

Dietary Exposure of the Chinese Population to Acrylamide*

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Abstract

Objective To assess the current status of the acrylamide in the Chinese food supply, the dietary acrylamide exposure in the Chinese population and to estimate the public health risks of the current consumption.

Methods The acrylamide content in the total diet study (TDS) food samples was analyzed using an LC-MS/MS method. Based on the analytical results, the dietary exposure calculations were performed using a deterministic method, combining mean acrylamide concentrations from the food group composite with their associated food consumptions.

Results Acrylamide was detected in 43.7% of all samples collected and acrylamide concentration varied from ND to 526.6 $\mu\text{g}/\text{kg}$. The estimated dietary intakes of acrylamide among Chinese general population given as the mean and the 95th percentile (P95) were 0.286 and 0.490 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$, respectively. The margins of exposure (MOEs) for the population calculated using both benchmark dose lower confidence limit for a 10% extra risk of tumors in animals (BMDL₁₀) 0.31 and 0.18 $\text{mg}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$, were 1069 and 621 for the mean dietary exposure, and 633 and 367 for the high dietary exposure respectively.

Conclusion These MOE values might indicate a human health concern on acrylamide for Chinese population. Efforts should continue to reduce acrylamide levels in food in order to reduce the dietary risks to the human health.

Key words: Acrylamide; Dietary exposure; Assessment; Health risk; Total diet study

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INTRODUCTION

Acrylamide, an important industrial chemical, is considered to have neurotoxic and carcinogenic effects *in vitro*. It is also listed as "probably carcinogenic to humans" under the Group 2A by the International Agency for Research on Cancer (IARC)^[1]. In 2002,

acrylamide was found to be present in certain fried, baked and deep-fried foods with high concentrations, and subsequently it was found in coffee which caused worldwide attention due to its probable carcinogenic effect^[2]. It is now well known that acrylamide is formed as a result of Maillard reaction between reducing sugars and free amino acids, such as asparagines, at high temperatures^[3-5].

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Before high levels of acrylamide being found in heat treated foods, a potential exposure of acrylamide had only been detected in drinking water induced by polyacrylamide in the water refining process^[6-7]. Although maximum limits for acrylamide in foods have not been established, the Australian National Health and Medical Research Council (NHMRC, NRMCC) guideline states that acrylamide in drinking water should not exceed $0.2 \mu\text{g/L}$ ^[8]. According to World Health Organization (WHO) guideline for acrylamide maximum level in drinking water is $0.5 \mu\text{g/L}$ ^[9]. Due to multiple sources of dietary exposure to acrylamide, a consultation was held in June 2002 by the Food and Agriculture Organization/World Health Organization (FAO/WHO) to discuss possible health risks due to acrylamide^[10]. Based on the expert committee evaluation, $1 \mu\text{g}\cdot\text{kg}^{-1} \text{ bw}\cdot\text{day}^{-1}$ is considered as an average exposure to acrylamide while $4 \mu\text{g}\cdot\text{kg}^{-1} \text{ bw}\cdot\text{day}^{-1}$ is considered as high exposure to acrylamide^[11]. The mean dietary exposure range to acrylamide is $0.2\text{-}1.0 \mu\text{g}\cdot\text{kg}^{-1} \text{ bw}\cdot\text{day}^{-1}$ for the general adult population while 95th percentile range is $0.6\text{-}1.8 \mu\text{g}\cdot\text{kg}^{-1} \text{ bw}\cdot\text{day}^{-1}$ ^[12]. Since 1990, China has conducted the Total Dietary Study (TDS), also known as the Market Basket Study. Up to now TDS has been conducted in four occasions^[13-14]. The purpose of the TDS is to monitor the trend of dietary exposure to various substances, involving mainly five steps: purchasing retail foods commonly consumed by the population, preparing them in table ready form, homogenizing and compositing them, analyzing a range of concerned chemicals, including acrylamide^[15], pesticide residues, industrial and elemental contaminants, nutrients in foods, and finally, the estimating intakes of these substances from diets. It also exhibited the exposure trends over time. The TDS has become an important tool for monitoring dietary exposure to chemicals and the associated risks to human health in China.

The purpose of the current study was to estimate the dietary exposure of Chinese general population to acrylamide and to assess its associated health risks. Finally, the intake data from this study is also compared with a previous survey performed in 2000.

MATERIALS AND METHODS

Sampling and Preparation

The sampling strategy of the current study is

consistent with WHO Total Diet Study (TDS)^[16-17].

