

Estimate of intake of sulfites in the Belgian adult population

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Food Additives and Contaminants



Estimate of intake of sulphites in the Belgian adult population

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3 4	1	Estimate of intake of sulfites in the Belgian adult population
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46 47 48 49 50 51 52 53 54 55 56 57 58 59 60	32 33 34 35	Running title: intake of sulphite in the Belgian population Keywords: food consumption survey, sulphite, Tier 2, Tier 3, adults, Belgium

38 Abstract39 An exposure ass

An exposure assessment was performed to estimate the usual daily intake of sulfites in the Belgian adult population. Food consumption data were retrieved from the national food consumption survey. In a first step, individual food consumption data were multiplied with the maximum permitted use levels for sulfites, expressed as sulphur dioxide, per food group (Tier 2). In a second step, on the basis of a literature review of the occurrence of sulfites in different foods, the results of the Tier 2 exposure assessment and available occurrence data from the control programme of the competent authority, a refined list of foods was drafted for the quantification of sulphite. Quantification of sulphite was performed by a high performance ion chromatography method with eluent conductivity detector in beers and potato products. Individual food consumption data were then multiplied with the actual average concentrations of sulfite per food group, or the maximum permitted levels in case actual levels were not available (partial Tier 3). Usual intakes were calculated using the Nusser method. The mean intake of sulfites was 0.34 mg/kg bw/day (Tier 2), corresponding with 49 % of the acceptable daily intake (ADI) and 0.19 mg/kg bw/day, corresponding with 27 % of the ADI (partial Tier 3). The food group contributing most to the intake of sulfites was the group of wines. The results showed that the intake of sulfites is likely to be below the ADI in Belgium. However, there are indications that high consumers of wine have an intake around the ADI.

77 Introduction

There are 3 ways of being exposed to SO_2 : by inhalation, by skin contact or by ingestion from the food chain (NPI, 2005). Sulfite (H₂SO₃) is formed by sulphur dioxide (SO₂) gas which is very soluble in water. The presence of sulphur dioxide in the environment is due to combustion processes of organic compounds, such as natural combustion from forest fires or combustion in energy or transformation plants. Sulfite is also present in the exhaust fumes from cars and trucks. Depending on the pH of the environment, different forms of sulfite can exist or coexist.

Sulfite has raised concerns because of its toxicity for the lungs and its ability to cause allergic reactions, especially in asthmatic persons (Valley et al., 2009). It was estimated that an exposure of 10 to 50 ppm during 5 to 15 min may have a measurable effect on humans. In the UK, 2 ppm (8 hours time weighted average TWA) or 5 ppm (10 min TWA) were set as maximum occupational exposure values (European Cooperation on School Education, 2004). In Belgium, the maximum allowed concentration of sulfite in the air is 350 μ g/m³ during maximum 1 hr or 125 µg/m³ during maximum 24 hr (European Community 1999a). In a review of the Health Council of the Netherlands sulfite was found to be responsible of chronic urticaria, skin allergy and bronchial asthma in some humans. The NOAEL in rats was set at 72 mg SO₂/kg bw/day (Health Council of the Netherlands, 2005).

98 The Acceptable Daily Intake (ADI) is the amount of SO₂, in food that can be ingested orally 99 over a lifetime without an appreciable health risk. The ADI is expressed per kg body weight 100 (abbreviated "bw") per day. In the case of sulfites (expressed as SO₂), the JECFA (Joint 101 FAO/WHO Expert Committee on Food Additives) has set the group ADI value to 0.7 mg/kg 102 bw/day (JECFA, 1999).

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Sulfite can be intentionally added to food for its antioxidant or antimicrobial properties. Sulfite inhibits the enzymatic activity of polyphenol oxidase, which is responsible for browning in fruits, vegetables and seafood products. It also prevents oxidation of food due to contact with the air and the browning effect occurring from the Maillard-type reactions. The aim of adding sulfite to wine is to prevent microbial growth and oxidation of volatile odorous compounds which may develop during the fermentation (Ough, 1986).

Sulfite is allowed as a preservative (E220-E228) in a list of different foods (excluding all
others) under the condition of respecting the maximum use levels, as specified by the
European Directive 95/2/EC as amended (European Community 1995) and for wine in annex
V of the Council regulation (EC) No. 1493/1999 (European Community 1999b).

The concentration in different foods is expressed as sulphur dioxide (SO_2) in mg/kg or mg/l, depending on the nature of the food and relates to the total quantity from all sources. The legislation mentions that an SO₂ content of not more than 10 mg/kg or 10 mg/l is not considered to be present. Directive 2000/13/EC on labelling of pre-packaged food requires labelling of products containing sulfite, if the SO₂ concentration is higher than 10 mg/kg or 10 mg/l, for the benefit of people with a food intolerance for sulfite (European Community 2000).

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Sulfite can be present in foods as free sulfite or react with a variety of organic compounds in foods to form reversibly or irreversibly bound products. In our digestive tract, the free and reversibly bound forms can be liberated (Wedizicha, 1992). García-Alonso et al., (2001) reported that the amount of added sulfite in fresh sausage was reduced by 26 % when S-sulphonate bounds (irreversibly bound) were formed and by 23 % when C—O—SO₂⁻ bounds (reversibly bound) were formed. Those percentages of loss may vary sharply from 10 to 49 % depending on the conditions when adding the sulfite to the meat (time of mixing, presence of oxygen, degree of mincing and fat composition of the meat).

