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The Risk Assessment of Sulphite Intake through Dried Fruit Consumption in Hamadan, Iran

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HIGHLIGHTS

- Eleven out of 18 whole-dried apricot samples and 3 out of 18 half-dried apricot ones had unacceptable level of sulphite.
- Daily intakes of sulphite through dried fruit in preschool children, females, and males were 0.53, 0.35, and 0.30 mg/kg body weight/day, respectively.
- Consumers should be advised to avoid excessive consumption of dried fruits, especially whole-dried apricot samples.

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Acronyms and abbreviations
ADI=Acceptable Daily Intake
EU=European Union
SO_2=Sulfur dioxide

ABSTRACT

Background: Dried fruit is one of the main sources of sulphur dioxide (SO_2) exposure. The major goal of the current study was to determine SO_2 concentration in some widely consumed dried fruits in Hamadan, Iran. In addition, an estimate of sulphite intake was made through the aforementioned dried fruit.

Methods: Totally, 126 dried fruit samples were randomly collected from local market in Hamadan, Iran. The sulfur dioxide, moisture contents, and pH of dried fruit samples were measured by iodometric titration, gravimetric method, and pH meter, respectively. Data analysis was performed using SPSS statistical software.

Results: The mean sulphite residue in raisin, sultana, half-dried apricot, whole-dried apricot, dried prune, peach fruit leather, and apricot fruit leather was 236.49±231.34, 485.84±217.55, 1204.00±750.21, 2257.78±1406.63, 597.32±401.82, 84.74±109.22, and 38.28±32.40 mg/kg, respectively. Eleven out of 18 (61.11%) whole-dried apricot samples and 3 out of 18 (16.66%) half-dried apricot samples had unacceptable level of sulphite. The moisture contents and pH of all dried fruit were at an acceptable level. The amount of daily sulphite exposure through the consumption of dried fruits for preschool children, adult females, and adult males were 0.53, 0.35, and 0.30 mg/kg body weight/day, respectively.

Conclusion: As the intake of sulphite through the dried fruit in Iran was lower than acceptable daily intake, it appears that the content of this food preservative in dried fruits consumed in Iran has no serious risk for Iranian consumers. However, consumers should be advised to avoid excessive consumption of dried fruits, especially whole-dried apricot.

Introduction

Sulfur dioxide (SO_2) and some compounds such as sodium sulphite, sodium bisulphite, calcium sulphite, etc. releasing SO_2 during their utilization, have been widely applied in the food and beverage

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industries (Silva and Lidon, 2016; Stražanac et al., 2019). These compounds are utilized as antioxidant agents for preventing oxidation and maintaining flavour and color; preservative agents for inhibiting the growth of deterioration organisms, such as various species of yeast and bacteria; and also anti-browning agents for controlling enzymatic and non-enzymatic (Maillard) reactions (Lou et al., 2017; Machado et al., 2009). The utilization of sulphite compounds such as SO_2 during the drying of fruits and vegetables facilitates the drying process and reduces drying time (Stražanac et al., 2019; Türkyılmaz et al., 2013). In addition, sulphur dioxide inhibits insect attacks for fruit, maintains the natural color of the dried product and impedes changes of color that would be caused by enzymes or chemicals during production and storage (Cantín et al., 2012; Guerrero and Cantos-Villar, 2015).

The SO_2 and sulphites are allergen components that could result in allergic reactions in asthma patients and individuals who their sulphite oxidase activity has been reduced (Soubra et al., 2007). In addition, these food additives can cause skin reactions (Vally et al., 2009) and DNA damage (Meng et al., 2005). Therefore, food safety organizations have considered the acceptable limit for SO_2 and sulphite in foodstuffs. Consequently, the mentioning of the name and concentration of sulphite compounds used in a food or beverage formulation on the food label is a legal requirement (Guido, 2016). In European Union (EU), the allowable limit for sulphite residue in various products is different. For dried apricots, peaches, grapes, prunes, and figs; sulphite residue should be lower than 2000 mg/kg while the maximum of its level in dried apples and pears was considered as 600 mg/kg and in other dried fruit and nuts were lower than 500 mg/kg (EFSA, 2016). Hence, Codex (1995) suggested different limits for the sulphur dioxide residue, including 1500 mg/kg in raisins and 2000 mg/kg in dried apricots. According to Iran National Standard, the maximum acceptable level of SO_2 for different types of grapes is different: 700 mg/kg in sultana raisins (ISIRI, 2012), 1500 mg/kg in golden raisins (angora; ISIRI, 2012), 200 mg/kg in peach fruit leather (ISIRI, 2018), and 2000 mg/kg in whole-dried apricots (ISIRI, 2014), as well as half-dried apricots (ISIRI, 2015).

