



## Food safety risk assessment for estimating dietary intake of sulfites in the Taiwanese population



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### ABSTRACT

The purpose of this study was to assess the health risk associated with dietary intake of sulfites for Taiwanese general consumers by conducting a total diet study (TDS). We evaluated the exposure of Taiwanese to sulfites in the diet and its associated health risk. This study used a list of 128 food items representing 83% of the total daily diet. Among the 128 food items, 59 items may contain sulfites. Samples of the 59 food items were collected and subjected to chemical analysis to determine the sulfur dioxide concentration. Health risk was assessed by calculating the ratio of exposure level to the acceptable daily intake (ADI) level of the analyte. For high-intake consumers, the HI of sulfites was 19.7% ADI for males over the age of three years at the 95th percentile; whereas for females over the age of 66, the HI was 17.8% ADI. The HI for high-intake consumers was above 10% ADI. This suggests that regulatory actions must be continued and that consumers should be advised to be aware of processed foods with relatively high contamination to avoid excessive exposure.

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## 1. Introduction

The total diet study (TDS) has been used as a national monitoring research tool in assessing the associated health risk from dietary exposure to specific analytes [27,15,6]. TDS can be used to determine the levels of various contaminants and nutrients present in foods and to estimate public health risk due to chronic exposure to

chemical substances [22]. They aim to estimate chronic risk to public health due to chemical substances. Approximately 33 countries, including Australia, New Zealand, Canada, China, France, Taiwan, The Netherlands, the United Kingdom, and the United States of America, have carried out TDS or TDS-like studies [7].

Sulfites are compounds that contain the sulfite ion ( $\text{SO}_2^-$ ). Sulfur dioxide has been used since ancient times for its cleansing, disinfecting, and purifying properties. In addition, sulfites have a number of technological uses, for example, as antioxidant, bleaching agents, flour treatment agents, and preservatives. Sulfites are permitted in various foods such as wine, cordials, and dried fruit and vegetables. They are used in the food industry to maintain food color, to prolong shelf life, and to prevent microbial growth [11,19,37,34]. Sulfites are also used in the production of some food packaging materials and as processing aids for sterilizing bottles prior to packaging food or drink. Food is therefore a major source of sulfites. Sulfites may be present in food as sulfurous acid, inorganic sulfites, and other forms bound to the food matrix.

Sulfur dioxide used as a food additive in food for human consumption is generally recognized as safe when used in accordance with good manufacturing practice [35]. However, sulfites can trig-

*Abbreviations:* ADI, acceptable daily intake; ADD, average daily dose; BW, body weight; CAC, codex alimentarius commission; C, concentration; CR, consumption rate; COX-2, cyclooxygenase-2; EGF, epidermal growth factor; EGFR, epidermal growth factor receptor; EFSA, European Food Safety Authority; FAO, Food and Agriculture Organization; FSANZ, Food Standards Australia New Zealand; FSAI, food safety authority of Ireland; HI, hazard index; IARC, International Agency for Research on Cancer; ISO, International Organization for Standardization; JECFA, Joint FAO/WHO Expert Committee on Food Additives; LOD, limit of detection; LOQ, limit of quantitation; NOEL, no observed effect level; NAHSIT, nutrition and health survey in Taiwan; TFDA, Taiwan Food and Drug Administration; SCF, The Scientific Committee for Food; TDS, total diet study; ND, undetected; USFDA, US Food and Drug Administration; WHO, World Health Organization.

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ger asthma and other symptoms of allergic responses such as skin rashes and irritations in sulfite-sensitive people [39,9].

Sulfites were selected for evaluation in the 21st Australian TDS. FSANZ results show that the mean estimated dietary exposure to sulfites for all population groups is well below the acceptable daily intake (ADI). However, the 95th percentile of estimated dietary exposures to sulfites exceed the ADI for most population groups assessed, ranging from approximately 80% of the ADI for girls aged 13–18 years to approximately 280% of the ADI for boys aged two to five years [10]. The results of the 21st Australian TDS show a potential public health and safety concern for individuals with above-average consumption.

The Codex Committee on Food Additives and Contaminants and the Codex Committee on Contaminants in Food at its 38th session placed a dietary exposure assessment of sulfites on the priority list for evaluation by the [17].

Additives categorized under sulfites in the current Taiwan “Standards for Specification, Scope, Application and Limitation of Food Additives” list are sulfur dioxide, sodium sulfite, potassium sulfite, sodium sulfite (anhydrous), sodium metabisulfite, potassium bisulfite, and sodium bisulfite [31]. These sulfites are added to foods such as processed dried fruit, wine, beer, fruit, vegetable juices, drinks, processed fish, and seafood for the major purposes of preservation and inhibition of browning reactions [1,25,32].

