



Occurrence of volatile organic compounds in foods from the Belgian market and dietary exposure assessment



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ABSTRACT

Because volatile organic compounds (VOCs) are ubiquitous in the environment and may represent health risks, it is important to know whether they are present in our diet and to identify possible sources in order to reduce daily intake. In the present study, 377 food samples from the Belgian market were screened for the presence of 21 VOCs. The most prevalent of these compounds and respective percentages of occurrence were: chloroform (97%), toluene (95%), ethyl benzene (80%), o-xylene (79%) and benzene (58%). Dietary intake by the Belgian adult population was calculated for these five most prevalent VOCs. The maximum probabilistic dietary intake was with 0.151, 0.645, 0.138, 0.066 and 0.118 $\mu\text{g kg bw}^{-1} \text{day}^{-1}$ for chloroform, toluene, ethyl benzene, o-xylene and benzene respectively below the minimal risk level (MRL) set for each of the VOCs for oral chronic/acute exposure and below the Reference Dose for Chronic Oral Exposure (RfD). The Margin of Exposure (MOE) for benzene was significantly larger than 100,000. Therefore, it was concluded that the dietary exposure to VOCs for the Belgian adult population does not impose health risks.

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1. Introduction

Volatile organic compounds (VOCs) are emitted to the environment from several potential sources including the combustion of fuels and wood and from industrial emissions (Biedermann, Grob, & Morchio, 1996). These compounds are also found in indoor environment due to the use of paints, resins, rubbers, and detergents. In addition, VOCs are present in cigarette mainstream smoke (Tang, Hemm, & Eisenbrand, 2000; Wallace et al., 1987). Due to their widespread presence in air, water and soil and moreover their lipophilic nature, these compounds may accumulate in food (Biedermann, Grob, & Morchio, 1995; Carrillo-Carrion, Lucena, Cardenas, & Valcarcel, 2007; Fabietti, Delise, & Bocca, 2000). Additional food contamination could occur from migration of packaging materials, and formation during food processing

(Bendall, 2007; Carrillo-Carrion et al., 2007; Guillard, Mauricio-Iglesias, & Gontard, 2010; Hodgson, Casey, Bigger, & Scheirs, 2000; Jickells, Crews, Castle, & Gilbert, 1990; Lachenmeier et al., 2010; Seo & Shin, 2010; Tang et al., 2000; Varner, Hollifield, & Andrzejewski, 1991). VOCs have been reported in cooking emissions due to the use of charcoal (Mugica et al., 2001) and the food ingredients used (Huang et al., 2011). Compounds such as benzene, toluene, ethyl benzene, o-xylene and chloroform are among the most common VOCs found in foodstuffs (Biedermann et al., 1996; Fabietti et al., 2000; Fleming-Jones & Smith, 2003; Heikes, Jensen, & Fleming-Jones, 1995; Hiatt & Pia, 2004; Vichi, Pizzale, Conte, Buxaderas, & Lopez-Tamames, 2007). Lachenmeier et al. (2010) reported benzene and toluene formation from several possible precursors found in foods such as, benzoic acid, pinene, limonene, carene, carotene and phenylalanine. Benzene, toluene, ethyl benzene and p/o-xylene have been found in thermoset polyester used for the production of plastic cookware due to the decomposition of t-butyl perbenzoate, which is applied as a polymerization initiator (Guillard et al., 2010; Jickells et al., 1990). The presence of

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chloroform in drinking-water has been previously linked to the chlorination of organic matter naturally present in raw water supplies (WHO, 2011). Suspected sources of exposure to this VOC are linked to drinking water and could involve drinking water and food ingestion, inhalation and dermal exposure during showering or bathing (Miyahara, Toyoda, Ushijima, Nose, & Saito, 1995; WHO, 2011).

Benzene is evaluated as carcinogenic to humans (Group 1) (IARC, 1987) whereas for the other mentioned VOCs toxicological data still remain limited in human studies. However there are indications that they may represent a potential danger to human health. The classification of the other VOCs is possibly carcinogenic to humans (Group 2B) for chloroform (IARC, 1999a) and ethyl benzene (IARC, 2000). Xylenes (IARC, 1999c) and toluene (IARC, 1999b) were not classified as carcinogens to humans (Group 3), although they still may result in neurological effects. It should be noted that their toxicity is usually evaluated individually or in simple mixtures, whereas very little is known about the toxic effects of complex mixtures. Thus, the presence of VOCs is undesirable in food and the ALARA principle (As Low As Reasonably Achievable) should be applied by food authorities. Currently there are no legal limits for the above mentioned VOCs in food, except for benzene, and that only relates to a limit established for drinking water ($1 \mu\text{g L}^{-1}$ by the European Council) (EC, 1998).

Because VOCs are ubiquitous in the environment and may present health risks, it is important to know to which extent they are present in our diet and possibly identify the sources to reduce the daily intake. This study monitored the occurrence of VOCs in foods on the Belgian market and estimated a quantitative dietary exposure of VOCs by the Belgian adult population.