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Cooking was shown to reduce the level of sulfite in beef and chicken burgers (Pena-Egido et al; 2005; Garcia-Alonso et al., 2001) the distribution of the 3 different forms of sulfite (free, reversibly bound and S-sulphonate) remained approximately the same after as before the cooking process. The losses reported for meat burgers were 31 % for the S-sulphonates, 29 % for the free sulfite and 28% for the total sulfite. This study also showed that the frying process reduced the amount of total sulfite on average by 41 % (± 13%). A study in shrimps (Armentia-Alvarez et al., 1994) showed that during cooking the loss of free and reversibly bound sulfite varied between 45 and 55 %. The level of total sulfite was about 5 times higher in the non edible parts compared to the edible parts. The authors concluded that the boiling step did not change the distribution of free and total sulfite in shrimps, but a reduction of about 40 % was observed for the total sulfite in the edible parts. From the boiled water it was

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143 shown that the loss of SO_2 in shrimps during the boiling process was not totally recovered in 144 the water due to oxidation of sulfite and reaction of sulfite with food components.

Another study on shrimps (Armentia-Alvarez et al., 1997) showed that during bad freezing storage (> -18° C), the free sulfite combined with the disulfide bounds of proteins denatured by the freezing process. The longer the storage time, the higher the amount of combined sulfite produced. Reduction of sulfite concentration after a storage period of 1 month was reported to be between 25 and 50 % for fish products, potatoes and dried fruits (Leclercq et al., 2000)

Finne et al., (1986) studied the residual sulfite content in shrimps after different days of conservation in ice. They demonstrated that, starting with a concentration of about 80 ppm, the residual content was reduced to less than 10 ppm after 6 days of storage on ice.

Directive 95/2/EC (European Community, 1995) requires that Member States of the EU monitor the intake and use of food additives in order to allow evaluation of intake trends. Measures can be taken to amend the legislation if intake concerns are identified in one or more Member States. The Report from the European Commission on Dietary Food Additive Intake in the European Union (European Commission, 2001) describes a harmonized, tiered method to estimate additive intakes. In the first step (Tier 1), theoretical food-consumption data (Hansen, 1979) are combined with the maximum permitted usage levels for the additive. This conservative estimate was performed at European level. For the additives exceeding the ADI in Tier 1, in a further step (Tier 2), national food consumption data are combined with maximum permitted usage levels. Such intake estimate will still overestimate the real intake, but can exclude additives, which do not pose a major risk from the most difficult estimate in Tier 3: a combination of actual national food consumption data with actual levels of the additive. The report from the Scientific Committee on Food (SCF) about Dietary Food Additive Intake in the European Union (1998) showed that, based on Tier 1, there were 10 additives in the European Union for which a more refined intake estimate was needed. Among those additives also sulfite was mentioned (Scientific Committee on Food). A Tier 2 exposure assessment in 7 countries (Belgium not included) showed that the range of estimated intakes was 20-266% of the ADI for adults (European Commission).

The objective of this study was to perform a refined exposure assessment for sulfite intake in
 the Belgian adult population, using the data from the national food consumption survey of

176 2004. Another objective was to identify the food groups contributing most to the total intake177 of sulfites in Belgium.

179 Materials and Methods

181 Study design

A semi-probabilistic approach was used to perform the exposure assessment. For the Tier 2 exposure assessment, maximum levels of sulphur dioxide in different foods were linked to the food consumption data of these specific foods, derived from the Belgian food consumption survey (2004). For some foods in the EU legislation, it is unlikely that they are consumed in significant quantities or they were too difficult to link with the data from the food consumption survey: burger meat with a vegetable or cereal content of minimum 4 %; breakfast sausages; Longaniza fresca and Butifarra fresca; dried salted Gadidae fish; sago; pearl barley; dried ginger; pulp of horseradish; pulp of onions, shallots and garlic; yellow paprika in brine; flavourings on the basis of lemon fruit juice; concentrated grape juice for the making of house wine; mostrarda di frutti; gelatinized fruit extracts or fluid pectin; glass preserves of cut lemon; white syrup and molasses; orange-, grapefruit-, apple- and pineapple juice for unpackaged sell in catering enterprises; concentrates on the basis of fruit juice with at least 2.5 % pearl barley; *capilhé groselha*; unfermented grape juice for religious use; wine without alcohol; fermented vinegar; marinated nuts; vacuum packaged sugar maize and distilled alcoholic beverages with whole pears. Consequently these products were not taken into account in the intake assessment. This could lead to an underestimation of the intake, but this was supposed to be limited. As stated in the introduction, cooking and freezing can decrease the sulfite concentration of foods. This was not taken into account either due to the lack of reliable processing factors. Consequently this could lead to an overestimation of the intake.

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In order to perform a more refined estimate of the sulfite intake (partial Tier 3 exposure assessment), on the basis of literature, available concentration values for products analyzed by The Belgian Federal Agency for Safety of the Food Chain (FASFC) and the results from the Tier 2 exposure assessment, it was decided to analyze concentrations of sulphur dioxide in beers and potato products. In these samples concentrations were determined using a high performance ion chromatography method with eluent conductivity detector, optimized and validated for this purpose. In the partial Tier 3 exposure assessment, analyzed actual levels of

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sulphur dioxide in different foods were linked to the food consumption data of these specific foods, derived from the Belgian food consumption survey (2004). For foods for which no analyzed levels were available, maximum concentrations were used, as in the Tier 2 exposure assessment. No illegal uses of sulfites in food groups in which no use of sulfites is authorised, were taken into account in this study.

