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Evaluation of dietary exposure to deoxynivalenol (DON) and its derivatives from cereals in China

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ABSTRACT

Probabilistic estimation of dietary exposure to DON, including its acetylated derivatives, and type B trichothecenes from cereals and cereal-based products in Chinese populations was investigated in the current work. Different cut-offs as proposed Maximum levels (MLs) for DON in various raw cereals and/or cereal-based foods were assessed based on the risk assessment results. Occurrence data was documented from 31 provinces of China over the year 2010–2013. Food consumption data was obtained from the National Diet and Nutrition Survey conducted in 2002. Dietary exposure was implemented by Monte Carlo (MC) simulations and Bootstrap resampling. The exposure distributions were specified as percentiles with confidence intervals (95% CIs) and compared with the group provisional maximum tolerable daily intake (PMTDI) of 1 μg/kg bw/day and the group acute reference dose (ARfD) of 8 μg/kg bw/day for DON and its acetylated derivatives. Results indicate that 75% of children and 90% of the general population and the adults are under the group PMTDI value, while 99% of the three populations are under ARfD value. However, under the assumptions of the proposed cut-offs, the P99.9 percentiles would decrease significantly. The probabilistic assessment in this study indicated that high-end exposure to DON and its derivatives should be concerned, especially for children. Rigorous formulation of maximum limits for DON and its derivatives in the relevant foodstuffs combined with increased monitoring should be considered as an effective way to reduce risk.

1. Introduction

Deoxynivalenol (DON) is a type B trichothecene mycotoxin produced by a variety of Fusarium fungi commonly found in cereals. DON exposure has been associated with the occurrence of acute gastrointestinal diseases, which is recognized along with aflatoxin, fumonisin, ochratoxin A, and zearalenone as one of the key mycotoxins of great public health concern (IARC., 2012; Ortiz, Van Camp, Mestdagh, Donoso, & De Meulenaer, 2013). International workshop was organized on trichothecenes with a special focus on DON, and a number of experts have reviewed the knowledge with respect to occurrence, prevention, analytical methodologies, surveillance and exposure assessments, toxicology and risk assessment (Larsen, Hunt, Perrin, & Ruckenbauer, 2004). The level of DON exposure in humans depends on the extent of contamination in the consumed food items and the amount of the food ingested (Srey, Kimanya, Routledge, Shirima, & Gong, 2014). Recently, occurrence and exposure of DON have been assessed in many countries (Escobar et al., 2013; Gratz, Richardson, Duncan, & Holtrop, 2014; Mankeviciene, Jablonskyte-Rasce, & Maiksteniene, 2014; Mishra, Ansari, Dwivedi, Pandey, & Das, 2013; Ortiz et al., 2013; Raad, Nasreddine, Hilan, Bartosik, & Parent-Massin, 2014; Rodríguez-Carrasco, Fattore, Albrizio, Berrada, & Mañes, 2015; Srey et al., 2014), where DON was frequently detected with a number of samples exceeding the maximum limits and the estimated dietary exposure exceeded the toxicological or health reference value.

In addition to DON, the most abundant trichothecene, 3-acetyl-deoxynivalenol (3AcDON) and 15-acetyl-deoxynivalenol (15AcDON) are naturally occurring fungal secondary metabolites and contribute to the total DON-induced toxicity (JECFA., 2010a). Furthermore, DON-3-glucoside, a naturally occurring conjugate of DON recently detected in wheat, corn and barley, may also be

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metabolized to release the parent DON in humans and animals (Berthiller, Schuhmacher, Adam, & Krska, 2009; Sasanya, Hall, & Wolf-Hall, 2008). Consequently, the DON exposures may be underestimated without consideration of exposure to the DON derivatives.

In China, a case study on risk assessment of co-occurring DON and its acetyl derivatives in wheat and maize was conducted in Shanghai, suggesting a potential cumulative health risk to the local population and the necessity of co-occurring risk assessment (Han et al., 2014). A larger-scale study on co-exposure of DON and zearalenone was reported from domestic wheat flour and corn-based products, indicating children at a high risk to both mycotoxins and the Chinese regulatory limit should be evaluated based on the risk assessment results, especially specific tolerance limits need to be established in foods intended for children (Wang, Zhu, Shao, & Li, 2015). Compared with the Maximum level (ML) of 2 mg/kg for DON in raw wheat, maize and barley recommended by Codex Alimentarius Commission (CAC), European Union proposed more rigorous criteria that ML in cereals and cereal flour, bran and germ as end product served for general people was 0.75 mg/kg and ML in cereal-based processing foods and baby foods for infants and young children was 0.2 mg/kg (Commission Regulation (EC) No 1881, 2006). The national mandatory ML is 1 mg/kg in China for wheat, maize, barley and foods derived from them, and whether it needs revision or be conformance with CAC or EU for food regulation needs to be evaluated based on risk assessment.

