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中国两大优势产区小麦重金属镉含量调查与膳食暴露评估

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摘要：【目的】分析中国两大小麦优势产区小麦重金属镉的污染水平和特点，明确不同消费人群食用小麦引起的镉膳食暴露风险，以期为中国农产品质量安全监管提供科学依据。【方法】在中国黄淮海和长江中下游两大小麦优势产区的 8 个省（市）采集 2013 年收获的小麦样品 393 份，利用低温消解进行前处理，通过石墨炉原子吸收分光光度计测定全部小麦样品中镉浓度，分析不同区域不同品种小麦镉的含量差异。结合中国不同消费人群的小麦消费数据和体重信息，利用非参数概率方法对小麦镉的膳食暴露量进行评估。【结果】中国两大小麦优势产区的小麦镉含量均值为（0.032±0.051）$mg·kg^{-1}$，97.5%的小麦样本镉含量低于国家限量要求。长江中下游优势产区的小麦镉含量均值为（0.060±0.091）$mg·kg^{-1}$，黄淮海优势产区的小麦镉含量均值为（0.024±0.025）$mg·kg^{-1}$，长江中下游小麦优势产区的小麦镉含量水平显著高于黄淮海小麦优势产区的小麦镉含量水平，不同品种、同一品种的小麦镉含量也存在一定的差异。中国不同消费人群对于小麦镉的膳食暴露风险不同，总体表现为：农村高于城市，低龄高于高龄，18 岁以下的消费人群呈现女性高于男性，18 岁以上的消费人群则呈现男性高于女性的特点。中国不同消费人群对于小麦镉的平均暴露量为国际食品添加剂联合专家委员会（JECFA）推荐的 PTMI（暂定每月耐受摄入量：25 $μg·kg^{-1}$ bw）的 9.0%—16.8%，P97.5 百分位数暴露量为 PTMI 的 28.9%—53.8%，暴露量最高值出现在 4—7 岁女童，暴露量最低值出现在 60—70 岁女性。中国城市消费人群对于小麦镉的膳食暴露风险表现为：低龄高于高龄，男性高于女性，但 60—70 岁的城市消费人群中女性偏高于男性，并且城市女性消费人群中 14—18 岁和 45—60 岁两个年龄段的暴露量偏低，中国城市消费人群对于小麦镉的平均暴露量为 PTMI 的 7.3%—13.1%，P97.5 百分位数暴露量为 PTMI 的 23.4%—42.2%，暴露量最高值出现在 4—7 岁男童，暴露量最低值出现在 14—18 岁女性。中国农村消费人群对于小麦镉的暴露量呈现与全国类似的规律，平均暴露量为 PTMI 的 9.6%—17.9%，P97.5 百分位数暴露量为 PTMI 的 30.9%—57.4%，暴露量最高值出现在 4—7 岁女童，暴露量最低值出现在 60—70 岁女性。【结论】中国黄淮海小麦优势产区和长江中下游小麦优势产区的小麦受到的重金属镉污染程度较轻，区域间和品种间差异都较为明显。中国不同消费人群对于小麦镉的膳食暴露风险不同，食用这些区域生产的小麦引起的镉暴露量较低，且风险在可接受的水平。

关键词：小麦；小麦优势产区；镉；膳食暴露

Survey and Dietary Exposure Assessment of Cadmium in Wheat from Two Main Wheat-Producing Regions in China

