Study of the Main Constituents of Some Authentic Hazelnut Oils

COLIN CREWS,*,† PATRICK HOUGH,† JOHN GODWARD,† PAUL BRERETON,†
MICHELLE LEES,§ SEBASTIEN GUIET,§ AND WILFRIED WINKELMANN#

Central Science Laboratory, Sand Hutton, York YO41 1LZ, United Kingdom; Eurofins Scientific,
Rue Pierre Adolphe Bobierre, B.P. 42301, 44323 Nantes Cedex 3, France, and Eurofins/
Wiertz-Eggert-Jorissen, Stenzelring 14 b, D-21107 Hamburg, Germany

This paper describes the composition of authentic hazelnut oils obtained from nuts collected from five countries that are major suppliers of hazelnut oil. Oils were analyzed using standard methods for fatty acids, fatty acids in the triacylglycerol 2-position, tocopherols and tocotrienols, triacylglycerols, sterols, steradienes, and iodine value. The results were generally in good agreement with those of other publications. Tocotrienols, previously unreported in hazelnut oil, were detected in one sample. There were no major differences in the composition of oils from different countries. Roasting the nuts prior to pressing had little effect on oil composition.

KEYWORDS: Hazelnut oil; *Corylus avellana***; fatty acids; triacylglycerol 2-position; tocopherols; tocotrienols; triacylglycerols; sterols; steradienes; iodine value**

INTRODUCTION

Hazelnuts provide a ready source of a nutritionally valuable oil, the kernels containing ~60 wt % of oil, which can be recovered effectively by pressing. Hazelnut oil is relatively rich in mono- and polyunsaturated fatty acids, which are believed to have beneficial effects in the prevention of coronary heart diseases, and regular consumption of nuts has proven to reduce the incidence of such diseases and to increase longevity (*1, 2*). About 700 000 metric tons of hazelnuts are produced worldwide every year (*3*). Approximately 70% of world production is from Turkey, with smaller quantities from the United States, Italy, France, and Spain. Production in Turkey has increased considerably in recent years (*4*).

Hazelnuts can be obtained from various species of *Corylus*. In Turkey, the major producing country, different varieties of the genus *Corylus* are used, principally the common hazelnut *C. avellana* var. *pontica* and also the giant hazelnut *C. avellana* × *maxima* Mill and the Turkish hazelnut *C. colurna* var. *glandulifera*. Wild varieties interbreed with cultivated ones, and different varieties are frequently grown and harvested together.

Hazelnut oils are often sold to the speciality product market due to its distinctive flavor; the oil is therefore in demand as a salad oil and for flavoring confectionery. The flavor of hazelnut oil can be increased significantly by roasting the nuts prior to pressing; thus, the hazelnut oils sold on the market can be crude products from roasted or unroasted nuts, refined oils from unroasted nuts, or mixtures of these.

The composition of edible vegetable oils has been studied for many years with a view to obtaining information to improve the quality of the product in terms of storage stability, flavor, taste, and nutrition and also to ensure the authenticity of the material.

A number of analytical methods have been developed for the analysis of vegetable oils, and those most frequently applied have been tested by in-house validation or collaborative trial and have been adopted as official procedures by bodies such as the Association of Official Analytical Chemists, the American Oil Chemists' Society, and the International Olive Oil Council.

Many tables describing the composition of the major and selected minor components of vegetable oils have been produced and published in standard texts. The Codex Alimentarius Committee on Fats and Oils has published standards for a number of named vegetable oils, which are based on tables of composition (*5*). Data for the composition of an oil sample can be compared with the ranges published in the Codex standard to help to judge whether the sample is authentic and of sufficient purity for trading purposes. However, the usefulness of the trading standards is limited by the accuracy of the data contained in the tables, and the scientific literature occasionally provides evidence of data from authentic oils that falls outside the Codex ranges.

There are likely to be many reasons for differences between genuine results and published data. The two most important are, first, that the number of samples taken for analysis can be insufficient to cover the variety of composition found in nature and, second, that advances in technology continually improve the qualitative and quantitative performance of the measuring instruments. The latter is best illustrated in the increase in

* Author to whom correspondence should be addressed (e-mail c.crews@csl.gov.uk; fax +44 1904 461 111).
† Central Science Laboratory.
§ Eurofins Scientific.
# Eurofins/Wiertz-Eggert-Jorissen.

resolution of chromatographic separation systems used in many analyses over the past 40 years.

The purpose of this survey was to provide data for the composition of hazelnut oil, which is not well represented in terms of sample numbers in the current Codex standard. To this end, samples of known origin were obtained from specific oilseed-producing countries and extracted by means as similar as possible to those used in industry. The oils were analyzed using standardized methods incorporating modern quality assurance procedures.

A similar study carried out in 1995 reported the results of a survey of the composition of hazelnuts (*6*). The survey was limited to 10 samples of nuts collected from eight geographical locations with the oil obtained by solvent extraction in the laboratory. Various analyses were carried out using British Standard and IUPAC methods and the results compared with some values published in the literature.

For the survey reported here a larger number of samples (32) were collected from a reduced number of countries (five) that provided 85% of the world's hazelnut production in 1993 (*3*) and the major market share for oil production. A full audit trail was established to ensure the authenticity of the samples. Oil was extracted by pressing under industry conditions and analyzed using official methods with full quality assurance procedures in place.

To observe any effect of roasting on composition, portions of five of the hazelnut samples were roasted in the laboratory under conditions that simulated industry practice. These samples were subjected to the same analyses as the oils from unroasted nuts.

MATERIALS AND METHODS

Hazelnuts. It was not possible to confirm the identity of any nut sample in terms of variety, and it was evident from the size and shape of nut and the thickness of the shell that various varieties were present. However, each batch of samples appeared to be homogeneous in terms of shell size, shape, and color, and all nuts were in good condition on delivery.

Hazelnuts, in batches of 4 kg each, were collected from Croatia (one sample), France (nine samples), Italy (eight samples), Spain (eight samples), and Turkey (six samples). Two samples, one each from France and Croatia, were collected from the year 2000 harvest and the remainder from 2001. To guarantee the authenticity of the oils, the nuts were collected by the authors in the country of origin. It was ensured as closely as possible that the grade and variety were intended and suitable for oil production, that is, that they was not a type normally used for direct consumption or confectionery manufacture.

