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Estimate of intake of sulfites in the Belgian adult population

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3 36
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5 37
6 38 **Abstract**
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9 40 An exposure assessment was performed to estimate the usual daily intake of sulfites in the
10 41 Belgian adult population. Food consumption data were retrieved from the national food
11 42 consumption survey. In a first step, individual food consumption data were multiplied with
12 43 the maximum permitted use levels for sulfites, expressed as sulphur dioxide, per food group
13 44 (Tier 2). In a second step, on the basis of a literature review of the occurrence of sulfites in
14 45 different foods, the results of the Tier 2 exposure assessment and available occurrence data
15 46 from the control programme of the competent authority, a refined list of foods was drafted for
16 47 the quantification of sulphite. Quantification of sulphite was performed by a high
17 48 performance ion chromatography method with eluent conductivity detector in beers and
18 49 potato products. Individual food consumption data were then multiplied with the actual
19 50 average concentrations of sulfite per food group, or the maximum permitted levels in case
20 51 actual levels were not available (partial Tier 3). Usual intakes were calculated using the
21 52 Nusser method. The mean intake of sulfites was 0.34 mg/kg bw/day (Tier 2), corresponding
22 53 with 49 % of the acceptable daily intake (ADI) and 0.19 mg/kg bw/day, corresponding with
23 54 27 % of the ADI (partial Tier 3). The food group contributing most to the intake of sulfites
24 55 was the group of wines. The results showed that the intake of sulfites is likely to be below the
25 56 ADI in Belgium. However, there are indications that high consumers of wine have an intake
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77 Introduction

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

86
87 Sulfite has raised concerns because of its toxicity for the lungs and its ability to cause allergic
88 reactions, especially in asthmatic persons (Valley et al., 2009). It was estimated that an
89 exposure of 10 to 50 ppm during 5 to 15 min may have a measurable effect on humans. In the
90 UK, 2 ppm (8 hours time weighted average TWA) or 5 ppm (10 min TWA) were set as
91 maximum occupational exposure values (European Cooperation on School Education, 2004).
92 In Belgium, the maximum allowed concentration of sulfite in the air is 350 µg/m³ during
93 maximum 1 hr or 125 µg/m³ during maximum 24 hr (European Community 1999a). In a
94 review of the Health Council of the Netherlands sulfite was found to be responsible of chronic
95 urticaria, skin allergy and bronchial asthma in some humans. The NOAEL in rats was set at
96 72 mg SO₂/kg bw/day (Health Council of the Netherlands, 2005).

97
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).

103
104 Sulfite can be intentionally added to food for its antioxidant or antimicrobial properties.
105 Sulfite inhibits the enzymatic activity of polyphenol oxidase, which is responsible for
106 browning in fruits, vegetables and seafood products. It also prevents oxidation of food due to
107 contact with the air and the browning effect occurring from the Maillard-type reactions. The
108 aim of adding sulfite to wine is to prevent microbial growth and oxidation of volatile odorous
109 compounds which may develop during the fermentation (Ough, 1986).

110

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3 111 Sulfite is allowed as a preservative (E220-E228) in a list of different foods (excluding all
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5 112 others) under the condition of respecting the maximum use levels, as specified by the
6
7 113 European Directive 95/2/EC as amended (European Community 1995) and for wine in annex
8
9 114 V of the Council regulation (EC) No. 1493/1999 (European Community 1999b).

10
11 115 The concentration in different foods is expressed as sulphur dioxide (SO₂) in mg/kg or mg/l,
12
13 116 depending on the nature of the food and relates to the total quantity from all sources. The
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15 117 legislation mentions that an SO₂ content of not more than 10 mg/kg or 10 mg/l is not
16
17 118 considered to be present. Directive 2000/13/EC on labelling of pre-packaged food requires
18
19 119 labelling of products containing sulfite, if the SO₂ concentration is higher than 10 mg/kg or 10
20
21 120 mg/l, for the benefit of people with a food intolerance for sulfite (European Community
22
23 121 2000).

24 122

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26 123 Sulfite can be present in foods as free sulfite or react with a variety of organic compounds in
27
28 124 foods to form reversibly or irreversibly bound products. In our digestive tract, the free and
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30 125 reversibly bound forms can be liberated (Wedizicha, 1992). García-Alonso et al., (2001)
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32 126 reported that the amount of added sulfite in fresh sausage was reduced by 26 % when S—
33
34 127 sulphonate bounds (irreversibly bound) were formed and by 23 % when C—O—SO₂⁻ bounds
35
36 128 (reversibly bound) were formed. Those percentages of loss may vary sharply from 10 to 49 %
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38 129 depending on the conditions when adding the sulfite to the meat (time of mixing, presence of
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40 130 oxygen, degree of mincing and fat composition of the meat).

