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Estimation of Sulphite Levels in Food Products Available in Delhi, India

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Abstract

Sulphites used as preservatives in foods might pose a risk for sensitive individuals, causing broncho-constriction, urticaria and dermatitis. The present work aimed at determining sulphite concentration using Optimized Monier-Williams method. A total of 357 food products were collected from different markets of Delhi and analysed for sulphites. The food products were classified into eight categories namely preserves, dried fruits, beverage concentrates, sugars, confectionery items, ready to serve beverages, bakery products and miscellaneous products. The mean recovery for 8 major food matrices was found to be 82.02% with relative standard deviation % RSD_R of 3.22 and % RSD_r of 1.72. The sulphite levels, expressed as sulphur dioxide, ranged from <10.0 mg/kg to 406 mg/kg. A total of 21% food products had sulphite concentration below the quantifiable limit of 10 mg/kg and in 30% of food products sulphites were not-detectable. Though most food samples presented sulphite levels below 50% of Maximum Permissible Level (MPL), a few local brands or un-branded samples of products like jam (n=1), sugar (n=3), fruit bars (n=3), digestives (n=1) and fruit beverages (n=1) had sulphite concentration 106% - 136% of the MPL. Beverages are likely to be major contributors to sulphite intake and brand loyal children are most likely to exceed their acceptable daily intake (ADI). Using the average concentration of sulphite obtained, 200 ml of beverage like squash will provide approximately 71% of the ADI for a child weighing 20 kg. Data generated can be utilized to estimate dietary exposure to sulphites and associated risk. Small scale manufacturers need to be sensitized about good manufacturing practices.

Keywords: Sulphiting agents; Food additives; Preservatives; Processed foods; Dietary exposure; Risk assessment

Introduction

Sulphur dioxide (SO_2) and its salts are collectively known as sulphiting agents. These salts include potassium metabisulphite (INS No. 224); sodium metabisulphite (INS No. 223); potassium bisulphite (INS No. 225); sodium bisulphite (INS No. 222) and sodium sulphite (INS No. 221). The above mentioned salts are white crystalline powders having the pungent odour of SO_2. They are freely soluble in water and insoluble in alcohol. They are used as food additives because of their ability to perform a number of roles [1]. The functions and applications of sulphites include inhibition of non-enzymatic browning and enzyme reactions, and antimicrobial action. It is also used as an antioxidant, dough conditioner, acidulant and in extraction of corn starch. Sulphur dioxide functions as a bleaching agent in the manufacture of refined sugar [2].

The MPLs of sulphites in food products like preserves, sugar, confectionery items, ready to serve beverages, beverage concentrates, dried fruits, processed meat products, dehydrated vegetables, bakery wares etc. are given by the Codex body - General Standards for Food Additives (GSFA). In India, their addition in food products is regulated by Food Safety and Standards Authority of India (FSSAI). As per GSFA, sulphites in solid food products can be used at a level ranging from 15-500 mg/kg food product and in liquid food products in the range of 50-200 mg/kg of food product [3].

SO_2 and its salts were assigned an acceptable daily intake (ADI) value of 0-0.7 mg/kg b.w. /day by Joint FAO/WHO Expert Committee on Food Additives (JECFA). The ADI was based on a long-term three-generation study of reproductive toxicity in rats [4]. The most commonly reported adverse reaction to sulphites is that of broncho-constriction in a sub-group of asthmatic subjects [5-8]. In another case reported by Prenner and Stevens [9], an "anaphylactic response" was observed after an oral challenge of 10 mg total dose of sodium bisulphite. Other idiosyncratic adverse reactions reported include contact dermatitis [10] and hypotension [11]. Neurotoxic [12,13], cytogenic [14-16] and genotoxic [17,18] effects have been found to be associated with high levels of exposure to sodium metabisulphite.

National data on sulphite intake based on the measurement of sulphite levels in foods and beverages, is lacking in India which makes it difficult to estimate exposure levels. The present work aimed to determine the sulphite content in commonly consumed food products available in the markets of Delhi, the capital city of India. This data will be further used to estimate the contribution of different types of food products to the intake of sulphites.

Materials and Methods

The Optimized Monier-Williams method, which is a distillation-titration procedure, is most widely used for the analytical determination of sulphite in foods and beverages [19,20]. Hillery et al. [21] in an intra-laboratory study established that the optimized Monier- Williams procedure is capable of determining sulphites in foods with an overall %RSDr ranging from 3.8 to 9.8.

Reagents

All chemicals were of analytical grade and nitrogen gas used was 99.9% pure which was purchased from Industrial Medical Engineers (India). The solutions used for the sulphite determination were purchased from Merck Specialities Pvt. Ltd (Germany).

