

Does Frequent Replenishment with Fresh Monoenoic Oils Permit the Frying of Potatoes Indefinitely?

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The frying life of two monoenoic oils, extra virgin olive oil (EVOO) and high oleic acid sunflower oil (HOSO), used to fry potatoes following a domestic frying model, by replenishing the oil in the fryer with fresh oil after each use to maintain the oil volume/fresh potato ratio, was studied by measuring polar content (PC) and triglyceride oligomer (TO) content. A combination of column and high-performance size-exclusion chromatography was used. Changes in the PC and TO content of both oils according to the number of frying uses were adjusted to linear, logarithmic, and power equations. While all three equations reflected the alterations quite accurately ($p < 0.001$), the logarithmic and power equations defined them best. Frequent replenishment of both frying oils with fresh oil produced a stabilization of PC and TO levels after 20–40 uses. According to the linear adjustment equation, 321 ± 33 frying operations with EVOO and 290 ± 40 with HOSO would be needed to raise PC to 25%. To reach the 10% TO level set for discarding oil used in frying, EVOO and HOSO would have to be used 319 ± 35 and 241 ± 37 times, respectively. Using the power adjustment, however, the theoretical frying life of both oils would be much higher (at least 4460 frying uses before reaching a 25% PC and 538–1552 uses for the EVOO and 249–1319 uses for the HOSO to reach a 10% TO level). The frying life of EVOO and HOSO would be extended almost indefinitely (at least 170 000 uses) when the logarithmic adjustment was applied. These data suggest that frequent replenishment of used monoenoic oil with fresh oil permits one to fry sets of fresh potatoes a very high number of occasions.

Keywords: Column chromatography; deep frying; frying life; extra virgin olive oil; high oleic acid sunflower oil; HPSEC; polar content; triglyceride oligomers

INTRODUCTION

French-fried potatoes are very popular among North Americans and inhabitants of the European Union countries. During deep-fat frying, oils undergo oxidation, polymerization, hydrolysis, cyclization, and isomerization (Dobarganes and Márquez-Ruiz, 1993; Romero, 1998; Romero et al., 1998). Moreover, during frying, dehydration of the potato and penetration of fat into the food is known to occur (Garrido-Polonio et al., 1994; Romero, 1998; Varela, 1988). Thus, French fries obtained by frying in an abused oil may contain significantly high levels of thermoxidized and polymerized products that could be undesirable from a nutritional point of view (Cuesta et al., 1988; Friedman, 1991; López-Varela et al., 1995; Márquez-Ruiz et al., 1996). The choice of oil to use in frying is difficult as factors such as stability, price, and nutritive value must be considered. Oils with a high saturated fatty acid content present unique stability in frying applications (Arroyo et al., 1996). However, these oils are least desirable from a nutritional and human health standpoint (Cuesta et al., 1988; Frankel et al., 1991; Oubiña Rumeu, 1998; Sebedio et al., 1988).

Throughout recent decades in Spain, the use of the traditional olive oil has sharply declined in favor of that of other oils for frying purposes and snack food preparation (Moreiras et al., 1990; Romero 1998). However, the debate on deep-fat frying is primarily focused on the point at which any oil used for frying should be discarded (Firestone, 1996) and on whether frequent replenishment of used oil with fresh oil lengthens or shortens its frying life (Romero et al., 1998; Sánchez-Muniz et al., 1993). Several authors (Cuesta et al., 1993; Friedman, 1991; and Romero et al., 1998) report that the profile of used frying oil can be improved or at least maintained by replenishing it with fresh oil, as a certain blend of unaltered and altered oil in the fryer offers optimum cooking properties. However, other authors (Quaglia, 1996) indicate that mixing fresh with used oils accelerates alteration of the oils through a catalytic process.