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43 132 Cooking was shown to reduce the level of sulfite in beef and chicken burgers (Pena-Egido et
44
45 133 al; 2005; Garcia-Alonso et al., 2001) the distribution of the 3 different forms of sulfite (free,
46
47 134 reversibly bound and S-sulphonate) remained approximately the same after as before the
48
49 135 cooking process. The losses reported for meat burgers were 31 % for the S-sulphonates, 29 %
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51 136 for the free sulfite and 28% for the total sulfite. This study also showed that the frying process
52
53 137 reduced the amount of total sulfite on average by 41 % (± 13%). A study in shrimps
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55 138 (Armentia-Alvarez et al., 1994) showed that during cooking the loss of free and reversibly
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57 139 bound sulfite varied between 45 and 55 %. The level of total sulfite was about 5 times higher
58
59 140 in the non edible parts compared to the edible parts. The authors concluded that the boiling
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141 step did not change the distribution of free and total sulfite in shrimps, but a reduction of
142 about 40 % was observed for the total sulfite in the edible parts. From the boiled water it was

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3 143 shown that the loss of SO₂ in shrimps during the boiling process was not totally recovered in
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5 144 the water due to oxidation of sulfite and reaction of sulfite with food components.
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9 146 Another study on shrimps (Armentia-Alvarez et al., 1997) showed that during bad freezing
10 147 storage (> -18°C), the free sulfite combined with the disulfide bounds of proteins denatured
11 148 by the freezing process. The longer the storage time, the higher the amount of combined
12 149 sulfite produced. Reduction of sulfite concentration after a storage period of 1 month was
13
14 150 reported to be between 25 and 50 % for fish products, potatoes and dried fruits (Leclercq et
15
16 151 al., 2000)

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18
19 152 Finne et al., (1986) studied the residual sulfite content in shrimps after different days of
20 153 conservation in ice. They demonstrated that, starting with a concentration of about 80 ppm,
21 154 the residual content was reduced to less than 10 ppm after 6 days of storage on ice.
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26 156 Directive 95/2/EC (European Community, 1995) requires that Member States of the EU
27 157 monitor the intake and use of food additives in order to allow evaluation of intake trends.
28 158 Measures can be taken to amend the legislation if intake concerns are identified in one or
29 159 more Member States. The Report from the European Commission on Dietary Food Additive
30 160 Intake in the European Union (European Commission, 2001) describes a harmonized, tiered
31 161 method to estimate additive intakes. In the first step (Tier 1), theoretical food-consumption
32 162 data (Hansen, 1979) are combined with the maximum permitted usage levels for the additive.
33 163 This conservative estimate was performed at European level. For the additives exceeding the
34 164 ADI in Tier 1, in a further step (Tier 2), national food consumption data are combined with
35 165 maximum permitted usage levels. Such intake estimate will still overestimate the real intake,
36 166 but can exclude additives, which do not pose a major risk from the most difficult estimate in
37 167 Tier 3: a combination of actual national food consumption data with actual levels of the
38 168 additive. The report from the Scientific Committee on Food (SCF) about Dietary Food
39 169 Additive Intake in the European Union (1998) showed that, based on Tier 1, there were 10
40 170 additives in the European Union for which a more refined intake estimate was needed.
41 171 Among those additives also sulfite was mentioned (Scientific Committee on Food). A Tier 2
42 172 exposure assessment in 7 countries (Belgium not included) showed that the range of estimated
43 173 intakes was 20-266% of the ADI for adults (European Commission).
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59 174 The objective of this study was to perform a refined exposure assessment for sulfite intake in
60 175 the Belgian adult population, using the data from the national food consumption survey of

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3 176 2004. Another objective was to identify the food groups contributing most to the total intake
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5 177 of sulfites in Belgium.
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8 178
9 **Materials and Methods**
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11 180
12 **Study design**

13 182 A semi-probabilistic approach was used to perform the exposure assessment. For the Tier 2
14 183 exposure assessment, maximum levels of sulphur dioxide in different foods were linked to the
15 184 food consumption data of these specific foods, derived from the Belgian food consumption
16 185 survey (2004). For some foods in the EU legislation, it is unlikely that they are consumed in
17 186 significant quantities or they were too difficult to link with the data from the food
18 187 consumption survey: burger meat with a vegetable or cereal content of minimum 4 %;
19 188 breakfast sausages; *Longaniza fresca* and *Butifarra fresca*; dried salted *Gadidae* fish; *sago*;
20 189 pearl barley; dried ginger; pulp of horseradish; pulp of onions, shallots and garlic; yellow
21 190 paprika in brine; flavourings on the basis of lemon fruit juice; concentrated grape juice for the
22 191 making of house wine; *mostrarda di frutti*; gelatinized fruit extracts or fluid pectin; glass
23 192 preserves of cut lemon; white syrup and molasses; orange-, grapefruit-, apple- and pineapple
24 193 juice for unpackaged sell in catering enterprises; concentrates on the basis of fruit juice with
25 194 at least 2.5 % pearl barley; *capilhé groselha*; unfermented grape juice for religious use; wine
26 195 without alcohol; fermented vinegar; marinated nuts; vacuum packaged sugar maize and
27 196 distilled alcoholic beverages with whole pears. Consequently these products were not taken
28 197 into account in the intake assessment. This could lead to an underestimation of the intake, but
29 198 this was supposed to be limited. As stated in the introduction, cooking and freezing can
30 199 decrease the sulfite concentration of foods. This was not taken into account either due to the
31 200 lack of reliable processing factors. Consequently this could lead to an overestimation of the
32 201 intake.
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50 202
51 203 In order to perform a more refined estimate of the sulfite intake (partial Tier 3 exposure
52 204 assessment), on the basis of literature, available concentration values for products analyzed by
53 205 The Belgian Federal Agency for Safety of the Food Chain (FASFC) and the results from the
54 206 Tier 2 exposure assessment, it was decided to analyze concentrations of sulphur dioxide in
55 207 beers and potato products. In these samples concentrations were determined using a high
56 208 performance ion chromatography method with eluent conductivity detector, optimized and
57 209 validated for this purpose. In the partial Tier 3 exposure assessment, analyzed actual levels of