It seems that the consumption rate of some dried fruits may be relatively high in Iran which usually consumed as healthy snacks. The dried fruits contain low levels of fat while having high levels of fibre and carbohydrates; they are also suitable sources of various nutritional elements as well as vitamins (Heshmati et al., 2017; Heshmati and Mozaffari Nejad, 2015). There are several types of dried fruits prepared in Iran. For instance, fruit leather, fruit bar, or fruit slab is a chewy and flavourful substance prepared from fruit puree (Diamante et al., 2014). Raisin and sultan are dried vine fruit although raisins are made from seed or seedless grapes which their seed whether or not be removed during processing while sultanas are obtained from seedless grape (Grigoryan and Hakobyan, 2015).

Dried fruit is one of the main sources of sulphur dioxide exposure (Lien et al., 2016; Lou et al., 2017). Published information about SO_2 values in foodstuff consumed among Iranians is scarce. Thus, the major goal of the current study was to determine SO_2 concentration in some widely consumed dried fruits in Hamadan, Iran. In addition, an estimation of sulphite intake was made through the aforementioned dried fruit.

Materials and methods

Samples and material

One-hundred twenty six dried fruit samples, including raisin, sultana, half-dried apricot, Gheisi (local name of whole-dried apricot), dried prune, peach fruit leather, and apricot fruit leather (each 18 samples) were collected from local market in Hamadan, Iran from April to July 2018. All chemicals, including phenolphthalein, buffer solutions of pH meter, hydrochloric acid, and also sodium thiosulfate were bought from Merck (Darmstadt, Germany).

Moisture content determination

The moisture content of dried fruit samples was carried out according to the gravimetric method prescribed by AOAC (2000). The specimens were thoroughly crushed and mixed, then 2 g of the sample was weighted in crucible and placed in drying oven at temperature of 70±1 ºC and low pressure (<100 mm Hg or 13.3 kPa) for 6 h. The moisture content is expressed as weight/weight % (w/w %).

The pH content determination

The pH of different dried fruit specimens was determined using a pH meter (Denver Instruments, TX, USA). At first, 10 g of the sample was completely crushed. Then, 100 ml of distilled water was added to ground samples. After 24 h, the pH was measured. Before measurement, the pH meter was

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calibrated by buffer solutions of reference pH of 4.0 and 7.0.

SO_2 determination

The SO_2 contents of dried fruit were measured according to Iran standard method (ISIRI, 2017). This method is based on the sublimation of sulfur dioxide in the presence of heat, water, and acid. The released sulfur dioxide gases are collected in an Erlenmeyer flask, which its concentration was determined by the iodometric titration. About 32 g of each dried fruit and 250 ml of distilled water were poured into digestion balloon and heated to boil, and then 10 ml of hydrochloric acid (37%) was added. The acidified sample was refluxed for 55 min. Finally, the collected sulfur dioxide was titrated with the sodium thiosulfate 0.1 N. The concentration of sulfur dioxide was determined by following equation:

$C_{SO2}=(A-B)\times0.1\times64/(2\times M)$

C_{SO2}: concentration of sulfur dioxide (mg/kg)

A: the volume (ml) of sodium thiosulfate for titration of 25 ml of iodine solution (0.1 N)

B: the volume (ml) of sodium thiosulfate for titration of residual iodine in the collected solution

M: sample weight (g)

