Review

Dietary exposure assessment to mycotoxins through total diet studies. A review

Dionisia Carballo[a], Josefa Tolosa[b], Emilia Ferrer[b,*], Houda Berrada[b]

[a] Faculty of Agricultural Science, National University of Asunción, Paraguay
[b] Laboratory of Food Chemistry and Toxicology, Faculty of Pharmacy, University of Valencia, Avenue Vicent Andrés Estellés s/n, 46100, Burjassot, Spain

ARTICLE INFO

ABSTRACT

Mycotoxins are secondary metabolites of fungi that contaminate food in several stages and their increasing presence in food chain demand further control. Assessment of mycotoxins human exposure through processed diet is an important component of food safety strategies. The present review explores and summarises total diet studies (TDS) carried out in different countries focusing on mycotoxins determination. TDS were classified by samples preparation, mycotoxins analysis and dietary exposure evaluation. Most of reviewed TDS performed multi-mycotoxins analysis in composite samples mainly, prepared taking into account local culinary habits. High performance liquid chromatography coupled with fluorescence detector was the predominant and the most sensitive technique used for determination. Ochratoxin A was the most analyzed mycotoxin, followed by trichothecenes, aflatoxins, zearalenone, fumonisins, patulin, enniatins, and beauvericin respectively. *Alternaria* toxins and ergot alkaloids were also included. Food commonly analyzed were cereals, meat, vegetables, fruits, nuts and beverages. The findings in food were in below the current European legislation, except for some sporadic samples of wine and milk meaning less than 1% of total analyzed samples. Dietary exposure was evaluated, through the estimated daily intake mycotoxin evaluation and risk assessment concluded that relatively scarce toxicological concern was associated to mycotoxins exposure. However, a special attention should be paid to meat and cereal products high percentile consumers.

1. Introduction

In the last years, food safety policies based on scientific evidence are being implemented in food industries to protect consumer's interest (Lee et al., 2015). The European Food Safety Authority (EFSA) through the Council and European Parliament established several scientific advice and technical support in all areas impacting on food safety sharing information, data and best practices, for emerging risks identification and coherent communication development (European Commission, 178/2002; EFSA, 2009). The monitoring or surveillance data often focus on individual chemical substances in raw food commodities and don't often provide a direct assessment of dietary population exposure. There is scarce data in literature behaving toward ready-to-eat foods analysis, leading sometimes to an over-estimation of the amount of contaminants presents in food chain (EFSA, FAO & WHO, 2011). In this sense, De Nijs et al. (2016) suggested food consumption surveys, food monitoring studies, duplicate diet studies and total diet studies (TDS) as useful strategies to be used for mycotoxin exposure estimation.

Many countries regularly conduct TDS to evaluate population exposure to different food contaminants. (Moy and Vannoort, 2013; Betsy et al., 2012; Lee et al., 2015). In this sense, international committees EFSA, FAO & WHO (2011) published harmonized guidance document to improve dietary exposure assessment's efficiency and accuracy. Selection of representative food of the overall diet and culinary preparation of ready-to-eat dishes are critical steps in TDS implementation which very often combined analytical results with food consumption data for relatively accurate estimation of dietary exposure to such contaminants. The wide varieties of chemical substances present in diet make the establishment of priorities in TDS approach necessary. In particular, for mycotoxins, analysis is recommended in selected food items (Fig. 1) (WHO, 2002; WHO, 2015).

Mycotoxins are compounds produced by fungi growing mostly on plant products during production and storage. The Rapid Alert System for Food and Feed of the European Union reports mycotoxins on the second position according to the total number of hazard notifications (RASFF, 2017). The most important mycotoxins in food and animal feed are: aflatoxins (AFs), produced by *Aspergillus* species; Ochratoxin
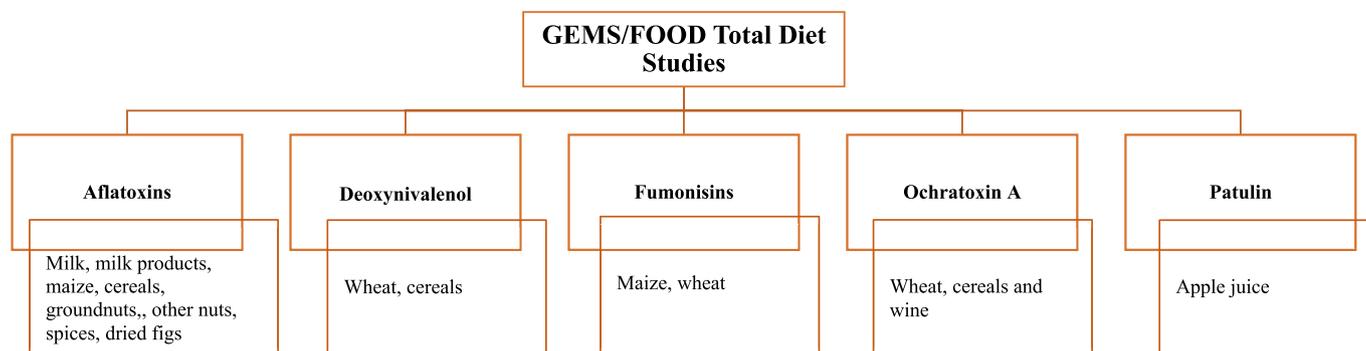
Fig. 1. Mycotoxins analysis suggested in the main food in TDS workshop (WHO, 2002; WHO, 2015).

A(OTA), produced by both *Aspergillus* and *Penicillium*; trichothecenes (TCTs) [HT-2, T2 toxin and deoxynivalenol (DON)], nivalenol (NIV), zearalenone (ZEA), fumonisins (FBs). Emerging mycotoxins such as fusaproliferin (FUS), moniliformin (MON), beauvericin (BEA), and enniatins (ENNs), altenuene (ALT), alternariol (AOH), alternariol methyl ether (AME), altertoxin (ALTs), and tenuazonic acid (TeA) produced mainly by *Fusarium* and *Aternaria* toxins (Kralj and Prosen, 2009; Yang et al., 2014; Marin et al., 2013).

The presence of mycotoxins has been highly investigated in different foodstuff such as cereals products (Juan et al., 2017; Saladino et al., 2017), coffee and tea beverages (Pallarés et al., 2017; García-Moraleja et al., 2015), vegetables (Dong et al., 2019; Rodríguez-Carrasco et al., 2016) and some studies have also been carried out in cooked food (Sakuma et al., 2013; Carballo et al., 2018).

Table 1 summarises mycotoxins subject of study in foodstuffs extracted from different reviewed TDS. Mycotoxin's exposure evaluation through TDS have been conducted in several countries; even countries in process of development such as Benin, Cameroon, Mali and Nigeria and some countries have conducted several TDSs like France, Spain and New Zealand (Ingenbleek et al., 2017). The number of mycotoxins varies from one mycotoxin (Tam et al., 2011) to 38 (López et al., 2016).

This review offers for the first time a compilation data about mycotoxin occurrence and dietary exposure assessment through TDS studies. Also it features an overview of the different methodologies reported on mycotoxin analysis in food, published in the last two decades, focusing on samples preparation, analytical techniques used and estimation of the potential contribution to the dietary exposure.

2. Methodology

A systematic literature review was conducted using the databases Web of Science, PubMed and Scopus with the focus on the following keywords: total diet studies, mycotoxin, occurrence, dietary exposure etc. The period of time framed was of last two decades. Eighteen articles, which met the criteria to be included into the study, were analyzed and classified. To facilitate data presentation four groups were established based on selection and collection of samples, samples preparation, analytical determination techniques and dietary exposure assessment. The information was double-checked to select bibliographies of relevant literature and summarize the information about, analytical methodology, studied mycotoxins, limits of detection and quantitation, incidence and concentration levels of mycotoxin. Finally, data available was used to estimate exposure dietary to mycotoxins.

3. Results and discussion

3.1. Selection and collection of samples

Several tools were applied to obtain population consumption data, such as food frequency questionnaires, 24-h recalls, and family budget surveys (Kim et al., 2015). Dietary intake data from national surveys was one of the most frequently used on TDS. Some countries disposed of detailed information available from national surveys for individual consumption, like France (Leblanc et al., 2005; Sirot et al., 2013), Ireland (Food Safety Authority of Ireland) (FSAI, 2016) and New Zealand (Australian National Nutrition Survey) (Food Standard Authority New Zealand, 2011).

In this sense, the number of collection sites is usually determined first within budget limit and the duration of the TDS is sometimes based on population size (national or regional/local), distance between sites and geographical conditions (Kim et al., 2015). Food is often collected three to four times a year and randomly from different locations (Betsy et al., 2012) and during various seasons of year (Elegbede et al., 2017).