Sampling and sample preparation

For the purpose of this study, the commonly available brands (multinational, national, local brands and un-branded) of food products in which the addition of sulphites is permitted, were purchased from supermarkets and specialized stores in five geographical zones of the city of Delhi, between February 2014 and March 2015. Some food products sold loose by pushcart retail vendors which could contain sulphites were also purchased for analysis. The food products were classified into eight categories namely preserves and accompaniments, dried fruits, beverage concentrates, sugars, confectionery items, ready to serve beverages, bakery products and miscellaneous products. Even products containing ingredients in which addition of sulphites is permissible and which can thus contribute to sulphite exposure were selected. A total of 357 food products were analysed for estimating the concentration of sulphites in them. Three different batches of the same type and brand of product were carefully homogenised before analysis. The samples were analysed in duplicate and analyses were carried out as quickly as possible to avoid the loss of sulphites.

Method of determination

The analyses were performed according to the AOAC Official Method 990.28 [20], which measures free sulphite and reproducible portion of bound sulphite. The sulphite levels, expressed as sulphur dioxide, were computed as follows:

SO_2 (mg/kg or mg/L)=(32.03 × V_B × M × 1000)/W

Where, 32.03=equivalent molar mass of SO_2 (g/mol)

V_B=volume (mL) of standardized NaOH solution required to reach the end point

M=molar concentration of standardized NaOH solution determined by standardization with potassium hydrogen phthalate (mol/L)

1000=converts grams to milligrams

W=Weight (g) or volume (ml) of sample introduced into the 1000 mL flask.

Recovery assays

The recovery assays were carried out by spiking samples of sulphite-free food matrices with standard solutions that were prepared fresh daily in the laboratory. The standard solutions were made by diluting sodium metabisulphite Granular AR (ACS) grade (Noba Chemie Pvt. Ltd. India) in distilled water. The amount of sulphite added, expressed as sulphur dioxide, ranged from 10 to 750 mg/kg depending on the maximum permissible levels in the food concerned. The repeatability (inter-day and intra-day variability) and external reproducibility (inter-laboratory variability) of the method was evaluated using % relative standard deviation (RSD) associated with the measurements of SO_2 obtained during the recovery tests at all levels of spiking. The inter-day repeatability was checked in triplicates for three days for different food matrices. The intra-day repeatability was checked in triplicates for a day. Data entry, management and statistical analysis were performed using SPSS 16.0 and Microsoft Excel 2010.

Results and Discussion

Recovery assays

Samples were spiked at levels ranging from 10 to 750 mg/kg of sulphur dioxide. The average recovery was 82.02% with a %RSD_R of 3.22 and %RSD_r of 1.72.

Its reproducibility i.e., inter-laboratory variability was estimated by reporting its mean recovery, standard deviation i.e., SD_R and %RSD_R. This was done by comparing the results obtained in our analytical laboratory with the results of an NABL accredited and FSSAI approved laboratory. Reproducibility was checked for food matrices like sugar, ready to serve beverage and beverage concentrate. The samples were checked in duplicate at both the labs at 10 ppm level. The results are shown in Table 1.

Food Matrix	Spike Level (mg/kg)	Mean Amount Recovered (mg/kg)	SD_R	%RSD_R	Recovery%
Sugar	10	8.292	0.1	1.15	81.2-82.9
Ready to serve beverage	10	8.272	0.18	2.16	81.1-83.6
	100	94.951	7.88	8.3	90.89-99.1
Beverage Concentrate	10	8.157	0.16	1.98	80.2-83.2
Potato Chips	100	70.536	1.78	2.52	68.66-71.12

Table 1: Inter-laboratory study showing reproducibility. SD_R-Standard deviation reproducibility; %RSD_R-%Relative standard deviation.

The SD_R for foods in the present study ranged from 0.095 to 0.179 and %RSD_R ranged from 1.15 to 2.16 which were well below the limit of %RSD i.e., 10 as calculated by Horwitz from 300 inter-laboratory collaborative trials for trace constituents at 10 ppm [22]. The recoveries obtained from selected food matrices like beverages, beverage concentrates and sugar spiked at the level of 10 mg/kg were above 80% as recommended by AOAC [20]. For a few foods like ready to serve beverage and potato chips, spiking was carried out at 100 ppm i.e., 100 mg/kg level in the two laboratories. It was found that the recoveries in case of beverage was above 80% and in case of potato chips was 70.5% and the %RSDR in these samples at this level were also below 10% in accordance with results of published inter-laboratory collaborative trials as shown in Table 1.