Exposure assessment

Dietary exposure to sulphite through dried fruit was estimated by considering mean sulphite in the collected dried fruits and per capita consumption of these products by Iranian people according to methods suggested in the previous study (Mischek and Krapfenbauer-Cermak, 2012). For estimation of risk of sulphite intake through dried fruit consumption, the mean sulphite level that preschool children, male, and female were exposed during a day, was divided to Acceptable Daily Intake (ADI) value of sulphite (0.78 mg/kg) and expressed as percentage. The average body weights were considered 20 kg for preschool children with aged of 3-6 years old, 60 kg for an adult female with aged of 19-65 years old, and 70 kg for an adult male with aged of 19-65 years old. In the current study, data of per capita consumption of dried fruit was obtained from a food frequency questionnaire form completed by 500 resident persons in Hamadan, Iran. Per capita consumption of raisin, sultana, half-dried apricot, whole-dried apricot, dried prune, peach fruit leather, and apricot fruit leather was considered 3.9, 3.9, 4.8, 4.8, 2.5, 0.6, 0.6 g/day for adults, respectively; and 1.95, 1.95, 2.4, 2.4, 1.25, 0.3, 0.3 g/day for preschool children, respectively.

Statistical analysis

Data analysis was performed using SPSS statistical software (version 15.0; SPSS, Inc., Chicago, IL, USA). All measurements were carried out in triplicate and average and standard deviation were reported. To compare the sulphite value with the permissible limit, one sample t-test was used. The comparison between SO_2, moisture, and pH value of the various type of dried fruit was done by Analysis Variance (ANOVA) followed by Tukey's post-hoc procedure. Pearson correlation coefficient was used to determine correlation among sulphite, moisture, and pH value. $P<0.05$ was considered as significant difference.

Results

The mean of sulphite concentrations in various dried fruit was showed in Table 1. There was a significant difference ($p<0.05$) between the sulphite concentrations of products. The highest and lowest mean sulphite levels were found in whole-dried apricot (Gheisi) (2257.78±1406.63 mg/kg) and apricot fruit leather (38.28±32.40 mg/kg), respectively. The mean sulphite value in other product, including raisin, sultana, half-dried apricot, dried prune, and peach fruit leather was 236.49±231.34, 485.84±217.55, 1204.00±750.21, 597.32±401.82, and 84.74±109.22 mg/kg, respectively.

As indicted in Table 2, the moisture contents and pH of all dried fruit were at an acceptable level. The highest values of moisture and pH belonged to dried prune (36.11±8.29%) and raisin (4.49±0.22%), respectively. Also, the lowest values of moisture and pH were detected in raisin (12.61±1.23%) as well as apricot fruit leather (2.81±0.35%), respectively, indicating a significant difference ($p<0.05$).

In collectively, SO_2 concentration had positive and significant correlation with moisture content (correlation coefficient=0.367 and $p<0.05$) as well as pH (correlation coefficient=0.228 and $p=0.01$) of samples (Table 3). However, it was only in raisin and sultana samples that the SO_2 concentration had a positive and significant correlation with moisture content ($p<0.05$). Among various dried fruit samples, SO_2 concentration of whole-dried apricot samples showed significant correlation ($p<0.05$) with pH.

The average estimated daily intake of sulphite through dried fruit was 0.53 mg/kg body weight/day for preschool children, 0.35 mg/kg body weight/day for female, and also 0.30 mg/kg body

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weight/day for male (Table 4). The total sulphite intake through dried fruit was 75% of ADI for pre-school children, 50% of ADI for female adults, and 42.86% of ADI for male adults.

Table 1: The concentration of sulfur dioxide residue in dried fruits in Hamadan, Iran

Samples	No.	Mean (mg/kg)	SD**	Min (mg/kg)	Max (mg/kg)	Acceptable limit (mg/kg) according to:			Sample number (%) containing SO_2 value more than acceptable levels		
						Iran standard	Codex	EU	Iran standard	codex	EU
Raisin	18	236.49 c*	231.34	16.12	720.95	1500	1500	2000	0	0	0
Sultana	18	485.84 c	217.55	159.65	951.23	700	1500	2000	2 (11.11)	0	0
Half-dried apricot	18	1204.00 b	750.21	96.00	2640.00	2000	2000	2000	3 (16.66)	3 (16.66)	3 (16.66)
Whole-dried apricot	18	2257.78 a	1406.63	144.00	5008.00	2000	2000	2000	11 (61.11)	11 (61.11)	11 (61.11)
Dried prune	18	597.32b c	401.82	48.09	1440.68	2000	1000	2000	0	2 (11.11)	0
Peach fruit leather	18	84.74 c	109.22	16.08	385.54	200	-	-	2 (11.11)	-	-
Apricot fruit leather	18	38.28 c	32.40	14.40	112.35	200	-	-	0	-	-
Total	126	700.63	962.19	14.40	5008.00	-	-	-	-	-	-

