is carcinogenic. For the other dyes, EFSA concludes they are potentially genotoxic and possibly carcinogenic, but conclusive evidence is lacking (EFSA, 2005). Due to this lack of information, the toxicity of these dyes was classified as high (no evidence base). As a result of their prohibited use in the EU, several RASFF notifications have been reported since 2003 for the selected spices and herbs. Also, these dyes have a known history of adulteration for economic benefits (e.g. in paprika and chillies) (Johnson, 2014).

The medium priority group (yellow in Fig. 1) contains several pesticides: carbendazim, cypermethrin, dimethoate, endosulfan and ethion. These pesticides were either frequently reported in RASFF or found at levels above the MRL, but their toxicity is classified as less severe than for chlorpyrifos and triazophos. The environmental contaminants (heavy metals, dioxins and dl-PCBs and PAHs) were also classified in the medium priority group. Heavy metals were classified as severely or highly toxic due to their carcinogenic properties and/or severe chronic effects. The probability of occurrence of lead, mercury and arsenic was classified as high as there was no evidence base to categorize them (no MLs available and/or limited testing). For cadmium, less than 1% of the samples was above the ML (0.20 mg/kg wet weight for fresh herbs; EU regulation 1881/2006). Dioxins and dl-PCBs have a severe

Table 3
EU legal limits for the various compounds.

Compound	ML or MRL (if available)	Legislation
Aflatoxin B_1	Spices: 5 μg/kg	Reg. (EC) 1881/2006
Aflatoxin B_2	Spices: 10 μg/kg[a]	Reg. (EC) 1881/2006
Aflatoxin G_1	Spices: 10 μg/kg[a]	Reg. (EC) 1881/2006
Aflatoxin G_2	Spices: 10 μg/kg[a]	Reg. (EC) 1881/2006
Arsenic (inorganic)	No ML in spices and herbs	Reg. (EC) 1881/2006
Bifenthrin	Sweet peppers/bell peppers: 0.5 mg/kg	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 0.1 mg/kg^{2}	
	Nutmeg: 0.1 mg/kg^{2}	
	Basil and edible flowers: 0.05 mg/kg^{2}	
	Parsley: 0.05 mg/kg^{2}	
	Thyme: 0.05 mg/kg^{2}	
Cadmium	Spices: No MLs	Reg. (EC) 1881/2006
	Fresh herbs: 0.20 mg/kg (wet weight)	
Carbendazim	Sweet peppers/bell peppers: 0.1 mg/kg^{2}	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 0.1 mg/kg^{2}	
	Nutmeg: 0.1 mg/kg^{2}	
	Basil and edible flowers: 0.1 mg/kg^{2}	
	Parsley: 0.1 mg/kg^{2}	
	Thyme: 0.1 mg/kg^{2}	
Chlorpyrifos	Sweet peppers/bell peppers: 0.01 mg/kg^{2}	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 1.0 mg/kg	
	Nutmeg: 5.0 mg/kg	
	Basil and edible flowers: 0.05 mg/kg^{2}	
	Parsley: 0.05 mg/kg^{2}	
	Thyme: 0.05 mg/kg^{2}	
Cypermethrin	Sweet peppers/bell peppers: 0.5 mg/kg	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 0.1 mg/kg[b]	
	Nutmeg: 0.1 mg/kg[b]	
	Basil and edible flowers: 2.0 mg/kg	
	Parsley: 2.0 mg/kg	
	Thyme: 2.0mg/kg	
DDT	Sweet peppers/bell peppers: 0.05 mg/kg[b]	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 1.0 mg/kg	
	Nutmeg: 1.0 mg/kg	
	Basil and edible flowers: 0.05 mg/kg[b]	
	Parsley: 0.05 mg/kg[b]	
	Thyme: 0.05 mg/kg[b]	
Dimethoate	Sweet peppers/bell peppers: 0.02 mg/kg[b]	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 0.5 mg/kg	
	Nutmeg: 5.0 mg/kg	
	Basil and edible flowers: 0.02 mg/kg[b]	
	Parsley: 0.02 mg/kg[b]	
	Thyme: 0.02 mg/kg[b]	
Dioxins and dl-PCBs	No MLs in spices and herbs	Reg. (EC) 1881/2006
Endosulfan	Sweet peppers/bell peppers: 0.05 mg/kg[b]	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 5.0 mg/kg	
	Nutmeg: 1.0 mg/kg	
	Basil and edible flowers: 0.05 mg/kg[b]	
	Parsley: 0.05 mg/kg[b]	
	Thyme: 0.05 mg/kg[b]	
Estragole	No MLs in spices and herbs	Reg. (EC) 1334/2008
Ethion	Sweet peppers/bell peppers: 0.01 mg/kg[b]	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 5.0 mg/kg	
	Nutmeg: 3.0 mg/kg	
	Basil and edible flowers: 0.01 mg/kg[b]	
	Parsley: 0.01 mg/kg[b]	
	Thyme: 0.01 mg/kg[b]	
Fumonisin B_1	No MLs in spices and herbs	Reg. (EC) 1881/2006
Lead	No MLs in spices and herbs	Reg. (EC) 1881/2006
Mercury	Sweet peppers/bell peppers: 0.01 mg/kg^{2}	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 0.02[b] mg/kg^{2}	
	Nutmeg: 0.02 mg/kg^{2}	
	Basil and edible flowers: 0.01 mg/kg^{2}	
	Parsley: 0.01 mg/kg^{2}	
	Thyme: 0.01 mg/kg^{2}	
Metalaxyl	Sweet peppers/bell peppers: 0.5 mg/kg	Reg. (EC) 396/2005
	Peppercorn (black, green, and white): 0.1 mg/kg[b]	
	Nutmeg: 0.1 mg/kg[b]	
	Basil and edible flowers: 2.0 mg/kg	
	Parsley: 2.0 mg/kg	
	Thyme: 2.0 mg/kg	
Methyleugenol	No MLs in spices and herbs	Reg. (EC) 1334/2008
Myristicin	No MLs in spices and herbs	Reg. (EC) 1334/2008
Ochratoxin A		Reg. (EC) 1881/2006