Table 2 shows the detailed results obtained from the recovery studies for studying repeatability, which were performed with sugar, biscuit, and beverage concentrate, ready to serve beverage, fruit bar, potato chips, jam and raisins which did not have detectable level of sulphites. The maximum %RSDr was 5.98 for biscuits spiked at 10 ppm, which is below the maximum limit of 10% as given by Horwitz et al. [22]. The recoveries obtained at different levels of spiking are

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comparable with the findings obtained by FSANZ [23] and Machado and Toledo [24], who reported recoveries higher than 90% for sulphite added to foods such as sultanas, hominy, dried mangoes, and fruit juice, using the optimized Monier-Williams method. In case of biscuits and potato chips the recoveries at all levels of spiking obtained were lower than 80% i.e., 62-78% which were similar to those reported by Warner et al. [25]. This is not a limitation of the method but instead it is difficult to estimate sulphur dioxide in such food matrices by any technique of estimation [25]. However, it is within the acceptable level of recoveries i.e., 60-120% but lower than 80% as prescribed by the method (Table 2).

Food Matrix	Spike Level (mg/kg)		Recovery (%) Intra-Day	Recovery (%) Inter-Day	RSD_r (%)	RSD_r (%)
					Intra-Day	Inter-Day
Sugar	10	n	3	9	1.39	1.61
		Mean%	81.24	81.69		
		SD_r	0.113	0.132		
	50	n	3	9	3.17	1.51
		Mean%	85.09	85.71		
		SD_r	1.35	0.649		
	100	n	3	9	1.61	1.19
		Mean%	83.73	84.81		
		SD_r	0.641	1.015		
Jam	10	n	3	9	1	2.79
		Mean%	81.49	82.59		
		SD_r	0.081	0.23		
	50	n	3	9	1.59	0.74
		Mean%	83.41	84.83		
		SD_r	0.663	0.313		
	100	n	3	9	1.37	1.18
		Mean%	81.62	83.04		
		SD_r	1.119	0.98		
Ready to Serve Beverage	10	n	3	9	1.51	1.71
		Mean%	82.33	82.85		
		SD_r	0.124	0.141		
	50	n	3	9	2.19	1.52
		Mean%	91.74	90.71		
		SD_r	1.004	0.689		
	100	n	3	9	1.53	1.76
		Mean%	90.58	91.29		
		SD_r	1.389	1.606		
Beverage Concentrate	10	n	3	9	1	1.08
		Mean%	85.62	84.31		
		SD_r	0.084	0.091		
	50	n	3	9	2.53	3.09

		Mean%	88.88	90.78		
		SD_r	1.122	1.404		
	100	n	3	9	1.62	1.67
		Mean%	91.28	94.53		
		SD_r	1.476	1.582		
	350	n	3	9	0.45	0.86
		Mean%	98	98.45		
		SD_r	1.537	2.969		
Biscuit	10	n	3	9	5.98	3.42
		Mean%	66.15	65.06		
		SD_r	0.395	0.223		
	50	n	3	9	2.86	1.18
		Mean%	72.06	71.88		
		SD_r	1.029	0.425		
	100	n	3	9	3.02	4.78
		Mean%	72.49	70.69		
		SD_r	2.189	3.376		
Raisins	10	n	3	9	1.07	1.12
		Mean%	82.21	81.87		
		SD_r	0.088	0.095		
	50	n	3	9	2	4.28
		Mean%	84.72	86.92		
		SD_r	0.939	1.862		
	100	n	3	9	1.21	1.42
		Mean%	85.01	85.58		
		SD_r	1.031	1.212		
	500	n	3	9	0.392	0.81
		Mean%	87.21	87.88		
		SD_r	1.707	3.554		
	750	n	3	9	1.5	1.61
		Mean%	88.15	87.99		
		SD_r	9.917	10.559		
Fruit Bar	10	n	3	9	0.53	0.7
		Mean%	80.94	80.08		
		SD_r	0.043	0.057		
	50	n	3	9	1.88	3.24

		Mean%	84.21	84.11		
		SD_r	0.791	1.361		
	100	n	3	9	3.02	1.4
		Mean%	84.04	83.36		
		SD_r	2.538	1.171		
Potato Chips	10	n	3	9	2.39	2.34
		Mean%	70.2	70.73		
		SD_r	0.167	0.167		
	50	n	3	9	4.13	4.66
		Mean%	72.83	73.27		
		SD_r	1.504	1.705		
	100	n	3	9	2.66	2.54
		Mean%	74.98	76.81		
		SD_r	1.997	2.924		

Table 2: Repeatability of different food matrices. n=number of samples; SD_r=Repeatability Standard deviation; $\%RSD_r$=% Repeatability Relative standard deviation.

The recoveries obtained from selected food matrices spiked at the levels ≥ 10 mg/kg were above or equal to 80% for most of the matrices. These findings are in accordance with the AOAC recommendation that suggests recoveries of ≥ 80% for food matrixes spiked at the 10 mg/kg level making it the quantifiable limit, in order to ensure accurate data using the optimized Monier–Williams method [20,21]. The limit of quantification (LOQ) for these food matrices studied was hence established to be 10 mg/kg. Similar findings were reported by Machado and Toledo [24], Leclercq et al. [26] and Suh et al. [27].