The nuts were packed in secure heavy-duty polyethylene containers and sealed with numbered tamper-proof plastic tags with a unique number that was used at all stages from collection through analysis, allowing a detailed audit trail to be established. They were stored at 4 °C prior to removal of the kernels. The hazelnuts were shelled by hand as there was no commercial means of shelling nuts in the United Kingdom. The kernels were stored in sealed bags in the dark at 4 °C until transport to the oil-pressing plant. The samples and their origin are listed in **Table 1**.

Roasted Hazelnuts. To observe any effect of roasting on composition, portions of five of the hazelnut samples were roasted in the laboratory under conditions that simulated industry practice (*7*). Nuts were roasted in a laboratory oven (Gallenkamp Hotbox Size 1, Sanyo Gallenkamp, Loughborough, U.K.), which had been fitted with a rotating shaft and drum turned by an external drive shaft at ~30 rpm. The oven was preheated to 165 °C and held for 30 min. Hazelnut kernels were introduced quickly and roasted for 25 min at 165 °C. The roasted nuts were transferred to sealed bags and allowed to cool and then stored at 4 °C prior to pressing.

Table 1. Origin and Collection Date of Samples

country	region	collection date
Croatia	Unje, Black Sea coast	August 2000
France	Pillac	December 2000
France	Farranans, Isere	September 2001
France	Cancon	September 2001
France	Cancon	September 2001
France	Haute Pyrenees	September 2001
France	Cancon	September 2001
France	Cancon	September 2001
France	Cancon	September 2001
Italy	Sangano, Piedmont	August/September 2001
Italy	Busca, Piedmont	August 2001
Italy	Torino	September 2001
Italy	Petralia	September 2001
Italy	Provincia di Torino	September 2001
Italy	Provincia di Cuneo	September 2001
Spain	Porrera, Catalonia	September 2001
Spain	Reus, Catalonia	September 2001
Spain	Al Forga, Catalonia	August 2001
Spain	Riudons, Catalonia	August 2001
Spain	Espinaredo, Asturias	September 2001
Spain	Villaviciosa, Asturias	August 2001
Spain	Catalogne	September 2001
Turkey	Black Sea coast, Istanbul[a]	August 2001
Turkey	Black Sea coast, Istanbul	August 2001
Turkey	Eninonu, Istanbul	August 2001
Turkey	Eninonu, Istanbul	August 2001
Turkey	Istanbul	August 2001
Turkey	Istanbul[a]	August 2002

[a] Supplied as kernels, otherwise in the shell.

Hazelnut Oil. The oil was extracted from the hazelnut kernels by cold pressing in an industry pilot scale screw press (Statfold Seed Oils, Tamworth, U.K.). The batch size and processing rates were adjusted to maintain the sample temperature at ~37 °C. The oils were then filtered and stored in polypropylene bottles under nitrogen at 4 °C in the dark prior to analysis. Normal steam cleaning procedures were carried out between samples to ensure that there was no adventitious mixing of different samples.

Standards. Standards of the sterols stigmasterol (95%), and sitosterol (containing campesterol) and of fatty acid methyl esters were obtained from Sigma, Poole, U.K. The steradienes stigmasta-3,5-diene (69%) with campesta-3,5-diene (13%) and stigmasta-3,5,22-triene (97%) were obtained from Chiron AS, Trondheim, Norway. The internal standard cholesta-3,5-diene (95%) was obtained from Sigma. Tocopherols and tocotrienols (>95%) were obtained from Merk Biosciences, Schwalbach, Germany. The CBR reference sample RM162 (soy and maize oil) was purchased from the Community Bureau of Reference, Brussels, Belgium.

Determination of Sterols. The composition of the sterols (desmethyl sterols) was determined according to the International Standards Organization method (*8*). Gas chromatography (GC) was carried out with a Varian model CP3380 gas chromatograph (Varian Inc., Walton, U.K.) fitted with a JW DB-5 fused silica column (60 m × 0.25 mm i.d. × 0.1 μm film thickness, Fisher Scientific, Loughborough, U.K.). Helium carrier gas was used at a pressure of 30 psi, 1 mL/min. The injector and detector temperatures were 320 °C, and the oven was programmed from 240 to 255 °C at 4 °C/min. The injection volume was 1 μL, with a split ratio of 20:1.

For quality assurance purposes two reference materials were analyzed. These were a mixture of commercial standards comprising sitosterol, campesterol, and stigmasterol and a characterized in-house mixture of equal volumes of sunflower oil and rapeseed oil. Batches for GC comprised a reagent blank, the standard mixture, the reference oil, five samples, and one sample injected in duplicate. Acceptance criteria were that results for the samples injected in duplicate, the standards, and the reference oils had to agree to within ±10% of the mean values.

Determination of Steradienes. The steradienes campesta-3,5-diene, campesta-3,5,22-triene, stigmasta-3,5-diene, and stigmasta-3,5,22-triene

and their major isomers were determined according to the European Commission method for stigmastadienes in olive oil (*9*). The sensitivity and scope of the method were increased by the incorporation of mass spectrometric detection, the use of internal standardization, and the use of calibration graphs as published previously (*10*).

GC-MS was performed using a Voyager instrument (Thermo-Finnigan, Hemel Hempstead, U.K.) fitted with a JW DB-WAXETR column (30 m × 0.25 mm, Fisher Scientific) with a 0.25 μm phase. The GC used helium carrier gas at a pressure of 15 psi. The injector and transfer line temperatures were 260 °C, and the column oven temperature was held isothermally at 250 °C. Splitless injections of 1 μL were made. The mass spectrometer was operated in selected ion monitoring mode, recording the response for the molecular ions of the steradienes and, for confirmation purposes, the sterol ring fragment at *m/z* 255.