Table 3 (*continued*)

Compound	ML or MRL (if available)	Legislation
	Capsicum spp. (dried fruits thereof, whole or ground, including chillies, chilli powder, cayenne and paprika): 20 μg/kg *Piper* spp. (fruits thereof, including white and black pepper): 15 μg/kg *Myristica fragrans* (nutmeg): 15 μg/kg Mixtures of spices containing one of the outlined spices (see legislation): 15 μg/kg	
PAH	Dried spices and herbs: Benzo(a)pyrene: 10 μg/kg Sum of benzo(a)- pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene: 50 μg/kg	Reg. (EC) 1881/2006
Para Red	Unauthorised substance	Reg. (EU) 1129/2011
Profenofos	Sweet peppers/bell peppers: 0.01 mg/kg[b] Peppercorn (black, green, and white): 0.07 mg/kg[b] Nutmeg: 0.05 mg/kg[b] Basil and edible flowers: 0.05 mg/kg Parsley: 0.05 mg/kg Thyme: 0.05 mg/kg	Reg. (EC) 396/2005
Propamocarb	Sweet peppers/bell peppers: 3.0 mg/kg Peppercorn (black, green, and white): 0.05 mg/kg[b] Nutmeg: 0.05 mg/kg[b] Basil and edible flowers: 30.0 mg/kg Parsley: 30.0 mg/kg Thyme: 30.0 mg/kg	Reg. (EC) 396/2005
Safrole	No MLs in spices and herbs	REG. (EC) 1334/2008
Sterigmatocystin	No legislation in spices and herbs	
Sudan I	Unauthorised substance	Reg. (EU) 1129/2011
Sudan III	Unauthorised substance	Reg. (EU) 1129/2011
Sudan IV	Unauthorised substance	Reg. (EU) 1129/2011
Tefluthrin	Sweet peppers/bell peppers: 0.05 mg/kg Peppercorn (black, green, and white): 0.05 mg/kg Nutmeg: 0.05 mg/kg Basil and edible flowers: 0.05 mg/kg Parsley: 0.05 mg/kg Thyme: 0.05 mg/kg	Reg. (EC) 396/2005
Trans-anethole	No legislation in spices and herbs	
Triazophos	Sweet peppers/bell peppers: 0.01 mg/kg[b] Peppercorn (black, green, and white): 0.07 mg/kg[b] Nutmeg: 0.02 mg/kg[b] Basil and edible flowers: 0.01 mg/kg[b] Parsley: 0.01 mg/kg[b] Thyme: 0.01 mg/kg[b]	Reg. (EC) 396/2005
Trifluralin	Sweet peppers/bell peppers: 0.01 mg/kg[b] Peppercorn (black, green, and white): 0.05 mg/kg[b] Nutmeg: 0.05 mg/kg[b] Basil and edible flowers: 0.02 mg/kg[b] Parsley: 0.02 mg/kg[b] Thyme: 0.02 mg/kg[b]	Reg. (EC) 396/2005