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3 210 sulphur dioxide in different foods were linked to the food consumption data of these specific
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5 211 foods, derived from the Belgian food consumption survey (2004). For foods for which no
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7 212 analyzed levels were available, maximum concentrations were used, as in the Tier 2 exposure
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9 213 assessment. No illegal uses of sulfites in food groups in which no use of sulfites is authorised,
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11 214 were taken into account in this study.

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14 216 Although the regulation specifies maximum levels for specific groups, in this paper, the
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16 217 results of the analyses were grouped to ensure a clear data presentation. The grouping was
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18 218 performed according to the EU directive 95/2/EC (European Community, 1995) as follows:
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20 219 beers; wines; sugars and glucose-based products; condiments and dessert sauces; gelatine and
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22 220 meat substitutes; shellfish; cereal-based products (starches and biscuits); processed potatoes,
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24 221 processed fruits (including dried fruits, candied fruits, jams, jellies and marmalades, lemon
25
26 222 and lime juice, fruit fillings and fruits in vinegar, brine, oil) and processed vegetables
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28 223 (including dried tomatoes, mushrooms, vegetables in vinegar, brine and oil and white
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30 224 vegetables).

31 225
32 226 **Food consumption data**
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35 228 Consumption data from the national food consumption survey 2004 were used to perform the
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37 229 exposure assessment. Aims, design and methods of this survey are described elsewhere (De
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39 230 Vriese et al. 2005). The target population comprised all Belgian inhabitants of 15 years or
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41 231 older. The sample included 3245 participants randomly selected from the National Register,
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43 232 using a multi-stage stratified procedure.

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46 234 Information on dietary intake was collected by two non-consecutive 24-hour recalls in
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48 235 combination with a food frequency questionnaire. During the 24-hour recall interviews the
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50 236 respondent reported the quantity of all foods and beverages consumed during the preceding
51
52 237 day. The 24-hour recall was carried out using EPIC-SOFT software (Slimani & Valsta, 2002).
53
54 238 This program allows obtaining very detailed information about the foods consumed and the
55
56 239 recipes used in a standardized way.

57 240
58 241 3083 participants, of which 1537 women and 1546 men, completed two 24-hour recalls.
59
60 242 Participants were categorized into four age groups: 15-18 years (n=760), 19-59 years (n=830),
243 60-74 years (n=789) and 75 years or older (n=704).

244

245 Sulphur dioxide concentration of selected foods

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

250

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

262

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

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

271

272 Collection of samples was performed in 2007 in 5 major supermarket chains with a good
273 market share, which are visited by approximately 77.5 % of the Belgian population in search
274 for their food provisioning (CRIOC - OIVO). In total for both categories 67 samples (21 for
275 potato products and 46 for beers) were collected and analysed.

276

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3 277 The only existing ISO method including sulfite analysis is the method ISO 10304-3:1997. The
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5 278 HPIC/conductivity detector with eluent conductivity suppressor was used in this study to
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7 279 perform the analyses. Ion chromatography is an appropriate technique for the separation of
8
9 280 sulfite from other anions like phosphate and sulphate, and it has also been recognised for the
10
11 281 quantification of sulfite (Armentia-Alvarez A. et al., 1997; Kim 1989; Kim et al. 1987; Kim et al.
12
13 282 2000; Paino-Campa et al. 1991; Pena-Egido et al., 2005; Pizzoferrato et al. 1990) Pulsed
14
15 283 amperometric detection necessitates very specific material and is not so easy to use because of
16
17 284 possible fouling of the electrode by matrix compounds (McFeeters and Barish, 2003). The
18
19 285 most used detection method in ion chromatography is eluent suppressed conductivity. With
20
21 286 the carbonate/bicarbonate eluent, the produced species in the suppressor is H_2CO_3 (which
22
23 287 decomposes in CO_2 and H_2O).
24

288

289 ***Material***

290 For sample preparation, a Robocoup mixer 3000, a T25 Ultra-Turrax (IKA) and a centrifuge
291 Beckman Avanti J-25 operated at 7500rpm were used. All extracts were filtered through
292 0.2 μm PVDF syringe filters. The analytical system was composed of a Waters 717+ auto
293 injector, an HP 1050 pump, an Alltech Suppressor model 640 and an Alltech conductivity
294 detector model 650. The acquisition of the signal and data treatment (calibration curve and
295 extract concentration) were made by a computer equipped with an analogue/digital converter
296 and ChromQuest software version 4.1. The separation column was composed of a Dionex
297 RFIC analytical column type IonPac AS14A-5 μm 3x150mm (particle size of 5 μm) and
298 precolumn RFIC IonPac AG14A-5 μm 3x30mm.

