

## Dietary exposure assessment to sulfites (SO<sub>2</sub>) in the Moroccan adult population

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### Abstract

Sulfites are a group of chemical food preservatives widely used in the food industry and may cause sensitivities and health adverse. Thus, sulfites are subject to an Acceptable Daily Intake (ADI) of 0.7 mg/kg of body weight. An exposure assessment was conducted to estimate the potential daily intake of sulfites in the Moroccan adult population. Dietary exposure assessment to sulfites was performed by combining analytical concentrations of selected foods, according to the optimized Monier-Williams method, and sulfites-containing food consumption data derived from an online food frequency questionnaire. For the total population, the mean dietary intake of sulfites was only 18% of the ADI. For the consumer group, the mean intake was well below the ADI, accounting for 27% of the ADI. Dietary exposure to sulfites of the regular consumer group exceeds the ADI at the mean of 137%. Dietary exposure to sulfites was also assessed in the adult asthmatic population, where the ADI is exceeded only at the 95<sup>th</sup> percentile of female asthmatic consumers, up to 113% of the ADI. The highest food contributors to sulfites intakes were potato chips, olives, and barbecue sausage. The results showed that the risk of exceeding the ADI is observed for a limited part of the population (regular consumers). However, for the asthmatic population, possibly more sensitive to sulfites, the consumption of sulfites-containing foods should be limited and controlled.

## 1. Introduction

Sulfur dioxide (SO<sub>2</sub>) and sulfating agents (identified in Europe by codes E220-E228) are among the most widely used chemical preservatives in processed foods because of their antibacterial, antifungal, and antioxidant properties (Taylor *et al.*, 1986). These food additives are authorized in Morocco by national regulation in more than 100 foods with Maximum Permitted Levels (MPL). Moreover, any use of sulfites in foods at a level of 10 mg or more (per kg or liter) must be declared on the labeling. However, several studies have reported that sulfites can cause sensitivities and adverse reactions, including symptoms of allergic response in specific subsets of the population, particularly for asthmatics (Lester, 1995; Gad, 2014). Other adverse effects have been attributed to sulfites, including anaphylaxis, hives, diarrhoea, abdominal pain and cramps, nausea and vomiting, pruritus, fainting, headache, and skin rashes (Wilson and Bahna, 2005; Vally *et al.*, 2009; Taylor *et al.*, 2014). However, health adverse and sensitivity to sulfites are

strongly dependent on the degree of exposure from all sources (Gad, 2014).

The International Agency for Research on Cancer (IARC) published in 1992 a report evaluating the carcinogenic risks to humans of sulfur dioxide and sulfites. This evaluation concluded that there is inadequate evidence of the carcinogenicity of sulfur dioxide and sulfites in humans and limited evidence of carcinogenicity in experimental animals of sulfur dioxide (IARC, 1992). Therefore, the IARC considered that sulfur dioxide, sulfites, bisulfites and metabisulfites are not classifiable due to their carcinogenicity to humans (Group 3).

Assessment of dietary exposure to food additives is one of the four steps that make up the risk assessment process of food additives, and it constitutes the spine of this process. There are two data types must be available to achieve a dietary exposure assessment: food consumption data and the concentration of food additives

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in foods. The risk characterization must be obtained by comparing the resulting dietary exposure estimate with the acceptable daily intake (ADI) for each studied food additive (FAO/WHO, Codex Alimentarius, 1989). The Joint FAO/WHO Expert Committee on Food Additives (JECFA) has established an ADI for sulfites of 0.7 mg/kg body weight expressed as sulfur dioxide (JECFA, 1974). This ADI is based on studies conducted in laboratory animals.

Dietary exposure to sulfites has been estimated in many countries using different approaches. The JECFA noted in a technical report (JECFA, 2009) that in all countries for which data on dietary exposure to sulfites were available, the ADI for the whole population did not exceed the mean but likely exceeded at high percentiles of exposure. In the same report, dietary exposure reached twice the ADI in children and teenagers in some countries. The main food contributors to total dietary exposure to sulfites differ from one country to another. However, alcoholic beverages remain among the main contributors, particularly in Europe (JECFA, 2009).

It would be inconclusive to extrapolate the results of previous studies on dietary exposure to sulfites from other countries because dietary habits and industrial/commercial practices differ from one country to another. Therefore, an exposure assessment to sulfites in the Moroccan population using specific national data is necessary, especially in the absence of official data or published studies.

The objective of the present study was to assess dietary exposure to sulfites in the Moroccan adult population for the first time, using a deterministic approach based on mean analytical levels of sulfites in a selected list of foods combined with consumption data from a food frequency questionnaire.

## 2. Materials and methods

This study combines average analytical concentrations of sulfites in food products and individual food consumption data derived from an online food frequency questionnaire. This combination has led to estimating dietary exposure to sulfites. The obtained results were then compared with the ADI values established by the JECFA to characterize the level of public health risk.

The selected foods are analyzed to determine their average concentration of sulfites. Only foods with a concentration above the limit of quantification (10 mg/kg of SO<sub>2</sub>) are included in the food frequency questionnaire. According to the Moroccan Regulation (Ministry of Agriculture/Ministry of Health, 2014) and Regulation

(EC) No. 1333/2008 (EC, 2008), an SO<sub>2</sub> content of not more than 10 mg/kg or 10 mg/L is not considered to be present.

