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Analytical Methods

# Optimised off-line SPE–GC–FID method for the determination of mineral oil saturated hydrocarbons (MOSH) in vegetable oils

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ABSTRACT

An optimised off-line SPE–GC–FID method based on the use of silver-silica gel was developed for the determination of mineral oil saturated hydrocarbons (MOSH) in vegetable oils, including olive pomace oil. The method is specific in not including the aromatic hydrocarbons. The performance of different silica gels (untreated, activated and treated with silver nitrate) was compared in terms of capacity to retain fat and retention of interfering olefins present in particularly large amounts in refined olive oils. A coefficient of variation of 9% was obtained performing six replicate analyses of an extra virgin olive oil fortified with an amount of MOSH near the estimated LOQ (15 mg/kg). Recoveries were close to 100%. The use of activated aluminium oxide as an additional tool to eliminate interference by endogenous long-chain *n*-alkanes, is discussed.



## 1. Introduction

Edible oils, like many other foodstuffs, are often contaminated by mineral oil products (Biedermann, Fiselier, & Grob, 2009; Moret, Populin, & Conte, 2009). Mineral oil saturated hydrocarbons (MOSH), consisting of open chain paraffins and cyclic naphthenes, are distinguished from mineral oil aromatic hydrocarbons (MOAH). MOSH and MOAH form "humps" of unresolved components. The sharp peaks on the top of the hump of the MOSH mostly represent *n*-alkanes of plant origin ($C_{21}$–$C_{35}$), recognised by the prevalence of odd carbon numbers. MOSH concentrations in different types of edible oils were reported by Fiorini et al. (2008), Moret, Populin, Conte, Grob, and Neukom (2003), and Wagner et al. (2001).

Since the MOSH are the predominant part of the mineral oil and more easily analysed than the MOAH, they are usually analysed as markers for the presence of mineral oils. Capillary GC is used with flame ionisation detection (FID). FID enables quantitative determination without calibration by the mineral oil determined in the samples, since it provides virtually the same response for all hydrocarbons.

The isolation of the MOSH from the lipids can be achieved with on-line (coupled HPLC–GC or HPLC–HPLC–GC) or off-line techniques. Coupled techniques are highly reproducible, allow processing a large number of samples per day and are less susceptible to contamination during sample preparation, but corresponding instrumentation is available only in few laboratories. A method for separate analysis of MOSH and MOAH, involving on-line HPLC–GC, was described by Biedermann et al. (2009).

Various approaches have been described for off-line sample preparation of endogenous *n*-alkanes (e.g. for characterising vegetable oils) and mineral hydrocarbons. To isolate the hydrocarbon fraction from sunflower oil, Castle, Kelly, and Gilbert (1993) applied HPLC pre-separation, followed by saponification and solid phase extraction (SPE) on a silica gel cartridge. Guinda, Lanzón, and Albi (1996) and Koprivniak, Procida, and Favretto (1997) utilised saponification followed by column chromatography on silica gel to analyse natural *n*-alkanes. Wagner, Neukom, Galetti, and Grob (2001) validated a method for MOSH analysis retaining up to 100 mg edible oil in a column packed with 3.5 g aluminium oxide. Hydrocarbons were eluted with 2 mL hexane (without controlled MOSH/MOAH separation), of which 50 μL was injected into GC–FID by the on-column/retention gap technique. To avoid interference by olefins (such as squalene and its isomerisation products, mainly for refined olive oils), olefins were brominated to render them more polar and retain them on the sorbent. Later, the aluminium oxide was replaced by 2 g of silica gel activated at 400 °C (Fiselier & Grob, 2008), which increased the capacity to retain lipids to 250 mg and rendered bromination unnecessary in most cases. Fiorini, Paciaroni, Gigli, and Ballini (2010) used 2 g untreated silica gel. The reconcentrated eluate (hexane) was injected into the GC–FID in splitless mode. A narrower band was obtained when using non-activated silica gel, which could have been the result

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of the lower retention of the naphthenes (cyclic alkanes). However, a lower capacity to retain fat (maximum 150 mg of oil) was reported for non-activated compared to activated silica gel (Fiselier & Grob, 2008). To assure quantitative recoveries with the collected fraction (5 mL), the amount of oil loaded onto the column was adjusted to the amount of paraffins expected in the sample. When analysing highly contaminated oils (around 500 mg/kg of paraffins), a maximum of 30 mg oil could be loaded.