For quality assurance purposes an in-house mixture of virgin olive oil with refined olive oil blended to contain ~0.15 mg/kg stigmastadienes was analyzed as a reference material. Extraction batches comprised a reagent blank, the in-house reference mixture, and 6−10 sample oils. Analytical (GC-MS) batches comprised a reagent blank, standards plus one or two of the extraction batches above. Acceptance criteria were that the levels of individual steradienes in the blank did not exceed 0.05 mg/kg and that the stigmastadienes content of the reference oil fell within 0.1−0.2 mg/kg.

Determination of Fatty Acids (FA). The FA composition of the oils was determined according to the method the International Union of Pure and Applied Chemistry method (*11*, *12*). GC was carried out with a Varian model CP3380 gas chromatograph (Varian Inc., Walton, U.K.) and a fused silica column (50 m × 0.35 mm i.d.), coated with CP-Sil 88 (0.2 μm film thickness, Varian Inc.). Helium carrier gas was used at a pressure of 10 psi, giving a flow rate of 1 mL/min. The injector and detector temperatures were 250 °C, and the oven was programmed from 160 to 220 °C at 1 °C/min. The injection volume was 1 μL, with a split ratio of 20:1.

For quality assurance purposes a Community Bureau of Reference (CBR) sample, RM162 (soya and maize oil), was used. Each analytical batch included a reagent blank, a range of standards, the CBR reference oil, 10−14 samples, 3 samples prepared in duplicate, and 3 samples injected in duplicate. A midrange standard was re-injected at the end of the batch as a back-calibration to check instrument stability. Acceptance criteria for the batch were that the FA concentrations of the CBR and back-calibration standards had to fall between 95 and 110% of the nominal values. Results for the duplicate preparations had to fall within 2% of the mean values for palmitic acid (C16:0), stearic acid (C18:0), oleic acid (C18:1), and linoleic acid (C18:2) and within 10% of the mean values for myristic acid (C14:0), palmitoleic acid (C16:1), linolenic acid (C18:3), arachidic acid (C20:0), eicosenoic acid (C20:1), behenic acid (C22:0), and lignoceric acid (C24:0). Results for the samples injected in duplicate had to fall within 2% of the mean values for C16:0, C18:0, C18:1, and C18:2, within 5% of the mean values for C16:1 and C18:3, and within 10% of the mean values for C14:0, C20:0, C20:1, C22:0, and C24:0.

Determination of FA in the Triacylglycerol 2-Position. The composition of FA in the triacylglycerol (TAG) 2-position was determined according to IUPAC method 2.210 (*13*). GC was carried out with a Hewlett-Packard model 5890 gas chromatograph (Agilent Technologie, Waldbronn, Germany) and a fused silica column 60 m × 0.25 mm coated with BPX70 (0.25 μm film thickness, SGE, Darmstadt Germany). Hydrogen carrier gas was used at a pressure of 21 psi, giving a flow rate of 1.2 mL/min. The injector and detector temperatures were 270 °C, and the oven was programmed from 80 to 120 °C at 20 °C/min, then to 200 °C at 6 °C/min, and then at 4 °C/min to 225 °C and held for 8 min. The injection volume was 1 μL. For quality assurance purposes an in-house reference material (cocoa butter) was analyzed. Each batch included a reagent blank, the cocoa butter reference sample, and up to 20 samples. Two samples in each batch were analyzed in duplicate. Acceptance criteria were that the results for the cocoa butter reference material had to fall within 100 ± 10% of the nominal value, and the results for the duplicate preparations had to fall within 2% of the mean values for C16:0, C18:0, C18:1, and C18:2.

Determination of Tocopherols and Tocotrienols. The tocopherol and tocotrienol composition was determined according to IUPAC method 2.432 (*14*). The HPLC system comprised a Merck-Hitachi model L-6000A pump (Merck, Darmstadt, Germany) and a 250 mm × 4 mm LiChrospher 100 diol column (particle size = 5 μm, Merck). The eluent was 4% *tert*-butylmethyl ether in hexane at a flow rate of 2.3 mL/min. The detector was a Hitachi model L-4000 UV operated at 295 nm (Merck). Injections (50 μL) were made using a Bischoff model 728 autosampler (Bischoff Chromatography, Leonberg, Germany). Tocopherol and tocotrienol reference standards were obtained from Calbiochem-Merck Biosciences, Schwalbach, Germany. The reference material was a vapor condensate of mixed vegetable oils enriched in tocopherols and tocotrienols. Each batch included a reagent blank, a reference material, a standard mixture, and up to 20 samples. Two samples in each batch were analyzed in duplicate. Acceptance criteria were that results for the reference sample had to fall within 10% of the characterized value and that results for the duplicate analyses had to agree within 5% of the mean value.

Determination of the Triacylglycerol Carbon Number (TCN). The TCN was determined according to the AOCS method (*15*). GC was performed using a Carlo Erba HRGC 5300 Mega series (CE Instruments, Wigan, U.K.). The GC was fitted with a JW DB-5HT column (15 m × 0.32 mm with a 0.1 μm film thickness; Fisher Scientific) with a helium carrier gas pressure of 12 psi. Cooled on-column injections (1 μL) were made with the oven programmed from 100 °C (held 1 min) to 310 °C at 49 °C/min and then at 4 °C/min to 390 °C.

For quality assurance purposes a characterized in-house reference mixture of sunflower and rapeseed oils was used. Each batch comprised a reagent blank, a range of standards, the reference oil, 15 samples, and 1 sample analyzed in duplicate. Acceptance criteria were that results for the in-house reference oil and for samples injected in duplicate had to agree to within 10% of the mean values.

Determination of the Iodine Value. The iodine value was calculated from the FA composition according to the American Oil Chemists' Society method (*16*). The data were obtained from the analysis of all samples including the averaged results for three samples prepared in duplicate and the three injected in duplicate. Acceptance criteria were that the iodine values of the reference standard had to fall within 100 ± 10% of those calculated from the FA CBR composition data of the CBR reference material.

Statistical Analysis. The data were examined using the CSL-Metabolab visualization toolbox, a graphical user interface for the Matlab program (The MathWorks Ltd., Cambridge, U.K.) allowing multivariate and classical statistical analyses and methods for rapid integration and spectral visualization. The statistical methods employed were principal components analysis (PCA) for initial data exploration, one-way analysis of variance (ANOVA) to detect whether one or more oils were significantly different from the others for a given factor, and the *t* test for pairwise comparison of different oil types.