[a] Sum of aflatoxin B_1, B_2, G_1 and G_2.
[b] Indicates lower limit of analytical determination.

toxicity due to their adverse effects on several organ systems (EC, 2001). As there was only 1 RASFF notification and there were limited monitoring data, their probability of occurrence in the selected spices and herbs was classified as high. PAHs were classified as severe hazards due to the carcinogenic properties of benzoapyrene (IARC, 2015). Due to a lack of information on the presence of these compounds in spices and herbs, their probability was, by default, classified as high. Similarly, the probability of the plant toxins was classified as high. Both estragole and myristicin were classified in the medium priority group due to a high score for severity: estragole is probably carcinogenic (EMA, 2014) and for myristicin there is no evidence base on which to make the assessment.

The low priority group (green in Fig. 1) includes other plant toxins: methyleugenol, safrole and trans-anethole, pesticides: bifenthrin, DDT, metalaxyl, profenofos, propamocarb, tefluthrin and trifluralin as well as the mycotoxin sterigmatocystin. Some of these compounds have a high probability of occurrence (metalaxyl and profenofos), but have a low impact on human health. Other compounds may have severe effects on human health (tefluthrin), but have a low probability of occurrence in the selected spices and herbs.

4. Discussion

A risk matrix method was used to prioritize chemical hazards in the selected spices and herbs based on their characteristics and the data availability. The advantage of a risk matrix is that the methodology is easy to communicate and comprehend as the outcome can be visualised using an attractive colour grid ranging from low risks (green) to medium (yellow) and high risks. This facilitates the interpretation of the effect and exposure of the hazards. The disadvantage of a risk matrix is that it is a qualitative method, which is less accurate than semi-quantitative methods, such as a

Table 4
Results of the toxicological classification.

Compound	Acute toxicity		Chronic toxicity		Carcinogenicity		Overall classification
	ARfD (μg/kg bw/day)	Score	ADI/TDI (μg/kg bw/day)	Score	Group	Score	
Aflatoxin B_1	–	–	–	–	1[1]	Severe	Severe
Aflatoxin B_2	–	–	–	–	1[1]	Severe	Severe
Aflatoxin G_1	–	–	–	–	1[1]	Severe	Severe
Aflatoxin G_2	–	–	–	–	1[1]	Severe	Severe
Arsenic (inorganic)	–	–	–	–	1[1]	Severe	Severe
Bifenthrin	30[2]	High	15[2]	Medium	C[3]	Medium	High
Cadmium	–	–	TWI:2.5[4]	Severe	1[1]	Severe	Severe
Carbendazim	20[2]	High	20[2]	Medium	C[3]	Medium	High
Chlorpyrifos	5[2]	Severe	1[2]	High	E[3]	Low	Severe
Cypermethrin	200[2]	Low	50[2]	Low	C[3]	Medium	Medium
DDT (4,4′-Dichlorodiphenyltrichloroethane)	–	–	10[2]	Medium	2A[1]	High	High
Dimethoate	10[2]	High	1[2]	High	C[3]	Medium	High
Dioxins and dl-PCBs	–	–	TWI: 14 TEQ pg/kg bw/week[5]	Severe	3 (1 for 2378-TCDD)	Low/Severe	Severe
Endosulfan	20[2]	High	6[2]	High	E[3]	Low	High
Estragole	–	–	500[6]	Low	Probably carcinogenic[6]	High	High
Ethion	–	–	2[2]	High	E[3]	Low	High
Fumonisin B_1	–	–	PMTDI:2[7]	High	2B[1]	Medium	High
Lead	–	–	–	–	2B (2A for inorganic lead)[1]	Medium/High	High
Mercury			TWI: 4[8]	Severe	3[1]	Low	Severe
Metalaxyl	500[2]	Low	80[2]	Low	E[3]	Low	Low
Methyl eugenol			1000[9]	Low	2B[1]	Medium	Medium
Myristicin	Appears to be low[10]	Low	–	–	No information on carcinogenic potential[10]	High	High
Ochratoxin A	–	–	TWI: 0.12[11]	Severe	2B[1]	Medium	Severe
Para Red	–	–	–	–	Possibly carcinogenic, but there is a lack of data [12]	High	High
Polycyclic aromatic hydrocarbons (benzo[a]pyrene)	–	–	–	–	1[1]	Severe	Severe
Profenofos	1000[2]	Low	30[2]	Low	E[3]	Low	Low
Propamocarb	1000[2]	Low	290[2]	Low	E[3]	Low	Low
Safrole	–	–	–	–	2B[1]	Medium	Medium
Sterigmatocystin	–	–	–	–	2B[1]	Medium	Medium
Sudan I	–	–	–	–	Carcinogenic[12]	Severe	Severe
Sudan III	–	–	–	–	Possibly carcinogenic, but there is a lack of data [12]	High	High
Sudan IV	–	–	–	–	Possibly carcinogenic, but there is a lack of data [12]	High	High
Tefluthrin	5[2]	Severe	5[2]	High	E[3]	Low	Severe
Trans-anethole	2000[13]	Low	–	–	not indicative of any significant risk to human health[14]	Low	Low
Triazophos	1[2]	Severe	1[2]	High	Not carcinogenic[15]	Low	Severe
Trifluralin			15[2]	Medium	C[3]	Medium	Medium