Quantitative separation of aliphatic and aromatic hydrocarbons from petroleum by SPE on silica gel with silver nitrate was described by Bennet and Larter (2000). Monoaromatic steroid hydrocarbons (challenging the separation between paraffins and aromatics, due to their appreciable aliphatic character) were separated from aliphatic hydrocarbons.

Method validation and performance criteria for the analysis of mineral oil in edible oils was described by the Standing Committee on the Food Chain and Animal Health (SCFCAH, 2008) on request of the EU Member States to assist control authorities in the control of sunflower oil from Ukraine. In 2008, the Joint Research Centre (JRC) of the European Commission and the Institute for Reference Materials and Measurements (IRMM) organised a proficiency test on the determination of mineral oil in sunflower oil (JRC, 2009). Participating laboratories did not check for MOSH/MOAH separation and it remained unknown to which extent MOAH were included into the analysis. Of the 41 laboratories furnishing clean up details, 2 used on-line HPLC–GC or HPLC–HPLC–GC, 3 applied saponification followed by cleanup on silica gel, 10 utilised SPE cartridges packed with 2–3 g of silica gel (9) or aluminium oxide (1), while the remaining laboratories used columns of different size packed with silica gel (22), aluminium oxide (4) or Florisil (1). Five laboratories used silica gel treated with silver nitrate (15–18.5 g), known for increased retention of unsaturated compounds. Most laboratories used columns packed with large amounts of sorbent (10–30 g), requiring large volumes of solvent and a concentration step prior to GC analysis.

The aim of this work was to optimise and validate a simple and reliable SPE–GC–FID method for MOSH analysis in edible oils as an alternative to on-line HPLC–GC. The criteria were (i) minimal solvent consumption, (ii) minimised reconcentration through well exploiting the column capacity and (iii) avoidance of derivatisation for samples containing large amounts of potentially interfering olefins (i.e. olive pomace oil).

## 2. Materials and methods

Solvents were distilled. Glassware and other materials were rinsed with distilled acetone and hexane just before use. The standard mixture $C_{10}$–$C_{40}$ to check GC performance, single *n*-alkane standards ($C_{14:1}$, $C_{15}$, $C_{16}$, $C_{40}$), paraffin oil viscid (used as external standard), 5-α-cholestane (CHO), 1,3,5 tri-*tert*-butylbenzene (TBB), silica gel coated with ~10 wt.% silver nitrate (+230 mesh) and silica gel 60 (particle size 0.063–0.2 mm, 70–230 mesh) were from Sigma Aldrich (Milan, Italy). Silver-silica was not activated. Glass SPE cartridges (6 mL glass tubes with a frit) were from Macherey–Nagel (Chromabond, Düren, Germany).

### 2.1. Preparation of the SPE cartridge

No commercial silica gel cartridges were used because of contaminants interfering with GC analysis. Cartridges were packed by weighing in 1 g sorbent. They were placed into a vacuum manifold (VacMaster-10, Biotage, IST, Stepbio, Bologna, Italy) in the closed valve position and packed by mixing the dry sorbent with 2 mL distilled *n*-hexane (using the tip of a Pasteur pipette) to obtain an homogeneous slurry and release bubbles. The sorbent bed was allowed to soak with the solvent for 2–3 min and then settled by gentle vibration. Finally the solvent was drained by opening the valve and the cartridge conditioned with 5 mL *n*-hexane, taking the solvent level to the top of the packed bed. Drying of the sorbent bed during conditioning, sample loading and elution was avoided.