In general, samples were analyzed in form of individual food, composite samples and food groups samples (Leblanc et al., 2005; Sirot et al., 2013; Raad et al., 2014; Huong et al., 2016). Collected samples were from fast-food chain to supermarkets, representing typical products mostly consumed (Tam et al., 2011; López et al., 2016). The different food categories and the number of samples collected in reviewed TDS are listed in Table 1.

3.2. Samples preparation

As a wide variety of food is collected for TDS conduction, some samples require preparation such as peeling (e.g. orange or banana) and cooking for example. (e. g. beef and chicken) (Abbey and Mooney, 2013). Depending on TDS's approaches, namely the composite or individual food approach, steps for food preparation, as normally consumed in home, are recommended (Vannoort et al., 2013). The accuracy and comprehensiveness of the manual procedure is critical as it influences the manner in which food samples are handled, prepared and stored before analysis. These decisions can influence the quality and representativeness nature of the overall TDS (Abbey and Mooney, 2013).

3.3. Analysis of mycotoxins

3.3.1. Analytical methods

Instrumental determinations driven on TDS samples are largely characterized by their selectivity and sensitivity to meet the TDS challenge for measuring the trace levels of chemical residues in the most varied food matrices (Sack, 2013).

A list of the sensitivity of the methods and the instrumentation required was already provided (WHO, 2015) even a verification of the capabilities to replicate the key method's characteristics, including precision, limit of detection (LOD), limit of quantification (LOQ) and specificity, as well as ongoing monitoring of the accuracy during use is recommended.

Table 2 summarises the analytical methods used as well as LOD and LOQ reached for the reported mycotoxins. Liquid chromatography (LC)

Table 1
Total diet studies revised for mycotoxin analysis.

Country	Analyzed food	Composite (n)	Individual food (n)	Analyzed mycotoxins	Reference
Canada	Cereal and cereal products; alcohol drinks; coffee; tea; beans; fruits; sugars; chocolate; cheese; milk; eggs; dessert; meat; herb and spices; dried fruits; soya products; mixed dishes	140	-	OTA	Tam et al. (2011)
China	Cereal and their products; vegetables; legumes, nuts, seeds; fruits; meat and poultry; fats, oils; alcoholic beverages; non-alcoholic beverages; mixed dishes; snacks; sugars; condiments, sauces	600	1800	AFB_1, AFB_2, AFG_1, AFG_2, PAT, ZEA, DON, FB_1, FB_2, FB_3, ZEA, α-ZOL, βZOL, DON, 3AcDON, 15AcDON	Yau et al. (2016)
China	Cereal products; beans; potatoes; meat; eggs; aquatic products; milk; vegetables; fruits; saccharides; beverages; condiments	240	-	STC, CIT, CPA, MON, GLIO, MPA, Verru	Qiu et al. (2017)
France	Vegetarians food; biscuits; breakfast cereals; breads; pasta; rice; cakes; chocolates; desserts; nuts and oilseeds; vegetables; pulses; eggs; sugars; breads, buns; butter; dairy products; coffee; meat; offal; fruits; soft drinks; alcoholic beverages; pizzas, salt cakes, quiches; sandwiches; soup; prepared dishes; salads; compotes	456	2280	AFB_1, AFB_2, AFG_1, AFG_2, AFM_1, OTA, PAT, ZEA, FB_1, FB_2, DON, NIV, 3AcDON, 15AcDON, T-2 and HT-2, NEO, FUS-X, DAS, MAS	Leblanc et al. (2005)
France	Breads; breakfast cereals; pasta; rice; croissants; pastries; biscuits; cakes; milk; dairy products; eggs; butter; offal; delicatessen meat; vegetables; fruits; dried fruits; nuts and seeds; chocolate; non-alcoholic beverages; alcoholic beverages; coffee; pizzas; sandwiches; snacks; mixed dishes; desserts; compotes	577	1319	AFB_1, AFB_2, AFG_1, AFG_2, AFM_1, FB_1, FB_2, OTA, PAT, ZEA, DON, NIV, 3AcDON, 15AcDON, T-2, HT-2	Sirot et al., 2013
Ireland	Cereals; milk; dairy products; eggs; meat; fish; potatoes; vegetables; fruits; dried fruits; nuts; seeds; herbs spices; soups; sauces; sugars; beverages; fats; oils; snacks; pizza	141	1043	AFB_1, AFB_2, AFG_1, AFG_2, AFM_1, OTA, PAT, ZEA, FB_1, FB_2, DON, DAS, NIV, 3AcDON, 15AcDON, T-2, HT-2	FSAI (2016)
Lebanon	Breads and toasts; biscuits; croissants; cakes; pastries; pasta; pizza; pies; rice; pulses; olive oils, sesame oils; nuts, seeds, olives and dried dates; cheese; milk and milk-based beverages; milk-based ice cream and pudding; yogurt and yogurt-based products; caffeinated beverages; alcoholic beverages	47	705	AFB_1, AFM_1, OTA, DON	Raad et al. (2014)
New Zealand	Alcoholic and non-alcoholic beverages; cereal and cereal products; condiments; dairy products; eggs; fats and oils; fish; fruits; meat; nuts; seeds; snacks; sugars; vegetables; infant food	48	-	AFB_1, AFB_2, AFG_1, AFG_2, AFM_1, AFM_2	FSANZ (2001)
New Zealand	Alcoholic non-alcoholic beverages; cereal and cereal products; condiments; dairy products; eggs; fats; oils; fish; seafood; fish products; fruits; meat products; nuts and seeds; snacks; sugars; vegetables; infant food	65	-	AFB_1, AFB_2, AFG_1, AFG_2, OTA	FSANZ (2003)
New Zealand	Alcoholic and non-alcoholic beverages; cereal products; condiments; dairy products; eggs; fats; oils; fish; fruits; meat; nuts; seeds; snacks; sugars; vegetables; infant food, beverages; fast food	570	-	AFB_1, AFB_2, AFG_1, AFG_2, AFM_1, OTA, PAT, ZEA, FB_1, FB_2, DON	FSANZ (2011)
Spain	Cereal and cereal products; milk; cheeses; dried fruits; sweet corns; breakfast cereals; corn snacks; alcoholic beverages; coffee; vegetables; baby food; apple juice; jams and apple sauce; ethnics food; gluten-free food	1690	3447	AFB_1, AFB_2, AFG_1, AFG_2, AFM_1, OTA, PAT, ZEA, FBs, DON, T-2, HT-2	Cano-Sancho et al. (2012)
Spain	Milk; dairy products	60		AFM_1	Urieta et al. (1991); Urieta et al. (1996)
Spain	Cereal and cereal products; olives; pickles; apple; pear; eggs; milk; milk shakes; custards; soya products; cheeses; grapes; alcoholic beverages; juices; oils	240	-	AFB_1, AFB_2, AFG_1, AFG_2, OTA, ZEN, FB_1, FB_2, DON, NIV, 3AcDON, 15AcDON, T-2, HT-2, T-2 triol, NEO, FUS-X, DAS	Beltrán et al. (2013)
The Netherlands	Alcoholic, non-alcoholic beverages; sugars; dairy products; eggs; fish; fruits; cereal products; legumes; meat; offal; nuts; seeds; oils; fats; soy products; tuber; vegetables	88	-	AFB_1, AFB_2, AFG_1, AFG_2, AFM_1, AOH, AME, BEA, ENN A, ENN A_1, ENN B, ENN B1, OTA, PAT, ZEA, α-ZOL, β-ZOL, STE, FB_1, FB_2, FB_3, DON, DON 3G, FUS-X, NEO, DAS, NIV, 3AcDON, 15AcDON, T-2, HT-2, MON, MPA, NPA, PeA, ROC,	López et al. (2016); Sprong et al. (2016a); Sprong et al. (2016b)
Viet Nam	Rice; wheat and products; tubes root and products; beans products; tofu; oily seeds; vegetables; sugars; seasoning; fats; oils; meat products; egg and milk; fish; other aquatic products	42	1134	AFB_1, FBs, OTA	Huong et al. (2016)

Table 2
Analytical methods, limit of detection (LOD) and quantification (LOQ).