299 ***Reagents and solutions***

300

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

305 The aqueous solutions were prepared with a MilliQ Reagent Grade System's water with
306 conductivity lower than $18.2 \text{ M}\Omega\text{cm}^{-1}$ (from Millipore coupled after a Millipore Elix 100
307 deionised water production system). Stock solutions NaHCO_3 0.3 M, Na_2CO_3 0.2M and D-
308 Mannitol 0.5 M were prepared. Those solutions were kept in the fridge. The eluent used for
309 the anion separation was the buffer $\text{NaHCO}_3/\text{Na}_2\text{CO}_3$ 3mM/2mM. The sulfite extraction

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2
3 310 buffer was a mixture of NaHCO₃/Na₂CO₃/D-Mannitol 9mM/16mM/10mM. This resulted in a
4
5 311 final extraction pH between 8 and 10, as proposed in literature for free sulfite and reversible
6
7 312 bound sulfite extraction (Armentia-Alvarez A. et al. 1993; Wedzicha, 1992). The 1000 mg/l
8
9 313 sulphur dioxide standard stock solution was prepared by weighting 0.1968g anhydrous sulfite
10
11 314 and making a 100.0 ml solution with extraction buffer.

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13 315

15 316 *Sample collection, storage and pre-treatment*

17 317 After collection of the samples, they were identified by a unique identification number and
18
19 318 registered in a Microsoft Excel sheet. Following characteristics were added: brand name,
20
21 319 product description, lot number, temperature of conservation in the store and any relevant
22
23 320 information for the project (presence or absence of sulfite mentioned on the label or usage of
24
25 321 ingredients containing sulfite). The liquid samples where stored in a fridge and the solid
26
27 322 samples where stored in the freezer (-18°C or less) until pre-treatment or analysis.

28 323

30 324 The pre-treatment of the solid samples consisted of grinding at least 200 g collected sample
31
32 325 with a kitchen robot (Robot Coupe model R 301 Ultra) in order to get piece cuts less than 3~5
33
34 326 mm. Afterwards the prepared samples were stored deep frozen. All analyses where conducted
35
36 327 within 3 weeks after collection of the samples.

38 328 *Sample preparation*

40 329 For the preparation of the liquid samples 100 µl of D-Mannitol 0.5 M was added to about 100
41
42 330 ml of the sample before degassing in an ultrasound bath for 5 minutes; 5.0 ml of sample was
43
44 331 then diluted in the extraction buffer by a factor of 5 (final volume = 25.0 ml) and the extract
45
46 332 was filtered through a PVDF syringe filter with 0.2 µm pore size before injection on the HPIC
47
48 333 system. For the preparation of the solid samples, after a short period of unfreezing, about 5 g
49
50 334 of sample (weighted with at least 3 digits after the point) was weighted in a P.E. tube of 50 ml
51
52 335 to be centrifuged; 25.0 ml of extraction buffer (measured with a graduate cylinder of 25 ml)
53
54 336 was added to the sample; the sample was then homogenated in an Ultra-Turrax for a
55
56 337 minimum of time, so that the mixture looked like creamy (10 to 15 seconds suffice).

57
58 338 The tube was closed and set in a centrifuge at 7500 rpm for 10 minutes; the surnatant extract
59
60 339 was filtered through a PVDF syringe filter with 0.2 µm pore size before injection on the HPIC
340 system.

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5 342 ***Calibration curve - linearity***
6
7 343 Due to the range of maximum permitted levels in the different kinds of foods it was chosen to
8
9 344 use a calibration curve ranging from 0.1 to 200 mg SO₂/l.

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11 345 By diluting 1000 mg SO₂/l extraction buffer a series of eight dilutions was prepared (0.1, 0.5,
12
13 346 1. 5, 10, 50, 100, and 200 mg SO₂/l). Blank was used as 0 mg SO₂/l. The best fit was used in
14
15 347 order to maximize the coefficient of determination (> 98 %) and to minimise the residues at
16
17 348 each level.

18
19 349 ***Validation***

20
21 350 The method was validated for linearity, precision (repeatability, reproducibility), accuracy and
22
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 NaHCO₃/Na₂CO₃ 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
30
31 355 included). Different stabilization compounds (EDTA, D-Mannitol and formaldehyde) were
32
33 356 tested according to previous reports. D-Mannitol was chosen due to its best effect on the
34
35 357 sample extract. Some stability tests were made with a standard solution 10 mg SO₂/l. The
36
37 358 responses of a standard solution prepared in water and the same amount of standard solution
38
39 359 prepared in a pH~ 9.3 buffer with 10 mM D-mannitol were compared.