The sampling method used was described elsewhere, even though with some modifications^[13-14,18]. Briefly, since Chinese Diet is diverse among different ethnic groups and different geographical location, a multistage random cluster sampling method was used in this study. The whole country was divided into four area groups according to their geographical positions, dietetic habits and cooking styles. Each group comprised three provinces or a municipality, such as North 1, including Heilongjiang, Liaoning, and Hebei provinces; North 2, including Ningxia, Henan, and Shanxi provinces; South 1, including Shanghai municipality, Jiangxi, and Fujian provinces, and South 2, including Hubei, Guangxi, and Sichuan provinces. The food composite approach was used to investigate the total diet in above provinces and municipality which represented the typical dietary patterns in the Mainland China. Food samples were purchased from local markets, grocery stores, and local farms in sites in group during different seasons, from spring to autumn in 2007, and then prepared as consumed. The prepared foods were then mixed to form different composites (Table 1) similar as the average daily consumption for the respective populations. Therefore, twelve food composites were available for each province.

Food Consumption Data

The consumption data was collected by Chinese Center for Disease Control and Prevention (Chinese CDC) in 2000 using TDS's approach which were described previously^[18-19]. Briefly, the food consumption data in each of the twelve provinces or municipality were collected based on a 3-day household dietary survey and 24-h recalls. The average food consumption of a reference person (adult, male, 18-45 years old, 63 kg body weight and light physical activity) for each household was calculated based on the total household food consumption. High end consumption data was recorded by a single member of each surveyed household by 24-h recalls.

Chemical Analysis

Acrylamide analysis was carried out in the laboratory of Jiangsu Province Center for Disease Control and Prevention & Key Laboratory of Food Safety Risk Assessment, Ministry of Health. Acrylamide was extracted from food or water before a clean-up of the extracted on Solid Phase Extraction

combing Oasis HLB and BondElut Accucat cartridges as described by the United States Food and Drug Administration (US FDA) methodology^[20].

The final acrylamide extracts obtained were injected and analyzed by the Liquid Chromatography Tandem Mass Spectrometry (LC-MS-MS) method. The limits of detection (LOD) for liquid samples and solid samples were 0.3 and 1.2 µg/kg, respectively.

Quality Control

The method of acrylamide determination in food was validated. The laboratory participated in inter-comparison studies organized by Food Analysis Performance Assessment Scheme (FAPAS) of the Central Science Laboratory York (UK) analyzing a potato crisp (1404.0±140.4 µg/kg) test material in 2005 and received Z-score 0.5. Furthermore, analytical quality control was also implemented by the use of an in-house prepared reference material (potato crisps). Acrylamide in the in-house prepared reference material (potato crisps) was determined as 37.1±5.4 µg/kg (n=12). This material was used as a control in each series of analysis.

Dietary Exposure Estimates

The dietary exposures for Chinese general population were calculated using a deterministic assessment. For exposure calculations, levels of acrylamide below LOD were assumed to be equal to half of LOD for derive mean concentrations^[21]. The total intake was then calculated by summing the intakes from twelve food groups containing acrylamide. All estimated intakes were adjusted for the individual's self-reported body weight (bw) and expressed as daily intake in "µg·kg⁻¹ bw·day⁻¹".

Risk Assessment of Dietary Exposure to Acrylamide

JECFA (The Joint FAO/WHO Expert Committee on Food Additives) noted that the principal site of toxic action of acrylamide was the nervous system according to numerous studies in a range of laboratory animal species and epidemiological accounts of human industrial and accidental exposure^[12]. A No Observed Adverse Effect Level (NOAEL) for acrylamide induced morphological changes in nerves, detected by electron microscopy, was 0.2 mg·kg⁻¹ bw·day⁻¹ in subchronically exposed rats^[7]. Acrylamide is classified as a probable human carcinogen^[1]. No quantitative risk estimation was performed for that kind of substances and dietary intake should remain as low as possible. Mean

exposure and P95 were compared with BMDL₁₀ by calculating the corresponding MOEs for risk characterization purposes.

RESULTS

Levels of Acrylamide in Twelve Food Composites

Of the total 144 food composites samples, 43.7% were detected with acrylamide. Table 1 summarizes the results from 12 food groups. Acrylamide amounts ranged from ND to 526.6 µg/kg in all samples. The food groups with relatively high mean acrylamide concentrations were sugar (72.1 µg/kg; range: ND-526.6 µg/kg), followed by potatoes (31.0 µg/kg; range: 2.4-109.0 µg/kg), vegetables (22.3 µg/kg; range: 3.6-101.5 µg/kg), meats (12.3 µg/kg; range: 2.2-44.0 µg/kg), aquatic foods (11.4 µg/kg; range: ND-48.5 µg/kg), legumes and nuts (10.6 µg/kg; range: ND-45.7 µg/kg) and cereals (6.0 µg/kg; range: ND-33.0 µg/kg). Acrylamide was not detected in the eggs, milk, fruits, water, beverages and alcoholic beverages.