### 2.1 Sulfites concentrations in foods

#### 2.1.1 Samples

Sulfites use is permitted by Moroccan regulation (with a maximum level) in 20 food groups and over 100 food products (Ministry of Agriculture/Ministry of Health, 2014). This regulation also provides restrictions for foods that are not allowed to contain sulfites. To obtain a refined list of foods and beverages to be sampled, the list provided by the regulation was sorted by eliminating foods unavailable on the local market (such as sago and pearl barley, marinated nuts, horseradish pulp, bottled white heart cherries, rehydrated dried fruit, angelica and citrus peel).

Foods, not part of eating habits in Morocco (such as dried pears and peaches, breakfast sausages, and dried salted fish) or are unlikely to be consumed in significant quantities (such as syrups for pancakes, flavoured syrups for milkshakes and ice cream, fruit fillings for pastries, molasses, vacuum-packed sweet corn, lychees and vinegar) are also excluded from the sampling list.

A total of twenty types of food products potentially treated with sulfites are identified following the sorting and selection operation (6 foods where the addition of sulfites is prohibited and 14 foods where the addition of sulfites is permitted).

#### 2.1.2 Sampling and sample handling

Samples were collected from local groceries and supermarkets in the Beni Mellal region (north-central Morocco) from April to November 2021. The number of samples for each food is determined according to the number of brands available in the market, respecting a minimum representativeness rate of 80%. In the case of non-packaged foods (in bulk), a minimum of ten samples per food is applied. A unique code identifies each collected sample. Various data are recorded in a sampling form, such as place and date of sampling, taken samples portion, brand name, lot number, storage conditions, manufacturer name, and declaration or not of the presence of sulfites on the labelling.

The composite samples of the same food are then prepared for analysis. The weight of each sample to be included in the composite sample is determined based on the availability of the brand and the consumer's preferred purchase sources. As far as possible, composite samples were analyzed upon receipt and preparation at the laboratory.

All selected foods are analyzed as collected (without any cooking) except for meat products and shrimp prepared according to standard/traditional Moroccan recipes before analysis.

### 2.1.3 Analytical method

The determination of sulfites concentrations was performed according to the optimized Monier-Williams method (Association of the Analytical Chemists (AOAC), 2000). This method has a limit of quantification (LOQ) of 10 mg/kg. It allows the measurement of free sulfites and the reproducible portions of bound sulfites, such as carbonyl addition products in foods (AOAC, 2000).

A 50 g (mL for liquid samples) portion of the sample is ground in a blender with an ethanol/water mixture (5%/95%). The ethanol/water solution sample is transferred to a round-bottom flask and heated under reflux with a hydrochloric acid solution (90 mL) for 105 mins to transform the sulfites into sulfur dioxide (SO<sub>2</sub>). A stream of nitrogen injected into the refluxing solution allows the sulfur dioxide to be carried away to the neutral solution of hydrogen peroxide (3%) contained in a bubbler. Upon arrival in this solution, the sulfur dioxide is oxidized and transformed into sulfuric acid, titrated by a standard sodium hydroxide solution.

All samples were analyzed twice (two tests per analysis). Analysis results are reported with all the necessary information in an individual analysis report.

### 2.2 Food consumption data

An online food frequency questionnaire was developed using Google Forms (Google Docs Editors, Google Inc. USA) and sent randomly to the maximum number of adult participants over 18 years old living in all Moroccan regions from December 2021 to Mars 2022. No detailed national food consumption data were available.

This survey aimed to identify the part of consumers, regular and asthmatic consumers of sulfites-containing foods, the frequency of consumption, the way of consumption and the approximate consumed amounts. The food consumption survey is divided into two parts.

First, respondents were asked to fill out a general questionnaire with general information, such as area of residence, gender, age, estimated weight, current occupation and asthmatic and food allergy status. Then, they were asked to complete the second part of the survey, which focuses on the consumption of sulfites-containing foods. For each sulfites-containing food, the participant is asked to answer five questions. First,

whether or not they consume the food concerned, then if the answer is yes, they are asked whether they are regular consumers of this food (once a week). If the respondent is a regular consumer, the third question is about the frequency of consumption (once a week, twice, thrice, or more than thrice). The fourth question concerns how the participant consumes the food (cooked, raw). The last question was related to the approximate consumed amounts of this food by giving, if necessary, validated references to facilitate the choice (for example, soup spoon = 20 g, small bag = 30 g, large plate = 150 g). The daily consumed amount of each food is calculated by dividing the declared amount consumed at each meal by the frequency of consumption (e.g. if the respondent consumes 30 g of the food with a frequency of twice a week, the daily consumed amount in this case, is  $30 \times 02/07$ ).

Data collected from the survey were statistically analyzed using the integrated module on Google Forms combined with Microsoft Office Excel (version 2010, Microsoft, USA).

### 2.3 Exposure assessment to sulfites

Estimates of dietary exposure to sulfites were made by combining mean levels of sulfites in selected foods with food consumption data retrieved from an online food frequency questionnaire developed for this purpose. After the analytical determination of sulfites concentrations, foods for which all analyzed samples are below the LOQ are considered sulfites-free and are not included in the survey. The same approach was used in the 21<sup>st</sup> Australian Total Diet Study (Food Standards Australia New Zealand (FSANZ), 2005).

The estimated daily intake (EDI) of sulfites for the whole population (all respondents), the consumers, the regular consumers (at least once a week) and the asthmatic consumers was calculated by multiplying the average daily food consumption (g) for each sulfites-containing food by the mean analytical levels of sulfites (mg/g). The total intakes of sulfites from all foods were then summed. The average body weight reported by the total population was used to compare the EDI of sulfites with their respective ADI value (0.7 mg/kg bw/day) and get a dietary exposure in mg/kg bw/day. The 95<sup>th</sup> percentile values were also calculated for the regular consumers and asthmatic population.