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41 360
42 361 The final sulfite extraction buffer chosen after pH test in mixture with the sample matrix was
43
44 362 NaHCO₃/Na₂CO₃/D-Mannitol at concentration levels of 9 mM/16 mM/10 mM respectively.
45
46 363 The pH of this buffer was between 9 and 10. The extracts prepared with beer resulted in a pH
47
48 364 of 9.3 and of 8.7 when prepared with frozen French fries. NaOH/formol was tested for the
49
50 365 extraction of sulfite from beer. But the chromatographic separation gave very high
51
52 366 background noise after a few injections. The material was not able to work with an organic
53
54 367 modifier like acetone in high proportion like described in Metrohm application note S-95.

55
56 368
57 369 To test linearity it was opted for the weighted quadratic regression of the heights in function
58
59 370 of the extract concentration. This type of regression resulted in a determination coefficient
60
371 greater than 0.99 and residues lower than 15 % for the lowest concentration point.

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3 373 The range for the calibration was 0.1 to 100 mg/l extract (The injection of the point 200 mg/l
4
5 374 was kept in order to be able to quantify higher amounts from a new regression). The range in
6
7 375 the extract corresponded to a range in the sample going from 0.5 to 500 mg/kg (or mg/l,
8
9 376 depending of the nature of sample) if the dilution factor was taken into account.

10
11 377
12 378 In order to test the repeatability and reproducibility a sample (table beer) was fortified at 4
13
14 379 levels of concentration corresponding to 0, 5, 10 and 15 mg/l of the sample (which
15
16 380 corresponds to 0, 1, 2 and 3mg/l in the extract). The test was conducted in triplicate for each
17
18 381 level and repeated for each level at 3 different days. The non fortified sample did not result in
19
20 382 a detectable response and the obtained result of reproducibility was lower than 12.6 %.

21 383
22
23 384 The apparent recovery was, on average, for the 3 levels of concentration, 73 ± 6 at 3
24
25 385 concentration levels. A lower recovery may be explained by a possible reaction of SO₂ to
26
27 386 some compounds present in the matrix and/or by oxidation after the dissolution of ambient
28
29 387 oxygen into the extracts. Therefore, it was decided to correct the results for the recovery of
30
31 388 the control standard included in the series of analysis.

32 389
33 390 In beer sample, the sulfite peak was poorly separated from matrix interferences. The
34
35 391 minimum detectable concentration in the extract was about 0.5 mg/l. This corresponds to a
36
37 392 quantification limit (LOQ) of 1 mg/l extract which corresponds to 5 mg/l (or mg/kg) in the
38
39 393 sample. This is also the lowest tested concentration.

40 394

41 395 **Exposure assessment**

42 396 Only respondents with two completed 24-hour recall interviews were included in the analyses
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44 397 (n= 3083; 1546 men and 1537 women).

45 398

46 399 The individual intake of sulphur dioxide from a certain food product was estimated using the
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48 400 following equation:

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53 401
$$y_i (mg / kgbw / day) = \frac{c \times x_i}{bw_i}$$

54
55
56 402 where y_i is the intake of sulphur dioxide by individual i from a particular food (in mg per kg
57
58 403 bodyweight per interview day), c is the concentration of sulphur dioxide in that food (mg per
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60 404 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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3 406 group or per day, individual daily intakes of sulphur dioxide from different foods were added
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5 407 up.

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7 408 As mentioned before, two approaches were used for the calculation of sulphur dioxide intake.
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9 409 In the Tier 2 approach, c_i represents the maximum allowed concentration in each food. The
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11 410 maximum permitted levels of sulphur dioxide used in different foods are listed in the
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13 411 European Directive 95/2/EC (European Community, 1995). In the Tier 3 approach, c_i
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15 412 represents the actual average concentration of sulphur dioxide that is observed in a particular
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17 413 food.

18 414
19 415 The usual intake distribution for sulphur dioxide was estimated with the Nusser method
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21 416 (Nusser et al. 1996) using the C-side software (Iowa State University, 1996). Several
22
23 417 statistical methods are available to estimate usual intake distributions with the correct mean,
24
25 418 variance and skewness. These statistical procedures adjust for within-person or day-to-day
26
27 419 variability. Of all different statistical procedures, the Nusser method (Nusser et al., 1996) is
28
29 420 highly recommended because it eliminates the intra-individual variance and additionally
30
31 421 transforms the data to obtain approximately normally distributed data. The method is suitable
32
33 422 to estimate usual intake distributions in a population both for normally and non-normally
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35 423 distributed foods and nutrients. The usual intake distribution was weighted and adjusted for
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37 424 the age and sex distribution of the Belgian population and adjusted for day of the week and
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39 425 season.

40 426

41 427 **Results and discussion**

42 428
43 429 As an adult inhales about 15 m³ of air per day, this means that in the worst case the amount of
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45 430 SO₂ intake from the environment is 215 µg/m³ x 15 m³= 3225 µg/ day = 3.2 mg/day. 215
46
47 431 µg/m³ is the highest concentration of SO₂ measured in Belgium in 2005. Since the average
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49 432 body weight of a Belgian adult is around 70 kg, the final SO₂ intake from the environment is
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51 433 estimated to be 0.045 mg/kg bw/day. From those results it was decided to neglect the
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53 434 environmental contribution to SO₂ intake in Belgium.