Dietary Exposure Assessment to Acrylamide

Mean exposure as well as the P95 were calculated for general population. The daily exposures to acrylamide from each food group in the twelve provinces in four TDS regions are presented in Table 2. The acrylamide total intakes of twelve provinces ranged from 0.056 to 0.645 µg·kg⁻¹ bw·day⁻¹, with overall average of 0.286 µg·kg⁻¹ bw·day⁻¹. The P95 was found to be 0.490 µg·kg⁻¹ bw·day⁻¹.

Figure 1 showed the comparison of contribution of food groups in different regions. The vegetable group was the predominant contributor in each region with the percentage proportion ranging from 14% to 69%. The rest of dietary exposures were mainly from the cereals, the potatoes, meats and legumes and nuts groups with varying contributions from 1.8% to 52%.

As can be seen from Figure 2, for general population, the main food group contributors to exposure were vegetables products (48.4%), cereals (27.1%), potatoes (8.0%), meats (6.6%) and Legumes and nuts (5.8%).

Risk Assessment of Dietary Exposure to Acrylamide

Numerous animal experiment studies have shown that the nervous system was a principal site of the toxic actions of acrylamide^[11]. For the general

Table 1. Levels of Acrylamide in 12 Group Composites from 12 Provinces in Chinese TDS in 2007 (µg/kg)

Food Composites	North 1				North 2			South 1			South 2		Mean*
	Heilongjiang	Liaoning	Hebei	Shanxi	Henan	Ningxia	Shanghai	Fujian	Jiangxi	Hubei	Sichuan	Guangxi	
Cereals	13.2	ND	16.1	ND	ND	5.5	ND	33.0	ND	ND	ND	ND	6.0
Legumes and nuts	4.7	ND	22.5	ND	11.0	ND	4.8	45.7	15.9	4.5	2.7	14.1	10.6
Potatoes	6.1	14.2	109.0	3.6	33.0	19.9	2.4	87.8	36.0	19.9	28.3	11.6	31.0
Meats	7.5	7.9	8.3	2.2	7.5	5.7	22.3	44.0	13.0	5.3	14.4	9.9	12.3
Eggs	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.2
Aquatic foods	ND	ND	23.2	13.4	ND	ND	12.1	19.3	48.5	7.4	ND	9.3	11.4
Milk	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.2
Vegetables	8.1	4.0	4.3	8.6	3.6	54.7	20.9	5.9	101.5	20.9	23.0	11.9	22.3
Fruits	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.2
Sugar	ND	ND	267.9	14.5	ND	ND	15.9	8.0	13.7	ND	15.7	526.6	72.1
Water and beverage	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.2
Alcoholic beverage	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.2

Note.* For acrylamide, if measured concentration was less than LOD, then 1/2 LOD was used in calculations.

Table 2. Daily Intake of Acrylamide from Each Food Group in 2007 Chinese TDS (µg·kg⁻¹·bw·day⁻¹)

Food groups	North 1				North 2				South 1				South 2		Whole Country	
	Heilongjiang	Liaoning	Hebei	Shanxi	Henan	Ningxia	Shanghai	Fujian	Jiangxi	Hubei	Sichuan	Guangxi	Mean	P95		
Cereals	0.134	0.005	0.269	0.012	0.021	0.055	0.009	0.361	0.012	0.012	0.009	0.016	0.076	0.062		
Legumes and nuts	0.004	0.001	0.098	0.001	0.012	0.000	0.007	0.037	0.016	0.003	0.007	0.011	0.016	0.024		
Potatoes	0.005	0.033	0.076	0.004	0.024	0.049	0.001	0.028	0.008	0.012	0.027	0.004	0.023	0.073		
Meats	0.006	0.009	0.006	0.001	0.007	0.004	0.049	0.085	0.012	0.005	0.026	0.014	0.019	0.043		
Eggs	0.001	0.001	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.001		
Aquatic foods	0.000	0.000	0.006	0.001	0.000	0.000	0.012	0.042	0.007	0.005	0.000	0.002	0.006	0.027		
Milk	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000		
Vegetables	0.048	0.035	0.025	0.036	0.018	0.376	0.126	0.032	0.588	0.177	0.125	0.051	0.136	0.249		
Fruits	0.003	0.002	0.002	0.001	0.001	0.002	0.003	0.001	0.000	0.001	0.000	0.001	0.001	0.005		
Sugar	0.000	0.000	0.009	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.001	0.006	0.002	0.004		
Water and beverage	0.001	0.000	0.002	0.001	0.001	0.000	0.003	0.001	0.001	0.002	0.001	0.002	0.001	0.002		
Alcoholic beverage	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000		
Total	0.202	0.085	0.493	0.056	0.084	0.487	0.213	0.587	0.645	0.218	0.198	0.107	0.286	0.490		