*Different subscript letters indicate significantly difference within each column (p<0.05)
** SD: Standard Deviation

Table 2: The moisture content and pH in dried fruit samples in Hamadan, Iran

Sample	No.	Moisture (%)		Maximum acceptable moisture (%)	pH	
		Mean±SD	Range		Mean±SD	Range
Raisin	18	12.61±1.23 c*	11.33-16.92	19	4.49±0.22 a	3.85-4.78
Sultana	18	18.06±4.38 c	10.92-26.25	19	4.47±0.30 a	3.46-4.89
Half-dried apricot	18	18.28±6.90 c	9.00-33.33	20	4.21±0.28 a	3.69-4.75
Whole-dried apricot	18	26.08±8.83 b	12.33-39.33	25	4.21±0.33 a	3.46-4.87
Dried prune	18	36.11±8.29 a	12.03-55.67	35	3.28±0.15 b	3.08-3.69
Peach fruit leather	18	16.32±3.38 c	11.19-21.78	15	3.22±0.53 b	2.64-4.17
Apricot fruit leather	18	15.17±4.12 c	7.67-21.67	15	2.81±0.35 c	2.42-3.84

* Different subscript letters indicate significantly difference within each column (p<0.05)

Table 3: Correction of sulfur dioxide concentration with moisture and pH in dried fruit in Hamadan, Iran

Sample	pH		Moisture	
	Correlation coefficient	*P*-value	Correlation coefficient	*P*-value
Raisin	0.238	0.342	0.593	0.009
Sultana	0.429	0.076	0.730	0.001
Half-dried apricot	-0.134	0.595	0.147	0.560
Whole-dried apricot	-0.756	<0.001	0.429	0.076
Dried prune	-0.382	0.118	0.073	0.773
Peach fruit leather	-0.006	0.981	0.357	0.146
Apricot fruit leather	-0.170	0.500	0.010	0.967
Total	0.228	0.010	0.367	<0.001

Table 4: Estimated daily intake (mg/kg body weight per day) of sulphite through dried fruit and ADI% for different Iranian population groups

Sample	Mean (mg/kg)	Preschool children		Female		Male	
		Mean daily intake (mg/kg body weight per day)	ADI (%)	Mean daily intake (mg/kg body weight per day)	ADI (%)	Mean daily intake (mg/kg body weight per day)	ADI (%)
Raisin	236.49	0.02	3.29	0.02	2.20	0.01	1.88
Sultana	485.84	0.05	6.77	0.03	4.51	0.03	3.87
Half-dried apricot	1204.00	0.14	20.64	0.10	13.76	0.08	11.79
Whole-dried apricot	2257.78	0.27	38.70	0.18	25.80	0.15	22.12
Dried prune	597.32	0.04	5.33	0.02	3.56	0.02	3.05
Peach fruit leather	84.74	<0.01	0.18	<0.01	0.12	<0.01	0.10
Apricot fruit leather	38.28	<0.01	0.08	<0.01	0.05	<0.01	0.05
Total	700.63	0.53	75.00	0.35	50.00	0.30	42.86

* ADI: Acceptable Daily Intake

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Discussion

In the present work, sulphite residues were detected in some dried fruit samples distributed in Hamadan, Iran. The average concentration of sulphite in all dried fruits analyzed in the current study (700.63 mg/kg) was higher than the findings reported by Mischek and Krapfenbauer-Cermak (2012) in Australia (339.5 mg/kg) and Vandevijvere et al. (2010) in Belgian (179 mg/kg in dried fruits except apricots and 533 mg/kg in dried apricot). However, sulphite residues in dried fruit samples, analyzed by Bemrah et al. (2008) in France (1005.9 mg/kg) and Suh et al. (2007) in Korea (1070 mg/kg), was higher than our findings. The discrepancies between our results and other studies might be related to differences in the initial sulphite value used in the product, sulfitation processing, drying method, the duration between sulfitation and analysis time, moisture content of the samples, and measurement methods of sulphite (Leclercq et al., 2000; Lou et al., 2017; Machado et al., 2009; Mischek and Krapfenbauer-Cermak, 2012; Soubra et al., 2007). In addition, it is assumed that sulphite concentrations may be decreased during storage similar to other food contaminants. Because SO_2 is a volatile component and its amount might be reduced during longer storage time (Stražanac et al., 2019). In this regard, Ozturk et al. (2011) revealed that sulphite level was reduced in the dried apricot samples from 2174 to 1284 mg/kg after 1-year storage.