54 435

55
56 436 The usual sulfite intake of the Belgian adult population estimated by the Tier 2 approach was
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58 437 0.34 mg/kg bw/day (49 % of the ADI) at the mean level of consumption (Table 2). At the
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60 438 97.5th percentile of the population, the sulfite intake was estimated to be 1.1 mg SO₂/kg
439 bw/day (157 % of the ADI). It was observed that the ADI was exceeded beyond the 90th

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3 440 percentile of the population. At this percentile the contribution of wine consumption to the
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5 441 total intake was estimated to be 71 %. Including only wine consumers (55.1%), the ADI was
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7 442 reached at the 57.5th percentile of population (data not shown).
8
9 443

10 444 Wine contributed for more than 50 % to the total intake of sulfite. Sugar and glucose-based
11 445 products, beers, processed fruits and processed potatoes contributed each between 7-13 % to
12 446 the total intake, while shellfish and processed vegetables contributed each about 4 % to the
13 447 total intake. Condiments and cereal products contributed to less than 1 % to the total average
14 448 sulfite intake (Table 2).
15
16 449

17 450 Concentrations of sulfite in beer samples (n=46) were all under the detection limit (LOD) of 5
18 451 mg/l; these results were different from the concentrations found in other countries. For
19 452 example, in France (Mareschi et al. 1992) and Italy (Leclercq et al., 2000) the average sulfite
20 453 concentrations found in beer were respectively 7.5 and 15 mg SO₂/l. In these studies the
21 454 Monier-Williams method was used to perform the analyses. It is known that this method has
22 455 possible interferences with volatile compounds. The method used in the present study does
23 456 not have this disadvantage. With the limit of quantification of 5 mg SO₂/l it was not possible
24 457 to determine any residual SO₂ levels in the beers analyzed. Provided this and the results
25 458 reported by other countries obtained by more sensitive electrochemical methods of analysis
26 459 (Dvorak et al. 2006) it was decided not to perform an intake estimation of sulfite from beer
27 460 using a null concentration of sulfite in beers. It is known that sulfite could be produced during
28 461 the natural fermentation process in the bottle. Therefore half of the LOQ concentration was
29 462 used (medium bound approach) instead of the null concentration to perform the intake
30 463 estimation.
31
32 464

33 465 For potato products 6 samples out of 21 analyzed samples resulted in SO₂ levels above the
34 466 quantification limit (LOQ) of 5 mg/l and only one sample with no sulfite declared on the label
35 467 resulted in a concentration higher than the LOQ. This positive sample was a fresh pre-cut
36 468 product in which the concentration was 9.9 mg SO₂/kg. For the samples with a concentration
37 469 above LOQ, an average concentration value of 11.6 mg SO₂/kg was found and the results
38 470 were spread between 9.0 and 13.5 mg SO₂/kg. In total there were 83 samples checked for
39 471 sulfite on the label, while only 21 samples were analyzed. If all samples checked on the label
40 472 for sulfite would be considered (n=83) instead of only the analyzed ones, and if it is assumed
41 473 there are no frauds on sulfite usage, only 6 of 83 products were found to have a sulfite

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3 474 concentration above LOQ. For the evaluation of the sulfite intake from potato products it was
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5 475 decided to use the average concentration of the samples having a concentration above LOQ.
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7 476 This might result in an overestimation of the intake since only 7 % of selected and analyzed
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9 477 potato products had a SO₂ concentration above the LOQ. However, some consumers might be
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11 478 loyal to these particular kinds of brands. In addition this actual level used (11.6 mg/kg) is still
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13 479 far below the maximum permitted levels (50-100 mg/kg SO₂).
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16 481 From the Belgian Federal Agency for the Safety of the Food Chain (FASFC), sulfite analysis
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18 482 results for different kinds of foods sampled between 2003 and 2006 were obtained (Table 1).
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20 483 These results show that average actual levels found are often much lower than maximum
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22 484 levels.

23 485 After considering the data from different years, it was decided to take the average results from
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25 486 the last year only or from different previous years. For each food group, the average
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27 487 concentration of the samples having a concentration above LOQ (5 mg/l) was used, in order
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29 488 to account for consumers being loyal to certain brands of products. There was a drastic
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31 489 decrease (about factor 10) in SO₂ concentration in dried tomatoes from 2004 to 2005 due to
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33 490 the fact that in 2004 there were found levels above the maximum while no levels above the
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35 491 maximum were found in 2005; therefore only the 2 last year's results were taken into account
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37 492 (2005 and 2006). The available number of data for each kind of dried fruits, except dried
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39 493 apricots, is low; therefore it was decided to use the average concentration of sulfite in dried
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41 494 fruits (excluding dried apricots) (data from 2006) for dried fruits other than apricots. The
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43 495 concentration assigned to dried apricots was the average of the 2005 and 2006 measurements
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45 496 in dried apricots. For wines, the average concentration found by the FASFC in white and red
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47 497 wines for the years 2004 and 2006 was assigned.
48