Sulphites concentration data

A total of 357 food products were analysed for estimating the concentration of sulphites in them. The data shows that 51% (n=182) food products had either non-detectable levels of sulphites (n=107, 30%) or concentration levels below LOQ (n=75, 21%). Amounts below LOQ (10 mg/kg) were considered as “5 mg/kg” in other words, 50% of this limit. Similar protocol was used by Machado and Toledo [24], Leclercq et al. [26], and Suh et al. [27]. The concentration data has been presented in Table 3.

Food Products	**Total Number of Samples**	**Number of Not-detectable Food Samples (Nd)**	**Number of Samples having Sulphites Concentration <LOQ**	**[a]Mean Sulphite Concentration (mg/kg)**	**[b]Mean Sulphite Concentration (mg/kg)**	**Minimum Sulphite Concentration (mg/kg)**	**Maximum Sulphite Concentration (mg/kg)**
Dried Fruits							
Raisins	5	0	0	260.7	260.7	123.3	405.9
Sultanas	5	1	1	20.6	25.8	6.2	40.3
Apricots	5	0	0	267.4	267.4	200.2	333.9
Dates	4	0	0	102.9	102.9	71	138.3
Peanuts	2	2	0	-	-	-	-
Almonds	2	2	0	-	-	-	-
Cashewnuts	2	2	0	-	-	-	-
Preserves and Accompaniment							
Jams/ Marmalades	8	0	2	27.8	35.4	5.5	42.6

Pickles/ Murabbas	10	2	6	6.5	10.9	5.3	10.9
Jellies	9	2	3	9.3	15.1	3.1	24.1
Table Olives	4	4	0	-	-	-	-
Chutneys	5	5	0	-	-	-	-
Dessert Toppings	5	3	0	104.6	104.6	104.2	104.9
Sugars							
Raw Sugar (Khandasari)	2	0	0	66.9	66.9	65.3	68.5
Refined Sugar	10	1	3	20.4	28.1	5.3	42.7
Brown Sugar	4	4	0	-	-	-	-
Jaggery	5	0	0	22.7	22.7	20.2	24.1
Bura (Powdered Sugar)	5	0	0	30.3	30.3	15.4	42.7
Misri (Rock Candy)	5	5	0	-	-	-	-
Confectionery Items							
Fruit Bars	5	0	0	89.7	89.7	75.7	103.7
Digestives	5	0	2	68.5	110.9	5.5	115.6
Toffees	13	1	5	16.5	22.2	3.2	32.5
Sugar Coated Coloured Fennel	5	2	1	12.2	15.7	5.6	17.8
Chewing Gums	10	0	4	12.4	15.9	3.2	28.9
Chocolates	8	0	6	6.7	11.9	3.2	13.2
Brittles (Chikkis)	5	5	0	-	-	-	-
Candy Floss	5	0	3	10.9	19.6	7.4	19.6
Snow Balls (Chuski)	5	0	0	65.4	65.4	61.3	71.6
Beverage concentrates							
Syrups/ Sherbets	9	0	0	55.3	55.3	11.1	170.9
Fruit Cordials	4	0	0	274.3	274.3	243.1	305.6
Squashes/ Crushes	10	0	0	199.6	199.6	83.2	288.7
Soft Drink Powder	5	3	1	11.1	17.1	3.1	17.2
Ready To Serve Beverages							
Non-alcoholic Carbonated Beverages	9	0	4	14.8	39.9	5.5	60.5
Non-alcoholic Fruit Ready to Serve Beverages	12	0	8	21.8	56.6	3.2	95.4
Fruit Juices	9	5	3	50.8	93.2	5.6	93.2
Alcoholic Wines	4	0	0	129	129	99.4	171.5