Analytical technique	Mycotoxin	Foodstuffs	LOD (µg/kg)	LOQ (µg/kg)	Reference
HPLC-FD	AFs	Cereal products, dried fruit peppers, ethnic food	0.025–0.1	2.64	Cano-Sancho et al. (2012)
HPLC-FD	AFS	Chocolate	0.05	0.2	Sirot et al. (2013)
HPLC-FD	AFs	Cereal products, seeds, olives, dried dates	0.01	0.03	Raad et al. (2014)
HPLC-FD	AFM_1	Milk and milk based beverages	0.01	0.03	Raad et al. (2014)
HPLC-FD	AFM_1	Dairy products	–	0.025	Urieta et al. (1996)
HPLC-FD	OTA	Cereal products	0.25	1	Leblanc et al. (2005)
HPLC-FD	OTA	Cereal products, beer, dessert wine, coffee, peanuts, pistachios, baby food	0.003–1.160	–	Cano-Sancho et al. (2012)
HPLC-FD	OTA	Cereal products	0.01–0.06	0.05–0.025	Tam et al. (2011)
HPLC-FD	OTA	Cereal products, beverages	0.05	0.21	Raad et al. (2014)
HPLC-FD	ZEA	Cereal products, vegetables, nuts, oilseeds	0.25	1	Leblanc et al. (2005)
HPLC-FD	ZEA	Cereal products, beer, baby food, ethnics food	0.070–1.95	–	Cano-Sancho et al. (2012)
HPLC-FD	FBs	Corn flakes, corn snacks, baby food, gluten-free food, ethnics food	–	15.6	Cano-Sancho et al. (2012)
HPLC-FD	FBs	Beer	–	11.7	
HPLC-UV	AFs	Dried fruits, seeds	–	1	FSNAZ (2001)
HPLC-UV	OTA	Cereals, dairy products, eggs, vegetables, meat, fish, fruits, dried fruits, sugars, beverages	0.02–0.05	0.2	FSAI (2016)
HPLC-UV	FBs	Cereal products, vegetables, beverages, soft drinks, offal	10.0–20	20–40	Leblanc et al. (2005)
HPLC-UV	PAT	Fruits, alcoholic and non-alcoholic beverages	1.5–4	5	FSAI (2016)
HPLC-UV	DON	Cereals products	62.5	125	Raad et al. (2014)
HPLC-UV	PAT	Alcohol beverages, soft drinks	20	40	Leblanc et al. (2005)
HPLC-UV	PAT	Jams	–	6.25	Cano-Sancho et al. (2012)
UPLC-MS/MS	AFs	Cereal products, legumes, seeds, fats, oils	0.05	0.10	Yau et al. (2016)
UPLC-MS/MS	OTA	Cereals, legumes, seeds, meat, oil, mixed diches, sugars	0.05	0.10	Yau et al. (2016)
UPLC-MS/MS	OTA	Cereal products	0.09	0.29	Beltrán et al. (2013)
UPLC-MS/MS	ZEA	Cereals, meat, fats, oil, sugars, condiments	2.5	5	Yau et al. (2016)
UPLC-MS/MS	DON	Cereal products, non-alcoholic beverages, mixed diches, sugars	2.5	5	Yau et al. (2016)
UPLC-MS/MS	DON	Cereal products	0.7	2.4	Beltrán et al. (2013)
UPLC-MS/MS	FBs	Cereal products	2.5	5	Yau et al. (2016)
UPLC-MS/MS	FBs	Breakfast cereals	1.05	3.5	Beltrán et al. (2013)
UPLC-MS/MS	FBs	Beer	0.39	1.3	
LC-MS/MS	ZEA	Cereal products	1.5	5	Sirot et al. (2013)
LC-MS/MS	DON	Cereal products, mixed diches	3	10	Sirot et al. (2013)
LC-MS/MS	NIV	Rice, mixed diches	3	10	Sirot et al. (2013)
LC-MS/MS	HT-2	Pasta, dried fruits	3	10	Sirot et al. (2013)
LC-MS/MS	FBs	Breakfast cereals, biscuits	10	20	Sirot et al. (2013)
LC-MS/MS	PAT	Fruits, compotes	6	20	Sirot et al. (2013)
LC-MS/MS	OTA	Cereal products, meat	0.02	0.5	Sirot et al. (2013)
LC-MS/MS	OTA	Alcoholic beverages, coffee	0.01	0.05	Sirot et al. (2013)
LC-MS/MS	OTA	Beverages, sugars, milk, eggs, fish, fruits, cereal products, legumes, meat, offal, nuts, oils, soy products, tuber, vegetables	0.2	0.5	López et al. (2016)
ELISA	AFs	Cereals products, tubes, root, oils, vegetables, sugars, seasoning, meat, eggs and milk, fish, other aquatic products	2	–	Huong et al. (2016)
ELISA	AFM_1	Milk, yogurt	–	0.005–0.025	Cano-Sancho et al. (2012)
ELISA	OTA	Beans products, meat, eggs, milk, fish, other aquatic products	1.9	–	Huong et al. (2016)
HPLC-DAD	DON	Whole wheat breads, sweet corns, baby food, gluten-free food, ethnics food, beer	–	12.66	Cano-Sancho et al. (2012)
HPLC-DAD	T-2	Whole wheat breads, sweet corns	–	42–153	Cano-Sancho et al. (2012)
HPLC-DAD	HT-2	Whole wheat breads, sweet corns	–	9.0–61	Cano-Sancho et al. (2012)
GC-MS	DON	Cereal products, eggs, nuts oilseeds, prepared diches	15	60	Leblanc et al. (2005)
GC-MS	DON	Breads	–	12.66	Cano-Sancho et al. (2012)
GC-MS	NIV	Cereal products, nuts, oilseeds	30	60	Leblanc et al. (2005)
GC-ECD	DON	Pasta, corn snacks, corn flakes, wheat flakes	–	12.66	Cano-Sancho et al. (2012)
GC-ECD	T-2	Pasta, corn flakes, wheat flakes, corn snacks	–	42–153	Cano-Sancho et al. (2012)
GC-ECD	HT-2	Pasta, corn flakes, wheat flakes, corn snacks	–	9.0–61	Cano-Sancho et al. (2012)
GC-MS/MS	DON	Alcoholic beverages, cereal products, legumes, soy products, tuber	0.8	2.5	López et al. (2016)

and gas chromatography (GC) were widely used because of its superior performance and reliability, coupled to different detectors as fluorescence (FD), ultra-violet (UV), electron capture detector (ECD), diodearray (DAD), mass spectrometry (MS) and tandem spectrometry (MS/MS).

The most used technique for determination was HPLC-FD, achieving LODs between 0.003 and 1.95 µg kg^{-1} and LOQs of 0.03–15.6 µg kg^{-1} (Cano-Sancho et al., 2012), followed by HPLC-UV method with LODs between 0.02 and 62.5 µg kg^{-1} and LOQs 0.2–125 µg kg^{-1} (FSAI, 2016; Raad et al., 2014). UHPLC-MS/MS reached LODs from 0.05 to 2.5 µg kg^{-1} and LOQs 0.1–5 µg kg^{-1} (Yau et al., 2016; Beltrán et al., 2013) and LC-MS/MS LODs from 0.2 to 6 µg kg^{-1} and LOQs from 0.5 to

Table 3

Ochratoxin A: Occurrence and dietary exposure assessment.

Matrix	n	Incidence (%)	Mean (µg kg^{-1})	EDI (ng kg^{-1} bw day^{-1})	TDI (%)	Reference
Beans and products	108	50	9.68	0.2	1.176	Huong et al. (2016)
Meat and products	297	64	2.68	5	29.411	
Egg and milk	135	75	3.16	1.4	8.235	
Fish	54	50	2.24	1.4	8.235	
Other aquatic products	54	100	4.85	0.3	1.764	
Corn flakes	72	3	0.73	0.37–0.53	2.17–3.11	Cano-Sancho et al. (2012)
Wheat flakes	28	21	0.31			
Whole wheat breads	70	13	0.28			
Peanuts	72	42	0.21			
Pistachios	70	3	0.23			
Baby food	69	7	0.23			
Beer	71	89	0.02			
Dessert wine	141	57	2.85			
Red wine	120	15	0.51			
Coffee	72	49	2.17			
Bread and toast	45	–	0.29	0.59	3.47	Raad et al. (2014)
Biscuits and croissants	45	–	2.84	0.76	4.52	
Cakes and pastries	45	–	0.15	0.05	0.30	
Pizza and pies	45	–	0.223	0.15	0.88	
Rice	15	–	0.68	0.46	2.73	
Pasta and other cereal products	60	–	0.18	0.08	0.47	
Dried fruits	45	–	0.07	0.01	0.05	
Alcoholic beverages	15	–	1.472	0.64	3.80	
Caffeinated beverages	30	–	0.508	1.48	8.71	
Cereals and their products	76	38	0.07–0.10	1.3–5.4	7.3–31.7	Yau et al. (2016)
Legumes, nuts and seeds	24	20	0.05–0.09			
Meat and poultry	36	8	0.01–0.5			
Fats and oils	4	25	0.02–0.05			
Mixed dishes	44	11	0.01–0.06			
Sugars	4	100	0.22			
Cereal products	28	57	0.48	2.19[a]	12.90	Tam et al. (2011)
Breakfast cereals and bread	30	73	0.06–0.29	0.24–1.20[a]	1.4–7.05	Beltrán et al. (2013)
Bread rusk	18	39	0.43	0.71	4.176	Leblanc et al. (2005)
Breakfast cereals	12	17	0.29	0.02	0.117	
Biscuits	14	15	0.69	0.14	0.823	
Cakes	12	17	0.29	0.17	1	
Delicatessen meat	20	7	0.34	0.08	0.470	
Soft drinks	4	50	0.11	–	–	
Alcoholic beverages	18	28	0.04	0.05	0.294	
Coffee	14	7	0.04	0.08	0.470	
Breads	14	–	0.129–0.393	0.172–0.580	1.01–3.41	Sirot et al. (2013)
Breakfast cereals	6	–	0.033–0.25	0.003–0.014	0.01–0.08	
Pasta	4	–	0.1–0.35	0.006–0.120	0.03–0.70	
Rice and wheat products	6	–	0.067–0.3	0.007–0.084	0.04–0.47	
Delicatessen meat	80	–	0.047–0.253	0.017–0.099	0.1–0.58	
Alcoholic beverages	6	–	0.017–0.025	0.065–0.077	0.38–0.45	
Cereal products	–	–	1.5	0.47–3.11		FSAI (2016)
Pizza	–	–	0.20		2.7–18.2	
Seeds	–	–	1.8			
Coffee	–	–	1.1	0.73–1.01		López et al. (2016); Sprong et al., 2016a
Liquorice	–	–	1.3		4.29–5.9	
Legumes	–	–	1.7			