49 499 Measurements made in France (Mareschi et al., 1992) during the period 1989-1992, showed
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51 500 that the highest average SO₂ concentrations were found in salted fish fillets and dried fruits
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53 501 (more than 625 mg SO₂/kg), followed by mustard (225 mg SO₂/kg) and peeled potatoes (100
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55 502 mg SO₂/kg). Wines resulted in an average concentration of 75 mg SO₂/l (no differences were
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57 503 made between red and white wines). Other measurements made during the same period in the
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59 504 United States (Daniels et al. 1992) showed higher concentrations in dried fruits, mashed
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505 potatoes and wines. More recent (2000-2005) studies made in Italy (Leclercq et al., 2000) and

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3 506 Australia (Food Standards Australia New Zealand, 2005), showed that the sulfite level content
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5 507 in food tends to diminish in some kinds of foods like cider, beer, dried fruits and wines.
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9 509 The results of the partial Tier 3 based on the analyzed products and on the maximum levels
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11 510 for the other food groups, are given in Table 3. The mean intake of sulfites in the Belgian
12
13 511 adult population was estimated to be 0.19 mg/kg bw/day, corresponding to 27 % of the ADI.
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15 512 At the 97.5th percentile of the population the usual intake was 0.61 mg SO₂/kg bw/day (87 %
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17 513 of the ADI). It was found that for the average consumer wine contributed up to 14 % of the
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19 514 ADI). The sugar group still had a relative significant contribution to the intake, but actual
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21 515 concentrations were not available. It is known, however, that food producers try to avoid the
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23 516 need for allergen labelling of sulfite, by reducing the levels below 10 mg/kg.
24

25 517
26 518 Some groups of the population could be more exposed than others to sulfite intake, in
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28 519 particular wine consumers. The probability of approaching the ADI by wine consumers will
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30 520 depend on the particular choice of wine (sulfite concentration) and the consumption level.
31
32 521 This is hard to estimate. An exceeding of the ADI can not be excluded for a limited
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34 522 percentage of the population of wine consumers, who might consume wine with a higher
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36 523 average concentration than the average concentration used here in the tier 3 calculations, as
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38 524 can be shown from the tier 2 estimates (where the consumption of wine alone can reach the
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40 525 ADI).
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43 527 The fact that some food groups were not taken into account, could have led to a certain
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45 528 underestimation of the intake. However, the reduction that takes place during further
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47 529 preparation of foods was not taken in to account but is a factor of overestimation of the intake
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49 530 of sulfites. Further, the fact that no illegal uses of sulfites in certain food groups were taken
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51 531 into account, could also have led to an underestimation. In 2005 1002 tests with malachite
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53 532 green were performed in chopped meat by the FASCA of which 54 were positive. 43 of these
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55 533 were confirmed in the laboratory. The average sulfite content in the samples exceeding the
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57 534 LOQ was on average 474.1 +/-487.1 mg/kg and 1782.6 mg/kg at the 97.5 percentile.
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60 536 In 1999, JECFA reviewed the intake estimation of sulfite from different countries (JECFA,
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538 1999): Aus-NZ, China, Finland, France, India, Japan, Spain, United Kingdom and United
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and 1400 % of ADI. It should be noted that data modelling differed between countries. The

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3 540 different used models are the budget method, the poundage data method, the FBS/HES/sales
4 data method, model diets method and individual dietary records method. A summary of the
5 541
6 data method, model diets method and individual dietary records method. A summary of the
7 542 mean intake results *per capita* for those countries is given in Table 4. All of these results are
8 based on a Tier 2 like approach. Comparatively to those results, in the present study lower
9 543
10 544 intakes were found, except when compared to Finland, India and USA.

11 545
12
13 546 A more recent study in France (1998) performed a Tier 2 intake estimation (Verger et al. 1998).
14 547 The average sulfite intake was estimated to be 1.34 mg SO₂/kg bw/day (191 % of the ADI)
15 548 and the intake at the 95th percentile of the population to be 3.13 mg SO₂/kg bw/day (447 % of
16 549 the ADI). All of these estimates exceeded the ADI of 0.7 mg/kg bw/day and were comparable
17 550 to the earlier calculated data. The estimation of the intake of sulfite in France (Verger et al.
18 551 1998) on basis of actual concentrations for the global population was 20 mg SO₂/day *per*
19 552 *capita* in 1992 (48 % of the ADI). The authors concluded that the major contributors to the
20 553 intake of sulfite were alcoholic beverages (wine, cider and beer). They performed an intake
21 554 calculation for alcoholic beverage consumers only and for non alcoholic beverage consumers.
22 555 They found an intake of sulfite respectively of 31.5 mg SO₂/day (75.0 % of the ADI) and 1.96
23 556 mg SO₂/day (4.67 % of the ADI).