RESULTS AND DISCUSSION

Oil Extraction. The volumes of oil returned from the press varied considerably between samples, apparently as a factor of the physical nature of the kernel. They were generally smaller than would be expected from a full-scale oil-pressing plant. For several of the roasted nuts the yield of oil was very low. This was contrary to industry practice and was believed to be due to technical factors related to the size of the press and the softness of their kernels, and not to losses on roasting. As the quantity of oil obtained in most cases clearly differed from the actual volume present in the kernel, no data are presented for the oil content of the samples.

Quality Assurance. For the data reported all results for the replicate injection and analysis samples were within the warning limits of Shewart control charts (*17*), and all results for the reference materials were within the specified limits.

Sterol Composition. The sterol composition of hazelnut oils expressed in milligrams per 100 g of oil is given in **Table 2**.

Table 2. Sterol Composition of Hazelnut Oils (Milligrams per 100 g)

sample	unroasted					roasted		
	Croatia	France	Italy	Spain	Turkey	Croatia	Italy	Spain
no.	1	8	6	7	6	1	2	3
cholesterol	0.7	nd[a]–0.7	nd–0.5	nd–1.4	nd–0.7	nd	0.3–0.6	0.2–1.1
cholestanol	0.7	nd-0.7	nd	nd	nd	nd	nd	nd
brassicasterol	nd	nd	nd	nd	nd	nd	nd	nd
24-methylenecholesterol	nd	nd	nd	nd	nd	nd	nd	nd
campesterol	10	7.3–10.1	6.7–8.1	5.9–9.7	5.8–10.7	9.0	6.1–6.6	7.1–8.9
campestanol	0.6	0.4–0.9	nd–0.8	nd–0.7	nd–1.3	nd	0.3	nd–0.5
stigmasterol	1.6	1.4–2.7	2.0–5.4	1.5–1.9	1.5–2.8	1.6	1.6	1.5–1.8
Δ7-campesterol	nd	nd–1.1	nd–1.7	nd–0.6	nd–0.6	0.3	nd–0.5	nd–0.9
Δ5,23-stigmastadienol	nd	nd	nd-0.4	nd	nd	nd	nd	nd
clerosterol	1.6	1.5–2.4	1.8–2.8	1.9–4.0	1.8–3.8	1.4	2.7–2.9	2.2–2.7
β-sitosterol	134.1	112.5–161.2	116.4–138.8	92.9–149.1	84.5–143.1	122.5	110.8–118.2	119.3–144.5
sitostanol	4.8	3.2–6.7	3.2–7.6	2.9–6.2	3.3–11.7	4.1	4.0–6.0	3.9–6.7
Δ5-avenasterol	4.5	4.5–6.1	4.2–6.2	3.8–6.5	0.8–9.4	4.6	2.2–5.0	2.2–6.2
Δ5,24-stigmastadienol	0.4	0.3–1.5	nd–1.4	nd–0.8	nd–2.0	1	0.1	nd–0.7
Δ7-stigmastenol	2.4	nd–3.2	nd–2.4	nd–1.5	nd–2.1	2.5	nd	nd–0.3
Δ7-avenasterol	1.2	nd–0.8	nd–1.8	nd–0.7	nd–2.2	1.2	nd–1.3	nd–1.0
total	162.6	135.3–195.2	141.3–177.2	114.2–178.4	105.7–183.2	148.3	131.0–140.2	139.2–171.5

[a] nd, <0.1.

Table 3. Sterol Composition of Hazelnut Oils (Percent m/m)

sample	unroasted					roasted		
	Croatia	France	Italy	Spain	Turkey	Croatia	Italy	Spain
no.	1	8	6	7	6	1	2	3
cholesterol	0.4	nd[a]–0.5	nd–0.4	nd–0.8	nd–0.6	nd	0.2–0.4	0.1–0.8
cholestanol	0.4	nd–0.4	nd	nd	nd	nd	nd	nd
brassicasterol	nd	nd	nd	nd	nd	nd	nd	nd
24-methylenecholesterol	nd	nd	nd	nd	nd	nd	nd	nd
campesterol	6.1	4.8–6.3	4.6–5.1	4.9–5.5	4.2–6.3	6	4.3–5.0	4.9–5.2
campestanol	0.3	0.2–0.6	nd–0.5	nd–0.7	nd–0.8	nd	0.2	nd–0.3
stigmasterol	1.0	0.9–1.8	1.1–3.0	0.9–1.4	1.5	1	1.2	1.0–1.2
Δ7-campesterol	0.2	nd–0.7	nd–0.9	nd–0.4	nd–0.6	nd	nd–0.4	nd–0.6
Δ5,23-stigmastadienol	nd	nd	nd–0.3	nd	nd	nd	nd	nd
clerosterol	1.0	1.0–1.6	1.3–1.9	1.3–2.3	1.4–2.8	1	1.9–2.2	1.3–1.8
β-sitosterol	82.4	82.8–84.7	80.1–84.3	81.3–85.9	78.8–83.3	82	84.3–84.6	84.3–85.7
sitostanol	3.1	2.1–4.1	2.3–5.2	2.4–5.1	2.4–4.0	3	3.2–4.8	2.4–4.7
Δ5-avenasterol	2.9	3.0–4.2	3.0–4.6	2.6–5.0	2.8–5.1	3	1.6–3.1	1.7–3.3
Δ5,24-stigmastadienol	0.3	0.3–1.0	nd–0.6	nd–0.5	nd–1.2	1	0.1	nd–0.5
Δ7-stigmastenol	1.4	nd–1.6	nd–1.7	nd–1.2	nd–0.6	2	nd	nd–0.2
Δ7-avenasterol	0.7	nd–0.5	nd–1.3	nd–0.6	nd–1.8	1	nd–1.0	nd–0.6

[a] nd, <0.1.

The levels in oils from unroasted nuts ranged from about 135 to 200 mg/100 g. These values are lower than those of one earlier survey (6) but are in good agreement with the results other surveys (18−21). The dominant sterols were β-sitosterol and campesterol. Levels of sitostanol and Δ5-avenasterol were very variable, from about 3 to 12 mg/100 g and from 1 to 9 mg/100 g, respectively. The range of levels in oils from roasted nuts fell within that for the unroasted samples.