[a] The estimate daily intake was calculated in this study based on the mean concentration reported with consumption data of cereals products and assuming 70 kg as the average body weight for the Canadian and Spanish population.

20 µg kg^{-1} (Sirot et al., 2013). HPLC-DAD presented the highest LOQs reported from 9.0 to 153 µg kg^{-1}, being the less sensitive technique mainly for trichothecenes screening in cereal products (Cano-Sancho et al., 2012).

GC coupled to different detectors as ECD, MS and MS/MS presented LODs from 0.8 to 30 µg kg^{-1} and LOQs from 0.2 to 153 µg kg^{-1} (Leblanc et al., 2005; Cano-Sancho et al., 2012; López et al., 2016). Finally, enzyme-linked immunosorbent assay (ELISA), allowed specific evaluations of one mycotoxin or small related group of them, reached LOQs up to 0.005 µg kg^{-1} (Cano-Sancho et al., 2012), but none of them in multimycotoxin mode.

3.4. Mycotoxins occurrence in foodstuffs

OTA, DON, NIV, T-2 and HT-2 followed by AFs, ZEA FBs and PAT, were the most frequently reported mycotoxins (Tables 3–8).

Table 4
Deoxynivalenol: Occurrence and dietary exposure assessment.

Matrix	N	Incidence (%)	Mean (µg kg^{-1})	EDI (ng kg^{-1} bw day^{-1})	TDI (%)	Reference
Pasta	70	74	226	370–560	37–56	Cano-Sancho et al. (2012)
Corn flakes	65	75	109			
Wheat flakes	27	74	190			
Corn snacks	71	79	154			
Sweet corns	72	3	114			
Whole bread	72	17	68			
Breads	31	100	247			
Baby food	30	40	131			
Gluten-free food	18	11	216			
Ethnic food	35	40	406			
Cereals products	76	37	29.95–33.11	86.1–142.6	8.61–14.2	Yau et al. (2016)
Mixed dishes	44	22	10.57–14.49			
Sugars	4	25	3.65–7.40			
Non-alcoholic beverages	16	13	1.64–6.02			
Cereal products	60	95	2.4–203	9.94–841[a]	0.99–84.1	Beltrán et al. (2013)
Breads and toasts	45	–	524.17	1052	105.2	Raad et al. (2014)
Biscuits and croissants	45	–	340.33	92	9.2	
Rice	15	–	322	230	0.23	
Pizza and pies	45	–	121.16	80	0.8	
Cakes and pastries	45	–	109.67	37	0.3	
Pasta and other cereal products	60	–	62.50	28.8	2.82	
Dried fruits	45	–	62.50	12.55	1.25	
Alcoholic beverages	30	–	52.08	23.53	2.35	
Vegetarians food	35	9	16.3	–	–	Leblanc et al. (2005)
Breads	18	45	108.9	188	18.8	
Rice and semolina	8	25	58.8	9.51	0.951	
Viennese bread and buns	12	42	57.9	18.3	1.83	
Biscuits	14	29	46.4	9.52	0.952	
Nuts and oilseeds	22	5	16.2	0.83	0.083	
Pizzas, salt cakes and quiches	6	17	22.5	9.81	0.981	
Sandwiches	12	25	23.8	4.73	0.473	
Prepared dishes	24	6	15.4	22.2	2.22	
Breads	14	–	132.1–132.1	226.3–226.3	22.63–22.3	Sirot et al. (2013)
Breakfast cereal	6	–	8.5–10.8	0.5–0.6	0.05–0.06	
Pasta	4	–	56.3–56.3	30.6–30.6	3.06–3.06	
Rice	6	–	57.5–58.5	5.7–6.6	0.57–0.66	
Croissants	6	–	73.3–73.3	11.6–11.6	1.16–1.16	
Biscuits	8	–	58.4-61-9	6.7–7.1	0.67–0.71	
Pastries and cakes	18	–	54.2–55.0	38.2–38.5	3.82–3.85	
Pizza, quiches	4	–	101.3–101.3	23.8–23.8	2.38–2.38	
Sandwiches	28	–	83–83	16.1–16.1	1.61–1.61	
Mixed dishes	65	–	20.7–23.4	12.1–13.4	1.21–13.4	
Beer	–	–	9.1	95.8–107	9.58–10.7	López et al. (2016); Sprong et al., 2016a
Biscuits and cookies	–	–	55			
Breads	–	–	28			
Breakfast cereals	–	–	29			
Pasta	–	–	35			
Rye and maize products	–	–	30			
Nivalenol						
Vegetarians food	35	3	15.4	–	–	Leblanc et al. (2005)
Breads	18	–	15.8	27.7	2.30	
Nuts and oilseeds	22	–	16	0.79	0.06	
Rice and rice products	6	–	18.3–19.3	1.88–2.80	0.15–0.23	Sirot et al. (2013)
Mixed dishes	65	–	7.7–10.6	3.47–5.0	0.28–0.41	
T-2 Toxin						
Pasta	70	20	27	27–38	27–38	Cano-Sancho et al. (2012)
Corn flakes	65	11	24.4			
Wheat flakes	27	22	41.2			
Corn snacks	71	8	32.6			
Sweet corns	72	8	144.8			
Apple juice	–	–	14	2.6–10.3	2.6–10.3	López et al. (2016); Sprong et al., 2016a
HT-2 Toxin						

(continued on next page)

Table 4 (continued)

Matrix	N	Incidence (%)	Mean (µg kg^{-1})	EDI (ng kg^{-1} bw day^{-1})	TDI (%)	Reference
Pasta	70	46	24.8	27–38	27–38	Cano-Sancho et al. (2012)
Corn flakes	65	54	23.4			
Wheat flakes	27	52	40.3			
Corn snacks	71	34	82.8			
Sweet corns	72	11	31.6			
Whole wheat breads	72	22.2	39.2			
Pasta	4	–	3–10	1.67–5.55	2.78–9.25	Sirot et al. (2013)
Dried fruits	2	–	3–10	0.04–0.14	0.06–0.23	

[a] The estimate daily intake was calculated in this study based on the mean concentration reported with consumption data of cereals products (FAOSTAT, 2016) assuming 70 kg as the average body weight for the Spanish population.

3.4.1. Ochratoxin A

OTA has been reported in wide variety of agricultural commodities such as cereal products, oats, rye, beans, coffee, rice, peas and meat, and was notably present in wine, beer, grape juice and dried vine fruits (Alshannaq and Yu, 2017; Neme and Mohamed, 2017). OTA was linked to immunotoxicity, genotoxicity, neurotoxicity and teratogenicity and was classified by the International of Research on Cancer (IARC) in Group 2B as possible human carcinogen (IARC, 2012; Marin et al., 2013). EU established OTA limits in several foodstuffs, in the ranges of 0.50–10 µg kg^{-1} (European Commission, 1881/2006).

OTA was evaluated in 55% of the studied TDS, incidences and mean concentrations reported are listed in Table 3. (Leblanc et al., 2005; Cano-Sancho et al., 2012; Beltrán et al., 2013; Sirot et al., 2013; Raad et al., 2014; Yau et al., 2016; Huong et al., 2016; Tam et al., 2011; FSAI, 2016; López et al., 2016). The highest and lowest OTA prevalence were reported in Spain with 73% and 13% of breakfast cereals samples contaminated in Valencian and Catalonia region (Beltrán et al., 2013; Cano-Sancho et al., 2012). The highest concentration was reported in Viet Nam (Huong et al., 2016) with mean levels of 9.68 µg kg^{-1} in bean samples. The lowest concentration was reported in alcoholic beverages in French, ranging from 0.01 to 0.025 µg L^{-1} (Sirot et al., 2013).