Although the regulation specifies maximum levels for specific groups, in this paper, the results of the analyses were grouped to ensure a clear data presentation. The grouping was performed according to the EU directive 95/2/EC (European Community, 1995) as follows: beers; wines; sugars and glucose-based products; condiments and dessert sauces; gelatine and meat substitutes; shellfish; cereal-based products (starches and biscuits); processed potatoes, processed fruits (including dried fruits, candied fruits, jams, jellies and marmalades, lemon and lime juice, fruit fillings and fruits in vinegar, brine, oil) and processed vegetables (including dried tomatoes, mushrooms, vegetables in vinegar, brine and oil and white vegetables).

Food consumption data

Consumption data from the national food consumption survey 2004 were used to perform the exposure assessment. Aims, design and methods of this survey are described elsewhere (De Vriese et al. 2005). The target population comprised all Belgian inhabitants of 15 years or older. The sample included 3245 participants randomly selected from the National Register, using a multi-stage stratified procedure.

Information on dietary intake was collected by two non-consecutive 24-hour recalls in combination with a food frequency questionnaire. During the 24-hour recall interviews the respondent reported the quantity of all foods and beverages consumed during the preceding day. The 24-hour recall was carried out using EPIC-SOFT software (Slimani & Valsta, 2002). This program allows obtaining very detailed information about the foods consumed and the recipes used in a standardized way.

> 3083 participants, of which 1537 women and 1546 men, completed two 24-hour recalls. Participants were categorized into four age groups: 15-18 years (n=760), 19-59 years (n=830), 60-74 years (n=789) and 75 years or older (n=704).

Sulphur dioxide concentration of selected foods

The selection of the matrix types to be analyzed was based on the results of the Tier 2 calculation, literature and on the available sulfite concentrations obtained from the FASFC in different kinds of foods (Table 1). Both beers and pre-cut fresh potatoes or French fries and frozen prepared potato (precooked or not) products were sampled.

The Belgian law (Belgian Monitor) specifies 4 categories of beer (S and I to III) depending on the Plato degree. Beers are characterized according to the fermentation type, the bitterness, the colour, the wort density and the alcohol concentration. Those characteristics were important in the present study, for example in case of ale beers undergoing a second fermentation in the bottle where sulfite can be released by the fermentation process. The presence of fruits can also be a source of sulfite. Therefore, in this study, beers were categorized as follows: beer table blond, beer table brown, beer table triple blond, beer table non specified colour, beer pilsner, beer pilsner light, beer without alcohol, beer special blond, beer special amber, beer special brown, beer special with fruits, beer special unknown colour, beer gueuze, beer gueuze with fruits, beer white, beer white with fruits and beer unknown type and colour.

From the Belgian food consumption survey 2004 the frequencies and amounts of beer consumption for all beers were retrieved. Each brand was assigned to a group of beer, as defined earlier; and some simple calculations were made to determine the percentage of consumption for each beer category and each brand within each category. The number of samples was taken proportional to the brand distribution in the respective categories.

Samples were only taken for the brands with a percentage of consumption higher than 0.5 %, except for 1 brand of gueuze with fruits and 2 other brands of gueuze because they are representative in their specific categories and they were observed in different stores.

potato products and 46 for beers) were collected and analysed.

Collection of samples was performed in 2007 in 5 major supermarket chains with a good

market share, which are visited by approximately 77.5 % of the Belgian population in search

for their food provisioning (CRIOC - OIVO). In total for both categories 67 samples (21 for

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The only existing ISO method including sulfite analysis is the method ISO 10304-3:1997. The HPIC/conductivity detector with eluent conductivity suppressor was used in this study to perform the analyses. Ion chromatography is an appropriate technique for the separation of sulfite from other anions like phosphate and sulphate, and it has also been recognised for the quantification of sulfite (Armentia-Alvarez A. et al., 1997; Kim 1989; Kim et al. 1987; Kim et al. 2000; Paino-Campa et al. 1991; Pena-Egido et al., 2005; Pizzoferrato et al. 1990) Pulsed amperometric detection necessitates very specific material and is not so easy to use because of possible fouling of the electrode by matrix compounds (McFeeters and Barish, 2003). The most used detection method in ion chromatography is eluent suppressed conductivity. With the carbonate/bicarbonate eluent, the produced species in the suppressor is H_2CO_3 (which decomposes in CO_2 and H_2O).

289 Material

For sample preparation, a Robocoup mixer 3000, a T25 Ultra-Turrax (IKA) and a centrifuge Beckman Avanti J-25 operated at 7500rpm were used. All extracts where filtered through 0.2µm PVDF syringe filters. The analytical system was composed of a Waters 717+ auto injector, an HP 1050 pump, an Alltech Suppressor model 640 and an Alltech conductivity detector model 650. The acquisition of the signal and data treatment (calibration curve and extract concentration) were made by a computer equipped with an analogue/digital converter and ChromQuest software version 4.1. The separation column was composed of a Dionex RFIC analytical column type IonPac AS14A-5µm 3x150mm (particle size of 5µm) and precolumn RFIC IonPac AG14A-5µm 3x30mm.

299 Reagents and solutions

All reagents were at least from analytical grade. Sodium carbonate and sodium bicarbonate where purchased from VWR International. D-Mannitol, disodium salt of ethylenediamine tetra acetic acid (EDTA) and formaldehyde were purchased from Sigma-Aldrich and anhydrous sulfite was purchased from Fluka.

