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Combining headspace gas chromatography with mass spectrometry detection for confirmation of hydrocarbon residues in virgin olive oil following automatic screening

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Abstract

Hydrocarbon residues can be present in virgin olive oils either naturally or as contaminants. Presence of benzene, toluene, ethylbenzene, xylene isomers and styrene (BTEXS) in virgin olive oil, demanded the establishment of a cut-off level to discriminate oil samples containing these residues at normal levels from those at high levels caused by contamination. By introducing volatile components present in the headspace (HS) of the oil samples (without prior chromatographic separation) into the ionization source of a mass spectrometer, samples were classified as containing normal or high levels of BTEXS (recommendable or non-recommendable for human consumption). Confirmation and quantification of contaminated virgin olive oils were achieved by combining HS sampling with gas chromatography–mass spectrometry (GC–MS). No sample pretreatment was necessary for the chromatographic method, but the addition of 600 μl of ethyl acetate as chemical modifier for 10 ml of virgin olive oil. Detection limits ranged between 3 and 9 ng/ml. Oil analysis showed the presence of toluene in all samples, as well as the absence of benzene, ethylbenzene and *o*-xylene.
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Keywords: Virgin olive oil; Benzene; Toluene; Ethylbenzene; Xylene isomers; Styrene

1. Introduction

Benzene hydrocarbons are widely distributed in the environment, and can also be present in food, either naturally or as contaminants. BTEXS, abbreviation for the substances benzene, toluene, ethylbenzene, the isomers of xylene (*ortho*, *meta* and *para*) and styrene (also called vinylbenzene), are a subclass of volatile organic compounds (VOCs), with boiling points in the range 80–150 °C. The latter is frequently used in food industry to produce plastics by polymerization; those plastics are used as containers for many different food products, which could be contaminated by migration of styrene monomers; however, decarboxylation of the cinamic acid, naturally present in the olive pulp, can also produce the appearance of styrene residues in the olive oil. Natural sources of the rest of hydrocarbons include superior plant wax, algae, plankton and natural oil seepage; anthropogenic sources include domestic and industrial wastes, biomass and wood burning, incomplete fuel oil combustion and urban runoff [1,2]. Some studies have been carried out in Greek and German government's laboratories [3,4] as EU Commission expressed concern about dietary exposure to volatile aromatic compounds (BTEXS) [5]; the data reported by the cited studies showed that maximum concentrations found ranged between 50 μg/kg (ethylbenzene) and 300 μg/kg (xylene). Absence of an official method made the different laboratories work with different analytical techniques, what made more difficult direct comparison of the results. However, intakes from other sources, especially inhalation, are more important.

Available methods for determination of BTEXS residues are almost completely focused on environmental samples. Gas chromatography–mass spectrometry (GC–MS), using

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either static [6] or dynamic [7] headspace (HS) (purge and trap (P&T)) as sample introduction modules, are the analytical techniques of reference. However, some other techniques like LC [8] or near infrared (NIR) [9], as well as other sampling modules such as membrane introduction [10] and solid-phase microextraction (SPME) [11], have been already employed. The determination of these compounds in food samples has not been systematically considered; only one method exists in the literature [12] that used static headspace-GC–MS for the measurement of benzene-hydrocarbons in virgin olive oils.

The use of chromatographic techniques as traditional procedures for BTEXS determination allows separation and unequivocal identification of the particular hydrocarbon if mass spectrometric detection is employed; however, it means long time of analysis and high cost associated with the use of a chromatographic method. Sample screening methods allow reduction of these problems by providing a yes/no response before confirmation; in this regard, all samples are first analyzed by the sample screening method, which is easier and faster, and only positive results need to be confirmed by gas chromatography. Recently, our research group developed a new sample screening methodology to detect BTEXS residues in virgin olive oil by direct combination of headspace sampling and mass spectrometric detection [13]. This instrument has been successfully employed in the field of the olive oil analysis [14–16]. By using chemometric techniques, sample screening models are generated upon the volatiles profile of samples belonging to a training set; by application of these models to the oil samples, a yes/no response is offered (contaminated or no-contaminated samples). However, every positive result from the sample screening system requires confirmation by using a procedure of higher analytical rank.