The highest incidence and concentration of OTA were reported in

Table 5

Aflatoxins: Occurrence and dietary exposure assessment.

Matrix	n	Incidence (%)	Range (µg kg^{-1})	EDI (ng kg^{-1} bw day^{-1})	Reference
Rice and products	81	67	2.6	22.2	Huong et al. (2016)
Corn	27	50	3.2	1	
Tubes, root and products	54	50	2.1	0.1	
Beans and products	135	75	2.9	0.1	
Oily seeds	27	100	4	0.4	
Vegetables	54	100	2.4	1.6	
Sugars	135	100	4	0.1	
Seasoning	27	100	3.8	0.8	
Fats and oils	54	100	3.3	0.6	
Meat and products	297	91	4	7.7	
Eggs and milk	135	100	5.3	2.3	
Fish	54	50	2.3	1.4	
Other aquatic products	54	100	2.5	0.2	
Cereal products	76	3	0.05–0.24	0.2–2.8	Yau et al. (2016)
Legumes, nuts and seeds	24	19	1.34–1.50		
Fats and oils	4	44	0.41–0.52		
Sweet corns	71	3	0.87	0.058–0.182	Cano-Sancho et al. (2012)
Peanuts	72	11	2.66		
Pistachios	70	20	8.85		
Peppers	72	57	2.22		
Ethnics food	35	9	8.19		
Breads and toasts	45	–	0.242–0.275	0.51–0.52	Raad et al. (2014)
Cakes and pastries	45	–	0.105–0.115	0.037–0.038	
Pizza and pies	45	–	0.043–0.048	0.030–0.031	
Nuts, seeds, olives and dried dates	45	–	0.18–0.26	0.044–0.045	
Chocolate	10	–	0.03–0.07	0.001–0.005	Sirot et al. (2013)
Cereals, seeds	39	21	0.20–1.5	0.23–10.6	FSAI (2016)
Peanuts	9	–	6–38	0.42–2.66	FSANZ (2001)
Aflatoxin M1					
Dairy products	–	–	0.025	0.16	Urieta et al. (1996)
Milk	72	94	0.0097	0.036–0.043	Cano-Sancho et al. (2012)
Yoghurt	72	3	0.0038		
Milk and milk based beverages	30	–	0.11–0.18	0.21–0.22	Raad et al. (2014)

Table 6
Zearalenone: Occurrence and dietary exposure assessment.

Matrix	n	Incidence (%)	Mean (µg kg^{-1})	EDI (ng kg^{-1} bw day^{-1})	TDI (%)	Reference
Pasta	70	14	3.8	1.82–2.40	0.72–0.96	Cano-Sancho et al. (2012)
Wheat flakes	29	14	6.3			
Corn snacks	72	24	5.9			
Sweet corns	72	18	4.9			
Whole wheat breads	71	44	3.7			
Baby food	30	23	4.1			
Ethnics food	35	9	6.2			
Beer	71	11	3.1			
Cereal products	76	2	0.19–7.56	6.1–101.5	1.2–20.3	Yau et al. (2016)
Fats and oils	4	33	48.75–53.75			
Sugars	4	50	15.03–18.15			
Condiments	12	11	1.67–8.33			
Vegetarians food	35	3	8.7	–	–	Leblanc et al. (2005)
Breakfast cereals	12	17	22.7	4.5	2.25	
Vegetables	4	25	17	2.97	1.48	
Nuts and oilseeds	22	5	5.7	3.28	1.64	
Breads	14	–	1.5–5	2.41–8.03	1.205–4.015	Sirot et al. (2013)
Croissants	6	–	1.5–5	0.22–0.74	0.11–0.37	
Biscuits	8	–	3–5.8	0.29–0.64	0.145–0.32	
Pizzas, quiches	4	–	1.5–5	0.39–1.29	0.195–0.645	
Chocolates	10	–	1.55–4.1	0.14–0.31	0.07–0.155	

meat products (64%) at level of 2.68 µg kg^{-1} from Viet Nam (Huong et al., 2016), while the TDS performed in France in 2005 showed the lowest OTA incidence (7%) (Leblanc et al., 2005). The OTA prevalence data in alcoholic beverages, was only included in four TDSs (Leblanc et al., 2005; Cano-Sancho et al., 2012; Sirot et al., 2013; Raad et al., 2014). Cano-Sancho et al. (2012), reported the highest incidence in beer samples; about 89% were OTA contaminated and Sirot et al. (2013) reported the lowest concentrations ranging from 0.017 to 0.025 µg L^{-1}. Similar contents were detected in alcoholic beverages samples from France TDS at mean levels of 0.04 µg L^{-1} (Leblanc et al., 2005).

In coffee beverages, the highest incidence was found in Spain with 49% of coffee samples contaminated (Cano-Sancho et al., 2012) while lower incidences were observed in TDS performed in France, where only 7% of coffee samples contained OTA (Leblanc et al., 2005). All findings ranged from 0.04 µg L^{-1} (Leblanc et al., 2005) to 2.17 µg L^{-1}

(Cano-Sancho et al., 2012; Raad et al., 2014).

None reported coffee products did exceed the maximum limit (ML) established by EU for OTA of 10 µg L^{-1}. OTA was also reported in beans products at concentrations between 0.05 µg kg^{-1} to 9.68 µg kg^{-1} in 20% and 50% of the analyzed samples (Yau et al., 2016; Huong et al., 2016). Similar values were reported in legume composite samples at mean levels of 1.7 µg kg^{-1} (López et al., 2016). OTA was also detected at high incidence at sugar samples and aquatic product samples at mean levels of 0.22 µg kg^{-1} and 4.85 µg kg^{-1} respectively (Yau et al., 2016; Huong et al., 2016).

OTA was also detected in specific food such as fat and oil, mixed dishes with a incidence of 11%–25% respectively, ranging from 0.01 to 0.06 µg kg^{-1} in Chinese TDS (Yau et al., 2016). Relatively high OTA incidence at eggs and milk samples was described with mean contents of 3.16 µg kg^{-1} in TDS performed in Viet Nam (Huong et al., 2016). Concentrations lower than 0.25 µg kg^{-1} were reported by Cano-Sancho

Table 7
Fumonisins: Occurrence and dietary exposure assessment.

Matrix	n	Incidence (%)	Mean (µg kg^{-1})	EDI (ng kg^{-1} bw day^{-1})	Risk assessment (%)	Reference
Cereal products	76	4	2.58–9.76	1.6–97.3	0.08–4.9	Yau et al. (2016)
Corn flakes	72	39	78.9	100–103	5–5.15	Cano-Sancho et al. (2012)
Corns snacks	72	61	119.1			
Baby food	30	23	36.4			
Gluten-free food	18	33	10.4			
Ethnics food	35	51	202.9			
Beer	71	90	36.9			
Breakfast cereal	10	60	1.5–3.5	6.21–14.5	0.310–0.725	Beltrán et al. (2013)
Beer	10	100	1.3–13	3.79–37.14	0.189–1.857	
Offal	6	50	52.5	0.42	0.021	Leblanc et al. (2005)
Breakfast cereals	12	42	41.3	4.88	0.244	
Vegetables	4	50	47.5	3.11	0.1555	
Alcoholic beverages	4	25	7.4	50.1	2.505	
Soft drinks	2	50	8.2	1.17	0.0585	
Breakfast cereals	–	–	18	46.9	2.34	López et al. (2016); Sprong et al., 2016a
Dried fruits	–	–	10	1.17	0.0585	
Breakfast cereals	6	–	8–13.3	0.32–0.64	0.016–0.032	Sirot et al. (2013)
Biscuits	8	–	36.5–75	2.63–2.79	0.131–0.14	

Table 8
Patulin: Occurrence and dietary exposure assessment.

Matrix	n	Incidence (%)	Mean ($\mu g\,kg^{-1}$)	EDI ($ng\,kg^{-1}\,bw\,day^{-1}$)	Risk assessment (%)	Reference
Jams	77	5	13.5	4.28	1.07	Cano-Sancho et al. (2012)
Baby food	124	34	7.1			
Apple juice	71	42	8.1			
Fruits	39	–	0.04–6.09	0.28–10.59	0.07–2.6	Sirot et al. (2013)
Compotes and cooked fruits	6	–	1–8.33	0.24–1.58	0.06–0.39	
Non-alcoholic beverages	25	–	0.11–3.65	0.08–5.66	0.02–1.41	
Fruits	11	–	21.3		2.5–3	FSAI (2016)
Alcohol beverages	4	–	17	10.0–12.0		
Non-alcoholic beverages	13	–	31.9			
Cakes	6	33	31.7	5.45	1.362	Leblanc et al. (2005)
Soft drinks	2	50	20.5	0.78	0.195	
Alcohol beverages	2	50	19.5	3.54	0.885	

et al. (2012) in peanuts (42%), pistachios (3%) and baby food (7%).