The aqueous solutions where prepared with a MilliQ Reagent Grade System's water with
conductivity lower then 18.2 MΩcm⁻¹ (from Millipore coupled after a Millipore Elix 100
deionised water production system). Stock solutions NaHCO₃ 0.3 M, Na₂CO₃ 0.2M and DMannitol 0.5 M were prepared. Those solutions were kept in the fridge. The eluent used for
the anion separation was the buffer NaHCO₃/Na₂CO₃ 3mM/2mM. The sulfite extraction

buffer was a mixture of NaHCO₃/Na₂CO₃ /D-Mannitol 9mM/16mM/10mM. This resulted in a
final extraction pH between 8 and 10, as proposed in literature for free sulfite and reversible
bound sulfite extraction (Armentia-Alvarez A. et al. 1993; Wedzicha, 1992). The 1000 mg/l
sulphur dioxide standard stock solution was prepared by weighting 0.1968g anhydrous sulfite
and making a 100.0 ml solution with extraction buffer.

316 Sample collection, storage and pre-treatment

After collection of the samples, they were identified by a unique identification number and registered in a Microsoft Excel sheet. Following characteristics were added: brand name, product description, lot number, temperature of conservation in the store and any relevant information for the project (presence or absence of sulfite mentioned on the label or usage of ingredients containing sulfite). The liquid samples where stored in a fridge and the solid samples where stored in the freezer (-18°C or less) until pre-treatment or analysis.

The pre-treatment of the solid samples consisted of grinding at least 200 g collected sample with a kitchen robot (Robot Coupe model R 301 Ultra) in order to get piece cuts less than 3~5 mm. Afterwards the prepared samples were stored deep frozen. All analyses where conducted within 3 weeks after collection of the samples.

328 Sample preparation

For the preparation of the liquid samples 100 µl of D-Mannitol 0.5 M was added to about 100 ml of the sample before degassing in an ultrasound bath for 5 minutes; 5.0 ml of sample was then diluted in the extraction buffer by a factor of 5 (final volume = 25.0 ml) and the extract was filtered through a PVDF syringe filter with 0.2 µm pore size before injection on the HPIC system. For the preparation of the solid samples, after a short period of unfreezing, about 5 g of sample (weighted with at least 3 digits after the point) was weighted in a P.E. tube of 50 ml to be centrifuged; 25.0 ml of extraction buffer (measured with a graduate cylinder of 25 ml) was added to the sample; the sample was then homogenated in an Ultra-Turrax for a minimum of time, so that the mixture looked like creamy (10 to 15 seconds suffice).

The tube was closed and set in a centrifuge at 7500 rpm for 10 minutes; the surnatant extract
was filtered through a PVDF syringe filter with 0.2 μm pore size before injection on the HPIC
system.

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2 3 4	341	
5 6	342	Calibration curve - linearity
7 8	343	Due to the range of maximum permitted levels in the different kinds of foods it was chosen to
9 10	344	use a calibration curve ranging from 0.1 to 200 mg SO_2/l .
11 12	345	By diluting 1000 mg SO ₂ /l extraction buffer a series of eight dilutions was prepared $(0.1, 0.5,$
13	346	1. 5, 10, 50, 100, and 200 mg SO ₂ /l). Blank was used as 0 mg SO ₂ /l. The best fit was used in
14	347	order to maximize the coefficient of determination (> 98 %) and to minimise the residues at
16 17	348	each level.
18 19 20	349	Validation
20 21 22	350	The method was validated for linearity, precision (repeatability, reproducibility), accuracy and
23	351	limit of detection according to the procedure described by Van Loco et al (Van Loco and
24 25	352	Beernaert 91-94). After multiple tests for concentration ratios of the NaHCO ₃ /Na ₂ CO ₃ buffer
26 27	353	it was decided to use the composition 3 mM/2 mM. This composition permits the separation
28 29	354	of phosphate, sulfite, and sulphate with a total runtime period of 25 minutes (stabilization
29 30 31 32 33 34 35 36	355	included). Different stabilization compounds (EDTA, D-Mannitol and formaldehyde) were
	356	tested according to previous reports. D-Mannitol was chosen due to its best effect on the
	357	sample extract. Some stability tests were made with a standard solution 10 mg SO ₂ /l. The
	358	responses of a standard solution prepared in water and the same amount of standard solution
37 38	359	prepared in a pH~ 9.3 buffer with 10 mM D-mannitol were compared.
39 40	360	
40 41 42	361	The final sulfite extraction buffer chosen after pH test in mixture with the sample matrix was
43	362	NaHCO ₃ /Na ₂ CO ₃ /D-Mannitol at concentration levels of 9 mM/16 mM/10 mM respectively.
44 45	363	The pH of this buffer was between 9 and 10. The extracts prepared with beer resulted in a pH
46 47	364	of 9.3 and of 8.7 when prepared with frozen French fries. NaOH/formol was tested for the
48 ⊿9	365	extraction of sulfite from beer. But the chromatographic separation gave very high
50 51	366	background noise after a few injections. The material was not able to work with an organic
51 52	367	modifier like acetone in high proportion like described in Metrohm application note S-95.
53 54	368	
55 56	369	To test linearity it was opted for the weighted quadratic regression of the heights in function
57 59	370	of the extract concentration. This type of regression resulted in a determination coefficient
58 59	371	greater than 0.99 and residues lower than 15 % for the lowest concentration point.
60	372	

The range for the calibration was 0.1 to 100 mg/l extract (The injection of the point 200 mg/l was kept in order to be able to quantify higher amounts from a new regression). The range in the extract corresponded to a range in the sample going from 0.5 to 500 mg/kg (or mg/l, depending of the nature of sample) if the dilution factor was taken into account.

In order to test the repeatability and reproducibility a sample (table beer) was fortified at 4 levels of concentration corresponding to 0, 5, 10 and 15 mg/l of the sample (which corresponds to 0, 1, 2 and 3mg/l in the extract). The test was conducted in triplicate for each level and repeated for each level at 3 different days. The non fortified sample did not result in a detectable response and the obtained result of reproducibility was lower than 12.6 %.