Although a number of European countries have passed specific laws and regulations concerning culinary oil use in frying, none are yet in effect in the United States (Firestone, 1996). Most European countries assess used frying oil according to its polar content (PC). Belgium, the Czech Republic, France, Hungary, Italy, the Scandinavian countries, and Spain have set a maximum PC level of 25%, while other countries, including Austria and Germany, have established a PC cut point of between 20% and 27%. Moreover, among European countries TO content (triglyceride dimers and

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Table 1. Major Fatty Acid Composition (%) of Extra Virgin Olive Oil (EVOO) and High Oleic Sunflower Oil (HOSO)

fatty acids	EVOO ^a	HOSO ^a
16:0	9.91 ± 0.07	4.38 ± 0.08
18:0	3.73 ± 0.07	4.10 ± 0.02
18:1	80.60 ± 0.26	78.45 ± 0.10
18:2	4.59 ± 0.12	11.00 ± 0.04
18:3	0.56 ± 0.12	<0.1
20:0	0.31 ± 0.01	0.34 ± 0.00
20:1	0.20 ± 0.02	0.23 ± 0.01
22:0	nd	1.06 ± 0.03
24:0	<0.1	0.37 ± 0.02

^a Average of fourth samples ± SD. Nd: Not detected.

polymers) is another criterion used for evaluating used frying oil. Belgium and the Czech Republic have legislated a 10% maximum TO content, while The Netherlands permits up to 16% of these compounds in oil used for frying.

The goal of this study was to evaluate the frying life of extra virgin olive oil (EVOO) and high oleic acid sunflower oil (HOSO), used in discontinuous deep-fat frying of potatoes and frequently replenished with fresh oil, by measuring their polar and triglyceride oligomer contents. The number of uses needed to reach the critical PC level of 25%, and a TO content of 10% or 16%, was also considered.

MATERIALS AND METHODS

Materials. Samples. EVOO (Patrimonio Comunal Olivarero, Mora, Toledo, Spain), refined HOSO (VIPA, Andújar, Jaén, Spain), and fresh potatoes (Kennebec variety, Xinzo de Limia, Galicia, Spain) were purchased at a local store. Fatty acid composition of the unused oils is given in Table 1.

Methods. Frying Performance. Frying variables and conditions including the material and capacity of the fryers, oil temperature, number of frying times, total frying time, oil replenishment rate, food type, food/oil ratio, and total amounts of food fried are shown in Table 2. Fifteen batches of five frying operations were conducted, maintaining the oil volume constant by adding fresh oil after each frying. Ten frying operations were performed every day, five in the morning and five in the afternoon, leaving the oil to reach room temperature between batches (Figure 1). More extensive details of the frying method have been described previously (Romero et al., 1995b). Replenishment of oil lost throughout the 75 frying operations implied the addition of a total of 4.25 L of fresh HOSO and 4 L of fresh EVOO. Average temperature changes in the fryer bath during a frying day are shown in Figure 1.

Polar Content. Total PC of the oil was determined by silica column chromatography (Waltking and Wessels, 1981) with the following modification: an accurately weighed sample of 1 ± 0.01 g of each oil was dissolved in 20 mL of hexane/diethyl ether, using 87:13 v/v when fresh oil was analyzed and 90:10 v/v when the oil tested had been used for frying. The same proportion of hexane/diethyl ether was used to fill the column and to elute the nonpolar fraction, to obtain a sharper separation (Dobarganes et al., 1984). Two samples each of fresh EVOO and HOSO and of both oils from the first, fourth, eighth, twelfth, sixteenth, twentieth, thirtieth, fortieth, fiftieth, sixtieth, seventieth, and seventy-fifth uses were analyzed. Separation of nonpolar and polar fractions was checked by thin-layer chromatography on 0.5 mm thick 60 F 250 silica gel plates (20 × 20 cm glass). Polar and nonpolar fractions were diluted 50-fold (w/v) in hexane/diethyl ether (80:20 v/v). Samples were applied as 20 μL spots with a 705 Hamilton microsyringe. Plates were developed with hexane/diethyl ether/acetic acid (80:20:1 v/v/v) in a lined tank for ca. 25 min (ca. 17 cm) and then removed to let the solvent evaporate. Spots were visualized by exposure to iodine vapors.