In general, OTA contents reviewed were very similar although a slight difference was observed in prevalence.

3.4.2. Trichothecenes

TCs are structurally related mycotoxins produced by *Fusarium* species. DON is probably the most commonly detected trichothecene in cereals followed by T-2 and HT-2 toxins (Pereira et al., 2014; Covarelli et al., 2015; Bryła et al., 2018). DON is mainly associated to toxicological effects on gastrointestinal tract, immune, and endocrine systems (Juan-García et al., 2013). For instance, EU has established limits for DON in several foodstuffs, in the range of 200–1750 $\mu g\,kg^{-1}$ (European Commission, 1881/2006).

DON was found contaminating a variety of foodstuffs; mainly cereal based products with an overall incidence between 3% and 100% (Table 4). The higher incidence of DON was reported in TDS from Spain, 100% of bread samples (Cano-Sancho et al., 2012) and the highest concentration was published in Lebanese TDS, with mean level of 524.17 $\mu g\,kg^{-1}$ in bread and toast samples (Raad et al., 2014). However, the lowest incidence of DON was observed in TDS conducted also in Spain with only 3% of corn samples contaminated (Cano-Sancho et al., 2012) and the lowest contents (between 8.5 and 10.8 $\mu g\,kg^{-1}$) were detected in breakfast cereals samples from TDS study performed in France (Sirot et al., 2013). DON reported values did not exceed the EU maximum limits of 750 $\mu g\,kg^{-1}$ for cereals (European Commission, 1881/2006).

DON was also detected in 22% of the mixed dishes at levels ranging from of 10.57–14.49 $\mu g\,kg^{-1}$ (Yau et al., 2016), even at Leblanc study, only 6% of the prepared dishes contained DON at mean concentration of 15.4 $\mu g\,kg^{-1}$ (Leblanc et al., 2005).

Concerning beverages, DON was detected in 13% of analyzed samples of non-alcoholic beverages at concentrations between 1.64 and 6.02 $\mu g\,L^{-1}$ (Yau et al., 2016). Furthermore, DON was also present in 5% of nuts and oilseed samples with mean content of 16.2 $\mu g\,kg^{-1}$ (Leblanc et al., 2005).

NIV was detected in rice, bread, nuts, oil seed and vegetarian food samples even at incidences and concentrations lower than 5% and 20 $\mu g\,kg^{-1}$ respectively (Table 4) (Sirot et al., 2013; Leblanc et al., 2005).

T-2 and HT-2 toxins were mainly reported in cereal products (Table 4). The most prevalence was observed in cornflakes samples and the highest concentration was quantified in sweet corn up to 144.8 $\mu g\,kg^{-1}$ (Cano-Sancho et al., 2012). The TDS study performed in Netherlands also referred that apple juice composite contained T-2 at mean level of 14 $\mu g\,L^{-1}$. Therefore, cross-contamination during manufacturing of composites preparation could explain the presence of T-2 in apple juice samples (López et al., 2016). Therefore, for TCTs the

highest incidences and concentrations were observed for DON.

3.4.3. Aflatoxins

Fungi producing AFs grow on variety of food mainly cereal products, dried fruits spices, beer and milk and dairy products (Alshannaq and Yu, 2017). AFs linked to genotoxicity, carcinogenic, teratogenic, hepatotoxic, mutagenic and immunosuppressive effects (Theumer et al., 2018). AFB_1 is classified by IARC in Group 1 as carcinogen to human (IARC, 2012). For instance, EU has established limits AFs in several foodstuffs, in the ranges of 4–15 $\mu g\,kg^{-1}$ (European Commission, 1881/2006).

AFs were reported in variety of foodstuffs mainly cereal products, seed and dried fruit, ranging from 3% to 100% (Table 5). A relatively high incidence (67%) was found in rice and products samples (Huong et al., 2016). However, only 3% of cereals and their products were AFs positives at levels ranging from 0.05 to 0.24 $\mu g\,kg^{-1}$ (Yau et al., 2016). Incidence of 9% was reported by Cano-Sancho for ethnic food analysis even at mean concentrations of 8.19 $\mu g\,kg^{-1}$ (Cano-Sancho et al., 2012). In other TDS study performed in Ireland, 21% of the analyzed food, including cereals, nuts and pizza were contaminated by AFs even at levels lower than 1.5 $\mu g\,kg^{-1}$ (FSAI, 2016). AFs were reported in 20% of dried fruits at concentrations ranging from 2.66 to 8.85 $\mu g\,kg^{-1}$ (Cano-Sancho et al., 2012). However, lower contents ranging from 0.180 to 0.260 $\mu g\,kg^{-1}$ were detected in nuts, seed, and dried date (Raad et al., 2014). TDS study performed in New Zealand showed also that roasted salted peanuts samples contained AFB_2 and AFB_1 at mean levels of 6 $\mu g\,kg^{-1}$ and 38 $\mu g\,kg^{-1}$ respectively exceeding the maximum permitted concentrations fixed in 15 $\mu g\,kg^{-1}$ (FSANZ, 2001; European Commission, 1881/2006).

100% of vegetables and seasoning samples were contaminated by AFs at mean levels of 2.4 $\mu g\,kg^{-1}$ and 3.8 $\mu g\,kg^{-1}$ respectively in Viet Nam's TDS (Huong et al., 2016). Spain's TDS also reported AF's incidence of 57% for pepper samples even at mean level of 2.22 $\mu g\,kg^{-1}$ (Cano-Sancho et al., 2012). AFs were also present in fats and oil products; AFB_1 incidence of 100% were reported for oil seed and oil fat samples in Viet Nam and 44% in fats and oils in Hong Kong at mean levels lower than 4 $\mu g\,kg^{-1}$ and (Huong et al., 2016; Yau et al., 2016). 50% of fish and 100% of other aquatic products were also reported AFB_1 contaminated at mean level of 2.3 and 2.5 $\mu g\,kg^{-1}$ respectively, while 91% of meat samples contained AFs at mean concentrations of 4 $\mu g\,kg^{-1}$ (Huong et al., 2016). In this same study AFs were also reported in 100% of sugar samples at mean contents of 4 $\mu g\,kg^{-1}$. AFs were also detected in dark chocolate samples in French TDS even at trace levels (Sirot et al., 2013).

AFM_1 was reported in high incidence (94%) in milk samples even at mean contents of 0.0097 $\mu g\,L^{-1}$, while it was only detected in 3% of yogurt samples with mean levels of 0.0038 $\mu g\,kg^{-1}$ (Cano-Sancho et al.,

2012). Similar levels of AFM_1 were reported in Lebanon in milk based beverages 0.11–0.18 μg L^{-1}, exceeding the maximum permitted concentrations established in 0.05 μg kg^{-1} for milk (Raad et al., 2014; European Commission, 1881/2006).

3.4.4. Zearalenone

ZEA occurs in many agricultural products, including cereals, mixed feeds, rice, and corn silage. The most frequently contaminated crop was corn, followed by wheat (Stanciu et al., 2015). The toxicity of ZEA is associated with reproductive problems in specific animal species and possibility in humans (Marin et al., 2013; Zatecka et al., 2014). EU had established limits for ZEA in several foodstuffs, in the ranges of 20–200 μg kg^{-1} (European Commission, 1881/2006) (Table 6).

In cereal products, the ZEA prevalence of 44% were reported by Cano-Sancho et al. (2012) in whole bread while the maximum concentrations were reported in breakfast cereal samples with mean levels of 22.7 μg kg^{-1} (Leblanc et al., 2005). ZEA was also detected in 9% of ethnic food samples at mean concentrations of 6.2 μg kg^{-1} (Cano-Sancho et al., 2012).

The occurrence of ZEA in fats and oils has been monitored in TDS from Hong Kong; 33% of the sample resulted contaminated by ZEA in ranging from 48.75 to 53.75 μg kg^{-1} (Yau et al., 2016). ZEA also was investigated in nuts and oil seed, 5% of the analyzed samples resulted positive at mean levels of 5.7 μg kg^{-1} (Leblanc et al., 2005).

The prevalence of ZEA in sugar was about 50% at levels between 15.03 and 18.15 μg kg^{-1} (Yau et al., 2016) while ZEA quantified in chocolate ranged between 1.55 and 4.1 μg kg^{-1} (Sirot et al. (2013).

Alcoholic beverages also resulted contaminated by ZEA; 11% of beer samples were positive at mean levels of 3.1 μg kg^{-1} (Cano-Sancho et al., 2012).