Sulfur dioxide is traditionally used as an antioxidant and preservative in many foodstuffs. The TFDA surveys of commercially available foods found that the illegal use of preservatives and addition of bleaching agents is very common in Taiwan [29,18]. A local Taiwanese health bureau, for instance, analyzed sulfur dioxide from February to December in 2006. Among the 377 samples they collected, 15.1% tested positive for the bleaching agent sulfur dioxide. For example, zongzi (a glutinous rice dumpling wrapped in bamboo leaves), small dried shrimp, dried mushroom, and zongzi leaves have been found to contain more sulfur dioxide than what is allowed [38].

Sulfur dioxide is very often found to exceed permissible limits for their use. This problem not only poses a risk to public health, but also endangers the trade economy. Because of the growing concern on sulfur dioxide in processed food exceeding its ADI, we investigated sulfur dioxide concentrations and estimated the exposure of populations. As dietary composition and intake patterns in Taiwan are distinctly different from those in western countries where similar studies have been reported, the specific objectives of this study include the following:

- (1) To determine the concentrations of sulfur dioxide in Taiwanese foods as consumed.
- (2) To assess exposure of and risk to population groups of various ages.
- (3) To identify which food items pose the greatest exposure risk to consumers.
- (4) To provide recommendations for further follow up and monitoring of sulfur dioxide.

## 2. Materials and methods

### 2.1. Hazard identification

In a chronic-exposure experiment on three generations of animals that lasted nearly three years, rats were given drinking water with 750 ppm sulfur dioxide. Its report indicates no effect on growth, intake of food and fluid, fecal output, fertility, weight of the newborn, and frequency of tumor development [11]. In studies on long-term toxicity of sulfite via feeding and multigenerational studies in rats, metabisulfite levels of 1% and above led to pathological

changes in the stomach [33]. Chronic overexposure to sulfur dioxide by inhalation may cause chronic bronchitis with emphysema and impaired pulmonary function [12,24]. Swallowing the liquid causes burns and tissue destruction of the esophagus and digestive tract, which may be fatal [23]. Sulfur dioxide could increase the expression of epidermal growth factor (EGF), epidermal growth factor receptor (EGFR), and cyclooxygenase-2 (COX-2) at transcription and translation levels in the lungs and tracheas in asthmatic rats. This increase might be one of the mechanisms by which sulfur dioxide pollution aggravates asthma [20]. The toxicity of the sulfites is generally low; evaluations by the Scientific Committee for Food (SCF) and by JECFA have led to the conclusion that for most consumers, sulfites in foods are of low health concern, although single, large oral doses of sulfites can produce gastrointestinal disturbances [28]. However, a small section of the population, mainly people suffering from asthma, responds to sulfites with allergy-like reactions. In sulfite-sensitive people, sulfites can provoke asthma and other symptoms of an allergic response such as skin rashes and irritations. Sensitivity to sulfites in food is dependent on how much a person is exposed to sulfur dioxide or sulfites from all sources. The pathogenesis of adverse reactions to sulfites has not been clearly documented but it is unlikely that sulfite reactions are allergic and immunity-mediated or produce anaphylactic reactions. Labeling of foods containing sulfite at concentrations of 10 mg/kg or more is required in the European Union, although the threshold for sensitivity reactions may be even lower [5].

An evaluation by the International Agency for Research on Cancer (IARC) shows that there is inadequate evidence of carcinogenicity of sulfur dioxide, sulfites, bisulfites, and metabisulfites in humans [13]. There is inadequate evidence for the carcinogenicity due to sulfites, bisulfites, and metabisulfites in experimental animals. The JECFA has established an ADI of 0–0.7 mg/kg body weight (BW)/day for sulfur dioxide JECFA, 2009. The no observed effect level (NOEL) that was established at the highest experimental dose at which no adverse effects were observed based on long-term (lifetime) studies in rats was 70 mg/kg BW/day. In establishing the ADI, a safety factor of 100 was applied to the NOEL to take into account species differences and individual human variation. The terms “sulfites” and “sulfating agents” usually refer to sulfur dioxide gas, sodium sulfite, potassium sulfite, and calcium sulfite, as well as hydrogen sulfites and metabisulfites. In this study, the concentration of sulfites in food is expressed as sulfur dioxide. Sulfur dioxide, sodium sulfite, potassium sulfite, bisulfites, and metabisulfites are collectively referred to as sulfites for the purposes of this study.

### 2.2. Core food list for the Taiwan TDS

Food consumption data from the 2005–2008 Nutrition and Health Survey in Taiwan (NAHSIT) are available [26]. The sample population consisted of 6189 participants randomly selected from 48 counties. Information on dietary intake was collected by two nonconsecutive, 24 h recalls in combination with a food frequency questionnaire. The preparation method for the core food list was the same as that used in our previous studies [21]. To construct the food list for the Taiwan TDS, 268,431 raw data entries were first obtained from 6104 questionnaires and then consolidated into 11,182 different food items that were grouped into 12 major categories and 47 subcategories on the basis of their nutrient content [3,2]. We selected 128 food items from the 47 subcategories to form a core list of food items to represent 83% of the total diet for Taiwanese individuals aged three and above.