### 2.2. Sample preparation

Into a 2 mL volumetric flask, 1.0 g oil was weighed and diluted to volume with *n*-hexane. As an option for better control of the process, standards were added before and after SPE: 200 μL of a solution of $C_{14:1}$, $C_{16}$ and $C_{40}$ (50 μg/mL each) was added to the oil before dilution with *n*-hexane. Of the sample solution, 250 μL was loaded onto the packed cartridge. The first 1 mL *n*-hexane eluate corresponded to the dead volume and was discarded. The following 1.5 mL fraction contained the MOSH and was collected in a cone-shaped vial.

For performance control, 25 μL of an *n*-$C_{15}$ solution (50 μg/mL) was added to the eluate. The *n*-$C_{16}$/*n*-$C_{40}$ ratio checked for possible GC discrimination (more common when using splitless injection), the *n*-$C_{16}$/*n*-$C_{15}$ ratio for losses during sample preparation (recovery) and the absence of $C_{14:1}$ for efficient separation between MOSH and olefins.

### 2.3. GC–FID analysis

Of the collected fraction, 40 μL was introduced into GC–FID by the on-column/retention gap technique (Grob, 1987), using manual injection at 5 μL/s. The 5160 Mega series gas chromatograph (Carlo Erba Instruments, Milan, Italy) was equipped with FID, the base block being thermostatted at 330 °C. The 10 m × 0.25 mm i.d. separation column was coated with cross-linked PS-255 (0.15 μm; MEGA, Milan, Italy). A 5 m × 0.53 mm i.d. uncoated precolumn was attached by a press-fit connector. The oven temperature was programmed at 25 °C/min from 65 °C (4 min; solvent evaporation) to 320 °C (10 min). The carrier gas (helium) inlet pressure was 90 kPa (flow rate of 4 mL/min at 30 °C).

Data was acquired and processed by ChromCard (Fisons, Milan, Italy). The MOSH area was determined by the integration of the whole hump and subtraction of the peaks on the top of the hump (mainly endogenous *n*-alkanes). The position of the baseline was determined by injecting solvent. Quantification was performed by external calibration (paraffin oil at 100 mg/kg). The area of the hump can be also determined manually on the printed chromatogram by approximation with simple geometrical forms and comparison with the area of the calibration standard (Wagner et al., 2001).

## 3. Results and discussion

The starting point of this work was a manual method developed for a workshop (Fiselier & Grob, 2008). The amount of sorbent used to isolate the MOSH was reduced from 2 to 1 g in order to reduce solvent consumption. To avoid band broadening, the sample was loaded with a minimum amount of solvent (125 mg of oil in a total volume of 250 μL). The elution volume was minimised to obtain quantitative recovery at minimum dilution. The performance of silica gel activated overnight at 400 °C was compared with that of non-activated silica gel and silica containing silver nitrate.

### 3.1. Capacity to retain fat

The capacity of silica gel to retain fat depends on the amount of sorbent, its activation and the mobile phase used for elution, being maximum with a pure alkane, such as pentane or hexane (Grob,

**Table 1**
Experiments on the capacity to retain fat.

| | Untreated silica gel[a] | | Activated silica gel | | Silver-silica gel | |
|---|---|---|---|---|---|---|
| | Oil loaded (mg) | Fat breakthrough | Oil loaded (mg) | Fat breakthrough | Oil loaded (mg) | Fat breakthrough |
| Extra virgin olive oil | 75 | No | 125 | No | 125 | No |
| | 100 | Yes | 150 | No | 150 | No |
| | | | 175 | Yes | 175 | Yes |
| Extra virgin olive oil spiked with 500 ppm of paraffin oil | 75 | No | 125 | No | 125 | No |
| | 100 | Yes | 150 | No | 150 | No |
| | | | 175 | Yes | 175 | Yes |
| Olive oil | | | 125 | No | 125 | No |
| | | | 150 | No | 150 | No |
| | | | 175 | Yes | 175 | Yes |
| Olive pomace oil | | | 125 | No | 125 | No |
| | | | 150 | Yes | 150 | Yes |
| | | | 175 | Yes | 175 | Yes |

[a] Exposed to lab air overnight.