The absolute level of sterols is not of great value in authenticity testing compared to the relative composition, where the variation between samples of the same type is smaller. A considerably higher level of Δ5-avenasterol (21 mg/100 g) has been reported in one sample of hazelnut oil (22); however, the desmethyl fraction was not separated prior to GC, and the peak identified as Δ5-avenasterol might have included coeluting compounds. Others have reported similarly wide ranges of levels of Δ5-avenasterol, Δ7-avenasterol, and Δ7-stigmastenol (19).

The sterol composition of hazelnut oils expressed in percent m/m of total sterols is given in **Table 3**.

Values for the percentage composition of sterols were generally in good agreement with the results published elsewhere (18, 20, 21, 23−25). Lower campesterol and stigmasterol but higher sitosterol levels have been reported in 20 Spanish hazelnut oils (26), possibly as a consequence of lower chromatographic resolution.

Values for most sterols were similar to those found in the earlier survey (6). β-Sitosterol ranged from 78.8 to 85.9% (average = 82.7%), at the upper end of the range (74.9−84.4%, average = 81.1%) reported, as were the proportions of Δ5,24-stigmastadienol (nd−1.2%).

Similar values for cholesterol, campesterol, stigmastenol, and sitosterol have been reported in four Italian hazelnut oils (27) but with slightly higher levels of Δ7-stigmastenol (1.2−1.6%), possibly unresolved from neighboring peaks as no other sterols were measured.

Levels of the later eluting minor sterols were lower than most results reported elsewhere (6). For Δ7-stigmastenol, 14 samples (50%) were below the previous minimum value of 0.2%, and for Δ7-avenasterol, 20 (71%) were below the minimum of 0.4%. Higher levels of these minor sterols (Δ5-avenasterol at 6.1− 8.5%, Δ7-avenasterol at 1.9−3.3%, and Δ7-stigmasterol at 1.1− 2.9%) were found in six New Zealand oils (28), and very high

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J. Agric. Food Chem., Vol. 53, No. 12, 2005   **4847**

Table 4. Fatty Acid Composition of Hazelnut Oils (Percent m/m)

| | unroasted | | | | | roasted | | |
sample	Croatia	France	Italy	Spain	Turkey	Croatia	Italy	Spain
no.	1	8	6	7	6	1	2	4
myristic acid, C14:0	nd[a]	nd–0.1	nd	nd–0.1	nd	nd	nd	nd
pentadecylic acid, C15:0	nd	nd	nd	nd	nd	nd	nd	nd
palmitic acid, C16:0	5.1	5.0–6.3	5.8–6.6	5.3–6.1	5.1–6.3	5.2	6.2–6.6	5.2–6.2
palmitoleic acid, C16:1	0.2	0.2–0.3	0.2–0.3	0.2–0.3	0.1–0.3	0.2	0.2–0.3	0.2
margaric acid, C17:0	nd	nd–0.1	nd–0.1	nd	nd	nd	0.1	nd
heptadecenoic acid, C17:1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	n-0.1
stearic acid, C18:0	2.7	1.6–2.9	2.4–2.8	1.9–2.5	2.0–3.1	2.7	2.4–2.6	2.1–2.5
oleic acid, C18:1	82.6	75.6–81.9	80.3–83.4	75.3–81.8	75.7–82.9	81.1	83.3–83.8	78.5–82.3
linoleic acid, C18:2	8.7	9.4 to17.1	6.2–10.6	10.1–15.9	8.1–15.4	9.7	6.5–7.2	9.8–12.0
linolenic acid, C18:3	0.2	nd–0.4	0.1–1.0	0.1–0.2	0.1–0.3	0.6	0.1	0.1–0.5
arachidic acid, C20:0	0.1	0.1	0.1–0.2	0.1	0.1	0.2	0.1	0.1
eicosenoic acid, C20:1	0.2	0.1–0.2	0.1–0.2	nd–0.2	0.1–0.2	0.2	0.1	0.1–0.2
behenic acid, C22:0	nd	nd	nd	nd–0.1	nd	nd	nd	nd
docosenoic acid, C22:1	nd	nd	nd	nd	nd	nd	nd	nd
lignoceric acid, C24:0	nd	nd	nd	nd	nd	nd	nd	nd

[a] nd, <0.1.

Table 5. Iodine Values of Hazelnut Oils

| | unroasted | | | | | roasted | | |
sample	Croatia	France	Italy	Spain	Turkey	Croatia	Italy	Spain
no.	1	8	6	7	6	1	2	4
iodine value	87.0	87.1–95.4	84.7–88.1	88.0–93.0	85.8–91.9	88.5	83.9–84.7	88.2–89.8

proportions of Δ5-avenasterol were found in a single Spanish sample (*22*).

There was no evidence of a significant effect of roasting on the desmethylsterol composition, with the range of sterols falling within the ranges of the oils from unroasted nuts except for Δ5-avenasterol, which was reduced in the roasted samples.

Steradiene Composition. No steradienes (stigmastadiene, campestadiene, stigmastatriene, and campestatriene) were detected in any hazelnut oil before or after roasting, subject to a limit of detection of 0.02 mg/kg. There were no reports of the analysis of hazelnut oils for sterol degradation products in the literature. Steradienes are formed by dehydration of sterols, notably during refining, and would not be expected to be formed in oils that had not undergone severe treatment in processing.

FA Composition. The FA composition of the hazelnut oils is given in **Table 4**. The values obtained were in very close agreement with the ranges reported in the literature (*4, 6, 18–23, 25, 27, 29–36*).

One sample, from Italian nuts, had a C18:3 value of 1.0%, which was notably higher than the other samples. The C18:0 value for another sample from French nuts (1.6%) was lower than for any other sample, but higher than a single oil of 17 Spanish oils, which contained 1.4% (*18*). The C18:0 value for one sample from Turkish nuts was high (3.1%) but within the range reported elsewhere (*18*).