Beers	5	0	1	11.3	12.8	8.9	14.6
Flavoured Milk	3	3	0	-	-	-	-
Bakery Products							
Biscuits/ Crackers	16	0	7	23.6	42.2	6.2	43.5
Cakes/ Pastries	8	4	4	9.6	11.3	6.5	12.5
Breads	4	4	0	-	-	-	-
Miscellaneous Snack Items							
Breakfast Cereals	5	1	2	10.8	16.7	7.5	23.1
Potato Chips	8	0	1	36.1	39	5.3	48.9
Namkeens	5	5	0	-	-	-	-
Ice Creams	15	5	6	7.9	12.3	3.1	17.1
Sweet Meats (Mithaais)	15	2	10	7.8	17.1	3.2	17.7
Frozen Potato Snacks	6	3	1	7.7	15.3	7.2	20.2
Soup Powders	2	2	0	-	-	-	-
Instant Noodles and Pasta	4	4	0	-	-	-	-
Flavoured Yoghurt	3	3	0	-	-	-	-
Miscellaneous Food Ingredients							
Corn Flour/ Starch	5	1	1	9.8	11.4	9.3	12.2
Icing Sugars	3	0	1	8.5	10.2	8.3	10.4
Glazed Fruits	1	1	0	-	-	-	-
Desiccated Coconut Powder	2	0	0	14.6	14.6	13.3	15.8
Gelatin	2	0	0	93.2	93.2	91.9	94.5
Liquid Glucose	1	0	0	26	26	26	26
Corn Syrup	1	0	0	15.9	15.9	15.9	15.9
Caramel Color	1	0	0	317.1	317.1	317.1	317.1
Cocoa Powder	2	0	0	70.9	70.9	63.9	77.8

Table 3: Concentration of sulphites (mg/kg) in various food products. Nd- not detectable where sulphites were absent i.e., not detected so concentration was taken as 0. [a]Mean was derived assuming samples with detectable levels of sulphites and those with concentration below LOQ as having a value of 5 mg/kg. [b]Mean was derived only for samples in which sulphite levels were above LOQ.

In dried fruits, the concentration of sulphites was found to be in the range of <10 mg/kg to 405.9 mg/kg. The average concentration of sulphites in sultanas and apricots as reported in Australia and New Zealand [23] were 76 mg/kg and 2097 mg/kg respectively. In countries like Austria [28] and Spain [29], the concentration of sulphites in dried fruits was found to be 339.5 mg/kg and 1434 mg/kg respectively. In the present study the average concentration of sulphites in sultanas and apricots were found to be 20.6 mg/kg and 267.4 mg/kg respectively, which was much lower than the values reported elsewhere.

In case of preserves, in the present study the average concentration of sulphites in jams/ marmalades was found to be 27.8 mg/kg which was higher than the concentration of <10 mg/kg as reported by countries like Australia and New Zealand [23], Italy [26] and Korea [27] and of 13.5 mg/kg as reported in Austria by Mischek and Cermak [28]. In some of the products like pickles / murabbas (a fruit or vegetable immersed in spice flavored sugar syrup), jellies, chutneys and dessert toppings, sulphites were not labelled on the packet but were detected, which could be due to the presence of possible contributing ingredients like sugar in pickles/ murabbas; sugar, liquid glucose, fruit concentrates in jellies; sugar, liquid glucose, fruit concentrates, cocoa powder and caramel color in chutneys or dessert toppings in which addition of sulphites is permissible. In products like chutneys and table olives, sulphites were not detected. The concentration of sulphites in pickles was found to be <10 mg/kg which was lower than the concentration of 13.4 mg/kg and 30.3 mg/kg reported in countries like Spain by Urtiaga et al. [29] and in China by Zhang et al. [30] respectively.

Sulphur dioxide is widely used for sugar manufacture. In raw sugar manufacture, it prevents browning during the evaporation and crystallization processes. In processing of cane sugar, the extracted fruit juice is treated with lime and sulphur dioxide and the clarified extract is then crystallized. Besides this sulphur dioxide has several functions in case of sugar manufacture like inhibition of enzymic oxidation of polyphenols, inhibition of maillard reaction, precipitation of calcium ions as calcium sulphite and liberation of free acid from calcium salts of organic acids [31]. The amount of sulphite was higher in the raw sugar samples as compared to the samples of refined sugar, brown sugar, rock candy, jaggery and powdered sugar (bura). The average concentration of sulphites in refined sugars was found out to be 20.4 mg/kg which was higher than the concentration of <10 mg/kg as reported in countries like Italy and Korea by Leclercq et al. [23] and Suh et al. [27] and 16.4 mg/kg as reported in China by Zhang et al. [30] respectively. In products like brown sugar and misri (Rock candy), sulphites were not detected.

In case of confectionery items, sulphites were not labelled on the packet of the product but were detected, which could be due to the presence of certain possible contributing ingredients like sugar, liquid glucose, corn syrup, cocoa solids, caramel color and fruit syrups / concentrates in which the addition of sulphites is permissible. The average concentration of sulphites in fruit bars was found to be 89.7 mg/kg which was higher than the concentration of <10 mg/kg as reported in Australia and New Zealand by FSANZ [23]. In case of chewing gums, in the present study the concentration of sulphites was found to be 12.4 mg/kg which was similar to the concentration of 12.4 mg/kg as reported in Korea by Suh et al. [27].