ZEA reported values in cereal products did not exceeded the EU maximum limit of 200 μg kg^{-1} (European Commission, 1881/2006).

3.4.5. Fumonisins

FBs occur naturally at significant levels in maize and a variety of maize-based human foodstuffs and animal feeds (Silva et al., 2009; Oldenburg et al., 2017; James and Zikankuba, 2018), but in low concentrations in other cereal products (Piacentini et al., 2015; Stanciu et al., 2015). FB_1 is the most prevalent FBs in human food and also is classified in Group 2B as probably carcinogenic (IARC, 2012). For instance, EU has established limits for sum FBs in several foodstuffs, in the ranges of 200–2000 μg kg^{-1} (European Commission, 1881/2006).

The incidence of FBs ranged from 4% to 100% (Table 7). A high incidence up to 60% was found in breakfast cereal samples (Beltrán et al., 2013). Similar values were reported by Cano-Sancho et al. (2012) who detected FBs in 51% of ethnic food samples. Leblanc et al. (2005) quantified also FBs in 42% of the breakfast cereal samples. However, only 4% of cereal products were reported FBs contaminated in by Yau et al. (2016).

A mean concentration of 202.9 μg kg^{-1} was observed in ethnic food samples (Cano-Sancho et al., 2012). FBs were also detected in biscuits samples ranging from 36.5 to 75 μg kg^{-1} (Sirot et al., 2013). Similar values were reported by Leblanc et al. (2005) who quantified FBs in breakfast cereals samples at mean concentrations of 41.3 μg kg^{-1}.

These reported values of FBs in cereal products did not exceed the EU maximum limits of 400 μg kg^{-1} for corn-based food for direct human consumption (European Commission, 1881/2006).

High incidence of FBs was detected in beer alcoholic beverages; Cano-Sancho et al. (2012) and Beltrán et al. (2013) detected FBs in 90% and 100% of beer samples respectively, ranging from 1.3 to 36.9 μg L^{-1} while, Leblanc et al. (2005) quantified FBs in 25% of the alcoholic beverages. 50% of soft drink and vegetables samples resulted also contaminated by FBs at mean concentrations of 8.2 μg L^{-1} and 47.5 μg kg^{-1}, respectively (Leblanc et al., 2005).

3.4.6. Patulin

PAT occurs mainly in fruits, in particular in apple based products like juice and cooked apples (Iqbal et al., 2018; Abrunhosa et al., 2016). PAT exposure is related to acute human toxicity effects including nausea, vomiting, ulceration and haemorrhage (Alshannaq and Yu, 2017). EU has established limits for PAT in several foodstuffs, in the ranges of 10–50 μg kg^{-1} for infantile food and fruit juices respectively (European Commission, 1881/2006).

PAT was detected mainly in fruit based products (Table 8). In the first French TDS, 50% of alcohol and soft drink samples were contaminated with PAT (Leblanc et al., 2005). Cano-Sancho et al. (2012) also reported presence of PAT in 42% and 34% of apple juice samples and baby food respectively, from Spain. Concerning concentrations, the highest levels were reported in non-alcoholic beverages and cakes with contents of 31.9 μg L^{-1} and 31.7 μg kg^{-1} respectively (FSAI, 2016; Leblanc et al., 2005), while the smallest concentrations were detected in fruits ranging from 0.04 μg kg^{-1} to 6.09 (Sirot et al., 2013). None findings exceed the EU maximum limits (European Commission, 1881/2006).

3.4.7. Others mycotoxins

López et al. (2016) detected emerging mycotoxins, sterigmatocystin, Alternaria toxins and ergo alkaloids in TDS performed in the Netherland. Enniatins, mainly ENN B and ENN B_1, were quantified in apple sauce and cereal-based composites at 6.6 and 58 μg kg^{-1} respectively. BEA was quantified in nuts, dried fruits and tomato products by 2 μg kg^{-1}. Sterigmatocystin was also present in chocolate composite at LOQ level (0.5 μg kg^{-1}). Alternaria toxins (AME, AOH) were detected in a large variety of 88 composite samples analyzed; seven composite samples were positive for AOH, which varied from wine (1.0 μg kg^{-1}) to tomato products (8.9 μg kg^{-1}), and nine composite were contaminated with AME, which varied from pasta and breakfast cereals (3.2 μg kg^{-1}) to dried fruit (8.1 μg kg^{-1}). Ergot alkaloids only occurred in grain-based composites in concentrations ranging from 15 to 47 μg kg^{-1}.

In other TDS conducted in China, Qiu et al. (2017) analyzed seven mycotoxins in 240 mixed dietary samples. Cyclopiazonic acid was detected in bean samples ranging from 0.47 to 1.57 μg kg^{-1} and in alcohol samples at levels between 0.19 and 2.26 μg kg^{-1}. Moreover the contents of citrinin in fruit were 5.31 μg kg^{-1} and the content of moniliformin in milk of 3.60 μg L^{-1}.

3.5. Dietary exposure assessment

TDS are performed to assess dietary exposure to chemical substances in food to assess the effectiveness of risk management strategies to anticipate public health problems (EFSA, 2010; Petersen, 2013). The most commonly used methodology for estimating intake is by combining levels of contamination and food consumption data (Join FAO/WHO Expert Committee on Food Additives, 2001). Although, the estimated exposure in a TDS is rather complicated because of the diversity of food and variations in dietary (Kim et al., 2015).

The health risk characterization of each mycotoxin is performed then by comparing the estimate dietary intake (EDI) previously calculated with the tolerable daily intake (TDI) established (ng kg^{-1} bw day^{-1}) of the respective mycotoxins as reported in Tables 3–7

Most studies reviewed had considered lower and upper bound scenarios for assessment of dietary exposure. In the lower bound approach, the undetected values were replaced by zero and the un-quantified values were replaced by LOD. While, in the upper bound approach, the undetected values were substituted by LOD and the un-quantified values were replaced by LOQ (Sirot et al., 2013; Raad et al., 2014; Yau et al., 2016; Sprong et al., 2016b; Cano-Sancho et al., 2012; FSAI, 2016). However, Huong et al. considered medium bound as additional scenario. In this case, the undetected values were replaced by LOD/2 to estimate dietary exposure (Huong et al., 2016). Others authors used the

mean levels of mycotoxins for dietary intake calculation (Leblanc et al., 2005; Tam et al., 2011; Beltrán et al., 2013; Urieta et al., 1996; FSANZ, 2001).

3.5.1. Ochratoxin A

The EDIs reported ranged between 0.003 and 5 ng kg bw^{-1} (Leblanc et al., 2005; Sirot et al., 2013; Huong et al., 2016; Raad et al., 2014; Cano-Sancho et al., 2012; Tam et al., 2011; FSAI, 2016 and Sprong et al., 2016b) (Table 3). OTA Tolerable Weekly Intake (TWI) set is 120 ng bw^{-1} (JECFA, 2001). Risk assessment values (%TDI) were ranged from 0.01 to 29.41% (Tam et al., 2011; Huong et al., 2016). The main contributor to OTA dietary exposure was meat, with 29.41% of the TDI from Viet Nam TDS followed eggs, milk and fish with 8.2% (Huong et al., 2016). However, lowest values were reported in TDS performed in France by breakfast cereals consumption contributing from 0.01 to 1% of TDI (Sirot et al., 2013). Caffeinated beverages, contributed to OTA risk assessment from 0.47% reported in the first French TDS to 8.71% reported in Lebanon TDS (Leblanc et al., 2005; Raad et al., 2014). The findings suggested that caffeinated beverages did not represent a health risk for the average adult consumer since the estimated EDI of OTA was lower than the TDI established by JECFA (2001).

3.5.2. Trichothecenes

Cereals products were the main contributors to DON dietary exposure (Table 4). The EDI values were between 0.5 and 1052 ng kg^{-1} bw day^{-1} (Beltrán et al., 2013; Raad et al., 2014). TDS performed in France and Spain reported EDIs ranging from 0.83 to 560 ng kg^{-1} bw day^{-1} (Leblanc et al., 2005; Cano-Sancho et al., 2012), while the lowest values were observed in The Netherlands, with daily intake percentile 50 between 95.8 and 107 ng kg^{-1} bw day^{-1} (Sprong et al., 2016b).

The risk assessment was assessed comparing the previously calculated EDI with the tolerable daily intake (TDI) (1000 ng kg^{-1} bw day^{-1}) (JECFA, 2001). TDI values for DON were between 0.05% and 105.2% (Sirot et al., 2013; Raad et al., 2014). These results highlighted that DON exposure might be important, exceeding sometimes the toxicological reference values for average consumers. The main contributors to DON dietary exposure were bread and toast reaching a 105.2% of the TDI (Raad et al., 2014). However, breakfast cereals consumption was associated with low contribution to DON dietary exposure with 0.05% of TDI (Sirot et al., 2013). TDS performed in France, China and Spain, notified also comparable scenarios concerning DON dietary exposure through consumption of different foodstuffs commonly consumed in those countries (Leblanc et al., 2005; Sirot et al., 2013; Cano-Sancho et al., 2012; Yau et al., 2016).