Since 1996, when the Commission of the European Union alerted about the presence of benzene-hydrocarbons in virgin olive oils, several studies have been performed to establish the concentration level of these compounds. The main problem relies on the absence of an official method of analysis which, applied in all the laboratories, could lead to homogeneous conclusions. Since the problem is taken from the point of view of a higher sensitivity and selectivity, implicated laboratories employ purge-and-trap with GC–MS; however, many problems are associated with this technique (trap contamination, presence of artefacts, etc.), including low precision (for concentrations lower than 0.01 mg of BTEXS/kg of oil, variation coefficients were ca. 50%) [12]. In the present work, besides a classification method (according to the normal concentrations of BTEXS in oil), a conventional confirmatory method by HS-GC–MS is proposed, more robust than P&T–GC–MS, and more accessible to routine laboratories. The unique reference appearing in the literature about the determination of these compounds in olive oil by HS-GC–MS is a simple application note [12] in which experimental conditions were not optimized; however, that work presents a rigorous study of the parameters (chemicals and instrumentals) related to the determination of BTEXS in virgin olive oil by HS-GC–MS, with high sensitivity and precision, what could be proposed as an official method of analysis.

2. Experimental

2.1. Chemicals and standards

All reagents were of analytical grade or better. Benzene, toluene and ethylbenzene were purchased from Sigma–Aldrich (Madrid, Spain), the isomers of xylene, *ortho*, *meta* and *para*, were purchased from Riedel-de-Haën (Seelze, Germany) and styrene was purchased from Merck (Darmstadt, Germany). HPLC-grade organic solvents (ethyl acetate, isopropanol and methanol) were obtained from Panreac (Barcelona, Spain). Refined olive oil (uncontaminated with BTEXS) was obtained from a Spanish olive oil manufacturer company. Ten and 20 ml glass flat-bottomed vials, as well as PTFE–silicone seals, were purchased from Supelco (Madrid, Spain).

Stock standard solutions of each analyte were prepared in methanol at a concentration of 1.0 mg/ml and stored in glass-stoppered bottles in the dark at 4 °C. A standard solution containing benzene, toluene, ethylbenzene, *o*-, *m*- and *p*-xylene and styrene (BTEXS) at individual concentration of 10 μg/ml was prepared in methanol by appropriate dilution of the stocks. Working standard solutions were prepared as needed by spiking refined olive oil samples (blank olive oil) with the standard solution.

2.2. Apparatus

Sample screening method was carried out by using a Chemical Sensor 4440 system (ChemSensor 4440, Gerstel, Mülheim an der Ruhr, Germany), a direct combination of a headspace autosampler and a mass spectrometric detector. The autosampler had capacity for 44 headspace vials, and was composed of a robotic arm, an oven for sample heating/headspace generation, and a six-port injection valve (IV) with a 3 ml loop. Hydrodynamic injection was employed with helium as carrier gas (5.0 grade purity, Air Liquide, Seville, Spain). The operating conditions of the HS were as follows: vial equilibration time, 25 min; oven temperature, 95 °C; vial pressurization time, 12 s; loop fill time, 9 s; loop temperature, 110 °C. The interface of both modules (HS and MS) was an inert transfer line heated at 120 °C. All tubing of the instrument, as well as the transfer line, were passivated with SilicoSteel. The mass spectrometric detector was operated in full scan mode, with a scanned mass range between *m*/*z* 75 and 110. Electron impact ionization (EI) was operated with an ionization energy of 70 eV. The source and quadrupole temperatures were kept at 230 and 150 °C, respectively.

Confirmatory analyses were carried out on an Hewlett-Packard 7694 headspace autosampler (Agilent Technolo-

gies) directly coupled to an HP6890 gas chromatograph equipped with an HP5973 mass spectrometric detector based on a quadrupole analyser and a photomultiplier detector. Gas chromatographic separation was achieved on an HP-5MS fused silica capillary column (45 m × 0.32 mm i.d., and 0.25 μm of film thickness) coated with 5% phenyl–95% methylpolysiloxane. Chromatographic conditions were as follows: inlet temperature, 200 °C; inlet mode, split operation with split ratio 1:15; oven temperature, 40 °C (3 min), raised up to 60 °C at 5 °C/min, and up to 200 °C at 20 °C/min (2.0 min); column flow, constant flow of 1.1 ml/min of helium as carrier gas. The temperatures of the different zones of the detector were maintained as in the sample screening method. The MS detector was operated in full scan mode between m/z 50 and 350; the base peaks of each compound were used for quantification, namely: 78 for benzene, 91 for toluene, ethylbenzene ($m + p$)-xylene and o-xylene, and 104 for styrene. System control was achieved with an HP1701CA MS ChemStation (Agilent Technologies). Total ion current chromatograms were acquired and processed using G1701BA Standalone Data Analysis software (Agilent Technologies) on a Pentium II computer that also controlled the whole system.