2. Experimental

2.1. Supplies and reagents

Benzene (99.9%), benzene- d_6 with isotopic purity of 99.9% and EPA volatile organic calibration mix 502/524 (Supelco, USA) were purchased from Sigma–Aldrich (Bornem, Belgium) and stored at 4 °C. Sodium hydroxide was obtained by BDH (Poole, England) and boric acid was supplied by Merck (Darmstadt, Germany). Antifoam silicone 411 Rhodorsil[®] was purchased from VWR (Leuven, Belgium). Deionized water (Milli-Q, Millipore Corp.) of $18.0 \text{ M}\Omega \text{ cm}^{-1}$ resistivity was used throughout.

2.2. Determination of VOCs content in foodstuffs

In a recent study, we analyzed 455 food samples for the quantification of benzene in food by distillation and isotope dilution with Headspace Gas Chromatography coupled to Mass Spectrometry (HS-GC/MS). The sampling procedure was extensively described by Medeiros Vinci et al. (2012). Briefly, the samples were bought in local supermarkets based on their risk on benzene contamination: (1) foods with added benzoate; (2) foods which may naturally contain benzoic acid; (3) foods possibly related to benzene through environment/process contamination and (4) other foods. 377 samples were randomly selected out of these 455 food samples and were screened for the presence of 20 additional VOCs (Table 1). The analyzed foodstuffs belonged to the following categories: 1-non alcoholic drinks, 2-alcoholic drinks, 3-sauces, 4-dairy products, 5-fruits and vegetables, 6-meat and meat products, 7-fish and fish products, 8-cereal products, 9-cookies and cakes, 10-fats and oils, 11-ready to eat meals, 12-sugar and confectionary, 13-miscellaneous and 14-fresh eggs.

2.2.1. Sample preparation

Sample preparation was performed as described in Medeiros Vinci et al. (2010). Briefly, solid and fat containing samples were analyzed by HS-GC/MS after distillation, while liquid non fatty samples were directly analyzed by HS-GC/MS. Precautionary measures were taken to avoid sample contamination in the lab, such as storage and sample preparation in a segregate area from lab used for standards preparation and sample analysis.

2.2.2. HeadSpace-GC/MS analysis

Samples were injected on the GC–MS system using a G188A Headspace sampler (Agilent Technologies, Palo Alto, USA). The analyses were performed on an Agilent 6890N gas chromatograph coupled to an Agilent 5973N mass selective detector. Full automation was achieved using Agilent MSD ChemStation data acquisition and data handling software. After incubation of the sample (5 mL in a 10 mL headspace vial) at 85 °C for 20 min, 1 mL of the vapor phase was injected into the GC/MS system in a split injection mode (split ratio 6.8:1). Two different chromatographic methods were used for the determination of benzene and the other VOCs (Table 1) in food samples.

The chromatographic method used for benzene analysis was described in Medeiros Vinci et al. (2010). The method used for the analysis of the remaining VOCs was the following. The temperatures of the headspace loop, transfer line and EPC volatiles interface were 100, 120 and 160 °C, respectively. Substances were separated on a VF-624 capillary column ($60 \text{ m} \times 0.32 \text{ mm}$ (i.d.); $1.8 \mu\text{m}$ film thickness) (Varian, Walnut Creek, USA). The oven temperature was programmed from 45 °C (held for 20 min) to 250 at $25 \text{ }^\circ\text{C min}^{-1}$ and held for 10 min. High-purity helium (Alphagaz[™] 2, Air-Liquide) was used as carrier gas at a flow rate of 1.5 mL min^{-1} . The temperatures of the injection port, ion source, quadrupole and interface were set at 160, 230, 150 and 280 °C, respectively. The mass spectrometer was operated in the selected ion monitoring (SIM) mode. Retention times and recorded m/z values for the different VOCs are described in Table 1.

Quality control of analysis was described in Medeiros Vinci et al. (2012). Identification criteria of VOCs were based on comparison between retention time and the m/z of most abundant ion for each one of the VOCs with the standard EPA volatile organic calibration mix 502/524. The concentration of the VOCs was estimated by

Table 1
Criteria (retention time and ion m/z) used for identification of VOCs in food samples analyzed by distillation and HS-GC/MS.

Compound	Retention time (min)	Ion m/z in SIM mode
Benzene	7.19	78, 77, 51
1,1-Dichloroethane	10.58	63
Chloroform	14.68	83
1,1,1-Trichloroethane	15.47	97
Trichloroethylene	21.62	130
Bromodichloromethane	23.15	83
Toluene	24.80	91
Tetrachloroethylene	25.75	166
Chlorobenzene	27.20	112
Ethyl benzene	27.34	91
o-Xylene	27.52	91
Styrene	28.07	104
Cumene	28.50	105
Bromobenzene	28.92	77
Propylbenzene	29.01	91
2-Chlorotoluene	29.13	91
4-Chlorotoluene	29.27	91
1,2-Dichlorobenzene	30.10	146
Butylbenzene	30.38	91
1,3-Dichlorobenzene	30.48	146
1,2-Dibromo-3-chloropropane	31.30	157

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