Dried fruits are consumed in Morocco both raw and cooked. Results from our previous study on the effect of cooking treatments on the residual level of sulfites in dried fruits (Hajri et al., 2022) were used to obtain a more refined exposure estimate to sulfites from dried fruits. Therefore, the EDI was calculated with the

average sulfites concentrations of raw and cooked dried fruits.

### 3. Results and discussion

#### 3.1 Concentrations of sulfites in selected foods

A total of 232 samples from twenty foods were analyzed to determine their sulfites levels. Table 1 presents the mean concentrations of sulfites, the number of samples, the percentage of samples with a level above the limit of quantification (LOQ) and the MPLs for each food. Sulfites were detected, with an average concentration ranging from 20 to 2460 mg/kg, in 10 foods (50%), 3 of which are not authorized by national regulations to contain the food additive. The analyzed foods were shown in this study for all samples, either level above or below the limit of quantification. Thus, the World Health Organization (WHO) recommendations for evaluating low-level contamination of food (WHO, 1995) have not been applied.

Dried apricots contain the highest concentration of sulfites in all foods tested, with an average of 2460 mg/kg, which exceeds the national MPL of 2000 mg/kg. Other studies reported mean concentrations that exceed the MPL. For example, the 21<sup>st</sup> Australian Total Diet Study reported a mean concentration of 2097 mg/kg, with a maximum level of 3420 mg/kg (FSANZ, 2005).

Daniels *et al.* (1992) found a mean sulfites concentration of 2791 mg/kg, with a range of 2259-3722 mg/kg. However, cooking in boiling water can reduce sulfites by nearly 40% in dried apricots (Hajri *et al.*, 2022). This cooking method is widely used in Morocco to precook dried apricots before using them in traditional dishes. We used sulfites concentrations in cooked dried apricots to refine the sulfites dietary exposure estimates.

Mustard samples showed an average concentration of 499.2 mg/kg, close to the national MPL (500 mg/kg). Other studies reported average concentrations significantly lower than those found in our study. Ishiwata *et al.* (2003) reported a mean level of 67 mg/kg in mustard from Japan. Leclerc *et al.* (2000) found a mean level of 185 mg/kg in 3 samples from Italy, while Vandevijvere *et al.* (2010) reported only 18 mg/kg in a study from Belgium. All tested samples of raisins contain sulfites with a mean level of 374 mg/kg, lower than concentrations found in a study from France, which reported a mean level of 173 mg/kg (Mareschi *et al.*, 1992). As dried apricots, cooking reduced the initial sulfites content of raisins by nearly 40% (Hajri *et al.*, 2022).

Commercial potato chips contain a mean level of 680 mg/kg, widely above the national MPL (100 mg/kg). A study in France detected a mean level (without zero values) of 104.3 mg/kg in processed potatoes (Bemrah *et*

Table 1. Mean concentrations of sulfites (SO<sub>2</sub>) in selected foods (mg/kg) and the corresponding maximum permitted levels (MPL).

Food	No. of samples	Mean concentration (mg/kg)	% of samples > LOQ	National MPLs* (mg/kg)	CA MPLs** (mg/kg)
Dried apricots	12	2460	100	2000	1000
Mustard	4	499.2	100	500	250
Dried figs	12	< LOQ	0	2000	1000
Dried beans	14	< LOQ	0	50	50
Crackers	10	< LOQ	0	50	50
Raisins	20	373.9	100	2000	1000
Sugar	8	< LOQ	0	10	15
Dried plums	10	< LOQ	0	2000	1000
Potato chips	18	680.2	100	100	50
Confectionery	20	31.8	100	50	NP
Sausages and mortadellas	8	< LOQ	0	NP	NP
Fruit juice	9	< LOQ	0	250	50
Frozen fries	8	20	100	100	NP
Unpackaged olives	12	438.3	100	NP	NP
Canned olives	9	< LOQ	0	NP	NP
Fruit jams	16	< LOQ	0	100	100
Mayonnaise	8	37.3	100	NP	NP
Fresh barbecue sausage	14	332.2	100	NP	NP
Shrimp	8	42.2	100	150	100
Ground beef	12	< LOQ	0	NP	NP
Total	232	-	-	-	-

CA: Codex Alimentarius. MPLs: maximum permitted levels. NP: not permitted. LOQ: limit of quantification

\*Ministry of Agriculture/Ministry of Health, Morocco (2014). \*\*FAO/WHO, Codex Alimentarius (2021).

al., 2009). In Lebanon, Soubra *et al.* (2006) found a 195 mg/kg mean analytical level in 8 samples of potato chips. Industrial confectionery products in Morocco (such as candies, lollypops, and gum) contain an average of 32 mg/kg, while frozen fries have an average content of 20 mg/kg. Shrimps (peeled, frozen) have been tested after pan-frying and showed an average concentration of 42.2 mg/kg. Raw fresh shrimps showed an average sulfites concentration of 78.4 mg/kg in a study conducted in Morocco (Dahani *et al.*, 2018).

Moroccan regulation does not permit sulfites use in olives, fresh sausage (Moroccan merguez) and mayonnaise. However, sulfites were detected in all composite samples of these three foods. For olives (purchased in bulk from different specialized stores), an average content of 438 mg/kg was found. In the relevant recent studies, sulfites concentrations analysis was rarely performed. For example, sulfites have not been detected in olives in the 21<sup>st</sup> Australian Total Diet Study (FSANZ, 2005). However, in a study from the Basque Country, sulfites were found in 15% of the tested olives samples (Urriaga *et al.*, 2013). The high sulfites content of unpackaged olives was predicted. A preliminary survey revealed that unpackaged olives (prepared and marketed under uncontrolled conditions) might be illicitly treated with sulfites.