**Table 1**  
Food items listed for sulfur dioxide and concentrations of food sulfur dioxide residue.

Major Categories of food in Taiwan	Sub-Categories of food in Taiwan	Food items listed for sulfur dioxide analysis	Sulfur dioxide residue (g/kg)	
1. Grains	1.Rice and rice products	1. Rice milk	ND <sup>a</sup>	
		2.Rice dumpling	ND	
		3.Salty rice pudding	ND	
		4.Sushi	ND	
	2.Wheat and wheat products	5.Steamed rice cake with pig blood	ND	
		6.Noodles	ND	
		7.Bread bun	ND	
		8.Green onion pancakes	ND	
		9.Wheat flake and bran	ND	
	3.Starchy vegetable foods and products	10.Tapioca balls	ND	
		11.Potato starch	ND	
	4.Dry bean and starch	12.Konjac	ND	
		13.Corn	ND	
		14.Lotus seed	ND	
2. Oils	5.Vegetable oil	15.Salad dressing	ND	
	6.Animal oil	–	NA <sup>b</sup>	
	7.Nuts and nut products	16.Roasted and salted pumpkin seed	ND	
		17.Cashew nuts	ND	
3. Poultry and poultry products	8.Chicken and chicken products	–	NA	
	9.Duck and duck products	–	NA	
	10.Other poultry and the products	–	NA	
4. Meat and meat products	11.Pork and pork products	–	0.08	
	12.Beef and beef products	–	NA	
	13.Other meat and meat products	–	NA	
5. Fish and aquatic products	14.Freshwater fish	–	NA	
	15.Seawater fish	18.Dried fish	ND	
	16.Fish's viscera and the products	19.Kamaboko	ND	
		20.Dried shrimps	0.63	
	17.Other aquatic and the products	21.Shelled fresh shrimps	ND	
	22.Fish processed product	ND		
6. Other proteins	18.Egg and egg products	–	NA	
	19.Dairy foods	23.Cheese and condensed milk	ND	
		24.Butter	ND	
		25.Soybean milk and tofu pudding	ND	
	20.Soy bean and the products	26.Tofu	ND	
		27.Dried bean curd	ND	
		28.Bean product	ND	
	7. Fruits	21.Fresh fruits	–	NA
22.Fruit products		29.Preserved fruit	ND	
		30.Dried fruit	0.14	
23.Fresh fruits juices		–	NA	
8. Vegetables	24.Dark-colored vegetable	–	NA	
	25.Light-colored vegetable	–	NA	
	26.Bamboo shoot	–	NA	
	27.Cucurbits	–	NA	
	28.Legumes	–	NA	
	29.Fungus	–	NA	
	30.Other vegetable products	31.Dried day-lily	1.76	
	31.Preserved vegetables	32.Dried radish	0.05	
		33.Pickled vegetable	0.20	
	34.Bamboo shoot	–	ND	
9. Desert	32.Bread	35.Toast	ND	
	33.Cooked and snacks	36.Bread	ND	
		37.Cake	ND	
		38.Cookies	ND	
		39.Dried shredded squid	ND	
	34.Desert and candies	40.Deserts and candies	ND	
	35.Chinese deserts	41.Pastry	ND	
	36.Ice and beverages	42.Red bean cake	ND	
		37.Processed fruit juices	–	NA
		43.Fruit tea	ND	
	10. Alcoholic beverages	38.Alcoholic beverages	44.Beer	ND
		45.Rice wine	ND	
		46.Sorghum liquor	ND	
		47.Grape wine	ND	
		48.Other wine	ND	
11.Seasonings	39.Sugar	49.Fructose	ND	
		50.Crystal sugar	ND	
	40.Salts	–	NA	
	41.Sauces	–	NA	
	42.Other seasonings	–	NA	

Table 1 (Continued)

Major Categories of food in Taiwan	Sub-Categories of food in Taiwan	Food items listed for sulfur dioxide analysis	Sulfur dioxide residue (g/kg)
12. Others	43.Processed foods (meat)	51.Dumplings and pot stickers	ND
		52.Meat balls	ND
		53.Steam buns	ND
	44.Processed foods (others)	54.Steamed meatballs	ND
		55.Oyster omelet	ND
		56.Tempura	ND
	45.Processed foods (soup)	57.Prepared soup	ND
		46.Instant noodles	ND
	47.Ready-to-go food	58.Instant noodles	ND
		59.Thick soup	ND

<sup>a</sup> ND: Not Detected.

<sup>b</sup> NA: No analysis.