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Abstract: Objective The objective of this study is to analyze the variation and characteristics of cadmium levels in wheat of two main wheat-producing regions in China and assess the risk degree and law of cadmium in wheat from dietary exposure to different consumer groups. Results of this study could provide a scientific basis for the supervision of quality and safety of agro-products in China. Method A total of 393 wheat samples were collected from two main wheat-producing regions of Huang-Huai-Hai and the middle and lower reaches of the Yangtze River, including 8 provinces (cities). The wheat samples were treated with nitric acid and hydrogen peroxide solution by low-temperature digestion. The cadmium contents of the samples were detected by graphite furnace atomic absorption spectrophotometer (GFAAS). A non-parameter probabilistic model was used to assess the dietary exposure risk based on the information of wheat consumption and body weight of target populations. Result The mean of cadmium contents of wheat in China's two main wheat-producing regions was (0.032±0.051) mg·kg^{-1}, and cadmium contents in 97.5% wheat samples were below maximum allowable concentration (MAC). The average content of cadmium in wheat was (0.060±0.091) mg·kg^{-1} in the middle and lower reaches of Yangtze River region, while the level was (0.024±0.025) mg·kg^{-1} in Huang-Huai-Hai region. The cadmium levels of wheat in the middle and lower reaches of Yangtze River region were significantly higher than those in Huang-Huai-Hai region. There were different cadmium concentrations of wheat between different planting regions. Also it showed difference in the same variety from area to area. Considering the disparity of age and the difference of city and countryside, generally, differences were found in dietary exposure among the various consumer groups all over the country. Different consumption groups with exposure levels of cadmium in wheat had an overweighed ranking for the groups of countryside, younger ages, respectively. Considering the disparity of gender, the exposure levels of cadmium in wheat in females were higher than the levels in males (age 18 years old). When the age 18 years old, the opposite case presented. The percent ratio of the mean of the assessed dietary exposure value to PTMI recommended by JECFA (Joint FAO/WHO Expert Committee on Food Additives, Provisional tolerance monthly intake: 25 μg·kg^{-1} bw) was in the range of 9.0%-16.8%, the 97.5th percentile level was from 28.9% to 53.8%. The highest exposure level occurred in girls of 4-7 years old, and the lowest was found in women of 60-70 years old. In the city, considering the disparity of age and gender, the different consumption groups with exposure levels of cadmium in wheat have an overweighed ranking for the groups of younger ages and masculinity, respectively. Although the exposure level of females was higher than those of males in the age of 60-70 years old and there were lower exposure levels in the age of 14-18 and 45-60 years old in females in the city. The percent ratio of the mean of the assessed dietary exposure value to PTMI was in the range of 7.3%-13.1%, the 97.5th percentile level was from 23.4% to 42.2%. The highest exposure level occurred in boys of 4-7 years old, and the lowest was found in women of 14-18 years old. The assessed dietary exposure level of rural consumer groups had the similar rules of all over the country, the exposure value in the countryside to PTMI was in the range of 9.6%-17.9%, and the 97.5th percentile level was from 30.9% to 57.4%. The highest exposure level appeared in girls of 4-7 years old, and the lowest was found in women of 60-70 years old. Conclusion The study showed that the cadmium pollution in wheat was at lower level. There were significant differences in cadmium contents of wheat among the areas and within the varieties. The differences were found in dietary exposure among the various consumer groups. The probability of health risk of cadmium via wheat exposure was at the acceptable level.

Key words: wheat; main wheat-producing regions; cadmium; dietary exposure

0 引言

Cd

[1-3]

[4] 1970

Joint FAO/WHO Expert Committee on Food

Kamijoka Additives JECFA

[5-6] 16 33 41 55

61 64 73

33 JECFA

provisional tolerance weekly intake

PTWI 0.007 mg·kg^{-1} bw[7] 55
JECFA
[8] 64 JECFA
[9] 2010 JECFA
73
PTWI provisional
tolerance monthly intake PTMI 0.025
mg·kg^{-1} bw[10]

0.048 mg·kg^{-1}

6
3 [5] Corguinha [11]
15—83 μg·kg^{-1} Khan
[12]
0.1 mg·kg^{-1}
1
Bermudez [13]
0.017 mg·kg^{-1}

[14]
9.11% 0.25
mg·kg^{-1}[15] 68
2.9%
, [16]

ICRP

[17]