The mean concentration of sulfites in composite samples of fresh sausage (tested after cooking) was 332 mg/kg, similar to other studies, including in Australia (275 mg/kg; FSANZ, 2005), UK (322 mg/kg; Ministry

of Agriculture, Fisheries and Food, United Kingdom (MAFF), 1995) and the Basque Country (317.5 mg/kg; Urriaga *et al.*, 2013). The mean concentrations of sulfites found in mayonnaise were nearly 37 mg/kg.

All other foods tested (dried figs, beans, crackers, sugar, dried plums, sausages and mortadellas, fruit juice, canned olives and fruit jams) showed results well below the LOQ. Therefore, these food types are excluded from the list to be included in the food consumption survey.

### 3.2 Food consumption data

Responses from the online food frequency questionnaire were collected from October 2021 to February 2022, with 1155 respondents over 18 years old. This online survey provided general information (such as body weight and gender) and data on the consumption of sulfites-containing foods.

Table 2 describes the study population. Most respondents are from urban areas (86%) with a wide range of available foods. The female population (n = 653) represents 56.5%, while the male population (n = 502) represents 43.5% which is in line with the Moroccan demography. Home-prepared foods (including mayonnaise, olives, French fries and barbecue sausage) are automatically considered sulfite-free.

Table 3 presents food consumption data for all studied population categories. The total population consumes, on average, 25.5 g/day of sulfites-containing foods (24 g/day for males and 27 g/day for females),

Table 2. Description of the study population.

Gender	Respondents (%)	Average body weight (kg)	No. of asthmatic (%)	No. of food allergic (%)	Area of residence	
					Urban (%)	Rural (%)
Male	502 (43.5%)	70	18 (3.5%)	48 (9.5%)	409 (81.5%)	93 (18.5%)
Female	653 (56.5%)	60	39 (6%)	94 (14%)	583 (89.3%)	70 (10.7%)
Total	1155	-	57 (5%)	142 (12%)	992 (86%)	163 (14%)

Table 3. Food consumption (g/day) for all respondents, consumers, regular consumers, and asthmatics consumers.

Food	Sulfites-containing food consumption (g/day)								
	All respondents		Consumers		Regular consumers		Asthmatic consumers		P95
	Mean	%	Mean	%	Mean	P95	%	Mean	
Dried apricots	0.4	52.5	0.7	3.5	10.4	15.0	2.4	0.2	1.8
Mustard	0.4	41.0	1.0	12.3	3.5	7.4	3.0	0.4	3.9
Raisins	3.7	89.5	4.2	27.0	13.6	30.4	4.0	4.0	21.8
Potato chips	3.0	63.0	4.3	19.3	14.2	32.8	3.5	1.8	7.2
Confectionery	1.3	57.0	2.3	24.5	5.2	14.6	3.5	1.1	5.1
Frozen fries	5.4	78.0	6.9	34.3	15.8	28.5	3.4	4.3	15.7
Olives	5.0	73.5	6.9	36.0	14.0	33.3	3.6	4.8	17.7
Mayonnaise	1.3	65.5	2.1	23.3	5.6	12.9	3.3	0.8	3.8
Barbecue sausage	3.6	52.0	6.8	9.8	36.3	82.9	3.3	5.6	37.2
Shrimp	1.4	60.5	2.3	10.8	12.4	31.3	3.2	3.2	34.0
Total	25.5	-	37.5	-	131.0	289.2	-	26.2	148.2

while consumers take a mean of 37.5 g/day (37.2 g/day for males and 37.8 g/day for females). For regular consumers (at least once a week), an average of 131 g of sulfites-containing foods consumed per day (122 g/day for males and 140 g/day for females), while at the 95<sup>th</sup> percentile (P95), the consumed amount of sulfites-containing foods was 289 g/day (271 g/day for males and 307 g/day for females). Self-reported asthmatics take a mean of 26.2 g/day (25 g/day for males and 27.5 g/day for females), while at the P95, they consume a mean of 148 g/day (137 g/day for males and 159 g/day for females).

Thus, the amounts of potentially sulfites-containing foods are significant only for some regular consumers (average and P95). This category would be potentially exposed to a risk of exceeding the ADI. In addition, olives, barbecue sausage, and French fries (frozen) are the main contributors to the daily consumption of sulfites-containing foods.

### 3.3 Exposure assessment of sulfites in the Moroccan adult population

Dietary exposure estimates to sulfites were calculated separately for male and female respondents using the corresponding average body weight (60 kg for females and 70 kg for males). The estimation calculations concerned the total population (all respondents), consumers only, regular consumers, and the self-reported asthmatics population (Tables 4 and 5). The mean estimated daily intake of sulfites for the total population was 0.10 mg/kg bw/day for males, corresponding to only 14% of the recommended ADI, and 0.15 mg/kg bw/day for females, corresponding to 22% of the ADI. Similar results have been reported in France (Bemrah *et al.*, 2012), Ireland (Vin *et al.*, 2013), New Zealand (Cressey and Jones, 2009) and Australia (FSANZ, 2005). However, in a study conducted in Italy by Vin *et al.* (2013) using chemical data provided by the food industry, the estimated daily intake of sulfites for the total population was only 0.017 mg/kg bw/day (2% of the ADI). However, in Italy (Leclercq *et al.*, 2000), dietary exposure to sulfites using analytical levels in realistic sulfites-containing meals slightly exceeded the ADI for adults (120%) and children (110%).