Kaelin, & Artho, 1991). Table 1 reports results obtained by loading different amounts of various types of oil onto non-activated silica gel, silica gel activated overnight at 400 °C and silver-silica gel, using *n*-hexane as eluent. No difference was observed between activated silica gel and silver-silica gel: up to 150 mg oil (125 mg for olive pomace oil) could be loaded onto 1 g sorbent without breakthrough of fat. No capacity decrease upon fortification of an extra virgin olive oil with 500 mg/kg paraffin oil was observed. Lower and variable capacity was observed for non-activated silica gel (depending on humidity). Table 1 reports results obtained for silica gel left exposed to ambient air overnight.

Less than 4 mg oil was eluted with the MOSH when loading 175 mg extra virgin olive oil and 150 mg olive pomace oil onto the silver-silica column. A maximum load of 125 mg was chosen for all except olive pomace oil (100 mg), such that at most 75% of the column packing was exploited to retain the triglycerides, leaving sufficient capacity for retaining olefins and aromatics.

### 3.2. MOSH elution

To compare the elution profile of MOSH from silver-silica gel with that from silica gel activated overnight at 400 °C, an extra virgin olive oil spiked with 200 mg/kg paraffin oil was fractionated. After discharging the first millilitre, 250 μL fractions were collected and analysed by GC (Fig. 1). The MOSH fraction was slightly broader when using activated silica.

This was confirmed by a recovery experiment: after the dead volume (1 mL), the subsequent 1.5 mL and two 0.5 mL fractions were analysed. For the activated silica gel, 91% of the MOSH were recovered in the 1.5 mL fraction compared to 97% for the silver silica gel. Low coefficients of variation (⩽2.0%) were obtained for both the sorbents (3 replicates).

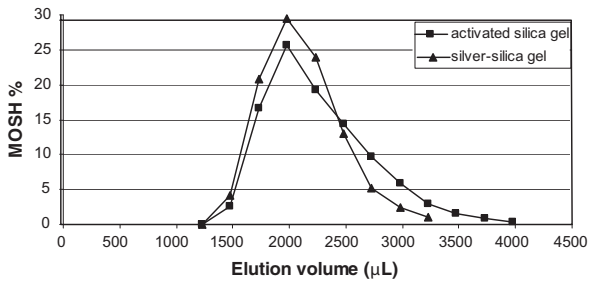


**Fig. 1.** Elution profiles of MOSH.

To verify an efficient separation between the MOSH and the MOAH, an extra virgin olive oil sample spiked with CHO (marking the end of the MOSH fraction) and TBB (marking the least retained MOSH), as described by Biedermann et al. (2009), was fractionated on silver-silica gel. Most of the CHO eluted with the first 1.5 mL of *n*-hexane (together with MOSH) and only traces with a further 0.5 mL of hexane. The subsequent fraction (0.5 mL of *n*-hexane/dichloromethane 40:60 v/v) was free from Cho and TBB. TBB started to be eluted with the following 0.5 mL of *n*-hexane/dichloromethane 40:60 v/v.

### 3.3. Retention of olefins

Using non-activated silica gel, olefins may be co-eluted with the MOSH fraction. Olive oils contain large amounts of squalene, which is partially isomerized after refining and forms a hump of unresolved isomers centred close to *n*-$C_{28}$. Other unsaturated hydrocarbons, such as sterenes and carotenoids, can also hinder quantitative MOSH determination (Biedermann et al., 2009).