The apparent recovery was, on average, for the 3 levels of concentration, 73 ± 6 at 3 concentration levels. A lower recovery may be explained by a possible reaction of SO₂ to some compounds present in the matrix and/or by oxidation after the dissolution of ambient oxygen into the extracts. Therefore, it was decided to correct the results for the recovery of the control standard included in the series of analysis.

In beer sample, the sulfite peak was poorly separated from matrix interferences. The minimum detectable concentration in the extract was about 0.5 mg/l. This corresponds to a quantification limit (LOQ) of 1 mg/l extract which corresponds to 5 mg/l (or mg/kg) in the sample. This is also the lowest tested concentration.

42
43395Exposure assessment

Only respondents with two completed 24-hour recall interviews were included in the analyses
(n= 3083; 1546 men and 1537 women).

 The individual intake of sulphur dioxide from a certain food product was estimated using thefollowing equation:

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$$y_i (mg / kgbw / day) = \frac{c \times x_i}{bw_i}$$

402 where y_i is the intake of sulphur dioxide by individual *i* from a particular food (in mg per kg 403 bodyweight per interview day), *c* is the concentration of sulphur dioxide in that food (mg per 404 kg), x_i is the consumption of a certain food by individual *i* (kg) and bw_i is the self-reported 405 body weight of individual *i* (kg). To estimate the total intake of sulphur dioxide per food

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group or per day, individual daily intakes of sulphur dioxide from different foods were added up.

As mentioned before, two approaches were used for the calculation of sulphur dioxide intake. In the Tier 2 approach, c_i represents the maximum allowed concentration in each food. The maximum permitted levels of sulphur dioxide used in different foods are listed in the European Directive 95/2/EC (European Community, 1995). In the Tier 3 approach, c_i represents the actual average concentration of sulphur dioxide that is observed in a particular food.

The usual intake distribution for sulphur dioxide was estimated with the Nusser method (Nusser et al. 1996) using the C-side software (Iowa State University, 1996). Several statistical methods are available to estimate usual intake distributions with the correct mean, variance and skewness. These statistical procedures adjust for within-person or day-to-day variability. Of all different statistical procedures, the Nusser method (Nusser et al., 1996) is highly recommended because it eliminates the intra-individual variance and additionally transforms the data to obtain approximately normally distributed data. The method is suitable to estimate usual intake distributions in a population both for normally and non-normally distributed foods and nutrients. The usual intake distribution was weighted and adjusted for the age and sex distribution of the Belgian population and adjusted for day of the week and season.

Results and discussion

As an adult inhales about 15 m³ of air per day, this means that in the worst case the amount of SO₂ intake from the environment is 215 μ g/m³ x 15 m³= 3225 μ g/ day = 3.2 mg/day. 215 $\mu g/m^3$ is the highest concentration of SO₂ measured in Belgium in 2005. Since the average body weight of a Belgian adult is around 70 kg, the final SO₂ intake from the environment is estimated to be 0.045 mg/kg bw/day. From those results it was decided to neglect the environmental contribution to SO₂ intake in Belgium.

The usual sulfite intake of the Belgian adult population estimated by the Tier 2 approach was 0.34 mg/kg bw/day (49 % of the ADI) at the mean level of consumption (Table 2). At the 97.5th percentile of the population, the sulfite intake was estimated to be 1.1 mg SO₂/kg bw/day (157 % of the ADI). It was observed that the ADI was exceeded beyond the 90th percentile of the population. At this percentile the contribution of wine consumption to the total intake was estimated to be 71 %. Including only wine consumers (55.1%), the ADI was reached at the 57.5th percentile of population (data not shown).

Wine contributed for more than 50 % to the total intake of sulfite. Sugar and glucose-based products, beers, processed fruits and processed potatoes contributed each between 7-13 % to the total intake, while shellfish and processed vegetables contributed each about 4 % to the total intake. Condiments and cereal products contributed to less than 1 % to the total average sulfite intake (Table 2).

Concentrations of sulfite in beer samples (n=46) were all under the detection limit (LOD) of 5 mg/l; these results were different from the concentrations found in other countries. For example, in France (Mareschi et al. 1992) and Italy (Leclercq et al., 2000) the average sulfite concentrations found in beer were respectively 7.5 and 15 mg SO₂/l. In these studies the Monier-Williams method was used to perform the analyses. It is known that this method has possible interferences with volatile compounds. The method used in the present study does not have this disadvantage. With the limit of quantification of 5 mg SO_2/l it was not possible to determine any residual SO₂ levels in the beers analyzed. Provided this and the results reported by other countries obtained by more sensitive electrochemical methods of analysis (Dvorak et al. 2006) it was decided not to perform an intake estimation of sulfite from beer using a null concentration of sulfite in beers. It is known that sulfite could be produced during the natural fermentation process in the bottle. Therefore half of the LOQ concentration was used (medium bound approach) instead of the null concentration to perform the intake estimation.