In case of beverage concentrates, the average concentration of sulphites in fruit cordials was found to be 274.3 mg/L which was higher than the concentration of 10 mg/L as reported in Australia and New Zealand by FSANZ [23]. In case of ready to serve beverages, the average concentration of sulphites in non-alcoholic carbonated beverages and ready to serve fruit beverages was found to be 14.8 mg/L and 21.8 mg/L respectively which was relatively higher than the concentration of 10 mg/L as reported in Australia and New Zealand by FSANZ [23] and of 6.5 mg/L as reported in Austria by Mischek and Cermak [28] respectively. In fruit juices, the average concentration of sulphites was found to be 50.8 mg/L. On an average the concentration was much higher than the concentration of 2 mg/L as reported in Australia and New Zealand by FSANZ [23] and <10 mg/L as reported in China [30], Brazil [24], Korea [27] and in Italy [26]. In wines in the present study, the average concentration of sulphites was found to be 129 mg/L which was higher than the concentration of 22.7 mg/L, 122 mg/L, 92.4 mg/L, 123 mg/L, 14.2 mg/L and 92 mg/L respectively as reported in China [30], Spain [29], Austria [28], Australia and New Zealand [23], Korea [27] and in Italy [26] respectively but lower than 150 mg/kg as reported in Brazil by Machado et al. [32]. In beers, the average concentration of sulphites was found to be 11.3 mg/L which was higher than the concentration of <10 mg/L as reported in Australia and New Zealand by FSANZ [23] but lower than the concentration of 12.7 mg/L and 16 mg/L as reported in Spain and Italy by Urtiaga et al. [29] and Leclercq et al. [26] respectively.

Using the average concentration of sulphite obtained in the study, a glass (200 ml) of beverage like squash prepared using a 50 ml of concentrate will provide approximately 71% of the ADI for a child weighing 20 kg. If the child always selects the same brand with maximum concentration of sulphite detected in the present study, then he/ she would be consuming approximately 103% of the ADI. A 200 ml serving of a ready to serve beverage like a carbonated drink will provide 21% of the ADI for the child. If the child is loyal to the brand having the maximum concentration of sulphite detected in the study, he/ she would be consuming 136% of the ADI. A 100 ml glass of wine will contribute to 41% of the ADI for sulphites for an adult weighing 60 kg.

In case of bakery products, the average concentration of sulphites in case of biscuits / crackers was found to be 23.6 mg/kg which was higher than the concentration of <10 mg/kg as reported in China and Italy by Zhang et al. [30] and Leclercq et al. [26]. In case of cakes / pastries, sulphites were not labelled on the packet of the product. But they were detected which could be due to the presence of certain possible contributing ingredients like sugar, liquid glucose, desiccated coconut, glazed fruits, dried fruits, icing sugar, corn starch, corn syrup, cocoa solids, caramel color and fruit syrups / concentrates in which the addition of sulphites is permissible. The average concentration of sulphites in case of cakes / pastries was found to be <10 mg/kg which was similar to the concentration reported in Australia and New Zealand by FSANZ [23].

In case of miscellaneous snack items like potato chips, frozen potato snacks, breakfast cereals, ice creams, sweet meats and corn flour, sulphites were not labelled on the packet of the product. But they were detected in some of them, which could be due to the presence of certain possible contributing ingredients like dehydrated potato or corn starch in potato chips and frozen potato snacks; sugar, liquid glucose, glazed fruits, dried fruits in breakfast cereals; sugar, liquid glucose, glazed fruits, dried fruits, icing sugar, corn starch, corn syrup, cocoa solids, desiccated coconut, caramel color and fruit syrups / concentrates in ice creams and sweet meats in which the addition of sulphites is permissible. In ice creams, the average concentration of sulphites was found to be <10 mg/kg which was similar to those reported in Australia and New Zealand by FSANZ [23] and in Italy by Leclercq et al. [26]. In potato chips, the average concentration of sulphites was found to be 36.1 mg/kg which was higher than the concentration of <10 mg/kg as reported in Australia and New Zealand by FSANZ [23] but relatively below the concentration of 40 mg/kg as reported in Italy by Leclercq et al. [26]. In breakfast cereals, the average concentration of sulphites was found to be 10.8 mg/kg which was almost similar to the concentration of <10 mg/kg as reported in Australia and New Zealand by FSANZ [23] and in Italy by Leclercq et al. [26]. In frozen potato products, the average concentration of sulphites was found to be <10 mg/kg which was similar to the concentration of <10 mg/kg as reported in Australia and New Zealand by FSANZ [23] and in Italy by Leclercq et al. [26]. In corn flour/ starch, the average concentration of sulphites was found to be <10 mg/kg which was similar to the concentration of <10 mg/kg as reported in Korea by Suh et al. [27].