2.3. Sample screening procedure

The proposed sample screening method works as follows: 20 ml headspace glass vials were filled with 10 ml of virgin olive oil, plus 800 μl of isopropanol as chemical modifier and, tightly sealed with PTFE/silicone septa, placed in the autosampler; then, the oil samples were heated in the oven at 95 °C with mechanical stirring, to ensure the equilibration of the BTEXS residues between the gaseous phase and the liquid sample; afterwards, by pressurizing (18.0 psi; 1 psi = 6894.76 Pa) and venting (4.0 psi) the sample vial (hydrodynamic injection), the loop connected to the injection valve was filled with the headspace of the sample. By switching the IV, a second helium stream carried the BTEXS residues inside the loop (3 ml) to the ionization chamber of the mass spectrometer via the transfer line.

2.4. Confirmatory procedure

For the confirmatory method, 10 ml of standard refined olive oil (blank olive oil) or virgin olive oil samples containing between 0.01 and 1.0 μg/ml of each BTEXS were placed in 20 ml headspace glass vials and sealed with PTFE/silicone septa; 600 μl of ethyl acetate were added as chemical modifier. Those samples providing a positive result in the sample screening method were analyzed by coupling a headspace generator autosampler to a gas chromatograph equipped with a mass spectrometer. Headspace conditions were as described for the sample screening method; the volatile fraction of the sample containing the BTEXS residues, retained in the loop of the injection valve (3 ml) was introduced into the injection port of the gas chromatograph.

2.5. Chemometric tools for sample screening method

For sample screening purposes, appropriate multivariate models were created able to predict unknown oil samples as uncontaminated or as contaminated with BTEXS residues, according to its concentration. For this purpose, a training set of samples was analysed by the ChemSensor in order to create adequate classification models with the generated data. Several pattern recognition techniques were employed for this purpose: cluster analysis (CA) and principal component analysis (PCA) were employed as unsupervised classification techniques to check the internal structures of the data and possible clustering of the samples, prior to the creation of the models. Afterwards, k-nearest neighbours (KNN) and soft independent modelling of class analogy (SIMCA) were applied to the data of the training set to generate adequate classification models. Several oil samples, unfortified and fortified with BTEXS at concentrations over 0.2 μg/ml (cut-off value), were analyzed as prediction set by the ChemSensor; the models generated upon the training set data were applied to predict these samples in order to validate the proposed sample screening method. When applied to commercial oil samples, the results obtained should be further confirmed by HS-GC–MS, as described in the previous section.

All chemometric techniques employed were obtained from the statistical software package "Pirouette: Multivariate Data Analysis" (v. 3.01), developed by Infometrix Inc. (Woodinville, WA, USA).

3. Results and discussion

The sample screening method was based upon the classification of the oil samples in contaminated or uncontaminated, depending on if its concentration of BTEXS residues was higher or not than a cut-off level established in 0.2 μg/ml what could indicate if the presence of the BTEXS in the olive oil follows natural degradation/transformation processes, or contamination. That value was fixed in previous studies, in which it was found out that, for a total of 75 virgin olive oil samples, the normal values of BTEXS ranged between 0.03 and 0.18 μg/ml. The analysis of samples was performed on a ChemSensor, without chromatographic separation, and by using chemometry as a powerful tool to create valid pattern recognition models. As no chromatographic separation existed, all the volatile compounds reached the detector at the same time, so providing a global response used as chemical fingerprint for the sample characterisation.

3.1. Headspace-gas chromatographic–mass spectrometric confirmation

The confirmatory procedure involved analysis of the oil samples with a headspace sampling module, directly coupled to a gas chromatograph with mass spectrometric detection; almost no sample pre-treatment existed but the addition of

a chemical modifier to the oil sample in order to improve the release of BTEXS residues to the gaseous phase of the sample. The injection of the volatile fraction of the sample into the chromatographic column was carried out by means of an autosampler; therefore, the use of an internal standard is not necessary.

Those chemical and instrumental variables affecting the headspace generation of the oil samples, as much in the screening as in the confirmatory method, were preliminarily optimized in order to obtain the best separation among the signal of uncontaminated and contaminated oil samples. For this purpose, uncontaminated refined olive oil samples were spiked with 0.5 μg/ml of individual BTEXS.