–: data not available or not applicable.
References: [1](IARC, 2015); [2](EC, 2015); [3](US EPA, 2014); [4](EFSA, 2009a); [5](EC, 2001); [6](EMA, 2014); [7](WHO., 2011); [10](EFSA, 2010); [8](EFSA, 2012b); [9](EFSA, 2012a); [10](Hallström & Thuvander, 1997); [11](EFSA, 2006); [12](EFSA, 2005); [13](IPCS, 1999); [14](Truhaut et al., 1989); [15](IPCS, 2002).

scoring method (van der Fels-Klerx et al., 2015). Various authors have used scoring methods to prioritize hazards using multiple factors for severity and probability that are combined to obtain a final score (Bietlot & Kolakowski, 2012; Taxell et al., 2013; van Asselt et al., 2013; VRC, 2015). The UK Veterinary Residue Committee, for example, uses hazard (A), potency (B), diet (C), Use (D), exposure (E) and Residue (F) into a formula $(A + B) \times (C + D + E + F)$ to obtain a table with numeral outputs to classify risks (VRC, 2015). As these methods comprise various factors for which information needs to be obtained, it is more laborious than qualitative methods such as risk matrices or decision trees. As indicated by van der Fels-Klerx et al. (2016), the most appropriate risk ranking method depends on the available time, budget, the risk manager's preferences and the available data.

In order to make the risk rankings as objective as possible, the nominal categories for severity and probability were linked to quantifiable references as recommended by Duijm (2015). Therefore, classification of toxic effects was based on available toxicological reference levels as well as information on carcinogenicity rather than on a description of the toxic effects. Furthermore, probability was based on historical data from available monitoring data and RASFF notifications and followed the classification as has been developed by the UK Veterinary Residue Committee (VRC, 2015). Additionally, a criterion on the potential for economic adulteration was included analogous to Hanlon et al. (2015). Their classification was made more concrete by including known examples of economic adulteration in spices and herbs or in other food products and the approved status of use within the EU. Nevertheless, a risk matrix also includes subjective elements such as the establishment of thresholds for the classification of low, medium, high and severe scores for severity and probability as well as the establishment of the colour grid within a risk matrix (Cox, 2008). As recommended by Hong (2012), we used a three colour grid rather than a four colour grid, since an extra colour (indicating for

Table 5
Results of the classification on likelihood of occurrence.