For potato products 6 samples out of 21 analyzed samples resulted in SO₂ levels above the quantification limit (LOQ) of 5 mg/l and only one sample with no sulfite declared on the label resulted in a concentration higher than the LOQ. This positive sample was a fresh pre-cut product in which the concentration was 9.9 mg SO₂/kg. For the samples with a concentration above LOQ, an average concentration value of 11.6 mg SO₂/kg was found and the results were spread between 9.0 and 13.5 mg SO₂/kg. In total there were 83 samples checked for sulfite on the label, while only 21 samples were analyzed. If all samples checked on the label for sulfite would be considered (n=83) instead of only the analyzed ones, and if it is assumed there are no frauds on sulfite usage, only 6 of 83 products were found to have a sulfite

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474 concentration above LOQ. For the evaluation of the sulfite intake from potato products it was 475 decided to use the average concentration of the samples having a concentration above LOQ. 476 This might result in an overestimation of the intake since only 7 % of selected and analyzed 477 potato products had a SO₂ concentration above the LOQ. However, some consumers might by 478 loyal to these particular kinds of brands. In addition this actual level used (11.6 mg/kg) is still 479 far below the maximum permitted levels (50-100 mg/kg SO₂).

From the Belgian Federal Agency for the Safety of the Food Chain (FASFC), sulfite analysis
results for different kinds of foods sampled between 2003 and 2006 were obtained (Table 1).
These results show that average actual levels found are often much lower than maximum
levels.

After considering the data from different years, it was decided to take the average results from the last year only or from different previous years. For each food group, the average concentration of the samples having a concentration above LOQ (5 mg/l) was used, in order to account for consumers being loyal to certain brands of products. There was a drastic decrease (about factor 10) in SO₂ concentration in dried tomatoes from 2004 to 2005 due to the fact that in 2004 there were found levels above the maximum while no levels above the maximum were found in 2005; therefore only the 2 last year's results were taken into account (2005 and 2006). The available number of data for each kind of dried fruits, except dried apricots, is low; therefore it was decided to use the average concentration of sulfite in dried fruits (excluding dried apricots) (data from 2006) for dried fruits other than apricots. The concentration assigned to dried apricots was the average of the 2005 and 2006 measurements in dried apricots. For wines, the average concentration found by the FASFC in white and red wines for the years 2004 and 2006 was assigned.

² 498

Measurements made in France (Mareschi et al., 1992) during the period 1989-1992, showed that the highest average SO₂ concentrations were found in salted fish fillets and dried fruits (more than 625 mg SO₂/kg), followed by mustard (225 mg SO₂/kg) and peeled potatoes (100 mg SO₂/kg). Wines resulted in an average concentration of 75 mg SO₂/l (no differences where made between red and white wines). Other measurements made during the same period in the United States (Daniels et al. 1992) showed higher concentrations in dried fruits, mashed potatoes and wines. More recent (2000-2005) studies made in Italy (Leclercq et al., 2000) and

Australia (Food Standards Australia New Zealand, 2005), showed that the sulfite level content
in food tends to diminish in some kinds of foods like cider, beer, died fruits and wines.

The results of the partial Tier 3 based on the analyzed products and on the maximum levels for the other food groups, are given in Table 3. The mean intake of sulfites in the Belgian adult population was estimated to be 0.19 mg/kg bw/day, corresponding to 27 % of the ADI. At the 97.5th percentile of the population the usual intake was 0.61 mg SO₂/kg bw/day (87 % of the ADI). It was found that for the average consumer wine contributed up to 14 % of the ADI). The sugar group still had a relative significant contribution to the intake, but actual concentrations were not available. It is known, however, that food producers try to avoid the need for allergen labelling of sulfite, by reducing the levels below 10 mg/kg.

Some groups of the population could be more exposed than others to sulfite intake, in particular wine consumers. The probability of approaching the ADI by wine consumers will depend on the particular choice of wine (sulfite concentration) and the consumption level. This is hard to estimate. An exceeding of the ADI can not be excluded for a limited percentage of the population of wine consumers, who might consume wine with a higher average concentration than the average concentration used here in the tier 3 calculations, as can be shown from the tier 2 estimates (where the consumption of wine alone can reach the ADI).

39 526

 The fact that some food groups were not taken into account, could have led to a certain underestimation of the intake. However, the reduction that takes place during further preparation of foods was not taken in to account but is a factor of overestimation of the intake of sulfites. Further, the fact that no illegal uses of sulfites in certain food groups were taken into account, could also have led to an underestimation. In 2005 1002 tests with malachite green were performed in chopped meat by the FASCA of which 54 were positive. 43 of these were confirmed in the laboratory. The average sulfite content in the samples exceeding the LOQ was on average 474.1 +/-487.1 mg/kg and 1782.6 mg/kg at the 97.5 percentile.

55 535

In 1999, JECFA reviewed the intake estimation of sulfite from different countries (JECFA,
1999): Aus-NZ, China, Finland, France, India, Japan, Spain, United Kingdom and United
States. The estimated range of intake of sulfite (expressed as sulphur dioxide) was between 10
and 1400 % of ADI. In should be noted that data modelling differed between countries. The

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540 different used models are the budget method, the poundage data method, the FBS/HES/sales 541 data method, model diets method and individual dietary records method. A summary of the 542 mean intake results *per capita* for those countries is given in Table 4. All of these results are 543 based on a Tier 2 like approach. Comparatively to those results, in the present study lower 544 intakes were found, except when compared to Finland, India and USA.

A more recent study in France (1998) performed a Tier 2 intake estimation (Verger et al. 1998). The average sulfite intake was estimated to be 1.34 mg SO₂/kg bw/day (191 % of the ADI) and the intake at the 95th percentile of the population to be 3.13 mg SO₂/kg bw/day (447 % of the ADI). All of these estimates exceeded the ADI of 0.7 mg/kg bw/day and were comparable to the earlier calculated data. The estimation of the intake of sulfite in France (Verger et al. 1998) on basis of actual concentrations for the global population was 20 mg SO₂/day per *capita* in 1992 (48 % of the ADI). The authors concluded that the major contributors to the intake of sulfite were alcoholic beverages (wine, cider and beer). They performed an intake calculation for alcoholic beverage consumers only and for non alcoholic beverage consumers. They found an intake of sulfite respectively of 31.5 mg SO₂/day (75.0 % of the ADI) and 1.96 mg SO₂/day (4.67 % of the ADI).