As chemical variables, sample volume and the use of a chemical modifier were evaluated. Sample volume was studied within the interval 8–16 ml using 20 ml vials; optimum signal was maximum for 10 ml, slightly decreasing over this value; the same sample-headspace volume ratios were evaluated by using 10 ml vials, but lower signals were yielded by the detector; so, 10 ml of oil sample in 20 ml vials were fixed as optimum. For the sample screening method, different organic solvents of variable polarity, namely: methanol, ethanol, isopropanol, *n*-hexane and ethyl acetate were assayed as chemical modifiers, as it has been proved that the release of volatile compounds from the sample matrix, including BTEXS residues, is favoured by its presence [17,18]. Variable amounts (200–800 μl) of the above mentioned organic solvents were evaluated. The best results corresponded to isopropanol, being 800 μl the optimum volume. For the confirmatory method, the same solvent employed for screening was tested, but the chromatographic resolution of the peaks was affected, being so obtained broad large peaks which quantification was difficult. So, trying to improve the chromatographic behaviour of the BTEXS residues and its retention on the GC column, variable volumes of *n*-hexane, ethyl acetate and methanol were evaluated to establish the optimum modifier and its volume. Ethyl acetate was selected as it offered the best peak resolution, which enabled the quantification of each BTEXS through the corresponding peak area; the optimum volume of modifier was further studied between 200 and 800 μl. As volumes above 600 μl did not improve the signals obtained, it was selected as optimum.

Instrumental variables were also optimized as influenced the signal obtained; heating time and oven temperatures were evaluated in the intervals 10−40 min and 70−110 °C, respectively. Signal slightly increased as raised equilibration time of the sample inside the oven, but time values higher than 25 min kept signal constant; as expected, BTEXS signal increased with oven temperature up to 95 °C. Therefore, 30 min of sample heating time and 95 °C of oven temperature were stated as optimum. As explained before, volatile compounds released from the sample filled the loop coupled to the injection valve by a two-step process that included pressurization and further vent of the vial. Pressurization and vent times of the vial were also optimized due to its influence on the amount of sample reaching the detector. Intervals assayed varied between 0.1 and 0.4 min for pressurization time and between 0.03 and 0.3 min for vent time. Optimal values were found to be 0.2 and 0.15 min for pressurization and vent times, respectively.

Table 1
Figures of merit for the determination of BTEXS compounds

Compound	Linear range (ng/ml)	Detection limit (ng/ml)	R.S.D. (%)
Benzene	10–1000	2.8	4.5
Toluene	10–1000	3.5	4.9
Ethylbenzene	20–1000	6.0	6.5
m + *p*-Xylene	20–1000	6.9	7.0
o-Xylene	20–500	7.4	7.5
Styrene	25–1000	8.8	8.1

3.2. Calibration, sensitivity and precision

Analytical curves for refined olive oil (blank) containing benzene, toluene, ethylbenzene, *m*-, *p*- and *o*-xylene and styrene, at different concentrations (between 0.01 and 1.0 μg/ml), were obtained by plotting the peak area against the analyte concentration for each compound. *m*-Xylene and *p*-xylene were identified and quantified together as were coeluted and presented the same retention times. The figures of merit of the calibration graphs (correlation coefficient ranged from 0.998 to 0.999) are summarized in Table 1. Detection limits were calculated for refined olive oil (blank) samples that were spiked with a 10 ng/ml concentration of each BTEXS as no blank signal was obtained ($n = 11$). The precision of the method, expressed as relative standard deviation (R.S.D.) was checked on 11 oil standards containing the seven analytes studied at a concentration of 50 ng/ml. As can be seen in the table, limits of detection ranged between 2.8 (benzene) and 8.8 (styrene), with an average R.S.D. value (expressed as reproducibility) of 6.4%.

In order to demonstrate the validity of the proposed confirmatory HS-GC–MS method, a recovery test was carried on different oil samples, namely: refined oils (corn and olive) and virgin olive oil. It was performed by fortifying each type of oil in triplicate at two different concentration levels by using external standards (50 and 100 ng/ml of each BTEXS in olive and corn refined oil, none of which containing BTEXS naturally, and 20 and 50 ng/ml in virgin olive oil, which contains BTEXS naturally). As no certified reference material was available for the recovery, the calculation of each BTEXS concentration in real samples was necessary; thus, these concentrations were previously calculated by using each calibration graph and by applying the standard addition method: 43 ± 2, 91 ± 6 and 43 ± 3 ng/ml of toluene, *m* + *p*-xylene and styrene, respectively, were obtained. The percent of recovery showed in Table 2 for virgin olive oil was calculated on the basis of the concentration added. All compounds were correctly identified and the average recoveries obtained (see Table 2) were acceptable for all types of oil samples, ranging between 93 and 97% (for analytes) and between 95 and 96% (for samples).