Some of the common food ingredients which could possibly lead to presence of sulphites were also analysed. This comprised of ingredients like icing sugars, glazed fruit, desiccated coconut powder, gelatine, liquid glucose, corn syrup, caramel color and cocoa powders in which addition of sulphites is permissible have also been depicted in Table 3. In cocoa powders, the average concentration of sulphites was found to be 70.9 mg/kg which was relatively higher than the range <10 mg/kg as reported in Korea by Suh et al. [27].

Besides these other products like tea leaves (n=2), vegetables (n=2), fruits (n=2), flour and grains (n=3), pulses and soy products (n=3), milk and milk products (n=5), hen egg (n=1), vegetable oil (n=1) and spices (n=2) were also analysed for sulphite which might be present in them naturally or might be present due to contamination. However, sulphite was non-detectable in these products.

The addition of sulphites in various permissible food products is regulated in India by national regulatory authority i.e., FSSAI. The data on the concentration of sulphites in various food products estimated in the present study has been compared with the MPLs given by FSSAI and General Standards for Food Additives (GSFA) Codex. This has been presented in Table 4.

However, it was interesting to note that for a few food products like apricots, dates, raw sugar, jaggery, bura, fruit bars, digestives, fruit juices, alcoholic wines, liquid glucose and corn syrup, the MPLs given by Indian regulations i.e., FSSAI were higher than the Codex GSFA standards. For a few food products like raisins, sultanas, jams/ marmalades, dessert toppings, pickles/murabbas, beer, refined sugar and corn starch, the MPLs given by GSFA were higher than FSSAI MPLs. No MPLs are given by FSSAI for food products like potato snacks, ice creams and cakes/ pastries. No MPLs are given by GSFA for food products like desiccated coconut powder, gelatine, caramel color, cocoa powder and its products, toffees, fennel, chewing gum and chocolates.

Food Products	**Maximum Permissible Level (MPL) for Sulphites (mg/kg) by FSSAI**	**Maximum Permissible Level (MPL) for Sulphites (mg/kg) by GSFA**	**% MPL**	
			FSSAI	**GSFA**
Dried Fruits				
Raisins	750	1000	34.8	26.1
Sultanas	750	1000	2.7	2.1
Apricots	2000	1000	13.4	26.7
Dates	50000	1000	0.2	10.3
Preserves And Accompaniment				
Jams/ Marmalades	40	100	69.5	27.8
Dessert Toppings	-	100	-	104.6
Pickles/ Murabbas	100	100	6.5	6.5
Jellies	40	40	23.3	23.3
Sugars				
Raw Sugar (Khandasari)	70	40	95.6	167.3
Refined Sugar	40	70	51	29.1
Jaggery	70	40	32.4	56.8
Bura (Powdered Sugar)	150	40	20.2	75.7
Confectionery Items				
Fruit Bars	100	30	89.7	299
Digestives	100	30	68.5	228.3
Toffees	2000	-	0.8	-
Sugar Coated Colored Fennel	2000	-	0.6	-
Chewing Gums	2000	-	0.6	-
Chocolates	150	-	4.5	-
Beverage Concentrates				
Syrups/ Sherbets	350	350	15.8	15.8
Fruit Cordials	350	350	78.4	78.4

Squashes/ Crushes	350	350	57	57
Soft Drink Powders	70	70	15.9	15.9
Ready To Serve Beverages				
Non-alcoholic Carbonated Beverages	70	70	21.1	21.1
Non-alcoholic Fruit Ready to Serve Beverages	70	70	31.1	31.1
Fruit Juices	350	50	14.5	101.6
Alcoholic Wines	450	250	28.7	51.6
Beers	70	250	16.1	4.5
Bakery Products				
Biscuits/ Crackers	GMP	50	-	47.2
Cakes/ Pastries	-	50	-	19.2
Miscellaneous Snack Items				
Potato chips	-	50	-	72.2
Frozen potato snacks	-	50	-	15.4
Ice creams	-	100	-	7.9
Miscellaneous Food Ingredients				
Corn flour/ starch	100	200	9.8	4.9
Icing sugars	20	20	42.5	42.5
Desiccated coconut powder	50000	-	0.1	-
Gelatin	1000	-	9.3	-
Liquid Glucose	40	20	65	130
Corn Syrup	450	40	3.5	39.6
Caramel Color	2000	-	15.6	-
Cocoa Powder	2000	-	3.5	-

Table 4: Comparison of sulphite concentration estimated in foods with national and international regulatory authority standards. MPL's given by FSSAI [2] and Codex GSFA [3].