High-Performance Size-Exclusion Chromatography (HPSEC). To obtain further information about hydrolytic and/or thermoxidative changes which occur during frying, polar fractions previously obtained by column chromatography were analyzed by HPSEC following the method of Dobarganes et al. (1988). Isolated polar fractions were analyzed in a Waters 501 chromatograph (Milford, MA) with a 20 μL sample loop. A Waters 410 refractive index detector and two 300 mm × 7.5 mm i.d. (5 μm particle size) pL gel (polystyrene-divinylbenzene) columns (Hewlett-Packard) with 0.01 and 0.05 μm pore size were connected in series and operated at 40 °C. High-performance liquid chromatography (HPLC) grade tetrahydrofuran served as the mobile phase with a flow rate of 1 mL/min. Sample concentration was 15–20 mg/mL in tetrahydrofuran. All eluents, as well as samples, were passed through a 2 μm filter. Two determinations of each polar fraction of fresh EVOO and HOSO and of the same oils from the first, fourth, eighth, twelfth, sixteenth, twentieth, thirtieth, fortieth, fiftieth, sixtieth, seventieth, and seventy-fifth frying uses were performed. The quantity of each polar compound was calculated as has been described previously (Cuesta et al., 1993). The TO level was the result of the triglyceride dimers and polymers quantified in the oils.

Statistical Methods. Changes in PC and TO content were adjusted to linear ($Y = a + bX$), logarithmic ($Y = a + b \ln X$), and power ($Y = aX^b$) equations by analysis of variance (ANOVA), where $Y =$ variable (PC or TO expressed in mg/100 mg of oil) and $X =$ number of frying uses. Linear, logarithmic, and power adjustment equations of EVOO and HOSO were compared by analysis of covariance (ANCOVA), taking into account their intercepts and slopes. The theoretical number of frying operations possible before having to discard the oil was obtained using the three equations.

RESULTS

Data on PC (mg/100 mg of oil) and alteration compounds of the polar fraction (mg/100 mg of polar fraction) of fresh EVOO and HOSO and of oils from the 75th frying operation are presented in Table 3. The percentages (mg/100 mg of polar fraction) of triglyceride polymers and oxidized triglycerides were higher in HOSO than EVOO. Table 4 shows the statistical comparison of the linear, logarithmic, and power adjustments of changes in PC and TO contents of the oils according to the number of uses. ANCOVA of the regression adjustments of PC and TO displays significant differences (at least $p < 0.05$) between EVOO and HOSO (Table 4). The graphic representations of the linear and logarithmic adjustments are displayed in Figures 2–4.

The frying life of the oils studied, calculated by taking into account the trends of PC and TO content and the different critical levels established for discarding cooking oil, is presented in Table 5 and Figures 2–4. The three adjustment equations considered suggest that HOSO displays a shorter frying life than EVOO when PC or TO levels are taken into account.

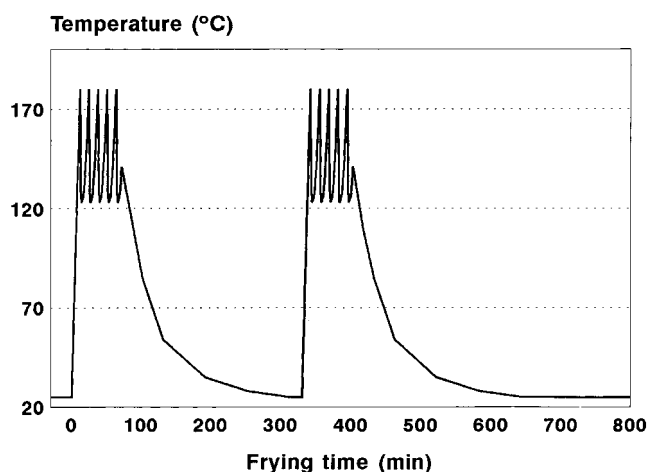
DISCUSSION

The two fresh oils (EVOO and HOSO) used in the study presented similar total PC levels (2.9–3.6 mg/100 mg of oil). These figures reflect the good quality of the oils, as PC of unused oils normally ranges between 0.4 and 6.4 mg/100 mg of oil (Lumley, 1988). Current study data are similar to other findings reported by our group (Arroyo et al., 1992; Cuesta et al., 1993; Garrido-Polonio et al., 1994) and by Dobarganes et al. (1993) and Pozo-Diez et al. (1995) in olive oil and HOSO. Discontinuous frying increased the polar content of the oils during the