Table 2
Percent recoveries (mean of six determinations ± S.D.) of BTEXS added to oil samples at concentration of 50 and 100 ng/ml

Compound	Recovery (%)		
	Refined olive oil	Refined corn oil	Virgin olive oil
Benzene	96 ± 4	89 ± 3	93 ± 6
Toluene	93 ± 5	97 ± 6	93 ± 5
Ethylbenzene	95 ± 5	95 ± 5	97 ± 6
m + *p*-Xylene	93 ± 6	97 ± 4	92 ± 4
o-Xylene	99 ± 4	95 ± 5	97 ± 5
Styrene	100 ± 4	94 ± 5	97 ± 6

3.3. Application to virgin olive oil

The proposed sample screening method was used to discriminate the presence of BTEXS residues at concentration levels higher than the cut-off value (0.2 μg/ml of total BTEXS). Oil samples were purchased from local markets in the area; geographical origin, olive variety and acidity grade were tried to keep as heterogeneous as possible with the aim to cover all types of virgin olive oils. Screening method was applied to 50 oil samples; only those containing BTEXS residues at concentration levels higher than 0.2 μg/ml gave positive responses (samples 3, 6, 9 and 12 in Table 3). In addition to the confirmation of the global positive response obtained from the screening system, the HS-GC–MS procedure was also used for the systematic quality control of the results provided in the screening step. For this purpose, several samples that generated an analytical signal lower than that associated to the cut-off level (classified as negative), were subjected to confirmation following the whole process in order to identify the analytes present in the sample and their concentration. The total concentration was calculated as the sum of the individual ones and it was compared with the cut-off. The results obtained for the selected samples are also listed in Table 3. As can be seen, all the results were consistent with those provided by the screening method. As it was showed in Table 3, the four positive samples contained toluene, together with *m* + *p*-xylene (two samples) or styrene (two samples); for the rest of BTEXS (benzene, ethylbenzene and *o*-xylene), concentrations lower than the detection limits were found. The higher styrene concentrations obtained for samples 9 and 12 (ca. 600 ng/ml) can be attributed to the plastic containers used for distribution of the samples at the markets and the potential migration of styrene monomers to the oil sample. Samples 3 and 6 were stored in glass bottles, what could explain the absence of styrene residues. Benzene, etylbenzene and *o*-xylene were also absent in the quality control of negative samples.

By way of example, Fig. 1 shows the chromatogram (A) for a refined olive oil (blank) fortified with 0.1 μg/ml of each BTEXS, and (B) for a virgin olive oil fortified with 0.05 μg/ml of each analyte. As can be seen, in both cases, all peaks are easily identified from the HS of the olive sample.

Table 3
Application of the screening and confirmatory methods to virgin olive oil samples

Sample	Screening response[a]	Concentration found by HS-GC–MS[b]		
		Toluene (ng/ml)	*m* + *p*-Xylene (ng/ml)	Styrene (ng/ml)
1	Negative	20 ± 2	n.d.	67 ± 4
2	Negative	15 ± 1	n.d.	68 ± 4
3	Positive	293 ± 15	24 ± 2	n.d.
4	Negative	n.d.	n.d.	n.d.
5	Negative	n.d.	27 ± 2	15 ± 1
6	Positive	290 ± 16	30 ± 2	n.d.
7	Negative	17 ± 1	n.d.	21 ± 3
8	Negative	30 ± 2	n.d.	40 ± 3
9	Positive	15 ± 1	n.d.	620 ± 30
10	Negative	22 ± 1	27 ± 2	25 ± 2
11	Negative	n.d.	n.d.	n.d.
12	Positive	29 ± 2	n.d.	602 ± 30
13	Negative	27 ± 1	n.d.	40 ± 3
14	Negative	30 ± 2	49 ± 3	23 ± 2
15	Negative	42 ± 3	93 ± 6	40 ± 3
16	Negative	18 ± 1	n.d.	36 ± 2
17	Negative	73 ± 4	46 ± 3	50 ± 3
18	Negative	29 ± 2	118 ± 6	31 ± 2
19	Negative	57 ± 3	80 ± 4	30 ± 2
20	Negative	19 ± 1	34 ± 3	53 ± 3
21	Negative	17 ± 2	n.d.	12 ± 1
22	Negative	38 ± 3	68 ± 4	79 ± 5
23	Negative	21 ± 1	n.d.	13 ± 1
24	Negative	14 ± 1	n.d.	18 ± 1
25	Negative	35 ± 2	94 ± 6	n.d.