In the present survey, the highest mean sulphite concentration was found in whole-dried apricot (Gheisi) samples followed half-dried apricot and dried prune. In addition, the sulphite levels in 11 out of 18 (61.11%) whole-dried apricot samples were more than maximum acceptable value (2000 mg/kg) according to Iran National Standard (ISIRI, 2014), Codex Alimentarius (Codex, 1995), and EU (EFSA, 2016). The most of whole-dried apricot samples are traditionally produced in Iran and the manufacturers are not familiar with the permitted dosage of sulphite compound. So, they might utilize the high amount of these food additives during the whole-dried apricot processing. The mean sulphite level in half-dried apricot samples in the current work (1204.00 mg/kg) was lower than those reported by Cressey and Jones (2009) in New Zealand (1554 mg/kg), FSANZ (2005) in Australia (2097 mg/kg), and Arslan et al. (2018) in Turkey (1987 mg/kg). Our reports indicated that the mean content of sulphite in sultana samples was significantly higher than that in raisin. Therefore, it could be stated that sulphite dosage used during sultana processing was probably higher than the raisin. In comparison, the sulphite value in sultana samples in Australia was reported as 76 mg/kg (FSANZ, 2005) that was too lower than our finding (485.84 mg/kg).

We could not find any published study in the literature regarding sulphite residue in apricot, peach, and fruit leather resulting in some limitations for data comparison. However, the possible reason for low sulphite content in our fruit leather samples might be related to the utilization of the lower sulphite dosage in the fruit leather production. Also, other compounds such as edible salt (sodium chloride) and vinegar are added in fruit leather formulation, which could lead to the reduction of sulphite level in the final product (Diamante et al., 2014).

In many previous studies, the risk assessment of sulphite intake through various groups of foodstuffs consumed in one special district has been reported as the percentage of ADI (Cressey and Jones, 2009; Lien et al., 2016; Machado et al., 2009; Vandevijvere et al., 2010). However, the estimation of sulphite intake through dietary exposure could be done with some other approaches, such as the individual dietary records, the model diets methods, etc. (Cressey and Jones, 2009; Leclercq et al., 2000; Mischek and Krapfenbauer-Cermak, 2012). In most similar investigations, the sulphite intake was often through the consumption of both wine and also dried fruits. However, in Muslim countries like Iran, the majority of people avoid from drinking of alcoholic beverages because of religious beliefs. Therefore, it appears that unlike most countries, dried fruits were the main contributors to sulphite exposure in Iranian population. In the current survey, the mean estimated daily intake of sulphite through dried fruit were 0.53, 0.35, and 0.3 mg/kg body weight/day for preschool children, adult female, and adult male, respectively that were lower than values found by Mischek and Krapfenbauer-Cermak (2012) in Austria. These researchers showed daily intake of sulphite through dried fruit as 0.12, 0.07, and 0.07 mg/kg body weight/day for Australian preschool children, adult females, and adult males, respectively. These variations seen in the sulphite intake value may be because of the differences in daily food consumption level, sulfite concentration in the analyzed samples, as well as the type of foodstuff included in the risk estimation formula.

Conclusion

In the current study, the mean sulphite residues in all analyzed dried fruits except whole-dried apricot samples were lower than the acceptable values according to the Iran standard, Codex, and EU. The moisture content and pH of all dried fruit was also at an acceptable level. Since the intake of sulphite through the dried fruit in Iran was lower than ADI, it appears that the content of this food preservative in dried fruits consumed in Iran has no serious risk for Iranian consumers. However, the other foodstuff sources might contain sulphite and their consumption with dried fruit could result in sulphite intake exceeded ADI. Therefore, consumers should be advised to avoid excessive consumption of dried fruits, especially whole-dried apricot samples. Also, regulatory agencies must continuously monitor the sulphite residue in various food products.

Author contributions

A.H. designed the study, analyzed the data, and wrote the manuscript; F.S. and M.K. conducted the experimental work. All the authors revised and approved the final manuscript.

Conflicts of interest

All the authors declared that they have no conflict of interests.

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