1 材料与方法

2012—2013

1.1 试验材料

1.1.1 小麦样品

8
393 110

1.1.2 试剂耗材

BV-
BV-
18.2 MΩ·cm 5 mg·mL^{-1}
50 mL
LabTech 1 000 mg·L^{-1}
GBW
10011
GBW8503b /
20% 24 h

1.1.3 仪器设备

AA-7000
1 Shimadzu EHD36
LabTech Milli-Q Academic
Millipore 1702
Sartorius

表 1 原子吸收分光光度计仪器工作参数
Table 1 The operating parameters of AAS

Parameter	Value
Wavelength (nm)	228.8
Spectral bandwidth (nm)	0.7
Lamp current (A)	8
Background corrector	Deuterium lamp
Sample size (μL)	20
/ Matrix developed agent/Sample size (μL)	$Pd(NO_3)_2$/ 5
Drying temperature ( )	150
Ashing temperature ( )	500
Atomization temperature ( )	1200

1.2 试验方法

1.2.1 样品采集与处理

393
2012 2013 110

8 180 1
2008—2015

71.1% 77.1%

4
2 8
5 3 1 m 5
1 kg 8
1
8 kg

3 80 4 h 425
μm

1.2.2 样品测定 0.25 g 50 mL
6 mL 110
1 mL 2 mL
0.5 mL 25 mL
GBW08503b
GBW10011 2
LOD 0.0003 mg·kg^{-1}
LOQ 0.001 mg·kg^{-1}

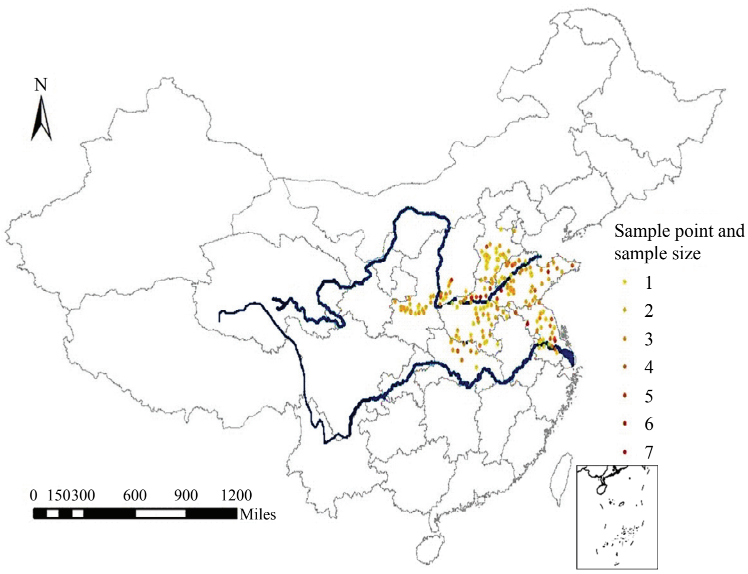


图 1 小麦抽样分布图

Fig. 1 The distribution of wheat samples

表 2 标准物质测定结果

Table 2 The determination results of standard substance

Standard substance		Element	Certified (mg·kg^{-1})	Measured (mg·kg^{-1})
GBW10011	GBW10011 wheat flour	Cd	0.018±0.004	0.020±0.002
GBW08503b	GBW08503b wheat flour	Cd	0.15±0.04	0.17±0.01

1.2.3 风险评估方法

1
[18]

[19-21]
JECFA
PTMI 0.025 mg·kg^{-1} bw
[22-23]

$$Y = \frac{30 \times X \times C}{W} \quad (1)$$

U Bootstrap

Y μg·kg^{-1} bw X Bootstrap

g C mg·kg^{-1} V U Bootstrap

W kg 100 000 Monte Carlo 100 000

——2002 [24]

——2002 U V 2 000

[25] 3 V 2 000

2

100 000 × 2 000=2 × 10^8 U

RAMA U 95% V

V P97.5 [27-28]