### 2.3. Food list for TDS of sulfites

Sulfites have a number of technological functions, including antioxidant, bleaching agent, flour treatment agent, and preservative. However, not all food items require sulfites. Only those containing high concentrations have a significant contribution to health risk. Therefore, a food list specific to sulfites was constructed from the core list of 59 items through the following adjustments: (1) omitting from the core list the food items that were not likely to contain sulfites; (2) adding to the list the food items that were known to contain especially high levels of sulfur dioxide, including those known to frequently violate the sulfite use regulations more than five times per year in a routine survey; and (3) adding to the list the food items that were legally permitted to contain sulfites. Thus, we established a list of 59 sulfite-containing food items (Table 1), which did not depend on levels of daily intake.

### 2.4. Purchase of food samples and preparation of selected foods

From the list of 59 food items, one to three food products were selected for collection and preparation of samples for chemical analysis. Food samples were obtained from 50 strategic sampling sites distributed among four cities and four counties in the northern, central, southern, and eastern parts of Taiwan. The purchased foods, food products, and foodstuffs were combined, washed, cut, and cooked with flavoring agents according to standard recipes in the laboratory. At least eight products for each item in the list were purchased from the sampling sites. Equal weights of food products collected from all sampling sites in two seasons were combined (spring and autumn). Each composite sample was homogenized and stored at  $-80^{\circ}\text{C}$  until analysis.

### 2.5. Analysis of sulfur dioxide

In the determination of free sulfur dioxide, 10 mL of 0.3% peroxide solution was added to an empty 50 mL pear-shaped flask to just above the mark indicated on the side. Dropper tubes were used to add three to four drops of mixed indicator solution (methyl red and methylene blue) to form a purple mixture. One to two drops of 0.01 N NaOH solution were added until the solution turned olive green. The pear-shaped flask was connected to an aeration distillation apparatus. Homogenized food matrix (5 g) was added to 20.0 mL of double-distilled water, 10 mL of 25% phosphoric acid solution, 2 mL of ethanol, and two drops of silicon oil in an empty 100 mL round bottom flask. The flask was quickly connected to an aspiration assembly, and the pump was switched on. The sample was then aspirated with 0.5–0.6 L/min of heat gas for 15 min. After 15 min, the aspiration was stopped, the pear-shaped flask carefully was disconnected, and the contents were titrated against 0.01 N NaOH until an olive green endpoint was achieved. The titer value was recorded.

### 2.6. Determination of bound sulfur dioxide

After the free sulfur dioxide was determined, the pear-shaped flask, including the contents and bubbler, was replaced. The sample (5 g) was added to 20.0 mL of ddH<sub>2</sub>O, 10 mL of 25% phosphoric acid solution, 2 mL of ethanol, and two drops silicon oil in a new empty 100 mL round-bottom flask to determine bound sulfur dioxide. To calculate the total sulfur dioxide content of the sample, free and bound values were added.

Analysis of food sulfur dioxide was conducted through the Method of Test for Sulfur Dioxide in Foods of [30]. All chemicals used were of analytical reagent grade. The test was performed by the Hungkuang University Testing and Analysis Center for Food and Cosmetics. This center has been certified for microbial, chemical, and genetic analyses of foods and cosmetics under the International Organization for Standardization (ISO) 17025 guidelines [14]. The measurement uncertainty was based on Guide, Quantifying Uncertainty in Analytical Measurement [4]. The performance of the method in sulfur dioxide analysis is shown in Table 2.

### 2.7. Hazard index (HI, %ADI)

The exposure assessment method was the same method used in our previous studies [21]. The HI values expressed as %ADI were calculated according to the following equation:

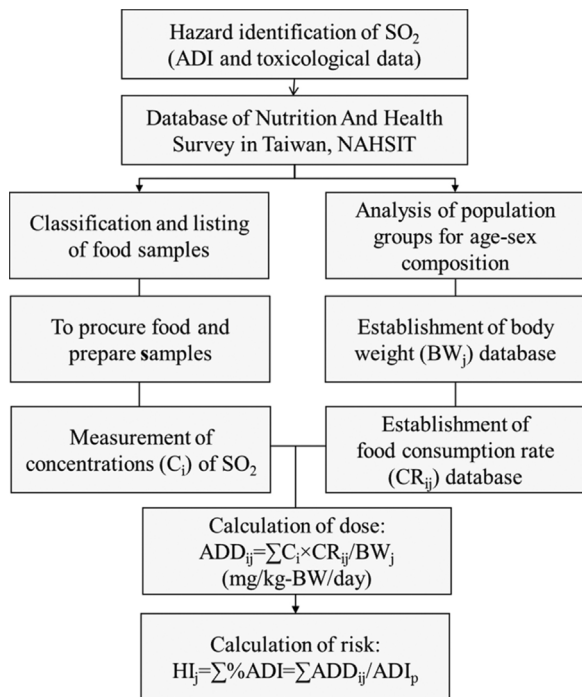
$$HI_j = \sum_{i=1}^n \%ADI_{ij} = \sum_{i=1}^n \frac{ADD_{ij}}{ADI_p} \times 100\% = \sum_{i=1}^n \frac{C_i \times CR_{ij}}{BW_j \times ADI_p} \times 100\% \quad (1)$$