Some workers have reported higher values of C20:0 and C20:1 than reported here (*34, 37*). In oil from 22 Spanish (Catalonia) nuts of six varieties C20:0 was found at 0.2−0.3%, twice the level reported here (*38*). Various other workers have published FA values similar to those reported here but with the proportions of some acids at slight variance with our results. Higher proportions of C18:3 were measured in eight Italian oils (*39*). Lower C18:0 and C18:1 and higher C18:2 and C18:3 levels were measured in 17 varieties in the United States (*40*), and lower C18:0 levels were measured in 6 oils from New Zealand (*28*).

Results for the oils from Turkish hazelnuts were in general agreement with other results for 10 oils from that country (*41*). Lower levels of C16:0 (4.6−5.3%) were reported from the analysis of the oils from 10 German varieties using a packed-column GC (*42*).

No marked changes were observed in the FA composition of oils from roasted nuts.

Iodine Value. The mean calculated iodine values of hazelnut oils are given in **Table 5**.

Three samples, two from French nuts and one from Spanish nuts, had iodine values slightly above the maximum of 92.8 reported earlier (*6*) at 93.1, 95.4, and 93.0, respectively; however, the spread of the results was comparable. Slightly lower values have been reported for eight Italian cultivars (*39*). The iodine values of oils from roasted nuts fell within the range of the oils from unroasted nuts.

FA in the Triacylglycerol 2-Position Composition. The triacylglycerol 2-position fatty acid composition of hazelnut oils is given in **Table 6**.

One Italian oil had very low C16:0 (0.2%) and very high C18:3 (1.0%). The high value for C18:3 was mirrored by that of the total FA composition result, but that for C16:0 was not.

For the oils from roasted nuts the values fell within those of the unroasted samples with the notable exception of the Italian oil described above, which surprisingly had very high C16:0 (4.3%) and 18:0 (1.9%) and very low C18:2 (7.7%).

TCN. The TCN composition of the hazelnut oils is given in **Table 7**.

The results were similar to those reported elsewhere (*6, 19*) except that for C56 was detected in a greater proportion of the samples, and two samples from Italian nuts and one each from Spanish and Turkish nuts had higher levels of C50.

There are few other reported investigations into the triacylglycerol profiles of hazelnut oils, although measurement of trilinolein is of relevance in detecting the presence of seed oils in olive oil. Published papers (*24, 43–45*) have reported the

Table 6. Triacylglycerol 2-Position Fatty Acid Composition of Hazelnut Oils (Percent m/m)

	unroasted					roasted		
sample	Croatia	France	Italy	Spain	Turkey	Croatia	Italy	Spain
no.	1	8	6	7	6	1	2	3
myristic acid, C14:0	nd[a]	nd	nd	nd	nd	nd	nd	nd
palmitic acid, C16:0	0.7	0.2–0.8	nd–0.3	0.3–0.7	0.3–1.3	0.4	0.3–0.4	0.3–0.5
palmitoleic acid, C16:1	0.2	0.1–0.2	0.1–0.2	0.1–0.2	0.1	0.1	0.1–0.2	0.1
margaric acid, C17:0	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
heptadecenoic acid, C17:1	nd	nd	nd	nd	nd	nd	nd	nd
stearic acid, C18:0	0.3	0.1–0.4	0.1–0.2	0.1–0.3	0.2–0.7	0.2	0.2–1.9	0.2–0.3
oleic acid, C18:1	86.1	75.6–85.3	84.3–89.6	76.5–84.5	78.7–88.2	84.3	85.8–88.4	80.2–84.0
linoleic acid, C18:2	12.5	13.9–23.6	9.0–14.9	14.5–22.1	11.0–20.0	14.4	7.7–10.7	15.3–19.0
linolenic acid, C18:3	0.1	0.1–0.3	0.1–1.0	0.1–0.2	0.1–0.2	0.6	nd–0.1	0.1–0.4
arachidic acid, C20:0	nd	nd	nd	nd	nd	nd	nd	nd
eicosenoic acid, C20:1	nd	nd	nd	nd	nd	nd	nd–0.1	nd
others	nd	nd–0.4	nd–0.3	nd–0.1	nd	0.1	nd–0.4	nd–0.4

[a] nd, <0.1.

Table 7. Triacylglycerol Carbon Number (TCN) Composition of Hazelnut Oils (Percent m/m)

	unroasted					roasted		
sample	Croatia	France	Italy	Spain	Turkey	Croatia	Italy	Spain
no.	1	8	6	7	6	1	2	3
TCN 48	nd[a]	nd	nd–0.1	nd	nd	nd	nd–0.1	nd–0.3
TCN 50	0.8	0.4–1.1	0.8–1.8	0.7–1.6	0.7–1.8	0.9	0.9–1.0	0.9–1.1
TCN 52	14.9	13.8–18.5	15.5–19.4	14.5–16.9	14.4–17.5	17.3	17.5–18.8	15.7–17.5
TCN 54	81.7	78.9–83.8	77.0–81.3	79.9–82.9	79.4–83.3	80.8	79.2–80.4	78.9–82.1
TCN 56	1.2	nd–0.9	nd–1.4	nd–1.0	nd–0.5	1	1–1.1	1.0–1.81
TCN 58	0.1	nd–0.1	nd	nd	nd	nd	nd–0.4	nd–0.4
TCN 60	nd	nd	nd	nd	nd	nd	nd	nd

[a] nd, <0.1.

Table 8. Tocopherol and Tocotrienol Composition of Hazelnut Oils (Milligrams per Kilogram)

	unroasted					roasted		
sample	Croatia	France	Italy	Spain	Turkey	Croatia	Italy	Spain
no.	1	8	6	7	6	1	2	3
α-tocopherol	340	343–545	118–440	314–590	319–492	340	346–362	368–548
β-tocopherol	nd[a]	nd–10	nd–10	nd–15	nd–10	77	nd–11	nd–12
γ-tocopherol	65	nd–10	nd–10	nd–42	nd–130	nd	nd–21	nd–52
Δ-tocopherol	nd	nd–10	nd–10	nd–10	nd	10	nd	nd
α-tocotrienol	nd	nd	nd–209	nd	nd	nd	nd	nd
β-tocotrienol	nd	nd	nd	nd-22	nd	nd	nd	nd
γ-tocotrienol	nd	nd	nd	nd-34	nd	nd	nd	nd
Δ-tocotrienol	nd	nd	nd	nd	nd	nd	nd	nd
total tocols	405	343–545	354–~500	314–613	351–554	427	367	372–610

[a] nd, <10.

triacylglycerol profiles as separated by triacylglycerol rather than TCN, and the coelution in the published profiles of some triacylglycerols with and without C16:0 acid means that conversion of published data to TCN values was not possible.