[26]

表 3　中国居民体重及小麦消费量

Table 3　The body weight and wheat intake of Chinese residents

Age	Nationwide				City				Countryside			
	Body weight (kg)		Dietary intake of wheat (g·d^{-1})		Body weight (kg)		Dietary intake of wheat (g·d^{-1})		Body weight (kg)		Dietary intake of wheat (g·d^{-1})	
	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female
4—7	21.09	20.18	81.8	84.0	22.31	21.21	76.0	72.0	19.72	18.91	83.5	87.6
7—11	29.94	29.24	99.7	99.7	32.11	31.02	89.5	83.7	27.80	27.33	102.6	104.2
11—14	40.48	40.6	126.0	117.1	42.75	42.29	107.9	96.7	39.46	39.78	130.9	122.7
14—18	53.64	49.7	151.8	134.8	57.62	51.63	132.1	97.6	52.25	48.92	159.2	149.9
18—30	63.29	53.68	175.5	133.1	66.04	54.48	144.9	112.4	62.28	53.36	189.6	142.7
30—45	64.24	56.64	166.8	136.7	67.78	58.65	145.6	116.6	62.62	55.71	175.9	144.9
45—60	62.48	56.55	159.2	133.5	67.17	60.05	141.2	115.8	60.04	54.32	166.0	140.3
60—70	60.83	53.52	151.5	122.7	65.35	57.40	132.5	117.5	56.93	50.01	158.6	124.8

2　结果

2.1　小麦中镉含量区域分布

393

100% GB 2762—2012 [29]

97.5%

0.032±0.051 mg·kg^{-1} 0.023 mg·kg^{-1}

8 [30]

0.05 mg·kg^{-1}

0.02 mg·kg^{-1}

4

P 0.001

0.060±0.091 mg·kg^{-1}

0.024±0.025 mg·kg^{-1}

[31]

2 25

2.2 不同小麦品种中镉含量

0.092 mg·kg-1 28

16 9023 0.059

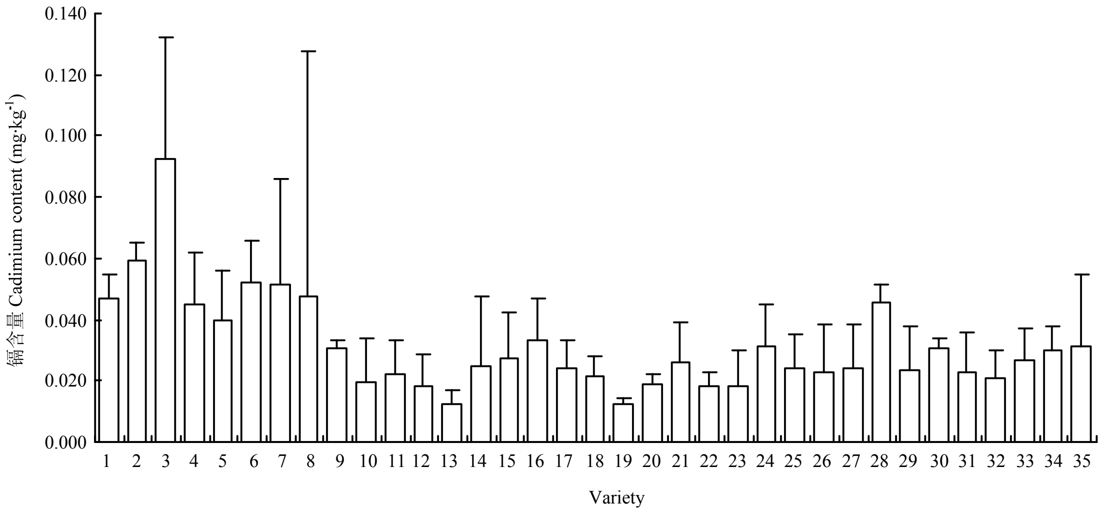
0.052 0.051 mg·kg-1

表 4 中国两大小麦优势产区小麦样品镉含量基本统计量

Table 4 The basic statistics of cadmium content in wheat samples from two main wheat-producing regions of China