Fig. 2 compares fractionating on activated silica and silver-silica for an olive pomace oil containing a large amount of isomerized squalene: on activated silica, olefins were partially co-eluted with MOSH (fraction I), whereas silver-silica selectively retained unsaturated hydrocarbons, rendering derivatisation unnecessary.

*n*-C14:1, added to monitor retention of olefins by the SPE, was completely absent from the MOSH fraction.

### 3.4. Selective retention of n-alkanes

Vegetable oils as well as other foods of plant origin may contain large amounts of natural paraffins, primarily odd-numbered *n*-alkanes. When these overload GC, it may be difficult to draw the upper contour line of the hump formed by the MOSH. Fiselier, Fiorini, & Grob (2009a, 2009b) proposed the use of aluminium oxide activated at 350–400 °C to selectively retain *n*-alkane with more than about 20 carbon atoms. Retention of *n*-alkanes is strong when using hexane as eluent, while small amounts of polar modifiers or impurities in the solvent cause it to irreversibly collapse. 300 mg aluminium oxide was usually sufficient to retain the *n*-alkanes of 20 mg oil with a high load of plant paraffins.

As an optional tool, 500 mg aluminium oxide activated overnight at 400 °C was weighted into a glass cartridge. The MOSH fraction from the silver-silica column was filtered through the dry aluminium oxide. The branched and cyclic MOSH were eluted with 1 mL *n*-hexane (2 times the dead volume of the sorbent bed). Fig. 3 shows the MOSH from an extra virgin olive oil fortified with

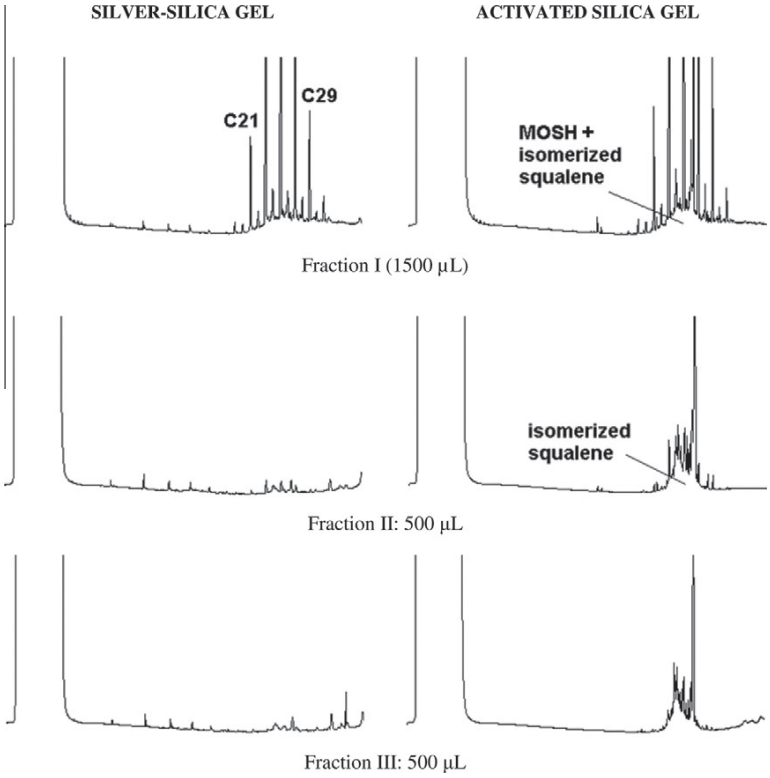


**Fig. 2.** GC–FID traces obtained by fractionating an olive pomace oil on activated silica gel or silver-silica gel.

100 mg/kg paraffin oil processed on silver silica and analysed before and after passage through aluminium oxide. The *n*-alkanes beyond about $C_{25}$ were completely retained, but also some 15% of the iso-paraffins and naphthenes.