The C54 proportions measured appeared to decrease in all but one sample on roasting, but these changes were not statistically significant when compared with the standard deviation of results for the reference oil.

Tocopherol Composition. The tocopherol composition of hazelnut oils is given in **Table 8**.

The major tocopherol present in all samples was α-tocopherol, the range of values for which was large, with one Italian sample having a relatively very low content (118 mg/kg). A broadly similar range (211−655 mg/kg) has been reported elsewhere for 17 hazelnut oils, and this also contained a similarly atypically low level (94 mg/kg) in one oil (*18*).

Levels of α-tocopherol were in good agreement with results published elsewhere (*19, 23, 41, 46−49*). Oils from six nuts grown in New Zealand were shown to have a wider range of α-tocopherol and contained β-, γ-, and Δ-tocopherols (*28*). Slightly lower levels of α-tocopherol were present in three samples grown in the United States, with a much lower level in a sample from the wild species *C. cornuta* (*35*). In fact, several workers have reported higher incidences of β- and γ-tocopherol than measured in this study (*23, 40, 46, 50, 51*). The possibility of using β- to γ-tocopherol ratios to detect hazelnut oil addition to olive oil has been suggested (*52*).

The wide range of γ-tocopherol, absent in several samples but present at up to 130 mg/kg, and the low incidence of samples with Δ-tocopherol or any of the tocotrienols to some extent reflected published values.

Tocotrienols were detected in two samples. A sample from Italian nuts contained α-tocotrienol as its principal tocol at 209 mg/kg, and one from Spanish nuts contained β- and γ-tocotrienols at 22 and 34 mg/kg, respectively. No tocotrienols
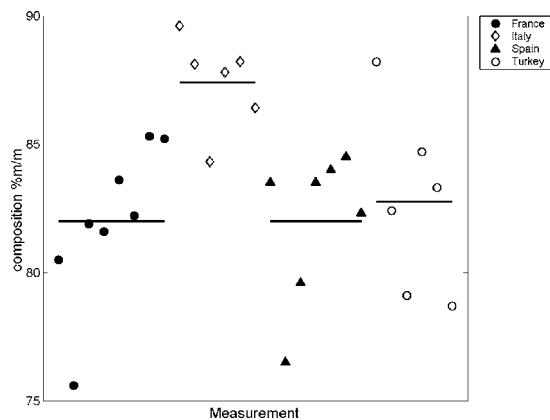
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Figure 1. ANOVA of values for C18:1 fatty acids.

Figure 2. ANOVA of values for 2-position C18:2.

were reported in a limited number of samples studied earlier (*6*, *46*).

Erratic changes were found in the levels of α- and γ-tocopherols when hazelnuts were roasted. There was also an increase in measured β-tocopherol in three of the six roasted samples. However, the significance of these changes is difficult to judge. Loss of vitamin E, mostly α-tocopherol, have been reported to occur with increasing roasting temperature over the range 180−300 °C (*53*).

Country of Origin. The composition of the Turkish samples agreed with published results for 10 varieties measured in terms of FA profiles but was lower in tocopherol content (*41*). The Spanish samples were lower in C18:0 and C20:0 FA than were two sets of nine cultivars analyzed earlier (*27*). Slightly higher proportions of β-sitosterol and Δ7-stigmasterol were present in one of the earlier sets; however, those values were probably enhanced by the small number of sterols (five) measured and by coelution of some sterols.

The Italian samples agreed well in FA composition with results for three samples analyzed in 1979 (*54*); again, peak coelution was probably responsible for higher β-sitosterol levels reported in the earlier work.

Although the number of samples analyzed was greater than that in most previous work, it remained limited in terms of the application of statistical treatment. However, the data were examined using the CSL-Metabolab visualization toolbox, which allowed multivariate and classical statistical analyses and rapid integration and spectral visualization.

In most cases the values within groups based on country of origin appeared to be normally distributed. However, there were certain cases when standard deviations differed between the countries, which violates the assumptions of ANOVA and *t* tests. Such tests can still provide useful indicators of countries of origin where there are strong differences between countries, but the *p* values do not have their usual statistical interpretations regarding significance levels.

The main focus was on the search for cases where there was some separation of ranges of results by country. ANOVA showed that most samples from Italy were separated from most samples from the other countries using data for the C18:1 FA (**Figure 1**), the 2-position FA (e.g., C18:2, **Figure 2**), and the triacylglycerol carbon number 56 (**Figure 3**). The latter data showed separation of Italian from Turkish oils but was less successful with the other countries.

Univariate statistics were performed using an ANOVA on all data looking for cases where at least one country had a significant difference and using pairwise *t* tests for all pairs of countries and all data looking for differences between individual
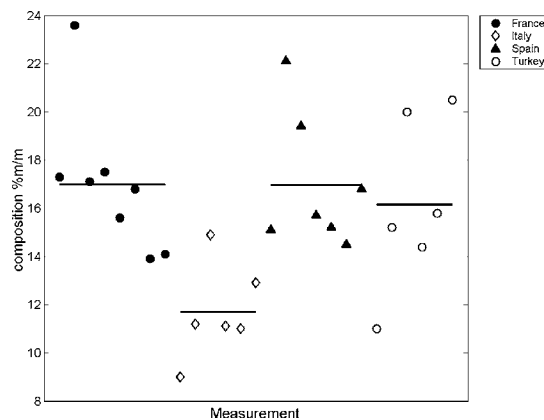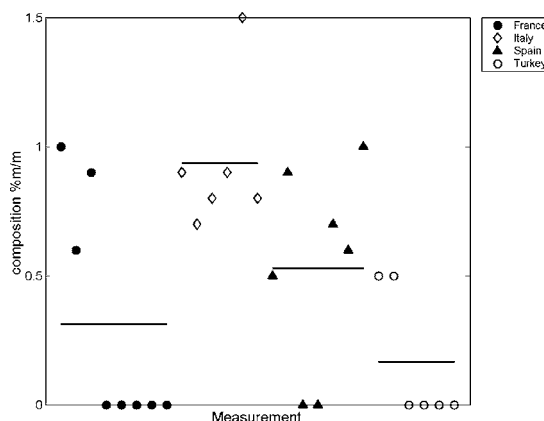