24 557
25
26 558 In Italy during 2000, exposure to sulfite was estimated for adults and children (Leclercq et al.,
27 559 2000). A first Tier 2 calculation resulted in an exposure of 0.82 mg SO₂/kg bw/day or 117 %
28 560 of the ADI, of which 32 mg was due to wine consumption (65 % of total intake), 8 mg from
29 561 dried fruits (16 %) and 2 mg from fish (4 %).

30 562
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32 563 Since the ADI is exceeded by the Tier 2 in Italy, calculation with actual data was performed
33 564 by the same authors. Leclercq *et al.* (2000) made 2 different modelled diets rich in sulfited
34 565 foods in order to calculate estimation of the intake on bases of actual concentrations in food.
35 566 The models differed for children and adults, where it was assumed that the body weight is 30
36 567 kg for a child and 60 kg for an adult. The ADI was not exceeded using the example diets
37 568 proposed for calculation, although nearly (more then 95 % of the ADI) for adults in the worst
38 569 case scenario. Real food consumption data would have increased the amount of information
39 570 and improved the intake estimates.

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42 572 A study conducted in Slovakia (Sinko & Janekova, 2006) demonstrated that the intake of
43 573 sulfite in a group of children aged between 7 and 10 years old (lowest body weight 21 kg;

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3 574 average weight 26 kg) was between 27 and 93 % of the ADI for a child of 21 kg. The study
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5 575 was based on the actual food consumption during 4 months and maximum allowed sulfite
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7 576 content in food.

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9 577
10 578 A recent survey in Australia (Food Standards Australia New Zealand, 2005) showed an
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12 579 average intake of sulfite in the population of 0.2 mg SO₂/kg bw/day (lower bound estimation;
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14 580 concentrations not detected set to zero). With the upper bound estimate, the intake obtained
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16 581 was the same (30 % of the ADI). The same calculation for the 95th percentile of the
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18 582 population showed an intake estimate of 0.9 mg SO₂/kg bw/day (lower bound or upper
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20 583 bound), that corresponded to 130 % of the ADI. It was shown that in Tier 2 approach the
21
22 584 intake estimated for high consumers was 1400 % of the ADI (JECFA) while with actual data,
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24 585 the intake for the high consumers became 130 %, which is more than 10-fold difference (yet
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26 586 still above the ADI).

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28 588 In this study, actual typical levels of sulfites were much lower than the maximum levels set in
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30 589 the legislation for several food groups. This resulted in average intake estimates going from
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32 590 0.34 mg/kg bw/day in Tier 2 to 0.19 mg/kg bw/day in the partial Tier 3. Wine showed a
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34 591 decrease in the sulfite intake estimate from 24 % (Tier 2) to 14 % (partial Tier 3) of the ADI
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36 592 for the mean consumer and from 116% (Tier 2) to 69% (partial Tier 3) for the 97.5 percentile.
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38 593 However, the choice of wine of a consumer might be different than average.

39 594

40 41 595 **Conclusion**

42
43 596 In conclusion, it was shown that the intake of sulfites in the Belgian adult population is likely
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45 597 to be below the ADI with the possible exception for some high consumers of wines with high
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47 598 sulfite levels. As a consequence, the intake of sulfites by wine consumers remains a point of
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49 599 attention. Further it was shown that for several food groups actual typical levels of sulfites are
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51 600 much lower than the maximum levels set in the legislation.

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54 602

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59
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606
607 606 occurrence data provided by the Federal Agency of the Safety of the Food Chain (FASFC).

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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)
Dried tomatoes	2005-2006	65	9	7.3	3.7	2.0	13.0	7	12.6	12.8
All dried fruits (defined or undefined; Apricots included)	2006	40	27	384	320	9.0	917	267	876	903
All dried fruits (defined or undefined; Apricots excluded)	2006	25	14	179	238	9.0	781			
Dried apricots	2005-2006	181	176	533	308	0.0	1448	478	1083	1182
Wines (Red, White or undefined)	2004-2006	83	82	114	45.4	30.0	240	113	180	184
of which Red	2004 & 2006	28	28	78.6	33.2	30.0	160	74.5	142	149
White	2004 & 2006	39	39	139	28.1	91.0	198	136	180	181
Raw Shrimps	2006	10	1	10.0	/	10.0	10.0	10	10.0	10.0
Mustard	2006	4	1	18.0	/	18.0	18.0	18	18.0	18.0
Mushrooms	2006	5	2	11.5	3.5	9.0	14.0	11.5	13.8	13.9
Jams	2006	10	10	16.0	11.6	1.0	33.0	14	32.6	32.8
Potato products	2006	16	9	3.8	3.4	0.3	10.0	3	9.6	9.8
Beers	2006	9	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 body weight/day)									% 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 CSIDE (no result produced)									
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 bodyweight/day)									% of ADI at average intake
	Mean (% of total)	SD	P25	P50	P75	P95	P97,5	P99		
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 [°]	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 ^{*°°}	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 reporting in CSIDE (no result produced)									
Condiments and dessert sauces ^{°°}	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

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

Table 4. JECFA summary of sulphite intake (Tier 2 exposure assessment) in 9 countries (JECFA, 1999)

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