Compound	Potential for economic adulteration		Historical data demonstrating presence of compounds		Overall classification
	Rationale	Score	Rationale	Score	
Aflatoxin B_1	Unintentionally present, environmental contaminants[1]	Low	• RASFF: >2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > ML in available monitoring programmes	Severe	Severe
Aflatoxin B_2	Unintentionally present, environmental contaminants[1]	Low	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > ML in available monitoring programmes	Severe	Severe
Aflatoxin G_1	Unintentionally present, environmental contaminants[1]	Low	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > ML in available monitoring programmes	Severe	Severe
Aflatoxin G_2	Unintentionally present, environmental contaminants[1]	Low	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > ML in available monitoring programmes	Severe	Severe
Arsenic (inorganic)	Unintentionally present, environmental contaminants[2]	Low	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: limited testing so no evidence on which to base the assessment	High	High
Bifenthrin	Intentionally used, agricultural chemical, approved in the EU[3]	Low	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: detected but at concentrations < MRL	Medium	Medium
Cadmium	Unintentionally present, environmental contaminants[4]	Low	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: detected > ML in <1% of the samples in the available monitoring programmes	High	High
Carbendazim	Intentionally used, agricultural chemical, not approved in the EU[3]	Medium	• RASFF: >2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > MRL in the available monitoring programmes	Severe	Severe
Chlorpyrifos	Intentionally used, agricultural chemical, approved in the EU[3]	Low	• RASFF: >2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > MRL in the available monitoring programmes	Severe	Severe
Cypermethrin	Intentionally used, agricultural chemical, approved in the EU[3]	Low	• RASFF: 1 notification between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > MRL in the available monitoring programmes	Severe	Severe
DDT (4,4′-Dichlorodiphenyltrichloroethane)	Intentionally used, agricultural chemical, not approved in the EU[3]	Medium	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: detected but at concentrations < MRL	Medium	Medium
Dimethoate	Intentionally used, agricultural chemical, approved in the EU[3]	Low	• RASFF: >2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > MRL in the available monitoring programmes	Severe	Severe
Dioxins and dl-PCBs	Unintentionally present, environmental contaminants with known examples of adulteration in other foods[5,6]	High	• RASFF: 1 notification in basil between 2004 and 2014 • Monitoring data: limited testing so no evidence on which to base the assessment	High	High
Endosulfan	Intentionally used, agricultural chemical, not approved in the EU[3]	Medium	• RASFF: 1 notification in pepper between 2004 and 2014 • Monitoring data: >1% of samples > MRL in the available monitoring programmes	Severe	Severe
Estragole	Unintentionally present, environmental contaminants[7]	Low	• RASFF: no notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: no testing so no evidence on which to base the assessment	High	High
Ethion	Intentionally used, agricultural chemical, not approved in the EU[3]	Medium	• RASFF: >2 notifications in chilli (pepper) between 2004 and 2014 • Monitoring data: >1% of samples > MRL in the available monitoring programmes	Severe	Severe
Fumonisin B_1	Unintentionally present, environmental contaminants[1]	Low	• RASFF: no notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: limited testing so no evidence on which to base the assessment	High	High
Lead	Unintentionally present, environmental contaminants[8]	Low	• RASFF: 1 notification in hot pepper between 2004 and 2014 • Monitoring data: detected in >1% of samples, but no ML specified so no evidence on which to base the assessment	High	High

(continued on next page)

Table 5 (*continued*)