Item	Middle and lower reaches of Yangtze River region [a]	Huang-Huai-Hai region [b]	Total
Province number	3	7	8
County number	42	138	180
Variety number	29	88	110
Sample number	89	304	393
Mean (mg·kg-1)	0.06	0.024	0.032
Median (mg·kg-1)	0.046	0.02	0.023
Standard deviation (mg·kg-1)	0.091	0.025	0.051
Maximum (mg·kg-1)	0.869	0.381	0.869
Minimum (mg·kg-1)	0.005	0.003	0.003
Detection rate (%)	100	100	100
[c] Pass rate (%)	91.0	99.3	97.5

[a] Middle and lower reaches of Yangtze River wheat-producing region Hubei (n=21) Henan (n=21) Jiangsu (n=47) [b] Huang-Huai-Hai wheat-producing region Hebei (n=36) Henan (n=76) Jiangsu(n=18) Shandong (n=107) Shanxi (n=27) Shaanxi (n=38) Tianjin (n=2) [c] The limited value of Cd in wheat: 0.1 mg·kg-1 (GB 2762—2012)



1 35 Hengguan 35 2 28 Huaimai 28 3 25 Xiangmai 25 4 4 Yangfumai 4 5 13 Yangmai 13 6 16 Yangmai 16 7 9023 Zhengmai 9023 8 58 Aikang 58 9 1 Fengdecunmai 1 10 22 Jimai 22 11 17 Jinan 17 12 66 Liangxing 66 13 99 Liangxing 99 14 8050 Linfen 8050 15 536 Linhan 536 16 4 Linmai 4 17 21 Lumai 21 18 502 Luyuan 502 19 1 Qingfeng 1 20 828 Shixin 828 21 18 Tainong 18 22 979 Xinong 979 23 22 Xiaoyan 22 24 26 Xinmai 26 25 30 Xumai 30 26 19 Yannong 19 27 24 Yannong 24 28 5158 Yannong 5158 29 49-198 Yumai 49-198 30 366 Zhengmai 366 31 7698 Zhengmai 7698 32 1 Zhongmai 1 33 16 Zhoumai 16 34 22 Zhoumai 22 35 Others

图 2 两大小麦优势产区主要小麦品种镉含量差异

Fig. 2 The cadmium content of different wheat varieties in two main producing regions

99 1
0.012 mg·kg^{-1} 66 979 22
0.018 mg·kg^{-1} 22 828
0.019 mg·kg^{-1}

58 25 9023
0.080 0.040 0.034 mg·kg^{-1}

2.3 不同消费人群小麦镉膳食暴露量

RAMA

5 4—70
8 48

18
18

PTMI 9.0%—16.8% P97.5
PTMI 28.9%—53.8%
4—7 60
—70

4—7
60—70

14—18 45—60

60—70

PTMI 7.3%
—13.1% P97.5 PTMI 23.4%—
42.2% 4—7
14—18

14—18

18

18

PTMI 9.6%—17.9% P97.5
PTMI 30.9%—57.4% 4
—7 60—70

4
—7

3 讨论

PTMI 10% Huang [14] Khan [12]
Bermudez [13]
Khan [32]
PTMI 30%
—40%
[33-39]

18
18

18

表 5 中国不同性别年龄组人群小麦镉膳食暴露评估

Table 5 Dietary exposures of cadmium in wheat for different age and gender groups of China