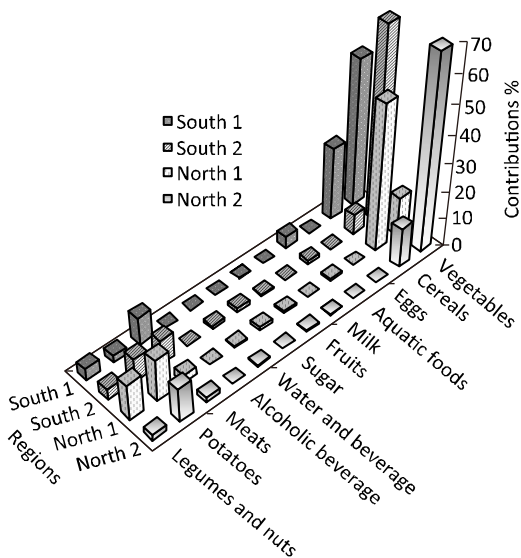


Figure 1. The comparison of contribution (% of Daily Intake) of food groups in different regions in 2007 Chinese TDS.

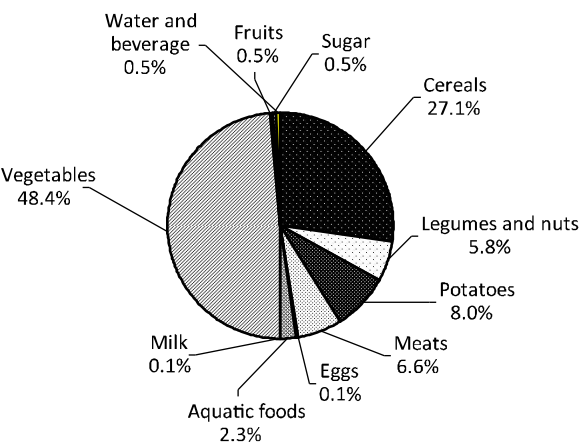


Figure 2. Contribution (% of Daily Intake) of various food groups to dietary acrylamide intakes for Chinese general population.

population and consumers with high exposure in China, the MOE values calculated relative to the NOAEL of $0.2 \text{ mg}\cdot\text{kg}^{-1} \text{ bw}\cdot\text{day}^{-1}$ (for the most sensitive non-carcinogenic end-point morphological changes in nerves, detected by electron microscopy, in rats) were 690 and 408, respectively^[12]. According to the principle of the most conservative estimate, when average and high dietary exposure were compared with the BMDL₁₀ of $0.31 \text{ mg}\cdot\text{kg}^{-1} \text{ bw}\cdot\text{day}^{-1}$ for the induction of mammary tumors in female rats^[12], the MOE values were 1069 and 633, respectively. For Harderian gland tumors in male mice, the BMDL₁₀

was $0.18 \text{ mg}\cdot\text{kg}^{-1} \text{ bw}\cdot\text{day}^{-1}$ ^[12] and the MOE values were 621 and 367 for average and high exposures, respectively.

DISCUSSION

As shown in Table 1, acrylamide levels in sugar varied significantly. The highest concentration of acrylamide in sugar was found from Guangxi province ($526.6 \text{ }\mu\text{g}/\text{kg}$), followed by Hebei province ($267.9 \text{ }\mu\text{g}/\text{kg}$) in this study. Most of samples from other provinces had low or even non-detected levels. The high acrylamide levels found in sugar may be explained by the use of polyacrylamide as flocculants in the sugar industry. The polyacrylamide itself contained a certain amount of unpolymerized acrylamide monomer and could contaminate the sugar during processing. A variety of factors may cause the degradation of polyacrylamide to low polyacrylamide or monomer^[22]. In addition, acrylamide was found in brown sugar but not white sugar by tracing back to the source. Brown sugar is unrefined product that consists of impurities, such as asparagine, which may possibly explain its high acrylamide content in brown sugar^[23]. Wei et al. detected the high acrylamide content in 4 brown sugars ranged from $347\text{-}1107 \text{ }\mu\text{g}/\text{kg}$ ^[23], where the value was consistent with the level found in this TDS. Similar results were also reported by JECFA^[12]. JECFA summarized that mean concentration of acrylamide in refined and unrefined sugar was 86 and $445 \text{ }\mu\text{g}/\text{kg}$, respectively; the reported maximum concentration was 438 and $2300 \text{ }\mu\text{g}/\text{kg}$, respectively from 2004 to 2009.