The passage through activated aluminium oxide represent an optional tool to reach lower detection limit and to lower measurement uncertainty when analysing sample containing large amounts of endogenous *n*-alkanes. Since the method proposed for routine analysis did not include this step, it was non-comprised in the validated procedure.

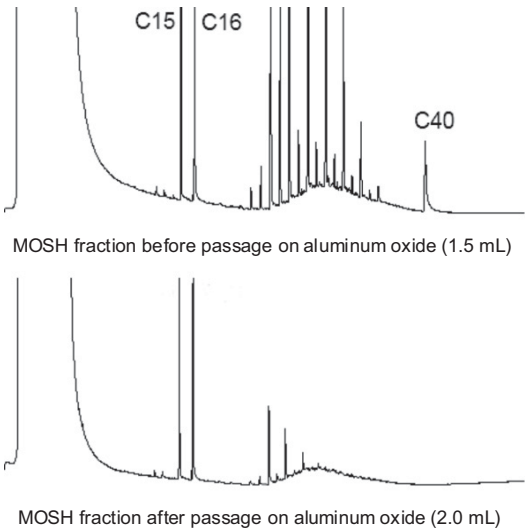


**Fig. 3.** Effect of processing an extra virgin olive oil spiked with 100 mg/kg paraffin oil on aluminium oxide.

### 3.5. Method performance

External calibration involved duplicate analysis of a blank extra virgin olive oil spiked with paraffin oil ranging from 10 to 500 mg/kg. Linear regression of the five-point calibration curve, obtained by plotting the concentration of paraffin oil against the average chromatographic area, provided a coefficient of determination of 0.998 ($y = 89498x + 123131$).

Precision and recovery were assessed by repeating the complete analysis of an extra virgin olive oil before and after fortification with 20 and 100 mg/kg paraffin oil 6 times on two different days (3 replicates per day). Table 2 reports averages, standard deviations and coefficients of variation of the chromatographic areas obtained for endogenous *n*-alkanes (unspiked extra virgin olive oil) and for the MOSH after spiking.

Coefficients of variation lower than 9% and 2% were found for spikes with 20 and 100 mg/kg of paraffin oil, respectively. Recoveries above 97% were found for both fortification levels. Comparing the hump of MOSH of a spiked sample with that of a standard solution of the same paraffin oil (100 mg/kg referring to the oil) showed virtually complete recovery (Fig. 4).

To assess accuracy, the contaminated crude sunflower oil, contaminated refined sunflower oil, spiked sunflower oil and blank sunflower oil used for the proficiency test on the determination of mineral oil in sunflower oil (JRC, 2009) were analysed with the proposed method. Table 3 compares the data (average of two replicates) obtained with assigned values reported in the final report of the proficiency test.

**Table 2**
Precision determined for an extra virgin olive oil with and without spiking with paraffin oil.

| | | Mean (area)[a] | SD | CV% |
|---|---|---|---|---|
| Extra virgin olive oil (not spiked) | C21 | 95607 | 3656 | 3.8 |
| | C22 | 144729 | 5411 | 3.7 |
| | C23 | 2190283 | 63262 | 2.9 |
| | C24 | 1057482 | 31619 | 3.0 |
| | C25 | 2012374 | 55847 | 2.8 |
| | C26 | 239543 | 5845 | 2.4 |
| | C27 | 1468990 | 37874 | 2.6 |
| | C28 | 178223 | 11527 | 6.5 |
| | C29 | 1150590 | 25606 | 2.2 |
| | C30 | 139177 | 3354 | 2.4 |
| | C31 | 828180 | 18390 | 2.2 |
| | C32 | 94830 | 2274 | 2.4 |
| | C33 | 398778 | 10026 | 2.5 |
| | C34 | 36777 | 865 | 2.4 |
| | C35 | 97308 | 2032 | 2.1 |
| | MOSH | – | – | – |
| Extra virgin olive oil spiked with 20 mg/kg of paraffin oil | MOSH | 1784612 | 160019 | 9.0 |
| Extra virgin olive oil spiked with 100 mg/kg of paraffin oil | MOSH | 8716661 | 163953 | 1.9 |

[a] Data are mean of 6 replicates.