where  $HI_j$  is the HI for age group  $j$ , and  $ADI_p$  is the ADI for chemical  $P$ .  $ADI_p$  is generally accepted as an intrinsic property of the target chemical  $P$ . ADI values of sulfur dioxide and sulfites used in this study were 0.7 mg/kg BW/day JECFA, 2009.  $ADD_{ij}$  is an analyte in the total diet from food item  $i$  for the specified age group  $j$ .  $C_i$  is the concentration (mg/kg) of the analyte in food item  $i$ ,  $CR_{ij}$  is the CR (g/person per day) of food item  $i$  by age group  $j$ , and  $BW_j$  is the average BW of age group  $j$ . ADD of sulfur dioxide was calculated on the basis of the daily food consumption rate (CR) and the concentration (C) of sulfur dioxide in food. The %ADI in each group for each population for the best probability distribution was calculated by using the data for BW, CR, and C and by applying Monte Carlo simulations and chi-square test (Crystal Ball<sup>®</sup>, Version 7.3, Decisioneering Inc., Denver, CO, USA). The results show that the sulfite exposure level of general consumers and high-intake consumers in Taiwan was in the 50th and 95th percentiles. The framework of this study is consistent with the principles and practices of food safety risk assessment generally accepted by international food authorities such as the WHO and FAO [15,36] and the [6]. The framework is laid out in Fig. 1.



**Table 2**  
Method performance for sulfur dioxide.

Analyte	LOD(ppm)	LOQ(ppm)	Recovery range (%)	RSD (%) (n=5)	Measurement uncertainty(ppm)	Accreditation Yes/No
Sulfur dioxide	2.0	3.2	91.3 ± 5.27	1.58	±0.064	Yes



**Fig. 1.** Flowchart of dietary exposure estimates for the sulfur dioxide in the total diet in Taiwan.

### 3. Results and discussion

#### 3.1. Concentration of sulfur dioxide

The concentrations of sulfur dioxide in the 59 food items in the total diet of the Taiwanese population aged three and above are listed in Table 1. The highest concentration of sulfur dioxide was found in dried day lily (1.76 g/kg). The detection rates for sulfur dioxide were approximately 6.2% (6/97) of the total samples analyzed. According to the recommendation of the WHO, the chemical is to be reported as undetected (ND) when C is less than the limit of detection (LOD). When the ND value is less than 60%, the data were replaced by 1/2 LOD. C values below the limit of quantitation (LOQ) but above the LOD were replaced by 1/2 LOQ. If the ND rates were over 60%, then C values were assumed to be zero [40]. In this study, the ND rate was greater than 60%. A concentration below the LOD was assumed to be zero, and the concentration of sulfur dioxide was regarded as 0 mg/kg when the daily intake was calculated. Regulations in Taiwan pertaining to the use of bleaching agents limit the sulfur dioxide concentration to 0.03–4.0 g/kg. In foods such as dried day lily, the maximum permissible concentration calculated as residual SO<sub>2</sub> is set at 4.0 g/kg. For foods such as golden raisin (1.5 g/kg), gelatin (0.5 g/kg), dried vegetables (0.5 g/kg), other dried fruits (0.5 g/kg), molasses (0.3 g/kg), starch syrups (0.3 g/kg), edible cassava starch (0.15 g/kg), syrup-preserved fruits (0.10 g/kg), shrimps (0.10 g/kg), shellfish (0.10 g/kg), and konjac (0.90 g/kg), it is 0.1–1.5 g/kg. For other processed foods except beverages (fruit juice excluded), wheat flour and its products (baked products excluded), the maximum permissible concentration is set at 0.03 g/kg [31]. In our studies, we found that the sulfur dioxide concentrations in

some samples of shrimps and sweet potato starch were above the regulation limit.

#### 3.2. Average daily dose (ADD) of sulfur dioxide

The ADD of food additives in each item was estimated by measuring the concentration of the target chemical in the food sample. For the CR of each food item of interest, data on individual 24 h recollections from the NAHSIT questionnaires were extrapolated to the number of sampled subjects by using the questionnaire weight of the population size in each specified group. A list of 59 sulfite-containing food items was selected to represent the total diet that may contain sulfur dioxide for the Taiwanese population three years and above (general consumers), according to three selection criteria: high relative CR, historically high level of occurrence of the analytes, and items for which the use of the analytes is permissible. Values for BW were derived from extrapolation of data obtained from the NAHSIT and the analyte concentration (mg/kg) in the food item. Results obtained by multiplying the concentration by the amount of food consumed are shown in Table 3. At the 50th percentile, males aged one to two years had the highest ADD of sulfur dioxide, with the exposure level reaching 0.5 mg/kg BW/day. In this group, participants who consumed the 59 food items comprised 94.6% of the 184 individuals in the group, meaning that only 10 people did not consume these food items during the 24 h recall interviews. Males aged 19–50 years and 51–65 years, as well as females aged over three years had the highest 95th percentile ADD of sulfur dioxide, with the exposure level reaching 0.8 mg/kg BW/day.