[a] Negative or positive according to the total concentration of BTEXS compounds, higher or lower than 200 ng/ml (cut-off level).
[b] Undetected (n.d.) benzene, ethylbenzene and *o*-xylene.

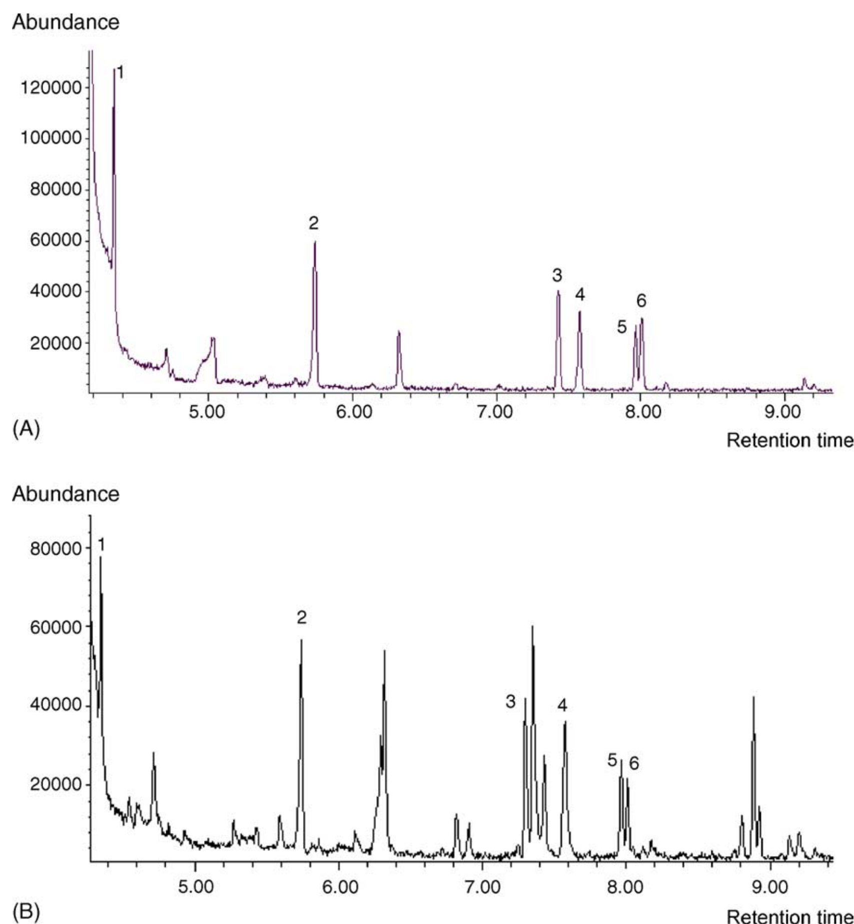

Fig. 1. Chromatogram for: (A) a refined olive oil fortified with 0.1 μg/ml of each analyte; and (B) a virgin olive oil fortified with 0.05 μg/ml of each analyte. Chromatographic peaks of interest: (1) benzene; (2) toluene; (3) ethylbenzene; (4) *m* + *p*-xylene; (5) styrene; and (6) *o*-xylene.

4. Conclusions

It has been proved that the sample screening method allows the discrimination of virgin olive oil samples containing BTEXS residues at normal concentrations (not toxic for human consumption) and samples with concentrations above the normal values as a consequence of contamination (although its consideration as recommendable or non-recommendable for human consumption has not been already defined by authorities due to the absence of a toxic level of BTEXS in these samples). The screening method is robust and reliable as it is based on the use of a fully automated instrument (HS-MS) and chemometric techniques which are able to classify samples correctly. On the other hand, the development of a confirmatory method (HS-GC–MS) allows validation of the results obtained by the sample screening method as no false negatives nor false positives were detected. Hence, the confirmatory HS-GC–MS method can be considered as robust, sensitive, precise and reliable, in such a way that could be proposed as an official method for the determination of BTEXS residues in edible oils.

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