The mean levels of acrylamide measured in sugar were the highest in the food groups. However, the sugar consumption remained very low, and therefore the contribution of the sugar was low in China (Table 2).

The higher mean acrylamide concentrations were also found in potatoes and vegetables. The highest concentration of acrylamide in potatoes was found from Hebei province ($109.0 \text{ }\mu\text{g}/\text{kg}$) while the highest concentration in vegetables was from Jiangxi province ($101.5 \text{ }\mu\text{g}/\text{kg}$). Different cooking method used may results in different acrylamide content in potatoes and vegetables. Starchy foods such as potatoes, bread, cereals and other carbohydrate-containing food or low protein plant foods when heated to over $120 \text{ }^{\circ}\text{C}$ tend to produce acrylamide^[24]. In brief, heating temperature, types and amounts of carbohydrates and amino acids, and other, partly as

yet unknown factors, cause a large variation in the amount of acrylamide formed in a particular food product. The acrylamide levels detected in cereals, legumes and nuts, potatoes, aquatic foods, vegetables especially the meats and sugar, were higher in 2007 compared to 2000. On the other hand its levels found in the alcohol beverage were lower in 2007 than those in 2000. This can be explained by the list of foods composited varied in the same categories of food groups in 2000 and 2007. The retained individual foods representing the same food category were aggregated into 12 general food groups (such as "cereals").

Results from the current survey were, in most cases, consistent with typical results from international studies, although there were variations in acrylamide levels in food group in 2000 and 2007. The food groups analysed from various countries and their mean acrylamide concentrations were shown as potato and potato products (532 µg/kg), cereals (273 µg/kg), fish and seafood (64 µg/kg), vegetables (52 µg/kg), meats and offals (42 µg/kg) and eggs (18 µg/kg)^[12]. In comparing global mean acrylamide levels for commodity groups with the levels obtained at the JECFA seventy-second meeting, the acrylamide level in Chinese diets except cereals and potatoes were in the same range as published in other Western countries^[12]. Different levels of acrylamide were observed in cereals and potatoes, which can be mainly ascribed to different cooking temperatures and different raw materials^[25]. Due to cereals and potatoes were mostly cooked by steaming at about 100 °C, lower levels were found in these two kinds of foodstuffs in China than other Western countries.

The vegetables and cereals were the major contribution to the food category of dietary acrylamide exposure in this study. The pattern of contributing foods in present study differed from other international studies. Its large daily consumption in vegetables and cereals explains its importance as source of acrylamide exposure in the Chinese diet. The major foods contributing to the total mean dietary exposure for most other countries were potato chips (10%-60%), potato crisps (10%-22%), bread and rolls/toast (13%-34%) and pastry and sweet biscuits (10%-15%)^[26-29]. Different patterns of food consumption may represent an additional influencing factor, especially regarding the consumption of cereals. Chinese dietary pattern belongs to the Oriental dietary patterns which are dominated by plant food

including cereals and vegetables. However, the proportion of grain and vegetables is low in the structure of the diet in Western countries. According to Matthys et al., the mean consumption of cereals by Chinese population is about threefold than that in Flemish adolescents^[30]. It should be noted that the common cooking method for vegetables are different in Western, they eat the salad or cook them in water for longer time.

The important contribution of vegetables category to the acrylamide exposure was based both on a high consumption of the foods and on a high content of acrylamide in this study. However, the contribution rate of the individual vegetable food to dietary acrylamide exposure was ambiguous when compared the combined with the classification of vegetables in the study. The reason was that in order to achieve more realistic dietary exposure estimates in the total diet study, the foods including vegetables composites were prepared and cooked to a "table ready" state according to local cuisine and then blended to form the respective group composites with weights proportional to the average daily consumption of each province. Vegetables were cooked when applicable (braised, fried, boiled etc.). The category, proportion and cooking method of vegetable might result different levels of acrylamide in vegetables composites sample. For example, the vegetables for inclusion in food list from Jiangxi province and percentage of consumed weight were fried long bean (23.5%), fried swamp cabbage (17.6%), boiled loofah soap (14.7%), fried eggplant (12.7%), fried green or red pepper (12.2%), fried cucumber (6.1%), raw tomato (3.7%), fried loofah (3.3%), fried dried radish (3.2%), fried tomato (1.6%), boiled seaweed soap (0.9%), and fried seaweed (0.6%), respectively. The vegetables for inclusion in food list from Henan province and percentage of consumed weight were fried beans (15.2%), raw cucumber (11.4%), fried little Chinese cabbage (10.1%), fried squash (10.9%), fried tomato, (9.0%), fried eggplant (8.9%), fried Chinese watermelon (8.5%), fried loofah (6.5%), stewed Chinese watermelon (5.3%), fried Chinese cabbage (4.9%), fried round green peppers (4.3%), fried white radish (2.0%), fried mushroom (1.6%), salted mustard, (1.2%), respectively. The kinds of the vegetables were more than 10, with weights proportional to the average daily consumption and cooking methods for inclusion in food list from other provinces. The classification of vegetables only provided a little information in this study and the further research on