In case of dried fruits, the concentrations of sulphites in all products were found to be below the MPL given by both FSSAI and Codex. The MPL's given by Indian regulatory body for apricots and sultanas were twice and fifty times higher than the Codex standards respectively. The ADI for sulphites is 0.7 mg/kg body weight/day. Theoretically for an Indian adult weighing 60 kg, the intake of sulphites can exceed the ADI, if he/she consumes 0.84 g of dates or 21 g of apricots (5 apricots) in a day if sulphites are present at MPL levels. For a child weighing 20 kg, the intake of sulphites can exceed the ADI, if he/she consumes 0.3 g of dates or 7 g of apricots (approximately 2) in a day. In preserves and accompaniments, for one local brand of jam the concentration of sulphites was found to be 106.5% of the MPL given by FSSAI. On an average the concentration was 69.5% of the MPL.

In case of sugars, the concentration of sulphites exceeded the MPL in all samples of raw sugars/ khandasari (local brands). It was found to be 167.3% of MPL given by Codex GSFA and almost 95% of FSSAI MPL. For refined sugars, the concentration of sulphites exceeded the FSSAI MPL for one of the un-branded samples (106.7% of MPL). The MPLs given by the Indian regulatory body for raw sugar, jaggery and powdered sugar (bura) were 1.75 times to 3.75 times higher than the Codex standards.

In case of confectionery items, the concentration of sulphites exceeded for fruit bars (n=3) including local brands and un-branded products with the highest being 103.7% and 346% of MPLs given by FSSAI and Codex GSFA respectively. For fruit digestives, the concentration of sulphites exceeded for one un-branded sample and was found to be 115.6% of the FSSAI MPL and 385.3% of the GSFA MPL. The MPLs given by Indian regulatory body for fruit bars and digestives were 3.33 times higher than the Codex standards.

In case of beverage concentrates and ready to serve beverages, the concentration of sulphites was found to be below the MPLs for all products. Only for one of the un-branded non-alcoholic ready to serve fruit beverage, the sulphite concentration was found to be 136.2% of the FSSAI MPL. The MPLs given by Indian regulatory body for alcoholic wines and fruit juices were two times to seven times higher than the Codex standards. Theoretically for an Indian adult weighing 60 kg, the ADI of sulphites can be exceeded if he/she consumes 120 ml of fruit juice or 93 ml of alcoholic wine in a day when sulphites is present at MPL levels of 350 ppm and 450 ppm in fruit juices and alcoholic wines respectively. For a child weighing 20 kg, the intake of sulphites can exceed the ADI, if he/she consumes 40 ml of fruit juice or a glass of beverage concentrate containing 40 ml of concentrate or a glass (200 ml) of a ready to serve beverage in a day when sulphites is present at the MPLs.

For bakery products and for miscellaneous snack items, the concentration of sulphites was well below the MPLs as given by Codex GSFA. FSSAI does not give MPLs for these products.

Some of the food ingredients like icing sugars, glazed fruit, desiccated coconut powder, gelatin, liquid glucose, corn syrup, caramel color and cocoa powders in which addition of sulphites is permissible were also tested. The concentration of sulphites exceeded only for liquid glucose which was found to be 130% of GSFA MPL. The MPLs given by Indian regulatory body for liquid glucose and corn syrup were twice to ten times higher than the Codex standards.

The study showed the presence of sulphites in food products like fruit bars, fruit digestives, jams, raw sugar, refined sugar, dessert toppings, ready to serve fruit beverage and liquid glucose at levels higher than the maximum levels permitted by the national or global legislations. These food products comprised of mostly local brands and un-branded products. This implies lack of awareness among small scale manufacturers about the appropriate amount of sulphite to be added to products.

Conclusion

Most of the food products analysed presented sulphite levels within the maximum permissible levels established by the Indian legislation and Codex GSFA. However, some of the food products like fruit bars, fruit digestives, jams, raw sugar, refined sugar, dessert toppings, ready to serve beverages and liquid glucose presented sulphite levels higher than the maximum levels permitted by the national or global legislations. These food products were mostly manufactured by small scale manufacturers or the un-organised sector. This highlights the need for setting up quality control mechanisms and spreading awareness about good manufacturing practices in this sector.

Theoretically, where sulphite is present at maximum permissible levels it is easy to exceed the ADI even if only one type of food product containing sulphite is consumed. This requires a relook at the MPLs prescribed for these food products. The levels of sulphite found in this study are much below the MPL. Hence when it is technologically feasible to add sulphur dioxide at lower levels why should the MPL levels be set at such high levels. MPLs prescribed by India also differed from those given by codex GSFA for some foods. To facilitate trade between countries, the issue of harmonization of standards needs to be examined more closely.

Codex Alimentarius Commission (CAC) has made it mandatory for declaring presence of sulphites on labels of food products in which the addition of sulphites is above 10 ppm. This is presently lacking in our country. This information could help individuals who are sulphite sensitive in selecting food products.

The data generated in this study will contribute to a databank on the levels of sulphites in foods and beverages in India as most brands which are available throughout the country have been analysed. This will facilitate the assessment of dietary exposure to sulphites among Indians.

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