The difference between males and females regarding mean dietary exposure to sulfites remains insignificant for the total population. It is mainly due to the body weight variance. However, in the Basque Country, Urriaga *et al.* (2013) revealed a considerable difference between the exposure to sulfites of men compared with women, mainly due to the high consumption of alcoholic beverages by men (0.48 mg/kg bw/day for men, corresponding to 69% of the ADI vs. 0.14 mg/kg bw/day

for women, corresponding to 21% of the ADI).

The average dietary exposure to sulfites for the consumers was well below the ADI, with 0.16 mg/kg bw/day (23% of the ADI) for male consumers and 0.22 mg/kg bw/day for female consumers (31% of the ADI). A close result is observed in a study conducted in New Zealand with no difference between males and females (0.197 mg/kg bw/day for females and 0.2 mg/kg bw/day for males (Cressey and Jones, 2009). A study carried out in Lebanon (Soubra *et al.*, 2006) reported mean dietary exposure to sulfites of 0.44 mg/kg bw/day (63% of the ADI) for the consumer group (students).

In the high-intake scenario (regular consumers), the ADI is exceeded in both the male and female populations, with a notable difference between them due to differences in body weight and the amounts of certain consumed foods. The mean daily intake of sulfites was 0.74 mg/kg bw/day for males (105% of the ADI) and 1.18 mg/kg bw/day for females (168% of the ADI). At the 95<sup>th</sup> percentile (heaviest regular consumers), dietary exposure to sulfites was 1.47 mg/kg bw/day for males (210% of the ADI) and 2.25 mg/kg bw/day for females (322% of the ADI).

The differences between male and female regular consumers regarding the mean dietary exposure to sulfites are due to average body weight (men have a higher average weight than women) and to the higher consumption of certain sulfites-containing foods by women, particularly potato chips. Exposure assessment to sulfites of high adult consumers in France (Bemrah *et al.*, 2012), Lebanon (Soubra *et al.*, 2006), Australia (FSANZ, 2005) and Austria (Mischek and Krapfenbauer-Cermak, 2012) was 120%, 215%, 130% and 243% of the ADI, respectively. However, the ADI did not exceed in studies from China (33% of the ADI; Zhang *et al.*, 2014) and Ireland (97% of the ADI; Vin *et al.*, 2013) in the high-intake scenario.

The ADI is not exceeded for the self-reported asthmatic subjects at the mean. Nevertheless, it is likely exceeded in the high-intake scenario (P95) for female asthmatic consumers. The mean dietary exposure to sulfites in this population category (considered as more sensitive to sulfites) was 0.10 mg/kg bw/day for males (14% of the ADI) and 0.14 mg/kg bw/day for females (19% of the ADI). At the P95, dietary exposure to sulfites was 0.50 mg/kg bw/day for males (72% of the ADI) and 0.79 mg/kg bw/day for females (113% of the ADI). No comparative data were found concerning the dietary exposure to sulfites for asthmatic subjects.

Table 6 presents dietary exposure estimates to sulfites and the corresponding ADI after considering the

Table 4. Dietary exposure to sulfites (SO<sub>2</sub>) in the Moroccan male adult population and percentage of the ADI.

Food	Average concentration (mg SO <sub>2</sub> /g)	Mean intake of all respondents (mg SO <sub>2</sub> /day)	Mean intake of consumers (mg SO <sub>2</sub> /day)	Intake of regular consumers (mg SO <sub>2</sub> /day)		Intake of asthmatic consumers (mg SO <sub>2</sub> /day)	
				Mean	P95	Mean	P95
Raw dried apricots	2.46	0.98	1.97	20.17	30.75	0.74	8.61
Mustard	0.50	0.15	0.50	1.60	3.50	0.30	1.50
Raw raisins	0.37	1.07	1.22	4.51	9.25	1.48	5.55
Potato chips	0.68	0.88	1.63	5.98	13.60	0.34	1.97
Confectionery	0.03	0.04	0.08	0.16	0.40	0.03	0.14
Frozen fries	0.02	0.11	0.15	0.31	0.57	0.08	0.29
Unpackaged olives	0.44	2.51	3.43	6.42	14.65	2.02	5.50
Mayonnaise	0.04	0.04	0.08	0.19	0.52	0.02	0.11
Fresh sausage	0.33	1.12	2.24	11.75	28.31	1.72	9.41
Shrimp	0.04	0.07	0.13	0.58	1.37	0.18	2.10
Total intake of SO <sub>2</sub> (mg/day)		7	11.4	51.7	103	7	35
Exposure to SO <sub>2</sub> (mg/kg bw/day)		0.1	0.16	0.74	1.47	0.1	0.5
% of the ADI		14%	23%	105%	210%	14%	72%

Notes: Acceptable daily intake for sulfites (ADI): 0.7 mg/kg bw/day (JECFA, 1974). Average body weight for males: 70 kg.

Table 5. Dietary exposure to sulfites (SO<sub>2</sub>) in the Moroccan female adult population and percentage of the ADI.