NIV dietary exposure was also assessed by cereal products, nuts, oilseed and mixed dishes consumption in first and second French TDS. Data ranged from 0.79 to 27.7 ng kg^{-1} bw day^{-1} by (Sirot et al., 2013; Leblanc et al., 2005) (Table 4). Tolerable daily intake (TDI) of NIV was 1200 ng kg^{-1} bw day^{-1} (Scientific Committee on Food, 2002). Bread was the main contributor to NIV dietary exposure with 2.30% of the TDI (Leblanc et al., 2005), while rice products contributed up to 0.23% of the TDI (Sirot et al., 2013).

EDI for the sum of T-2 and HT-2 toxins were also investigated and data obtained ranged between 2.6 and 38 ng kg^{-1} bw day^{-1} mainly by cereal products, dried fruit and apple juice consumption (Cano-Sancho et al., 2012; Sirot et al., 2013; Sprong et al., 2016b).

Regarding the risk assessment for T-2 and HT-2 toxins, TDI of 100 ng kg^{-1} bw day^{-1} by was used by Cano-Sancho et al. (2012) as a benchmark dose analysis that EFSA established and TDI of 60 ng kg^{-1} bw day^{-1} was used for French TDS as JECFA proposed as a Provisional Maximum Tolerable Daily Intake (PMTDI) for these toxins (EFSA, 2011; Sirot et al., 2013). The main contributors to T-2 and HT-2 toxin dietary exposure were pasta and cereal products with 2.78%–38% of TDI (Cano-Sancho et al., 2012; Sirot et al., 2013) (Table 4).

3.5.3. Aflatoxins

AFs are classified as carcinogenic and genotoxic compounds and even exposure levels as low as 1 ng kg^{-1} bw day^{-1} may induce liver cancer cases (American Cancer Society, 2011). Concentrations of AFs in food should be reduced As Low As Reasonable Achievable (ALARA) (EFSA, 2007).

The AFs exposure ranged from 0.001 to 22.2 ng kg^{-1} bw day^{-1} through dietary consumption in different countries: New Zealand, China, France, Lebanon, Spain, and Viet Nam (FSANZ, 2001; Leblanc et al. (2005); (Sirot et al., 2013; Cano-Sancho et al., 2012; Raad et al., 2014; Yau et al., 2016) (Table 5).

Rice products were pointed out to be the main source of AFB$_1$ exposure reaching 22.2 ng kg^{-1} bw day^{-1} and the association between high consumption of the AFs contaminated products and liver cancer for adult Viet Nam population was mentioned (Huong et al., 2016). EDI values of AFs in adult Ireland population were between 0.23 and 10.6 ng kg^{-1} bw day^{-1} through cereal products and seeds consumption (FSAI, 2016). AFB$_1$ EDI values up to 0.52 ng kg^{-1} bw day^{-1} for the adult Lebanese population were reported for bread and toast consumption (Raad et al., 2014).

AFM$_1$ exposure through milk and milk based beverages consumption obtained was ranged from 0.036 ng kg^{-1} bw day^{-1} to 0.22 ng kg^{-1} bw day^{-1} for adult Spanish and Lebanese population (Urieta et al., 1996; Cano-Sancho et al., 2012; Raad et al., 2014).

3.5.4. Zearalenone

The main contributors to ZEA dietary exposure reported in the TDS from China were cereal products, fats, oil, sugar and condiments reaching 20.3% of the TDI. Authors used PMTDI of 500 ng kg^{-1} bw day^{-1} for risk assessment (Yau et al., 2016). Nevertheless, values up 4% of the TDI were notified in the first and second French TDS for bread consumption, assuming TDI of 200 ng kg^{-1} bw day^{-1} (SCF, 2000a) (Sirot et al., 2013; Leblanc et al., 2005) (Table 6).

3.5.5. Fumonisins

FBs EDI ranged from 0.32 to 103 ng kg^{-1} bw day^{-1} through consumption of cereal products, gluten-free food, vegetable, offal, alcoholic beverages and soft drink, in TDS from France, Spain and China (Leblanc et al., 2005; Sirot et al., 2013; Cano-Sancho et al., 2012; Yau et al., 2016). Sprong et al. (2016b) reported EDIs in upper-bound scenario of 46.9 ng kg^{-1} bw day^{-1} through consumption of breakfast cereals and dried fruits.

The risk assessment (%TDI) was calculated by comparing the EDI previously estimated and TDI of 2000 ng kg^{-1} bw day^{-1} of TDI (JECFA, 2001).

The main contributor to the dietary exposure to FBs were cereal products contributing up to 5% of TDI for mean consumer for Catalonia region population (Cano-Sancho et al., 2012), even lower values were notified in the French TDS (0.016–0.14% of the TDI) (Sirot et al., 2013).

3.5.6. Patulin

The risk assessment for PAT was calculated by comparing the EDI estimated and TDI of 400 ng kg^{-1} bw day^{-1} (SCF, 2000b; Leblanc et al., 2005; Sirot et al., 2013; Cano-Sancho et al., 2012; FSAI, 2016). The main contributor to PAT dietary exposure were fruits, alcoholic and non-alcoholic beverages up to 3% of the TDI for mean consumer for Ireland population (FSAI, 2016; Leblanc et al., 2005; Sirot et al., 2013).

Summary of estimate dietary exposure to mycotoxins performed in the different TDS is provided in Table 9.

EDI for all mycotoxins were ranged between 0.001 ng kg^{-1} bw day^{-1} for AFs to 1052 ng kg^{-1} bw day^{-1} for DON; being cereal products the main contributor to DON the dietary exposure in Lebanese TDS reaching 105.2% of TDI (Sirot et al., 2013; Raad et al., 2014). Lower values were notified for OTA and FBs 0.01% each one reported in French TDS through breakfast cereals consumption (Sirot et al., 2013).

Table 9
Summary of estimated dietary exposure to mycotoxins presented in the different TDS.

Mycotoxin	TDI (ng kg bw day)	EDI (ng kg bw day)	TDI (%)	Reference
OTA	17	0.003–5	0.01–29.41	Huong et al. (2016); Cano-Sancho et al., 2012; Raad et al. (2014); Yau et al. (2016); Tam et al. (2011); Beltrán et al. (2013); Leblanc et al. (2005); Sirot et al. (2013); FSAI, 2016; Sprong et al., 2016a
DON	1000	0.5–1052	0.05–105.2	Cano-Sancho et al. (2012); Yau et al. (2016); Beltrán et al. (2013); Raad et al. (2014); Leblanc et al. (2005), Sirot et al. (2013); Sprong et al., 2016a
NIV	1200	0.79–27.7	0.065–2.30	Leblanc et al. (2005); Sirot et al. (2013)
T-2	60; 100	2.6–38	2.6–38	Cano-Sancho et al. (2012); Sprong et al., 2016a
HT-2	60; 100	0.04–38	0.06–38	Cano-Sancho et al. (2012)
AFs	–	0.0019–22.2	–	Huong et al. (2016); Yau et al. (2016); Cano-Sancho et al. (2012); Raad et al. (2014); Sirot et al. (2013); FSAI, 2016; FSANZ, 2001
AFM1	–	0.036–0.22	–	Urieta et al. (1996); Cano-Sancho et al. (2012); Raad et al. (2014)
ZEA	250	0.14–101.5	0.07–20.3	Cano-Sancho et al. (2012); Yau et al. (2016); Leblanc et al. (2005); Sirot et al. (2013)
FBs	2000	0.32–103	0.016–5.15	Yau et al. (2016); Cano-Sancho et al. (2012); Beltrán et al. (2013); Leblanc et al. (2005); Sprong et al., 2016a; Sirot et al. (2013)
PAT	400	0.08–12.0	0.02–3.0	Cano-Sancho et al. (2012); Sirot et al. (2013); FSAI, 2016; Leblanc et al. (2005)

4. Conclusions

This review on mycotoxins total diet summarized about eighteen studies performed in several countries highlighting the increasing interest for mycotoxin exposure through diet. HPLC-FD was the most widely used and OTA the most analyzed mycotoxin. Mycotoxin levels reported were relatively low in most of analyzed food except for milk and wine beverages which could approach the maximum limits. Despite the relatively high frequency of mycotoxins detected in the different food of habitual consumption, calculated EDIs were generally below the established TDIs, except for DON and OTA who amounted in some TDSs, appreciable ratio of their respective TDI through cereal and meat products.

Conflicts of interest

The authors declare no conflict of interest and informed consent was obtained from all individual participants included in the study.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fct.2019.03.033.

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