#### 3.3. HI (%ADI) of sulfur dioxide

The HI values of sulfur dioxide were calculated as a percentage of ADI (0.7 mg/kg BW/day) by using Eq. (1), and the ratio of ADD to ADI was expressed in terms of %ADI. HI (%ADI) of sulfur dioxide was calculated with the 50th and 95th percentiles of the different exposure populations (results are shown in Fig. 2). Males aged one to two years (9.8%) and females aged above 66 years (7.6%) at the 50th percentile had the highest %ADI for sulfur dioxide. At the 95th percentile, the upper end of the intake distribution is still below the ADI; females over 66 years of age (17.8%) and males over three years of age (19.7%) had the highest %ADI for sulfur dioxide. For general consumers (ages three and above), the 95th percentile HI of sulfur dioxide was 16.1% ADI for males and 19.7% ADI for females. A HI less than 100% indicates that no harm would result from dietary exposure to the analyte in the lifetime of any consumer. These results indicate that there is a large margin of safety in the use of sulfur dioxide in Taiwan.

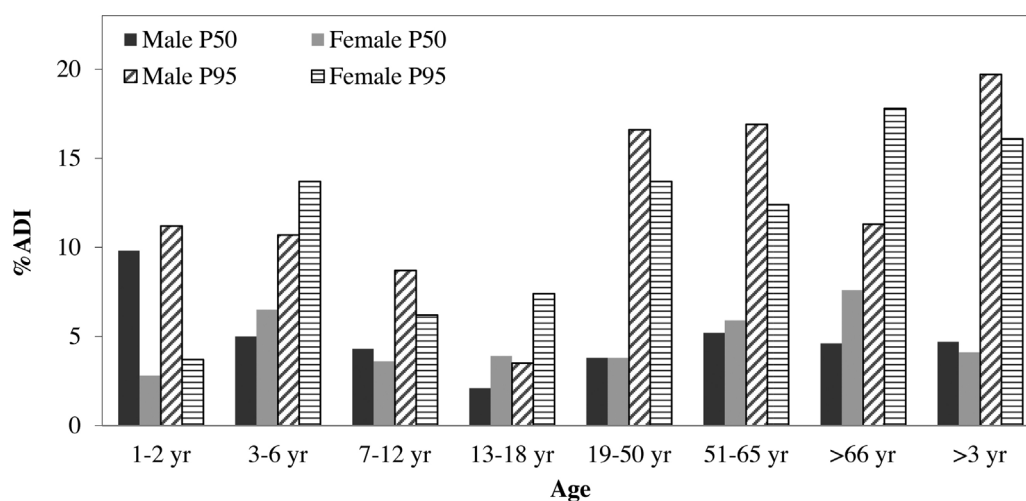
In a study carried out in the 21st Australian TDS [10], the %ADI values for sulfites, (the mean estimated dietary exposure for the population aged two years and above representing mean lifetime exposure), was approximately 35% of the ADI for males and 30% of the ADI for females. At the 95th percentile, the estimated dietary exposure to sulfites for the population aged two years and above (representing lifetime exposure for a high consumption of sulfites) was approximately 130% of the ADI for males and females.

In the 2001–2005 TDS carried out in Ireland [8], the sulfite intake was 94.3% of the ADI for above-average values (97.5th percentile) from lower-bound estimates. On the basis of the upper-bound estimate, the intake was 188.6% of the ADI. These results indicate that

**Table 3**  
ADD of sulfur dioxide for various exposure populations.

Exposure population	Whole participants		Participants (%) in 59 food items		Sulfur dioxide			
	Male	Female	Male	Female	50th ADD <sup>a</sup>		95th ADD	
					Male	Female	Male	Female
<1 Yr.	73	67	53.4	50.7	0.0	0.0	0.0	0.0
1–2 Yr.	184	170	94.6	95.3	0.5	0.1	0.6	0.2
3–6 Yr.	466	483	97.4	98.6	0.2	0.3	0.5	0.7
7–12 Yr.	1170	1018	97.4	98.6	0.2	0.2	0.4	0.3
13–18 Yr.	713	729	94.1	92.0	0.1	0.2	0.2	0.4
19–50 Yr.	1062	1082	98.6	97.5	0.2	0.2	0.8	0.7
51–65 Yr.	539	536	95.4	93.5	0.3	0.3	0.8	0.6
>66 Yr.	720	722	90.8	88.8	0.2	0.4	0.6	0.9
>3 Yr.	4670	4570	95.9	95.1	0.2	0.2	1.0	0.8

<sup>a</sup> ADD: average daily dose ( $\text{mg kg}^{-1} \text{bw day}^{-1}$ ).