the acrylamide contents in the vegetables should be needed.

Acrylamide dietary exposure was based on the combination of consumption data from the 2000 TDS food consumption survey and contamination data from 2000 and 2007 TDS in China. In previous 2000 Chinese TDS, the acrylamide intakes were estimated only for four regions rather than individual province. Therefore, summarization of the acrylamide intakes from composites foods of four regions were compared between 2000 and 2007 Chinese TDS (Table 3). The acrylamide total intake in people from South 1 (0.482 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$) was much higher than that from North 1, North 2, and South 2, which were 0.260, 0.209, and 0.174 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$, respectively in 2007 TDS. These results may be explained to the acrylamide intakes from vegetables and meats in South 1 were both higher than those in other three regions. With comparison to the result of 2000 TDS, the average total intake of acrylamide in the whole country increased by 53% in 2007. Such acrylamide exposure difference might be explained due to the increasing acrylamide levels in vegetables and meats over the past 7 years. Besides, the acrylamide levels in sugar, potatoes, aquatic foods, legumes and nuts and cereals also tended to increase but the levels in other types of food did not change significantly. In 2000 and 2007, the differences of the intake of acrylamide in some food mainly were various acrylamide levels in some food because the consumption data was the same.

Two highest acrylamide intakes were found in vegetables from Jiangxi (0.588 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$) and in cereals from Fujian (0.361 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$) (Table

2), mainly due to their higher content of acrylamide contaminant than that of other provinces. All estimated dietary exposure of twelve provinces were lower than the exposure estimated by JECFA in 2011 (for the general adult population mean intake 0.2-1.0 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$ and high intake 0.6-1.8 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$ ^[12]). The results revealed large difference in the acrylamide intakes among provinces, which is mainly due to differences in cooking conditions, dietary habits, culture, and economic level among provinces. Estimated mean and high percentile exposure for the Chinese general population were 0.286 and 0.490 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$, respectively (Table 2), which were lower than the values obtained in the JECFA Seventy-second meeting^[12]. The general population mean and high percentile exposure results were also lower than the results from other national estimations, such as France^[26], Ireland^[31], Netherlands^[32], Poland^[28], Spain^[29], Sweden^[33], and the United States^[34] (Table 4).

The MOE values in present study similar to the conclusion made at the JECFA Seventy-second meeting, the adverse neurological effects were unlikely at the estimated average and high exposure levels. For the general population and consumer with high exposure, the MOE was far greater than the conclusion made at the JECFA Seventy-second meeting^[12]. The calculated MOEs, based on BMDL defined for carcinogenic effects, kept a safe distance not big enough for the general Chinese population and high exposure consumer, indicating a health concern. Efforts should continue to reduce acrylamide levels in food in order to reduce the dietary risks to the human health.

Table 3. Comparative Daily Intake of Acrylamide from Each Food Group from 2000 and 2007 Chinese TDS ($\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$)

Food Groups	North 1		North 2		South 1		South 2	
	2000	2007	2000	2007	2000	2007	2000	2007
Cereals	0.040	0.136	0.013	0.030	0.009	0.127	0.132	0.013
Legumes and nuts	0.035	0.034	0.007	0.004	0.004	0.020	0.006	0.007
Potatoes	0.044	0.038	0.026	0.026	0.003	0.012	0.018	0.014
Meats	0.000	0.007	0.000	0.004	0.001	0.049	0.001	0.015
Eggs	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Aquatic foods	0.001	0.002	0.000	0.000	0.001	0.020	0.000	0.003
Milk	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Vegetables	0.146	0.036	0.158	0.143	0.039	0.248	0.061	0.118
Fruits	0.002	0.002	0.001	0.001	0.001	0.001	0.001	0.001
Sugar	0.000	0.003	0.000	0.000	0.000	0.001	0.000	0.002
Water and beverage	0.000	0.001	0.000	0.001	0.000	0.002	0.000	0.002
Alcoholic beverage	0.001	0.000	0.001	0.000	0.002	0.000	0.002	0.000
Total	0.270	0.260	0.207	0.209	0.052	0.482	0.221	0.174