Food	Average concentration (mg SO <sub>2</sub> /g)	Mean intake of all respondents (mg SO <sub>2</sub> /day)	Mean intake of consumers (mg SO <sub>2</sub> /day)	Intake of regular consumers (mg SO <sub>2</sub> /day)		Intake of asthmatic consumers (mg SO <sub>2</sub> /day)	
				Mean	P95	Mean	P95
Raw dried apricots	2.46	0.74	1.23	30.75	43	0	0
Mustard	0.5	0.25	0.55	1.85	4	0.10	2.40
Raw raisins	0.37	1.63	1.85	5.55	13.21	1.52	10.55
Potato chips	0.68	3.13	4.22	13.26	31.08	2.18	7.75
Confectionery	0.032	0.04	0.07	0.17	0.53	0.04	0.19
Frozen fries	0.02	0.10	0.13	0.32	0.57	0.09	0.34
Unpackaged olives	0.44	1.94	2.60	5.94	14.65	2.16	10.12
Mayonnaise	0.037	0.06	0.07	0.23	0.43	0.04	0.17
Fresh sausage	0.33	1.22	2.24	12.21	26.40	1.98	15.18
Shrimp	0.042	0.04	0.07	0.46	1.26	0.09	0.76
Total intake of SO <sub>2</sub> (mg/day)		9.15	13	70.75	135.2	8.2	47.46
Exposure to SO <sub>2</sub> (mg/kg bw/day)		0.15	0.22	1.18	2.25	0.14	0.79
% of the ADI		22%	31%	168%	322%	19%	113%

Notes: Acceptable daily intake for sulfites (ADI): 0.7 mg/kg bw/day (JECFA, 1974). Average body weight for females: 60 kg.

Table 6. Dietary exposure to sulfites and the corresponding percentage of the ADI after considering the reducing effect of cooking on the residual level of sulfites in dried fruits.

	Total intake of SO <sub>2</sub> (mg/day)		Exposure to SO <sub>2</sub> (mg/kg bw/day)		Percentage of the ADI	
	Males	Females	Males	Females	Males	Females
All respondents (mean)	6.16	8.22	0.09	0.14	13	20
Consumers (mean)	10.14	11.8	0.14	0.2	21	28
Regular consumers (mean)	41.43	55.65	0.59	0.93	85	133
Regular consumers (P95)	86.43	112	1.23	1.87	176	267
Asthmatic consumers (mean)	6.04	7.62	0.09	0.13	13	18
Asthmatic consumers (P95)	29.43	43.47	0.42	0.72	60	104

effect of cooking on the residual level of sulfites in dried apricots and raisins. The refined dietary exposure estimates to sulfites showed a slight decrease in the daily sulfites intake. However, the ADI is still exceeded for regular consumers. The results also showed a slight exceeding of the ADI (106%) for female asthmatic consumers at the high-intake scenario (P95), mainly due to body weight difference and high consumption of potato chips.

The ADI for sulfites in this study is exceeded in the high-intake scenarios (regular consumers) as in most other studies from different countries. Therefore, the potential health adverse due to the ingestion of sulfites-containing foods are insignificant for the total population. Nevertheless, national food risk managers must pay particular attention to the potential health risks from sulfites-containing foods, particularly for regular consumers who exceed the recommended ADI. Therefore, sulfites use and overuse control in primary and processed foods should be enhanced. Thus, the revision of national MPLs could eventually limit dietary sulfites exposure.

The list of foods selected in this study is not exhaustive. Other food categories in which sulfites may be used as food additives were not included in this study, which may lead to underestimating the dietary intake of sulfites. Sulfites intake from other non-food sources (such as environment and drugs) or naturally sulfites-containing foods such as onions (Brahimi *et al.*, 2022) may also lead to underestimating sulfites exposure. On the other hand, cooking treatments and storage conditions could reduce the sulfites concentrations in certain foods (Vandevijvere *et al.*, 2010). The sulfites exposure estimate from dried fruits was conducted based on raw dried apricots and raisins. However, these foods are also consumed after cooking in traditional dishes. In this case, dietary exposure to sulfites would be overestimated.

Further study on the consumption of sulfites-containing foods by asthmatic subjects (with a more significant number of respondents) and by children and teenagers is strongly recommended to assess further the dietary exposure to sulfites in these consumer categories and the current level of public health risk.

### 3.4 Contribution of food types to dietary exposure to sulfites

Table 7 presents the contribution percentage of all foods to the mean dietary intake of sulfites in the consumers and asthmatic consumers. Results showed that, for male consumers, sulfites intake was essentially due to the consumption of olives (30%), barbecue sausage (20%), and dried apricots (17%). For female consumers, potato chips (32%), olives (20%), and barbecue sausage (17%) were the significant contributors to sulfites exposure. Olives and barbecue sausage are still the major contributors to sulfites exposure for male and female asthmatic consumers, with a notable contribution from raisins and potato chips. Except for dried apricots and raisins, all other major contributors are not authorized to contain sulfites by national regulations (olives and barbecue sausage) or found to contain levels well above the maximum permitted levels (potato chips).

Dried apricots and dried fruits were also among the most contributors to sulfites intake in Australia (FSANZ, 2005), in France (Bemrah *et al.*, 2009), and for children in Austria (Mischek and Krapfenbauer-Cermak, 2012). In a study from the Basque Country (Urriaga *et al.*, 2013), children were mainly exposed to sulfites from meat products, especially fresh sausage. Processed potatoes were also significant contributors to the dietary intake of sulfites in France (Bemrah *et al.*, 2009) and Belgium (Vandevijvere *et al.*, 2010). Thus, dietary exposure studies in several countries show that alcoholic beverages, particularly wines, are the main contributors to sulfites intake in adults.

Table 7. Food type contribution (%) to mean estimated dietary exposure to sulfites.