**Fig. 2.** The food safety risk assessment hazard index (%ADI) of sulfur dioxide at 50th percentile (P50) and 95th percentile (P95) in eight exposure population.

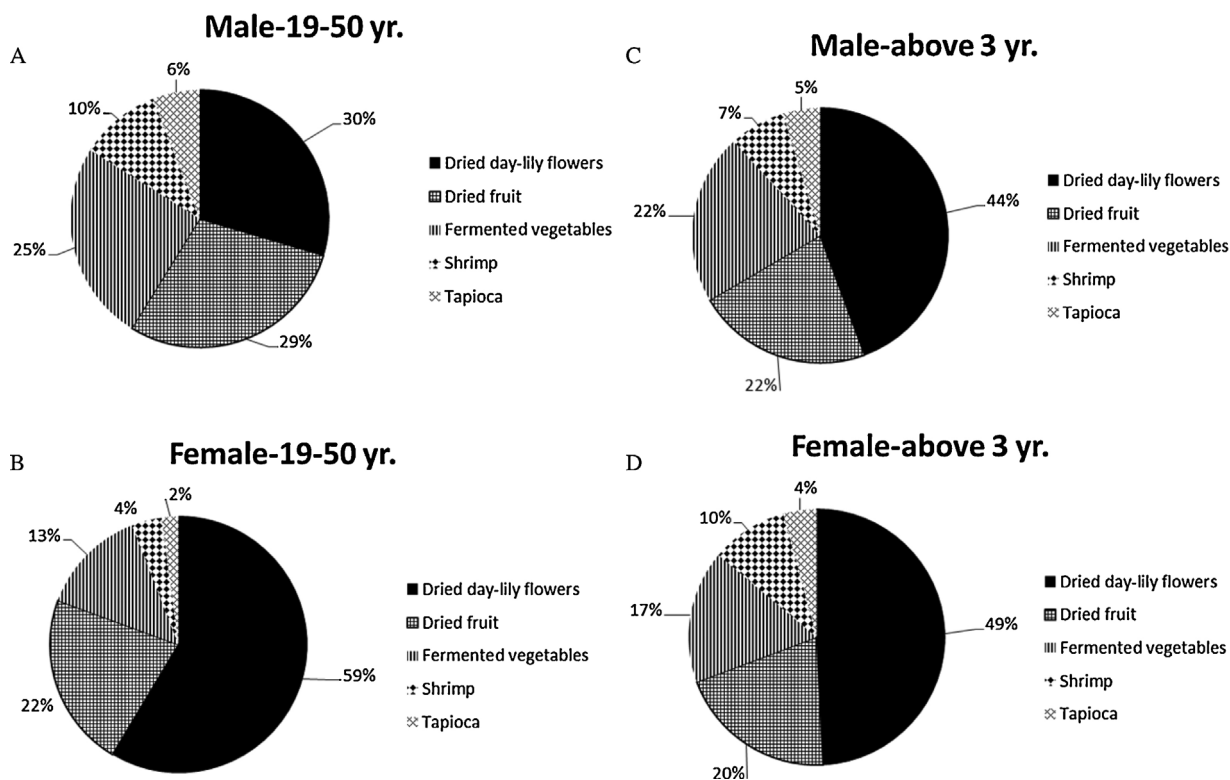
the ADI for sulfites may be exceeded by high-percentile adult consumers.

Compared with the results of the 21st Australian TDS and 2001–2005 Ireland TDS [10]; [8], the %ADI values for sulfur dioxide in our study were significantly lower than those in Australia and Ireland at the 50th and 95th percentiles. The marked difference in eating habits of the Taiwanese from those of the Australians and Irish may be the cause for the differences in %ADI between Australia, Ireland and Taiwan. Our results indicate that even in the 95th percentile, the sulfur dioxide exposure of the Taiwanese population is less than 20% of the ADI and is hence not at a harmful level. In establishing the ADI, we applied a safety factor of 100 to the NOEL to take into account species differences and individual human variation. This factor reduces the likelihood of adverse effects on humans. As the %ADI is not over 100%, we assume that sulfur dioxide poses no significant food safety risk in Taiwan under the current exposure scenario. However, HIs for the general population are close to 10% ADI, suggesting that regulatory actions must be implemented [16].