Table 2. Frying Operation Variables

frying variables	oils	
	extra virgin olive oil ^a	high oleic acid sunflower oil ^b
frying vessel	commercial electrical fryer with stainless steel vessel	commercial electrical fryer with stainless steel vessel
oil quantity (L/batch)	3	3
proportion of food to frying oil (g/L/batch)	500/3	500/3
food type	potatoes ^c	potatoes ^c
tot. oil replenishment (L)	4	4.25
temp (°C)	180	180
tot. frying time (h)	18	18
no. of frying times	15 discontinuous batches of five frying operation	15 discontinuous batches of five frying operation
tot. amt of food fried (kg)	37.5	37.5

^a From Patrimonio Comunal Olivarero (Mora, Toledo, Spain). ^b VIPA from the KOIPE Co. (Andújar, Jaén, Spain). ^c Kennebec variety (Xinzo de Limia, Galicia, Spain).

**Figure 1.** Average temperature changes during a frying day.

first 20–40 uses. Nevertheless, frequent replenishment stabilized subsequent total polar content in both oils. Despite different frying methods (null or frequent oil replenishment), Sánchez-Muniz et al. (1993) observed no differences in total PC up to the 30th operation in sunflower oil used for frying potatoes. Nevertheless, PC content was higher after the 50th use when oil was not replenished (24.1%) than when it was replenished frequently (18.9%). Present data show that PC increase more rapidly in HOSO than in EVOO during the initial frying operations (20–30) but eventually stabilized at a level ≈ 1.3 mg/100 mg of oil higher in HOSO than in EVOO. After 75 semi-discontinuous uses in frying fresh potatoes, the PC of both oils tested was much lower than the 25% level established by some countries as a cut point after which oils can no longer be used for frying (Firestone, 1996). This can be explained by much higher oil turnover rate obtained when frying potatoes in series of five frying operations compared to 75 discontinuous uses which would imply 75 heating–cooling periods, instead of only 15.

Total PC levels of both oils showed a very significant adjustment ($p < 0.001$) to linear, logarithmic, and power equations. However, logarithmic and power adjustments were higher than linear ones, due to the tendency of the PC to stabilize after a certain number of uses. Cuesta et al. (1993) found that changes in PC were highly adjusted to a cubic equation. Romero et al. (1995a,b) found that a second degree equation accurately and significantly reflected ($r > 0.99$; $p < 0.001$) the adjustment of the PC levels which varied according to the number of frying operations. However, polynomial

equations cannot explain what happens beyond the last frying operation. Although the future cannot be predicted, to compare the long-term behavior in frying of both oils we have made an approach extrapolating data to the adjustment equations obtained. According to the linear equation, and following the same experimental conditions, it can be suggested that 321 ± 33 uses of EVOO and 290 ± 40 of HOSO would be needed to raise the PC level to 25%. Logarithmic and power adjustments, which defined changes in PC even more accurately, indicate that an immensely greater number of uses appears possible before reaching the critical 25% level. The logarithmic adjustment equation suggests that, under the same experimental conditions, the addition of fresh oil after each frying operation, to keep the oil/food ratio at 3 L/500 g, would extend the frying life of both monoenoic oils almost indefinitely. Various factors may account for this finding, e.g. the addition of fresh oil after each use dilutes the concentration of alteration compounds formed during frying, while the addition of unaltered fatty acids and antioxidant compounds from the fresh oil helps to maintain the initial composition of the frying oil. In fact, the oil in the frying vat was completely replenished (3 L) before the 60th frying, thus explaining the tendency for PC to reach a near-steady state. The slight but significantly greater stability of EVOO, as compared to HOSO, seems mainly to be a consequence of the lower linoleic acid content of EVOO (4.5% vs 11.0% of total fatty acids in EVOO and HOSO, respectively). Fresh HOSO contains higher α -tocopherol levels than EVOO, while EVOO contains some polyphenols that are not present in HOSO (Sánchez-Muniz et al., 1998).