In this context, the purposes of this study were (1) to assess nationwide dietary exposure to free DON (fDON), DON including its acetylated derivatives (acDON) and the sum of DON, 3AcDON, 15AcDON, nivalenol (NIV), and 4-acetyl NIV (fusarenone X) (typeB trichothecenes, sumDON) from cereals and cereal-based products in Chinese populations by probabilistic model; (2) to evaluate the different cut-offs as proposed MLs for DON in various raw cereals and/or cereal-based foods based on the risk assessment results.

2. Materials and methods

2.1. Data collection

The nationwide occurrence of DON and its derivatives in cereal grains was well documented over the year 2010–2013 from 31 provinces of China. A total of 7356 retail cereal samples from 17 commodities were assessed for a range of trichothecenes by isotope dilution liquid chromatography tandem mass spectrometry (ID-LC/MS/MS) (GB/T 5009. 111-201x). Detailed food items included barley, buckwheat, maize, millet, oats, peanut, rice, sorghum, wheat and their products.

Food consumption data was obtained from the National Diet and Nutrition Survey conducted in 2002 by 24-h recall method on 3 consecutive days. The multistage and random cluster sampling method was used in the survey and most areas (30 provinces on the mainland) in China were covered. A total of 22,567 households with 65,915 consumers aged 2–100 years (the general population) remained for analysis, including 3701 children of 2–6 years old and 51,175 adults above 18 years old. Demographic information (age, weight, etc.) of the participants was collected all at once and used in the calculation (Table 1).

2.2. Probabilistic model of dietary exposure assessment

Probabilistic modeling of dietary exposure was implemented by Monte Carlo (MC) simulations. Daily consumptions were simulated

Table 1
Basic demographic information from the national nutrition and health survey 2002.

Population	Households	Persons	Person-days	Age (years) (mean± SD)	Weight (kg) (mean± SD)
General population	22,567	65,915	193,080	37.4 ± 19.8	53.4 ± 16.5
Male	–	32,072	93,994	36.9 ± 20.3	55.8 ± 18.1
Female	–	33,843	99,086	37.8 ± 19.4	51.0 ± 14.5
Children (2–6 years)	3406	3701	10,778	4.2 ± 1.4	16.4 ± 3.4
Male	–	2040	5945	4.2 ± 1.4	16.7 ± 3.4
Female	–	1661	4833	4.3 ± 1.4	16.1 ± 3.4
Adults (18 + years)	22,545	51,175	150,254	45.3 ± 14.8	59.7 ± 10.8
Male	–	24,152	70,963	45.9 ± 14.9	63.7 ± 10.7
Female	–	27,023	79,291	44.9 ± 14.7	56.1 ± 9.5

*Person-day: The Survey was conducted on 3 consecutive days (2 weekdays and 1 weekend, holidays excluded). Each study day for a participant means a person-day.

Table 2
Nationwide occurrence of DON and its derivatives over the year 2010–2013 in cereal grains in China (μg/kg).

Contaminant	Year	Sample number	Detectable rate (%)	<LOD = 0			<LOD = LOD/2			<LOD = LOD		
				Mean	Median	Max	Mean	Median	Max	Mean	Median	Max
fDON	2010	1722	49.5	96.1	0.0	4794.6	97.9	10.0	4794.6	99.7	15.5	4794.6
	2011	1557	66.0	94.7	14.9	3273.8	95.5	16.6	3273.8	96.2	20.0	3273.8
	2012	698	65.5	747.6	67.7	14,276.0	747.7	67.7	14,276.0	747.8	67.7	14,276.0
	2013	3265	69.6	218.9	97.6	6922.0	222.3	97.6	6922.0	225.6	109.0	6922.0
acDON	2010	1803	53.5	107.5	1.1	5042.2	111.9	10.0	5042.2	116.2	20.0	5042.2
	2011	1565	68.1	103.8	19.2	3462.5	105.3	22.0	3462.5	106.8	22.7	3462.5
	2012	698	65.6	769.9	70.6	14,604.2	770.3	70.6	14,604.2	770.7	70.6	14,604.2
	2013	3266	69.7	221.1	99.1	6922.0	224.6	99.1	6922.0	228.2	111.0	6922.0
sumDON	2010	1803	53.5	107.5	1.1	5042.2	111.9	10.0	5042.2	116.2	20.0	5042.2
	2011	1565	68.1	103.8	19.2	3462.5	105.3	22.0	3462.5	106.8	22.7	3462.5
	2012	698	65.6	785.1	71.9	15,066.3	785.6	71.9	15,066.3	786.2	71.9	15,066.3
	2013	3290	69.3	223.7	98.0	6922.0	227.4	98.0	6922.0	231.1	110.2	6922.0

* Over the 4 years for all mycotoxins, Trend test for detectable rate: $P < 0.001$; Kruskal–Wallis Test for contamination level: $P < 0.001$.