Compound	Potential for economic adulteration		Historical data demonstrating presence of compounds		Overall classification
	Rationale	Score	Rationale	Score	
Mercury	Unintentionally present, environmental contaminants[9]	Low	• RASFF: 1 notification in nutmeg between 2004 and 2014 • Monitoring data: limited testing	High	High
Metalaxyl	Intentionally used, agricultural chemical, approved in the EU[3]	Low	• RASFF: 1 notification in basil between 2004 and 2014 • Monitoring data: >1% of samples > ML in the available monitoring programmes	Severe	Severe
Methyleugenol	Unintentionally present, environmental contaminants[7]	Low	• RASFF: no notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: no testing so no evidence on which to base the assessment	High	High
Myristicin	Unintentionally present, environmental contaminants[7]	Low	• RASFF: no notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: no testing so no evidence on which to base the assessment	High	High
Ochratoxin A	Unintentionally present, environmental contaminants[1]	Low	• RASFF: >2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > ML in the available monitoring programmes	Severe	Severe
Para Red	Intentionally used, prohibited compounds with known examples of adulteration in spices and herbs[10]	Severe	• RASFF: >2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: no testing so no evidence on which to base the assessment	Severe	Severe
Polycyclic aromatic hydrocarbons (benzo[a]pyrene)	Unintentionally present, environmental contaminants with known examples of adulteration in other foods[11,12]	High	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: no testing so no evidence on which to base the assessment	High	High
Profenofos	Intentionally used, agricultural chemical, not approved in the EU[3]	Medium	• RASFF: 3 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: > 1% of samples > MRL in the available monitoring programmes	Severe	Severe
Propamocarb	Intentionally used, agricultural chemical, approved in the EU[3]	Low	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: detected > ML in <1% of the samples in the available monitoring programmes	High	High
Safrole	Unintentionally present, environmental contaminants[7]	Low	• RASFF: no notifications between 2004 and 2014 • Monitoring data: no testing so no evidence on which to base the assessment	High	High
Sterigmatocystin	Unintentionally present, environmental contaminants[1]	Low	• RASFF: no notifications between 2004 and 2014 • Monitoring data: no testing so no evidence on which to base the assessment	High	High
Sudan I	Intentionally used, prohibited compounds with known examples of adulteration in spices and herbs[10]	Severe	• RASFF: >2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: no testing so no evidence on which to base the assessment	Severe	Severe
Sudan III	Intentionally used, prohibited compounds with known examples of adulteration in spices and herbs[10]	Severe	• RASFF: > 2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: no testing so no evidence on which to base the assessment	Severe	Severe
Sudan IV	Intentionally used, prohibited compounds with known examples of adulteration in spices and herbs[10]	Severe	• RASFF: > 2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: no testing so no evidence on which to base the assessment	Severe	Severe
Tefluthrin	Intentionally used, agricultural chemical, approved in the EU[3]	Low	• RASFF: no notifications between 2004 and 2014 • Monitoring data: not detected in >100 samples	Low	Low
Trans-anethole	Unintentionally present, environmental contaminants[7]	Low	• RASFF: no notifications between 2004 and 2014 • Monitoring data: no testing so no evidence on which to base the assessment	High	High
Triazophos	Intentionally used, agricultural chemical, not approved in the EU[3]	Medium	• RASFF: >2 notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: >1% of samples > MRL in the available monitoring programmes	Severe	Severe
Trifluralin	Intentionally used, agricultural chemical, not approved in the EU[3]	Medium	• RASFF: No notifications between 2004 and 2014 in specified spices and herbs • Monitoring data: detected > ML in <1% of the samples in the available monitoring programmes	High	High

References: [1](Zain, 2011); [2](EFSA, 2009b); [3](EC, 2015); [4](EFSA, 2009a); [5](Hoogenboom et al., 2010); [6](Cheftel, 2011); [7](Williams & Mattia, 2009); [8](EFSA, 2010); [9](EFSA, 2012b); [10](Johnson, 2014); [11](EFSA, 2008); [12](Guillén & Sopelana, 2004).

PROBABILITY \ SEVERITY	LOW	MEDIUM	HIGH	SEVERE
SEVERE	Metalaxyl Profenofos	Cypermethrin	Carbendazim Dimethoate Endosulfan Ethion Para Red Sudan III Sudan IV	Aflatoxins Chlorpyrifos Ochratoxin A Sudan I Triazophos
HIGH	Trans-anethole Propamocarb	Methyleugenol Safrole Sterigmatocystin Trifluralin	Estragole Fumonisin B_1 Lead Myristicin	Arsenic (inorganic) Cadmium Mercury Dioxins & dl-PCBs PAHs
MEDIUM			Bifenthrin DDT	
LOW				Tefluthrin

Fig. 1. Risk matrix of chemical hazards in spices and herbs with red: high risk, yellow: medium risk and green: low risk. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

example medium-low risk) does not improve the performance of the risk matrix. According to Hong (2012), various colouring schemes are possible within a 4 × 4 risk matrix that fulfil the three axioms presented by Cox (2008): ‘weak consistency’, meaning that each hazard in the red category represents a larger risk than those in the green category; ‘betweenness’, meaning that red and green categories cannot be adjacent to each other and ‘consistent colouring’, meaning that hazards with (approximately) equal risk should have the same colour. We used the colouring scheme as presented by Hanlon et al. (2015), which fulfils these requirements. In this scheme, only 1 grid cell is identified as a high risk, coloured in red, which allows for prioritization of the most risky compounds that should be included in monitoring programs.