The polar compound distribution, determined by HPSEC, allowed differentiation of the main groups of compounds present and establishment of the relative influence of the three degradation pathways predominant in frying: hydrolysis, polymerization, and oxidation (Arroyo et al., 1992; Romero et al., 1995a,b; Romero et al., 1998; Sebedio et al., 1990). Despite the similar PC content level in the unused oils, distribution of the different polar compounds varied in these oils. The major compounds in the polar fraction of unused EVOO and HOSO were diglycerides. This finding has been reported in olive oils (Pérez-Camino et al., 1991; Pozo-Diez et al., 1995) and in some high oleic sunflower oils (Dobarganes et al., 1993; Pozo-Diez et al., 1995). Triglyceride polymers were only found in HOSO. This should be attributed to the refining process undergone by HOSO, which involves treating the oil at high

Table 3. Polar Content (mg/100 mg of Oil) and Altered Compound Composition (mg/100 mg of Polar Fraction) in Unused Extra Virgin Olive Oil (EVOO) and High Oleic Sunflower Oil (HOSO) and in Both Oils after Being Used in 75 Repeated Fryings of Potatoes^a

	EVOO		HOSO	
	unused	75th frying	unused	75th frying
polar content	2.76 ± 0.01	8.01 ± 0.00	3.60 ± 0.13	9.25 ± 0.05
triglyceride oligomers	0.00 ± 0.00	5.25 ± 0.14	0.79 ± 0.05	7.82 ± 0.01
triglyceride dimers	1.05 ± 0.20	26.60 ± 0.58	4.90 ± 0.01	28.60 ± 0.80
oxidized triglycerides	20.20 ± 0.63	37.40 ± 0.59	30.60 ± 0.34	38.90 ± 0.49
diglycerides	60.70 ± 0.03	23.50 ± 0.86	52.20 ± 0.01	20.20 ± 0.29
monoglycerides	0.00 ± 0.00	0.17 ± 0.04	0.00 ± 0.00	0.13 ± 0.04
free fatty acids	18.10 ± 0.86	7.10 ± 0.13	11.50 ± 0.15	4.37 ± 0.08

^a Results are the mean ± SD of 4 determinations.

Table 4. Linear, Logarithmic, and Power Adjustments of Polar Content Change and Triglyceride Oligomer Change (Both in mg/100 mg of Oil) in Extra Virgin Olive Oil (EVOO) and in High Oleic Acid Sunflower Oil (HOSO) and the Number of Frying Operations^a

		<i>r</i>	<i>a</i>	<i>b</i>	<i>P</i>	EVOO vs HOSO
Polar Content						
linear adjustment $Y = a + bX$	EVOO	0.9490	3.686 ± 0.270	0.067 ± 0.006	*	
	HOSO	0.9047	5.155 ± 0.377	0.069 ± 0.009	*	+++
logarithmic adjustment $Y = a + b \ln X$	EVOO	0.9571	2.133 ± 0.394	1.286 ± 0.123	*	
	HOSO	0.9781	3.506 ± 0.291	1.351 ± 0.091	*	+
power adjustment $Y = aX^b$	EVOO	0.9784	2.760 ± 0.052	0.244 ± 0.016	*	
	HOSO	0.9842	3.974 ± 0.037	0.204 ± 0.012	*	+++
Triglyceride Oligomers						
linear adjustment $Y = a + bX$	EVOO	0.9519	0.552 ± 0.117	0.030 ± 0.003	*	
	HOSO	0.8983	1.033 ± 0.219	0.038 ± 0.005	*	+++
logarithmic adjustment $Y = a + b \ln X$	EVOO	0.9664	-0.124 ± 0.155	0.574 ± 0.048	*	
	HOSO	0.9814	0.078 ± 0.151	0.764 ± 0.047	*	+++
power adjustment $Y = aX^b$	EVOO	0.9958	0.278 ± 0.049	0.529 ± 0.015	*	
	HOSO	0.9804	0.485 ± 0.099	0.484 ± 0.031	*	+++