In Italy during 2000, exposure to sulfite was estimated for adults and children (Leclercq et al.,
2000). A first Tier 2 calculation resulted in an exposure of 0.82 mg SO₂/kg bw/day or 117 %
of the ADI, of which 32 mg was due to wine consumption (65 % of total intake), 8 mg from
dried fruits (16 %) and 2 mg from fish (4 %).

Since the ADI is exceeded by the Tier 2 in Italy, calculation with actual data was performed by the same authors. Leclercq et al. (2000) made 2 different modelled diets rich in sulfited foods in order to calculate estimation of the intake on bases of actual concentrations in food. The models differed for children and adults, where it was assumed that the body weight is 30 kg for a child and 60 kg for an adult. The ADI was not exceeded using the example diets proposed for calculation, although nearly (more then 95 % of the ADI) for adults in the worst case scenario. Real food consumption data would have increased the amount of information and improved the intake estimates.

572 A study conducted in Slovakia (Sinko & Janekova, 2006) demonstrated that the intake of 573 sulfite in a group of children aged between 7 and 10 years old (lowest body weight 21 kg; average weight 26 kg) was between 27 and 93 % of the ADI for a child of 21 kg. The study
was based on the actual food consumption during 4 months and maximum allowed sulfite
content in food.

A recent survey in Australia (Food Standards Australia New Zealand, 2005) showed an average intake of sulfite in the population of 0.2 mg SO₂/kg bw/day (lower bound estimation; concentrations not detected set to zero). With the upper bound estimate, the intake obtained was the same (30 % of the ADI). The same calculation for the 95th percentile of the population showed an intake estimate of 0.9 mg SO₂/kg bw/day (lower bound or upper bound), that corresponded to 130 % of the ADI. It was shown that in Tier 2 approach the intake estimated for high consumers was 1400 % of the ADI (JECFA) while with actual data, the intake for the high consumers became 130 %, which is more than 10-fold difference (yet still above the ADI).

In this study, actual typical levels of sulfites were much lower than the maximum levels set in the legislation for several food groups. This resulted in average intake estimates going from 0.34 mg/kg bw/day in Tier 2 to 0.19 mg/kg bw/day in the partial Tier 3. Wine showed a decrease in the sulfite intake estimate from 24 % (Tier 2) to 14 % (partial Tier 3) of the ADI for the mean consumer and from 116% (Tier 2) to 69% (partial Tier 3) for the 97.5 percentile. However, the choice of wine of a consumer might be different than average.

Conclusion

596 In conclusion, it was shown that the intake of sulfites in the Belgian adult population is likely 597 to be below the ADI with the possible exception for some high consumers of wines with high 598 sulfite levels. As a consequence, the intake of sulfites by wine consumers remains a point of 599 attention. Further it was shown that for several food groups actual typical levels of sulfites are 600 much lower than the maximum levels set in the legislation.

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54 602

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 Table 1. Average sulfite concentrations * in different foods (data from the control program of the Federal Agency for Safety of the Food Chain, Belgium)

Matrix	Year	Number of samples	Number of samples exceeding the LOQ	Average sulphite concentration* (mg/kg) ¹	Standard deviation (mg/kg)	Minimum value (mg/kg)	Maximum value (mg/kg)	Median (mg/kg)	Percentile 95 (mg/kg)	Percentile 97.5 (mg/kg)
	2005 200		0	7.2	2.7	2.0	12.0	7	10 (10.0
Dried tomatoes	2005-2006	5 65 5 40	9	7.3	3.7	2.0	13.0	1	12.6	12.8
All dried fruits (defined or undefined; Apricots inclu	2000 2000	5 40	27	384	520	9.0	917	207	870	905
All dried fruits (defined or undefined; Apricots exclu	ded) 2006	o 25	14	179	238	9.0	781			
Dried apricots	2005-2006	5 181	176	533	308	0.0	1448	478	1083	1182
Wines (Red, White or undefined)	2004-2006	5 83	82	114	45.4	30.0	240	113	180	184
of which Red	2004 & 2006	5 28	28	78.6	33.2	30.0	160	74.5	142	149
White	2004 & 2006	5 39	39	139	28.1	91.0	198	136	180	181
Raw Shrimps	2006	5 10	1	10.0	/	10.0	10.0	10	10.0	10.0
Mustard	2006	5 4	1	18.0	/	18.0	18.0	18	18.0	18.0
Mushrooms	2006	5 5	2	11.5	3.5	9.0	14.0	11.5	13.8	13.9
Jams	2006	5 10	10	16.0	11.6	1.0	33.0	14	32.6	32.8
Potato products	2006	5 16	9	3.8	3.4	0.3	10.0	3	9.6	9.8
Beers	2006	59	3	1.3	0.6	1.0	2.0	1	1.9	2.0

* Including only samples exceeding the LOQ of 5 mg/L to calculate the average

Table 2

Usual intake of sulfites (mg/kg bw/day); results from the Tier 2 exposure assessment (national food consumption survey, 2004)