Age	Gender	P97.5 95% Mean and the 97.5th percentile values of monthly intake estimates and 95% confidence limits (μg·kg^{-1} bw)					
		Mean (nationwide)	P97.5 The 97.5th percentile (nationwide)	Mean (city)	P97.5 The 97.5th percentile (city)	Mean (countryside)	P97.5 The 97.5th percentile (countryside)
4—7	M	3.821 (3.373—4.558)	12.260 (9.075—13.843)	3.273 (2.908—3.881)	10.553 (7.811—13.467)	4.084 (3.604—4.856)	13.114 (9.707—14.757)
	F	4.190 (3.699—4.999)	13.446 (9.953—15.182)	3.260 (2.896—3.866)	10.512 (7.781—13.415)	4.468 (3.942—5.313)	14.346 (10.619—16.144)
7—11	M	3.308 (2.920—3.946)	10.615 (7.857—11.985)	2.677 (2.379—3.174)	8.633 (6.390—11.017)	3.560 (3.141—4.232)	11.429 (8.460—12.862)
	F	3.439 (3.037—4.104)	11.037 (8.170—12.462)	2.592 (2.303—3.073)	8.357 (6.186—10.665)	3.677 (3.244—4.372)	11.807 (8.739—13.286)
11—14	M	3.121 (2.755—3.724)	10.015 (7.414—11.308)	2.425 (2.154—2.875)	7.818 (5.787—9.977)	3.200 (2.823—3.804)	10.273 (7.605—11.561)
	F	2.917 (2.575—3.480)	9.360 (6.929—10.569)	2.196 (1.951—2.604)	7.082 (5.242—9.038)	2.975 (2.625—3.538)	9.553 (7.071—10.750)
14—18	M	2.864 (2.529—3.418)	9.192 (6.804—10.379)	2.202 (1.956—2.611)	7.100 (5.256—9.061)	2.939 (2.593—3.495)	9.438 (6.986—10.620)
	F	2.911 (2.570—3.473)	9.342 (6.915—10.548)	1.816 (1.613—2.153)	5.855 (4.334—7.471)	2.955 (2.608—3.514)	9.490 (7.024—10.679)
18—30	M	2.891 (2.553—3.450)	9.278 (6.868—10.476)	2.108 (1.872—2.499)	6.795 (5.030—8.672)	2.937 (2.591—3.492)	9.429 (6.979—10.610)
	F	2.565 (2.265—3.061)	8.233 (6.094—9.296)	1.982 (1.761—2.350)	6.391 (4.730—8.155)	2.580 (2.276—3.067)	8.283 (6.132—9.322)
30—45	M	2.643 (2.333—3.153)	8.481 (6.278—9.576)	2.064 (1.833—2.447)	6.654 (4.925—8.491)	2.710 (2.391—3.222)	8.700 (6.440—9.790)
	F	2.469 (2.180—2.946)	7.923 (5.865—8.946)	1.910 (1.697—2.264)	6.158 (4.558—7.858)	2.509 (2.214—2.983)	8.055 (5.963—9.065)
45—60	M	2.564 (2.264—3.060)	8.229 (6.091—9.292)	2.019 (1.794—2.394)	6.511 (4.819—8.309)	2.667 (2.353—3.171)	8.563 (6.338—9.636)
	F	2.394 (2.114—2.857)	7.684 (5.688—8.676)	1.852 (1.646—2.196)	5.973 (4.421—7.622)	2.491 (2.198—2.962)	7.999 (5.921—9.002)
60—70	M	2.516 (2.222—3.002)	8.075 (5.977—9.118)	1.948 (1.730—2.309)	6.280 (4.648—8.014)	2.688 (2.371—3.196)	8.629 (6.387—9.711)
	F	2.251 (1.987—2.685)	7.223 (5.346—8.155)	1.966 (1.747—2.331)	6.340 (4.693—8.091)	2.407 (2.124—2.862)	7.729 (5.721—8.698)

18

[40]

[41-43]

[44]

Monte Carlo Bootstrap

Bootstrap 2 000

[45]

4 结论

18

18

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