For the selected spices and herbs, aflatoxins, ochratoxin A, chlorpyrifos, triazophos and Sudan I were categorized in this top right corner. The latter is the result of fraudulent actions, which were notified in RASFF, primarily in 2004 and 2005. Since then, the number of notifications on the Sudan dyes and Para Red has decreased substantially probably due to the increased monitoring of these compounds in spices. The probability of occurrence, based on historical data, has thus decreased over the years. Nevertheless, as the potential for economic adulteration is included as a criterion for exposure, the dyes are expected to remain a top priority for the coming decade. Aflatoxins and ochratoxin A have severe toxic effects and were frequently encountered in monitoring programs and in the RASFF database. Therefore, they should have an increased monitoring frequency in national monitoring programs for spices and herbs compared to other chemical compounds. Especially since their presence is expected to be affected by climate change (Paterson & Lima, 2010). Only two pesticides, chlorpyrifos and triazophos, were seen as high priority compounds due to their severe acute toxic effects (low ARfD) and frequent probability of occurrence (historical data) in the selected spices and herbs. Pesticides are agricultural chemicals that are intentionally used in order to prevent crop damage due to fungi, insects etc. Some pesticides are not approved for use within the EU and, thus, should not be present above the limit of detection (LOD) in spices and herbs. This is the case for triazophos, which is encountered frequently in spices and herbs from India, Thailand and Brazil according to the RASFF database. Pesticides that are authorised for use in the EU need to comply with the specified MRLs (Regulation (EC) 396/2005). This means that pesticides may be used provided that Global Agricultural Practices (GAP) are followed. GAP indicates that prescribed withdrawal periods need to be applied in order to ensure that levels in the final products are below the MRLs. Although chlorpyrifos has been authorised in the EU, it has frequently been found in levels above the MRL in spices and herbs.

The medium priority group contains some compounds for which there was limited information to classify severity or probability. For example, there was a lack of data on the toxicological characteristics of the dyes Sudan III, IV and Para Red and the plant toxins estragole and myristicin, for which the carcinogenic potential is unclear. Plant toxins are naturally present in spices and herbs. For example, nutmeg contains myristicin and safrole and basil contains estragole (Williams & Mattia, 2009). Regulation (EC) 1334/2008 indicates that plant toxins such as methyleugenol and safrole cannot be added as such to food, but they may be present when they are naturally present in ingredients used as flavouring compounds. Further research is needed about the presence of plant toxins, their toxicological characteristics as well as their possible human health impact as spices and herbs are the predominant food products contributing to the exposure of these compounds (Williams & Mattia, 2009). Due to a lack of historical monitoring data on the presence of plant toxins in the selected spices and herbs, their probability of occurrence was also classified as high. Other compounds for which there was limited historical data available were dioxins and dl-PCBs, fumonisin B_1, sterigmatocystin, inorganic arsenic, mercury and PAHs. As a result, they were classified with a high score for probability. This precautionary approach might mean an over-classification of these compounds. In order to obtain a more realistic classification, more information should be gathered for these compounds. Furthermore, lead was classified with a high score on probability as this compound was detected in the selected spices and herbs, but no ML has been specified. This reveals that the systematic approach followed in this paper not only enabled the identification of the most risky compounds, but also revealed data gaps that need further attention. The compounds with limited historical data in most cases also had no ML, which is probably the reason why they are not incorporated in monitoring programs. Nevertheless, these compounds may be present in spices and herbs and, therefore, need further attention. A way forward would be to collect monitoring data for these compounds and provide these data to an international food safety authority, such as EFSA, in order to derive an ML based on these data and available ADIs/TDIs.

The prioritization method applied in this paper allows for classifying chemical compounds into low, medium and high frequency of monitoring. This helps to prioritize monitoring programs enabling an efficient allocation of monitoring budgets focusing on the high risk compounds. For spices and herbs, the mycotoxins aflatoxins and ochratoxin A should be included in monitoring programs, as well as the pesticides chlorpyrifos and triazophos and the dye Sudan I.

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