Figure 3. ANOVA of values for triacylglycerol carbon no. 56.

pairs of countries. The most significant comparisons (>99%) based on ANOVA *p* values were for 2-position FA, tocopherols, and CN 56. Although there were overlapping data points, most of the oils from Turkish nuts could be separated.

A PCA was performed using all variables. The data were both mean centered and scaled to unit variance, and 10 PC scores were calculated. There was reasonable but imperfect separation between oils from Italian nuts and those from Spain and France in PC scores 3 and 5.

Factors Affecting the Composition of Hazelnut Oil. The composition of vegetable oils, including hazelnut oil, is affected by variety, geographical location, climatic effects, and treatment, for example, addition of fertilizer, during growth as described below. The samples for this study were collected from sources used for commercial production, and taking into account the environment and treatments that the growing trees were exposed to was beyond the scope of the work.

The varietal effects on hazelnut oil composition have been studied with a view to providing commerce with nuts and oils rich in antioxidant components giving stability and health benefits (*41*). Significant differences in the tocopherol and sterol composition of oil from Spanish varieties of hazelnut grown in the United States have been reported (*18*). Higher levels of α-tocopherol were found in a New Zealand cultivar than in three U.S. and two European cultivars grown in New Zealand (*28*), but there were no significant differences in the FA or sterols. No significant differences was found in the α-tocopherol contents of 11 samples from 8 varieties of nuts grown in four regions of Turkey (*49*). It has been concluded (*33*) from measurements of single samples of 12 cultivars and varieties that they could be distinguished by virtue of their C18:2 acid levels, which varied to a greater extent (6.7−13.6%) than did the other acids.

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Linoleic acid levels are increased by irrigation and have been shown to vary with hazelnut variety and to different degrees within single varieties (*38*). Levels of C18:1 and C18:2 in particular differed between varieties of hazelnut originating in Spain and between these and an Italian variety grown in Spain (*44*). Similar differences were found between Spanish varieties grown in the United States (*18*).

Changes in the oil composition as the nut develops have been documented (*45, 52, 55, 56*). Levels of α-tocopherol increase with kernel development with a concomitant decrease in the levels of β- and Δ-tocopherols, whereas γ-tocopherol levels increase during the early stages of kernel development and then decrease. FA composition changes radically during kernel development, with C14:0, C16:0, C18:0, and C18:3 decreasing and C18:1 and C18:2 increasing over 3 months (*52*). Treatments such as irrigation and the application of fertilizer also affect the FA composition (*36*). The FA and tocopherol composition of hazelnut oil changes considerably with season and with stage of growth (*55*); however, as hazelnuts are harvested after falling from the tree, the samples in this survey may be assumed to be of similar maturity and to have been collected in the same season.

In a comparison of four varieties of hazelnut grown in two regions and sampled annually over three years (*23*) it was found that there were significant differences between years for almost all of the FA and significant differences between locations for C18:0, C18:1, and C18:2. The α-tocopherol content changed with harvest year but not variety (*46*). The triacylglycerol content of the same samples did not change with variety (*43*).

Handling of the nuts and oils can be assumed to affect their composition. The times between harvest of the nuts and their collection by analysts are in most cases not available. However, following heavy harvests in regions of high productivity such as Turkey, hazelnuts could be stored for up to a year before use. There are few published reports of changes of hazelnut oil composition on storage of nuts (*4, 28, 30, 37−38, 53, 58−59*); however, it has been reported (*57*) that tocopherols had good stability in vegetable oils at 4 °C, although other authors found α-tocopherol losses of ∼50% on storage for 36 weeks at 25 °C (*35*). Significant decreases (∼10%) have been found in the levels of C18:3 in hazelnuts stored for 12 months (*58*). Lesser decreases were found for C16:0 and C18:0 and increases (∼2%) in the levels of C18:2. The changes were similar regardless of the storage temperature (3−6 °C or 18−25 °C) and were slightly retarded by the use of a nitrogen atmosphere.

Awareness of the effects of factors such as the levels of pro- and antioxidant compounds and the saturated/unsaturated fatty acid ratios on the stability of harvested commercial hazelnuts has led to the breeding and testing of new varieties (*41*). Thus, it is most likely that the composition of hazelnut oils in commercial production will change as varieties having beneficial characteristics are introduced.

It is apparent from reviews of the literature that some changes in the pattern of lipid profiles were likely to be due to factors associated with the equipment and procedures used in different laboratories. Various approaches have been used for sampling, with sizes ranging from single kernels to kilograms. Samples have frequently been obtained from experimental plots, which do not necessarily represent commercial systems. Within the laboratory many techniques have been applied to the extraction and analysis of nut oils, ranging from pressing to extraction with organic solvents or supercritical fluids. Advances in separation technology have had a dramatic effect, allowing the resolution of chromatographic peaks previously reported as a single component into two or more constituents. These advances are particularly relevant in the determination of sterol and FA composition.

In this study attempts were made to follow the processing used in industry up to but not including refining steps such as washing, neutralization, winterization bleaching, and deodorization, some of which have a strong effect on the oil composition.

This paper adds to and updates the available compositional tables for hazelnut oils. The analyses have been performed using modern equipment and validated methods incorporating stringent quality control measures based on the use of fully characterized reference materials. The geographical origin of the samples has been confirmed by collection from the harvest site and the authenticity of the nuts and oil by the use of an audit trail. Industry conditions for the oil production were followed as closely as possible. The data obtained by these means will be the most relevant so far available for authenticity of traded hazelnut oils.

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