Food	% of contribution for consumers		% of contribution for asthmatic consumers	
	Male	Female	Male	Female
Dried apricots	17.2	9.4	10.7	0
Mustard	4.3	4.2	4.4	1.2
Raisins	10.7	14.2	21.4	18.5
Potato chips	14.3	32.4	4.9	26.6
Confectionery	0.7	0.5	0.5	0.5
Frozen fries	1.3	1	1.1	1.1
Olives	30	20	29.3	26.3
Mayonnaise	0.7	0.6	0.3	0.5
Barbecue sausage	19.7	17.2	24.8	24.2
Shrimp	1.1	0.5	2.6	1.1



#### 4. Conclusion

This study first showed that sulfites were detected in foods not permitted by Moroccan regulations (mayonnaise, olives, and barbecue sausage). Other tested foods contain sulfites levels exceeding the regulatory MPL (dried apricots and potato chips). Except for asthmatic subjects, dietary exposure to sulfites in Morocco must represent a low level of public health risk. However, estimated dietary exposure to sulfites of Moroccan adults showed that the risk of exceeding the recommended ADI is likely possible for regular consumers and a limited part of the asthmatic population. Thus, illegal practices (overuse and abuse) contribute significantly to excessive dietary exposure to sulfites in high-intake scenarios. The use of sulfites as food additives in local and imported foods should be controlled and supervised, especially in foods that contribute significantly to dietary exposure to sulfites.

#### Conflict of interest

The authors declare no conflict of interest.

#### References

- Association of the Analytical Chemists (AOAC). (2000). Official Method 990.28. Sulfites in Foods: Optimized Monier-Williams Method. 17<sup>th</sup> ed. Arlington, Virginia, USA: AOAC.
- Bemrah, N., Leblanc, J.C. and Volatier, J.L. (2009). Assessment of dietary exposure in the French population to 13 selected food colours, preservatives, antioxidants, stabilizers, emulsifiers and sweeteners. *Food Additives and Contaminants: Part B*, 1(1), 2-14. <https://doi.org/10.1080/19393210802236943>
- Bemrah, N., Vin, K., Sirot, V., Aguilar, F., Ladrat, A.C., Ducasse, C., Gey, J.L., Rétho, C., Nougadere, A. and Leblanc, J.C. (2012). Assessment of dietary exposure to annatto (E160b), nitrites (E249-250), sulphites (E220-228) and tartaric acid (E334) in the French population: the second French total diet study. *Food Additives and Contaminants: Part A*, 29(6), 875-885. <https://doi.org/10.1080/19440049.2012.658525>
- Brahimi, A., El Ouardi, M., Kaouachi, A., Boudboud, A., Hajji, L., Hajjaj, H. and Mazouz, H. (2022). Characterization of the biochemical potential of Moroccan onions (*Allium cepa* L.). *International Journal of Food Science*, 2022, 2103151. <https://doi.org/10.1155/2022/2103151>
- Cressey, P. and Jones, S. (2009). Levels of preservatives (sulfites, sorbate and benzoate) in New Zealand foods and estimated dietary exposure. *Food Additives and Contaminants: Part A*, 26(5), 604-613. [https://doi.org/10.26656/fr.2017.7\(6\).625](https://doi.org/10.26656/fr.2017.7(6).625)
- <https://doi.org/10.1080/02652030802669188>
- Dahani, S., Kandil, S. and Bouchriti, N. (2018). Évaluation du risque sulfites dans la filière des crustacés [Sulphites risk assessment in the crustacean sector]. *Revue Marocaine des Sciences Agronomiques et Vétérinaires*, 6(3), 391-401. [In French].
- Daniels, D.H., Joe, F.L., Warner, C.R., Longfellow, S.D., Fazio, T. and Diachenko, G.W. (1992). Survey of sulphites determined in a variety of foods by the optimized Monier-Williams method. *Food Additives and Contaminants*, 9(4), 283-289. <https://doi.org/10.1080/02652039209374074>
- European Community (EC). (2008). Regulation No. 1333/2008 of the European Parliament and of the Council of 16 December 2008 on food additives. *Official Journal of the European Union*, 354, 16-33.
- FAO/WHO, Codex Alimentarius. (1989). Guidelines for the simple evaluation of dietary exposure to food additives. CAC/GL 3-1989. Rome, Italy: FAO/WHO.
- FAO/WHO, Codex Alimentarius. (2021). General standard for food additives. CODEX STAN 192-1995. Rome, Italy: FAO/WHO.
- Food Standards Australia New Zealand (FSANZ). (2005). The 21<sup>st</sup> Australian Total Diet Study. A total diet study of sulphites, benzoates and sorbates. Canberra, Australia: FSANZ.
- Gad, S.C. (2014). Sulfites. In Wexler, P. (Ed.) *Encyclopedia of Toxicology*. 3<sup>rd</sup> ed., p. 416-419. Elsevier E-Book. <https://doi.org/10.1016/B978-0-12-386454-3.00932-5>
- Hajri, Z., Oussama, A., Chigr, M., Kzaiber F. and Boutoial, K. (2022). Effect of cooking and washing on sulfite (SO<sub>2</sub>) content of dried fruits, presented at the 1st International Conference on Chemical and Biological Sciences. Istanbul, Turkey: Sciendo.
- International Agency for Research on Cancer (IARC). (1992). Monographs on the evaluation of carcinogenic risks to humans: Occupational exposures to mists and vapours from strong inorganic acids and other industrial chemicals. Vol. 54. Lyon, France: IARC.
- Ishiwata, H., Nishijima, M. and Fukasawa, Y. (2003). Estimation of inorganic food additive, antioxidant, processing agent and sweetener concentrations in foods and their daily intake based on official inspection results in Japan in fiscal year 1998. *Journal of the Food Hygienic Society of Japan*, 44(2), 132-143. <https://doi.org/10.3358/shokueishi.44.132>
- Joint FAO/WHO Expert Committee on Food Additives