#### 3.4. Exposure contributor for sulfur dioxide

The intake contributor of sulfur dioxide is shown in Fig. 3. The most important contributor of sulfur dioxide was dried day lily for males and females, followed by dried fruit, fermented vegetables, shrimp, and tapioca. Major contributors of sulfur dioxide in the diet of Australians aged two years and above were white wine (approximately 20% for males; approximately 35% for females), beef sausages (approximately 25% for males; approximately 15% for females), and dried apricots (approximately 15% for males, approx-

imately 20% for females). In the 2001–2005 Ireland TDS, the main contributing food groups to dietary sulfite intake based on lower-bound measurements showed that alcoholic beverages (34.2%) and non-alcoholic beverages (32.4%) were the major contributing sources of sulfites in the Irish diet. Meat and meat products were also significant contributors, at 27.1% of intake, which is very different from findings in the Taiwan survey. Thus, the difference may be due to the different food CRs and eating habits. Sulfites are widely used in the preservation of fruits and vegetables and are added to many processed snacks, including cookies, pickles, soft drinks, meat, cereal bars, candy, margarine, and flour. It is also used in winemaking and is found in ingredients such as vinegar, corn syrup, cornstarch, and glucose syrup. Our results indicate that the average consumer's exposure to this food additive was within the safety limits and that there was no immediate health risk. The present safety regulations in Taiwan food only permit sulfites for bleaching agents to be used in the processing of 17 food items, in addition to other processed foods: wheat flour and its products (baked products excluded), edible cassava starch, shrimp and shellfish, syrup-preserved fruits, dried fruits, dried apricot, golden raisin, dried day lily, dried vegetable, beverages (fruit juice excluded), molasses, starch syrups, gelatin, konjac materials, and konjac products. Food items other than these 17 items are required to be free of any residues. Foods with a high contribution of sulfur dioxide are dried day lily flowers, dried fruit, fermented vegetables, shrimp, and tapioca. Therefore, consumers should be advised to be more aware of these processed foods with relatively high contaminated to avoid excessive exposure. By conservative assessment of daily exposure, we found that dried day lily flower and dried vegetables



**Fig. 3.** Pie charts showing the contribution of sulfur dioxide intake with normal consumption rates. (A) Male 19–50 years group. (B) Female 19–50 years group. (C) Male aged three and above group. (D) Female aged three and above group.

are the main contributors to sulfur dioxide exposure in Taiwan and hence require continuous monitoring.

### 3.5. Uncertainty analysis

Whenever information was missing in our risk assessment, we applied the most conservative conditions to obtain an upper estimate of risk exposure and to ensure protection of the public. There are some methodological limitations and uncertainties in our study that need to be noted.

#### 3.5.1. Errors in the estimation of CR

Major categories of food items were used to estimate CR of the overall population or of specific groups. Chinese foods are complex and dynamic, and it is very difficult to match exactly the food groups or items to be surveyed with the foods actually consumed.

#### 3.5.2. Mapping errors

A question that arises is whether the CR data and concentration data for each food item can be paired correctly. If the tested samples do not accurately represent each major category and show discrepancies with the major-category CR food types, then food matching problems may arise, contributing further to high uncertainty in the risk assessment results.

#### 3.5.3. Errors in CR surveys

Previous NAHSIT surveys in Taiwan have used the 24 h recall method [37], with each person providing information about their consumption over one day only. As there are large variations in the types and quantities of food that people eat on a daily basis, using the 24 h recall method to determine the ratio of consumption for each type of food results in a wide distribution and a large number of zero values. In addition, people's recollections of their consumption

are subject to error. Our study therefore represents a preliminary attempt to construct a “market basket” for the total diet in Taiwan.

For the above reasons, estimations of dose and risk entail a considerable degree of uncertainty and thus pose a limitation to the research. To improve the accuracy of our estimates, future efforts are focused on minimizing the aforementioned uncertainties and on validating the models and values of exposure factors in utilizing data resources from the NAHSIT.

## 4. Conclusions

The purpose of this study was to evaluate the HI (%ADI) of sulfite (sulfur dioxide) associated with ingestion and thus to establish a model for TDS on food additives in Taiwan by utilizing data available from a national nutritional status survey, the NAHSIT. TDS provides a model for future work to address specific additives in specific foods as needed by regulatory agencies for food safety risk. The calculated 95th percentile ratios of ADD to ADI of sulfite were less than 20% ADI, suggesting that the use of sulfites in food in Taiwan is within safe limits. However, sulfur dioxide is also known to be used on utensils in contact with food, such as chopsticks. This would likely increase consumers' exposure to the bleaching agent. The surveys revealed that the food items that contribute to sulfur dioxide exposure of populations aged three and above are dried day lily, soy-sauce-preserved vegetable, dried fruits, dried shrimp, and tapioca. Consumers should therefore be advised that when shopping for processed food products, especially those with a long shelf life, reading packaging labels is important. Excessive consumption of the above-mentioned food items that may contain sulfite should be avoided. In addition, we recommended assessment of the relative contributions of the food items if the government seeks to adjust the limit of sulfite use.

## Conflict of interest

The authors declare that there are no conflicts of interest.

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