^a *r*: regression coefficient. *a*: intercept ± SE. *b*: slope ± SE. *Y*: polar content or triglyceride oligomer content. *X*: number of frying times. *P*: significant adjustment to a linear equation (asterisk indicates $p < 0.001$, ANOVA test). Significant differences between EVOO and HOSO adjustment equations: +, $P < 0.05$; +++, $P < 0.001$ (ANCOVA test).

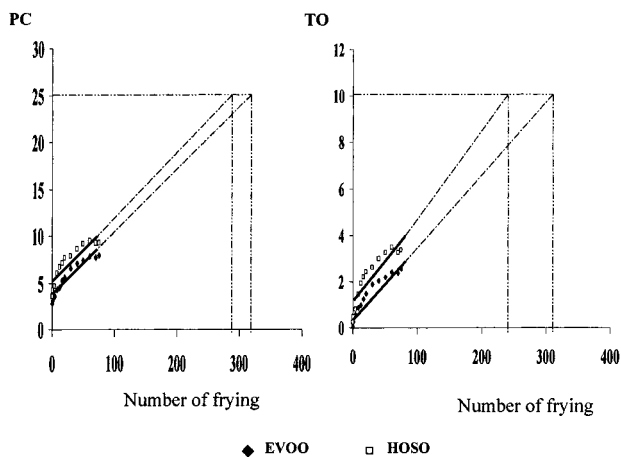


Figure 2. Linear adjustment ($Y = a + bX$) of polar content (PC) (mg/100 mg of oil) and triglyceride oligomer (TO) content (mg/100 mg of oil) vs number of frying times. X = number of frying times, Y = PC or TO content, a = intercept, b = slope, EVOO = extra virgin olive oil, and HOSO = high oleic acid sunflower oil.

temperatures (≈ 200 °C) (Dobarganes et al., 1993). After 75 uses, however, dimers and polymers and oxidized triacylglycerides were the major compounds found in EVOO and HOSO. Comparison of the oils offers interesting results: TO formation was slightly lower than that of oxidized triglycerides in EVOO, while, in HOSO, TO content was higher than that of oxidized triglycerides, suggesting that HOSO is more susceptible to polymerization during frying than EVOO. In a previous study (Sánchez Muniz et al., 1993), sunflower oil used to fry potatoes 50 times presented a higher TO

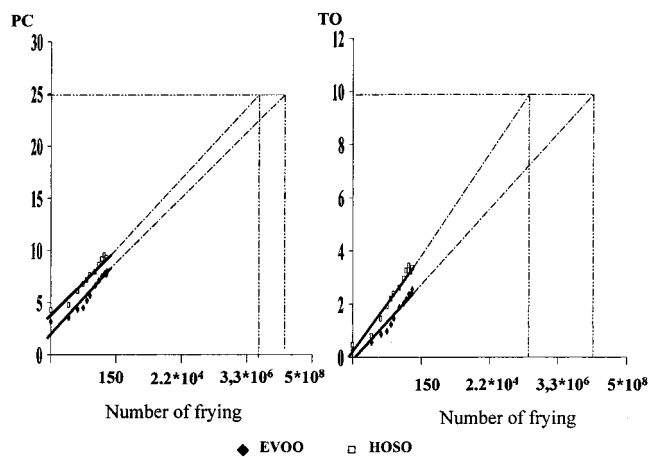


Figure 3. Logarithmic adjustment ($Y = a + \ln X$) of polar content (PC) (mg/100 mg of oil) and triglyceride oligomer (TO) content (mg/100 mg of oil) vs number of frying times. X = number of frying times, Y = PC or TO content, a = intercept, b = slope, EVOO = extra virgin olive oil, and HOSO = high oleic acid sunflower oil.

content when no fresh oil was added to the used frying oil than when the cooking oil was frequently replenished.