	Sulphite (mg/kg bo	% of ADI at average intake							
	Mean (% of total)	SD	P25	P50	P75	P95	P97.5	P99	_
Wines	0.17 (53%)	0.240	0.000	0.050	0.270	0.660	0.810	1.020	24
Sugars and glucose syrup based products	0.042 (13%)	0.063	0.000	0.014	0.064	0.168	0.216	0.283	6.0
Beers	0.033 (10%)	0.060	0.000	0.000	0.045	0.148	0.204	0.286	4.7
Processed fruits*	0.025 (7.6%)	0.061	0.000	0.063	0.022	0.107	0.166	0.283	3.5
Processed potatoes	0.024 (7.3%)	0.022	0.007	0.018	0.034	0.068	0.082	0.101	3.4
Processed vegetables**	0.014 (4.4%)	0.013	0.005	0.010	0.019	0.039	0.049	0.062	2.0
Shellfish	0.013 (4.2%)	0.019	0.000	0.006	0.002	0.052	0.065	0.083	1.9
Gelatine and meat substitutes	Error reporting in C	SIDE (no 1	esult pro	duced)					
Condiments and dessert sauces	0.0011 (0.35%)	0.0018	0.000	0.001	0.002	0.004	0.006	0.008	0.16
Cereal based products (starches and biscuits)	0.00045 (0.14%)	0.00087	0.000	0.000	0.001	0.000	0.000	0.004	0.064
Total intake	0.34	0.28	0.14	0.26	0.45	0.89	1.1	1.4	49
ADI acceptable daily intake									
SD standard deviation									

bw body weight

The usual dietary intakes were weighted and adjusted for the age and sex distribution of the Belgian population 2004 and adjusted for interview day and season.

Total number of consumption days is 6166; total number of consumers is 3083

The acceptable daily intake (ADI) for sulphite is 0.7 mg/kg bw/day

* including dried fruits (including dried apricots), candied fruits, jams, jellies and marmalades, lemon and lime juice, fruit fillings, fruits in vinegar, brine, oil.

** including dried tomatoes, mushrooms, vegetables in vinegar, brine and oil, and white vegetables

Table 3 Usual intake of sulfites (mg/kg bw/day); results from the partial Tier 3 exposure assessment (national food consumption survey, 2004)

	Sulphite	(mg/kg bodyw	eight/day)							% of
	Mean	(% of total)	SD	P25	P50	P75	P95	P97,5	P99	ADI at average intake
Wines	0.097	(51.1%)	0.141	0.000	0.028	0.151	0.383	0.483	0.620	13.9
Sugars and glucose syrup based products ^o	0.042	(22.1%)	0.063	0.000	0.014	0.064	0.168	0.216	0.283	6.0
Beers	0.008	(4.2%)	0.015	0.000	0.000	0.011	0.037	0.051	0.071	1.1
Processed fruits*00	0.012	(6.3%)	0.035	0.000	0.002	0.008	0.058	0.092	0.161	1.7
Processed potatoes	0.0024	(1.3%)	0.0020	0.00092	0.0021	0.0035	0.0062	0.0072	0.0086	0.3'
Processed vegetables****	0.014	(7.4%)	0.013	0.005	0.010	0.019	0.039	0.049	0.062	2.0
Shellfish ^{°°}	0.0097	(5.1%)	0.0148	0.000	0.0035	0.0138	0.0392	0.0513	0.0685	1.4
Gelatine and meat substitutes°	Error rep	orting in CSID	E (no result	produced)						
Condiments and dessert sauces ^{oo}	0.00017	(0.09%)	0.00019	0.000	0.000121	0.000275	0.000547	0.000658	0.000805	0.02
Cereal based products (starches and biscuits)°	0.00045	(0.24%)	0.00087	0.000	0.000	0.001	0.000	0.000	0.004	0.06
Total intake	0.190		0.159	0.074	0.146	0.260	0.505	0.611	0.753	27.1

ADI acceptable daily intake

SD standard deviation

bw body weight

The usual dietary intakes were weighted and adjusted for the age and sex distribution of the Belgian population 2004 and adjusted for interview day and season.

Total number of consumption days is 6166; total number of consumers is 3083

The acceptable daily intake (ADI) for sulphite is 0.7 mg/kg bw/day

* including dried fruits (including dried apricots), candied fruits, jams, jellies and marmalades, lemon and lime juice, fruit fillings, fruits in vinegar, brine, oil.

** including dried tomatoes, mushrooms, vegetables in vinegar, brine and oil, and white vegetables

° Maximum concentrations used instead of analyzed concentrations

^{oo} Combination of maximum concentrations and analyzed concentrations used depending on the availability of analyzed concentrations for the food items in the considered food groups

Country	Date	Survey	Intake of sulphites estimate (mg/kg bw per day)	% of ADI
Aus-NZ	1983	National, 24-h recall; adults, 25-64 years; sample, 6254	Aus-NZ permissions: 10	1400
China	1992	National household survey, 24-h recall; 30 provinces; sample, 91 818	Average consumer: 0.63 High consumer: 3.2	90 460
Finland	1994	poundage data	0.067	10
France	1993-94	Sales data	 0.70 (mean, corrected) 1.0 (mean, uncorrected) 2.2 (90th percentile, uncorrected) 3.2 (95th percentile, uncorrected) 	100 140 310 460
India	1995-96	Food balance sheet	0.35	50
Japan	1994	National nutrition intake survey	Average consumer: 0.033	
Spain	1993	Household survey	0.88	130
United Kingdom	1986-87	National; 7-day weighed record; adults, 16-64 years National; 7-day weighed record; children, 1.5-4.5 years	UK permissions/adult: 17 UK permissions/child: 77	2400 11000
United States	1982-88	14-day MRCA food frequency data (1982-87) combined with portion sizes from USDA/NFCS (1987-88); population ≥ 2 years	US permissions/mean: 0.30 US permissions/90 th : 0.73	40 100