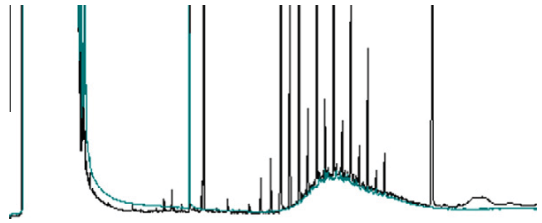

**Fig. 4.** GC–FID of a blank extra virgin olive oil spiked with 100 mg/kg of paraffin oil (black) and of the same amount of paraffin oil as used for spiking (grey).

According to Commission Regulation (EC) 1151/2009 regarding contaminated sunflower oil from Ukraine, referring to Commission Regulation (EC) 333/2007, analytical results on mineral oil contamination should be reported as $x \pm U$ (where $x$ is the analytical result and $U$ is the expanded measurement uncertainty), using a coverage factor of 2, which gives a level of confidence of approximately 95% ($U = 2u$, where $u$ is the standard relative uncertainty). A $U$ value of 18% was obtained for an extra virgin olive oil fortified with 20 mg/kg of MOSH (amount near the LOQ). For higher MOSH concentration (100 mg/kg), a lower $U$ value was obtained (4%).

Since the measurement uncertainty is mainly determined by the positioning of the baseline and the upper contour line of the hump against endogenous *n*-alkanes (Biedermann et al., 2009) and greatly depends on the molecular distribution of the MOSH, it depends on the sample analysed. Values tend to be higher for oils containing larger amounts of endogenous *n*-alkanes (e.g. pomace olive oil) and oils with MOSH covering a broad range of molecular mass.

**Table 3**
Accuracy determined by comparison with samples of a proficiency test.

| Sample | Assigned value (mg/kg) (proficiency test) | Average value (mg/kg) (proposed method) |
|---|---|---|
| Contaminated crude sunflower oil | 351[a] | 375 |
| Contaminated refined sunflower oil | 105[a] | 106 |
| Spiked sunflower oil | 114[b] | 117 |
| Blank sunflower oil | – | <LOD |

[a] Median value.
[b] Gravimetrically established.

A limit of detection (LOD) of approximately 5 mg/kg was derived from a blank extra virgin olive oil fortified with 10 mg/kg paraffin oil (lowest calibration point). The limit of quantification (LOQ) was around 15 mg/kg (3 times the LOD); it depends on the concentration of the interfering plant *n*-alkanes and the molecular mass distribution of the MOSH. Up to 10–15 times lower detection limits can be reached by concentrating the sample before injection. Losses of volatiles must be kept under control, primarily by not evaporating the sample to dryness (recoveries are quantitative starting from C10 to C12).

## 4. Conclusions

An off-line SPE–GC–FID method for the determination of MOSH in different types of edible oils was optimised as an alternative to on-line coupled techniques. It showed good performance characteristics and is applicable without derivatisation even to difficult oils, such as olive pomace oil. It could also be used to quantify endogenous *n*-alkanes for characterisation purposes. Compared to silica gel activated overnight at 400 °C, silver-silica gel better retains olefins and circumvents derivatisation. If required for MOSH analysis, additional passage through activated aluminium oxide eliminates interfering plant *n*-alkanes.

The method only determines the MOSH, which is usually around 70–80% of the mineral oil (depending on the source and the raffination). Therefore the result mostly underestimates the content of "mineral oil". This was also the case with the previous methods, but since the MOSH/MOAH separation was not controlled, it was probably also a source of systematic deviations obtained with differing methods. It will be a next step to extend the method to provide data also on the MOAH. Since toxicology is fundamentally different, it is probably more adequate to measure MOSH and MOAH separately.

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