The critical level of 10% in TO content was not surpassed in any oil used in the present study. Following the current experimental method and extrapolating results to the linear adjustment, one finds that 319 ± 35 frying operations with EVOO and 241 ± 37 with HOSO may be performed before the 10% level is reached. One interesting finding is the similar number of uses theoretically needed to reach the critical level

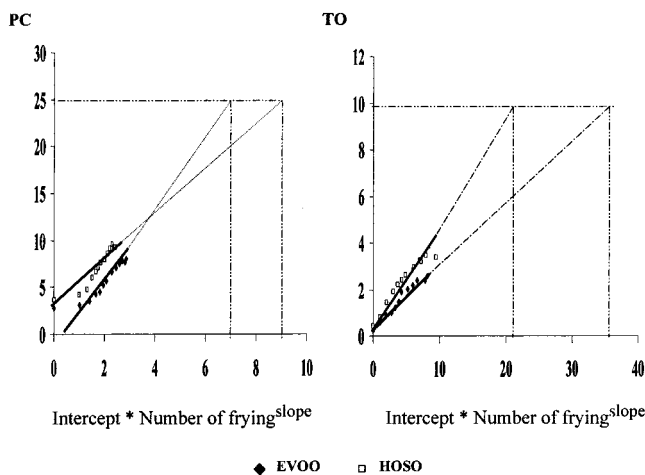


Figure 4. Power adjustment ($Y = aX^b$) of polar content (PC) (mg/100 mg of oil) and triglyceride oligomer (TO) content (mg/100 mg of oil) vs number of frying times. X = number of frying times, Y = PC or TO content, a = intercept, b = slope, EVOO = extra virgin olive oil, and HOSO = high oleic acid sunflower oil.

Table 5. Frying Life (Interval of Calculated Theoretical Maximum Number of Frying Times) of Extra Virgin Olive Oil (EVOO) and High Oleic Acid Sunflower Oil (HOSO) Based on a 25 mg/100 mg of Oil Polar Content Limit and a 10 mg/100 mg of Oil Triglyceride Oligomer Limit^a

	linear adjustment ^{b,e}	logarithmic adjustment ^{c,e}	power adjustment ^{d,e}
	Polar Content		
EVOO	288–354	$8.45 \cdot 10^6 - 485.58 \cdot 10^6$	4463–17129
HOSO	250–330	$2.43 \cdot 10^6 - 32.27 \cdot 10^6$	4776–15175
	Triglyceride Oligomers		
EVOO	283–354	$9.10 \cdot 10^6 - 30.57 \cdot 10^6$	538–1552
HOSO	203–278	$170772 - 1.26 \cdot 10^6$	249–1319

^a Values are frying limit \pm confidential limit. ^b $Y = a + bX$. ^c $Y = a + b \ln X$. ^d $Y = aX^b$. ^e Y = polar content or triglyceride polymers. X = number of frying times. a : intercept \pm SE. b : slope \pm SE.

of 25% in PC and 10% in TO. However, these figures differ greatly when the logarithmic and power adjustment equations are applied. According to the logarithmic adjustment the number of possible frying uses would be considerably higher in EVOO than in HOSO. As for the PC, TO shows a better adjustment to logarithmic and power equation models than to the linear ones. The relative high replenishment of fresh oil obtained in the present study may account for the tendency to reach a near-steady state for the TO oil content. With extrapolation of results to linear adjustments it can be suggested that 2160 frying uses would produce a 16% TO content in EVOO while 1395 uses would result in the same TO content in HOSO. According to linear adjustment calculations, a 16% TO content corresponds with 38% PC in EVOO and 32% PC in HOSO (an average PC of 35% if EVOO and HOSO are considered together). These findings indicate the need to unify the criteria of different countries and suggest that frying oil should be discarded when TO content reaches a maximum of 10% rather than 16%. In short, although a higher number of frying operations would be better in defining the long-term behavior of the oils studied, we can conclude that replenishment with fresh oil helps maintain the initial composition of monoenoic oils used in frying and effectively permits one to fry

fresh potatoes a very high number of occasions without discarding the oil.

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