Table 4. Daily Intake of Acrylamide ($\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$) in Different Countries by General Population

Country	Population Group	Average Exposure	95 th Percentile Exposure	Reference
China	Adults (≥ 15 years)	0.29	0.49	This study
France	Adults (> 15 years)	0.43	1.02	Sirost et al., 2012 ^[26]
Ireland	Adults (18-64 years)	0.6	na	Mills et al., 2012 ^[31]
Netherlands	Whole population (1-97 years)	0.48	0.6	Konings et al., 2012 ^[32]
Poland	Adults(19-96 years)	0.33	0.69	Mojska et al., 2012 ^[28]
Spain	Whole population	0.2	na	Rufian-Henares et al., 2012 ^[29]
Sweden	Population (18-74 years)	0.46 [*]	0.98	Svensson et al., 2012 ^[33]
The United States	Males (20- years)	0.39	0.91	Tran et al., 2012 ^[34]

Note. ^{*} Based on standard body weight of 63 kg.

In the risk assessment of acrylamide, animal experiments used to derive BMDL data, the large differences in the metabolism between the animals and human, limited investigations on which the intake calculation were based, and therefore were uncertainties. Validated biomarkers of internal exposure would provide a better exposure assessment^[12].

In conclusion, the present study estimated acrylamide dietary exposures in the general population based on the results of the fourth Chinese TDS. The average and P95 exposures estimated for the general population were 0.286 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$ and 0.490 $\mu\text{g}\cdot\text{kg}^{-1}\text{ bw}\cdot\text{day}^{-1}$ in 2007, respectively. The current dietary intake of acrylamide showed a notable increase compared with those in the previous study performed in 2000. The calculated MOE values might indicate a human health concerns due to carcinogenic effects of acrylamide in Chinese diet.

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REFERENCES

1. International Agency for Research on Cancer (IARC). Monographs on the Evaluation of Carcinogenic Risks to Humans: Some Industrial Chemicals No. 60.1994. IARC, Lyon.

2. Tareke E, Rydberg P, Karlsson P, et al. Analysis of acrylamide, a carcinogen formed in heated foodstuffs. J Agric Food Chem, 2002; 50, 4998-5006.

3. Zyzak DV, Sanders RA, Stojanovic M, et al. Acrylamide formation mechanism in heated foods. J Agric Food Chem, 2003; 51, 4782-7.

4. Becalski A, Lau BP, Lewis D, et al. Acrylamide in foods: occurrence, sources, and modeling. J Agric Food Chem, 2003; 51, 802-8.

5. Mottram DS, Wedzicha BL, Dodson AT, et al. Acrylamide is formed in the Maillard reaction. Nature, 2002; 419, 448-9.

6. Brown L, Rhead M. Liquid chromatographic determination of acrylamide monomer in natural and polluted aqueous environments. Analyst, 1979; 104, 391-9.

7. Burek JD, Albee RR, Beyer JE, et al. Subchronic toxicity of acrylamide administered to rats in the drinking water followed by up to 144 days of recovery. J Environ Pathol Toxicol, 1980; 4, 157-82.

8. National Health and Medical Research Council (NHMRC) and Natural Resource Management Ministerial Council (NRMCMC). Australian Drinking Water Guidelines Paper 6 National Water Quality Management Strategy. 2011. Commonwealth of Australia, Canberra.

9. WHO. Guidelines for drinking-water quality, third edition, incorporating first and second addenda, Vol.1, Recommendations. In 3rd ed. 2008. WHO, Geneva.

10.WHO. Health Implications of Acrylamide in Food: Report of a Joint FAO/WHO Consultation. 2002. WHO, Geneva.

11.The Joint FAO/WHO Expert Committee on Food Additives (JECFA). Evaluation of certain food contaminants: sixty-fourth report of the Joint FAO/WHO Expert Committee on Food Additives. 2006. WHO, Geneva.

12.The Joint FAO/WHO Expert Committee on Food Additives (JECFA). Evaluation of certain contaminants in food: seventy-second [72nd] report of the Joint FAO/WHO Expert Committee on Food Additives. 2011. WHO, Geneva.