- (JECFA). (1974). Toxicological evaluation of certain food additives with a review of general principles and of specifications: 17th report of the JECFA. Geneva: WHO.
- Joint FAO/WHO Expert Committee on Food Additives (JECFA). (2009). Safety evaluation of certain food additives. WHO food additives series: 60. Sixty-ninth meeting of the JECFA. Geneva: WHO.
- Leclercq, C., Molinaro, M.G., Piccinelli, R., Baldini, M., Arcella, D. and Stacchini, P. (2000). Dietary intake exposure to sulphites in Italy - analytical determination of sulphite-containing foods and their combination into standard meals for adults and children. *Food Additives and Contaminants*, 17(12), 979-989. <https://doi.org/10.1080/02652030010014402>
- Lester, R.M. (1995). Sulfites sensitivity: significance in human health. *Journal of the American Nutrition Association*, 14(3), 229-232. <https://doi.org/10.1080/07315724.1995.10718500>
- Mareschi, J.P., François-Collange, M. and Suschetet, M. (1992). Estimation of sulphite in food in France. *Food Additives and Contaminants*, 9(5), 541-549. <https://doi.org/10.1080/02652039209374108>
- Ministry of Agriculture and Ministry of Health, Morocco. (2014). Arrêté conjoint du ministre de l'agriculture et de la pêche maritime et du ministre de la santé n°1795-14 du 14 rejev 1435 (14 mai 2014) fixant la liste et les limites des additifs alimentaires autorisés à être utilisés dans les produits primaires et les produits alimentaires, ainsi qu'aux indications que doivent porter leurs emballages. Morocco : Ministry of Agriculture and Ministry of Health.
- Ministry of Agriculture, Fisheries and Food, United Kingdom (MAFF). (1995). Food Surveillance Information Sheet 65: Survey of sulphur dioxide and benzoic acid in foods and drinks. United Kingdom: MAFF.
- Mischek, D. and Krapfenbauer-Cermak, C. (2012). Exposure assessment of food preservatives (sulphites, benzoic and sorbic acid) in Austria. *Food Additives and Contaminants: Part A*, 29(3), 371-382. <https://doi.org/10.1080/19440049.2011.643415>
- Soubra, L., Sarkis, D., Hilan, C. and Verger, P. (2006). Dietary exposure of children and teenagers to benzoates, sulphites, butylhydroxyanisol (BHA) and butylhydroxytoluen (BHT) in Beirut (Lebanon). *Regulatory Toxicology and Pharmacology*, 47(1), 68-77. <https://doi.org/10.1016/j.yrtph.2006.07.005>
- Taylor, L.S., Bush, R.K. and Nordlee A.J. (2014). Sulfites. In Metcalfe, D.D., Sampson, A.H., Simon, A.R. and Lack, G. (Eds.). *Food allergy: adverse reactions to foods and food additives*, p. 361-374. New Delhi, India: Wiley. <https://doi.org/10.1002/9781118744185.ch29>
- Taylor, L.S., Higley, N.A. and Bush, R.K. (1986). Sulfites in foods: uses, analytical methods, residues, fate, exposure assessment, metabolism, toxicity, and hypersensitivity. *Advances in Food Research*, 30, 1-76. [https://doi.org/10.1016/S0065-2628\(08\)60347-X](https://doi.org/10.1016/S0065-2628(08)60347-X)
- Urriaga, C., Amiano, P., Azpiri, M., Alonso, A. and Dorransoro, M. (2013). Estimate of dietary exposure to sulphites in child and adult populations in the Basque Country. *Food Additives and Contaminants: Part A*, 30(12), 2035-2042. <https://doi.org/10.1080/19440049.2013.840930>
- Vally, H., Misso, N.L. and Madan, V. (2009). Clinical effects of sulphite additives. *Clinical and Experimental Allergy*, 39(11), 1643-1651. <https://doi.org/10.1111/j.1365-2222.2009.03362.x>
- Vandevijvere, S., Temme, E., Andjelkovic, M., De Wil, M., Vinkx, C., Goeyens, L. and Van Loco, J. (2010). Estimate of intake of sulfites in the Belgian adult population, *Food Additives and Contaminants: Part A*, 27(8), 1072-1083. <https://doi.org/10.1080/19440041003754506>
- Vin, K., Connolly, A., McCaffrey, T., McKevitt, A., O'Mahony, C., Prieto, M., Tennant, D., Hearty, A. and Volatier, J-L. (2013). Estimation of the dietary intake of 13 priority additives in France, Italy, the UK and Ireland as part of the FACET project. *Food Additives and Contaminants: Part A*, 30(12), 2050-2080. <https://doi.org/10.1080/19440049.2013.851417>
- Wilson, G.B. and Bahna, L.S. (2005). Adverse reactions to food additives. *Annals of Allergy, Asthma and Immunology*, 95(6), 499-507. [https://doi.org/10.1016/S1081-1206\(10\)61010-1](https://doi.org/10.1016/S1081-1206(10)61010-1)
- World Health Organization. (1995). Reliable evaluation of low-level contamination of food. Workshop in the Frame of GEMS/Food-EURO, Kulmbach, Germany. Geneva: WHO.
- Zhang, J.B., Zhang, H., Wang, H.L., Zhang, J.Y., Luo, P.J., Zhu, L. and Wang, Z.T. (2014). Risk analysis of sulfites used as food additives in China. *Biomedical and Environmental Sciences*, 27(2), 147-154. <https://doi.org/10.3967/bes2014.032>