13.Chen J, Gao J. The Chinese total diet study in 1990. Part I. Chemical contaminants. J AOAC Int, 1993; 76, 1193-205.

14.Li XW, Wu YN, Chen JS. The development and evolution of Chinese Total Diet Study. Chinese Journal of Epidemiology, 2011; 32, 456-9. (In Chinese)

15.WHO. 2005, Total Diet Studies: a recipe for safer food, available from:http://www.who.int/foodsafety/publications/chem/en/gems_brochure.pdf.

16.Global Environment Monitoring System - Food Contamination Monitoring and Assessment Programme (GEMS/Food). Introduction to GEMS/Food, available from:<http://www.who.int/foodsafety/chem/gems/en/>.

- 17.WHO. 2006, GEMS/Food Total Diet Studies; Food safety consultations. Report of the 4th International Workshop on Total Diet Studies Beijing, China 23-27 October 2006, available from: http://www.who.int/foodsafety/publications/chem/TDS_Beijing_2006_en.pdf.
- 18.Wu Y, Li X, Chang S, et al. Variable iodine intake persists in the context of universal salt iodization in China. *J Nutr*, 2012; 142, 1728-34.
- 19.Zhou P, Zhao Y, Li J, et al. Dietary exposure to persistent organochlorine pesticides in 2007 Chinese total diet study. *Environ Int*, 2012; 42, 152-9.
- 20.The U.S. Food and Drug Administration (FDA). Detection and Quantitation of Acrylamide in Foods, <http://www.fda.gov/Food/FoodSafety/FoodContaminantsAdulteration/ChemicalContaminants/Acrylamide/ucm053537.htm>, 2002.
- 21.WHO. GEMS/Food-EURO Second Workshop on Reliable Evaluation of Low-Level Contamination of Food: report on a workshop in the frame of GEMS/Food-Euro. 1995. WHO, Kulmbach.
- 22.Ou XH, Huang LH, He CJ, et al. Determination of Acrylamide in Sugar by Solid Phase Extraction High Performance Liquid Chromatography. *Food and Fermentation Industries*, 2007; 118-20. (In Chinese)
- 23.Wei CC, Ya MK, Daniel Yang, et al. Validation of an improved LC/MS/MS method for acrylamide analysis in foods. *J Food Drug Anal*, 2009; 17, 190-7, 231.
- 24.Stadler RH, Blank I, Varga N, et al. Acrylamide from Maillard reaction products. *Nature*, 2002; 419, 449-50.
- 25.Leung KS, Lin A, Tsang CK, et al. Acrylamide in Asian foods in Hong Kong. *Food Addit Contam*, 2003; 20, 1105-3.
- 26.Sirot V, Hommet F, Tard A, et al. Dietary acrylamide exposure of the French population: results of the second French Total Diet Study. *Food Chem Toxicol*, 2012; 50, 889-94.
- 27.Arisseto AP, Figueiredo Toledo MC. de, Govaert Y, et al. Contribution of selected foods to acrylamide intake by a population of Brazilian adolescents. *Food Science and Technology*, 2009; 42, 207-11.
- 28.Mojcka H, Gielecinska I, Szponar L, et al. Estimation of the dietary acrylamide exposure of the Polish population. *Food Chem Toxicol*, 2010; 48, 2090-6.
- 29.Rufian-Henares JA, Arribas-Lorenzo G, Morales FJ, Acrylamide content of selected Spanish foods: survey of biscuits and bread derivatives. *Food Addit Contam*, 2007; 24, 343-50.
- 30.Matthys C, Bilau M, Govaert Y, et al. Risk assessment of dietary acrylamide intake in Flemish adolescents. *Food and Chemical Toxicology*, 2005; 43, 271-8.
- 31.Mills C, Tlustos C, Evans R, et al. Dietary acrylamide exposure estimates for the United Kingdom and Ireland: comparison between semiprobabilistic and probabilistic exposure models. *J Agric Food Chem*, 2008; 56, 6039-45.
- 32.Konings EJ, Baars AJ, van Klaveren JD, et al. Acrylamide exposure from foods of the Dutch population and an assessment of the consequent risks. *Food Chem Toxicol*, 2003; 41, 1569-9.
- 33.Svensson K, Abramsson L, Becker W, et al. Dietary intake of acrylamide in Sweden. *Food Chem. Toxicol*, 2003; 41, 1581-6.
- 34.Tran NL, Barraj LM, Murphy MM, et al. Dietary acrylamide exposure and hemoglobin adducts-National Health and Nutrition Examination Survey (2003-04). *Food Chem Toxicol*, 2010; 48, 3098-108.