Sesame Seed Is a Rich Source of Dietary Lignans

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ABSTRACT: The variation in the contents of sesamin and sesamolin was studied in oils extracted from 65 samples of sesame seeds (*Sesamum indicum* L.) from plants with shattering ($n = 29$), semishattering ($n = 7$), and nondehiscent ($n = 29$) capsules. The oil content ranged from 32.5 to 50.6% and was greater in white than black seeds ($P < 0.001$). The sesamin and sesamolin contents in seeds ranged from 7 to 712 mg/100 g (mean ± SD, 163 ± 141 mg/100 g) and from 21 to 297 mg/100 g (101 ± 58 mg/100 g), respectively, with no difference between black and white seeds. Thus, there was a wide variation in the contents of sesamin and sesamolin, which were positively correlated ($R^2 = 0.66$, $P < 0.001$). There were negative correlations between the contents of sesamin and the contents of sesaminol ($R^2 = 0.37$) and sesamolinol ($R^2 = 0.36$) and between the content of sesamolin and those of sesaminol ($R^2 = 0.35$) and sesamolinol ($R^2 = 0.46$) ($P < 0.001$). Sesame seeds had an average of 0.63% lignans, making them a rich source of dietary lignans.

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KEY WORDS: HPLC analysis, lignan glucosides, oil-soluble lignans, sesame seeds, sesamin, sesamolin.

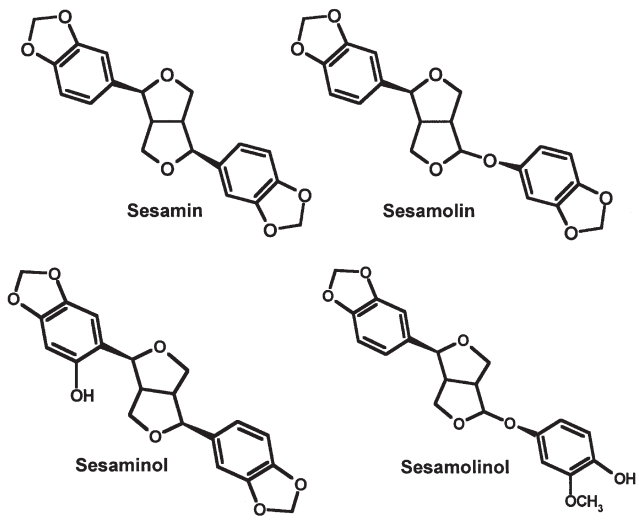
Sesame seeds, used as an oilseed since ancient time, contain 25% protein and 50% oil, the latter having unique chemical-physiological activities (1). Two major oil-soluble lignans, sesamin and sesamolin (Scheme 1), are considered responsible for the unique properties of sesame seed oil. Sesamin is known to reduce the absorption and biosynthesis of cholesterol in rats and plasma cholesterol in humans (2,3). Sesamin also elevates γ-tocopherol levels in humans (4). In addition, sesame seeds contain lignan glucosides, mainly sesaminol di- and triglucosides, sesamolinol diglucoside, and pinoresinol mono-, di-, and triglucosides (5–7), which are concentrated in the defatted sesame flour. Kang *et al*. (8) showed that sesaminol glucosides in defatted sesame flour can decrease susceptibility to oxidative stress.

It is important to understand the variation in the content of these physiologically active constituents of sesame seed for any potential incorporation in functional foods. This knowledge is interesting *inter alias* for sesame plant breeding that aims to increase the contents of both the oil and beneficial oil-soluble micronutrients in the seeds for nutritional purposes. A few studies have reported the contents of oil-soluble lignans in sesame seeds (9–14). However, there is no report that simultaneously provides data on the contents of both oil-soluble lignans and lignan glucosides in the same sesame seeds. Therefore, the aim of this study was to investigate the variability of the contents of sesamin and sesamolin, the oil-soluble sesame seed lignans, in the oils extracted from 65 different sesame seed samples. These samples were from plants with shattering, semishattering and nondehiscent capsules that were previously analyzed for lignan glucoside contents (5,6). The effect of capsule type on lignan contents was studied as well as the correlations between oil-soluble and glucosylated lignans. The total lignan and lignan glucoside contents of sesame seed were compared with those published for flaxseed, which is recognized as the richest source of dietary lignans (15–17).

MATERIALS AND METHODS

Chemicals and reagents. n-Hexane, 1,4-dioxane, methanol, isopropanol and γ-tocopherol (tocopherol standards, article no. 15496) used in this experiment were purchased from Merck (Darmstadt, Germany). They were of analytical grade and were used without further purification. Sesamin and sesamolin were kindly donated by Kalsec Inc. (Kalamazoo, MI).

Sesame seeds. Sixty-five different sesame seed samples, bred, grown, and harvested in Sesaco Corporation (Paris, TX) nurseries in Texas, USA, were used in this study. These seeds were white ($n = 47$), black ($n = 11$), and brown and yellow in colors ($n = 7$). Seeds from semishattering ($n = 7$) and nondehiscent ($n = 29$) plants were harvested when the plants were dry and the dehiscent types ($n = 29$) were harvested when half of the plant was dry. The plants were hand-selected, hand-cut, put



SCHEME 1

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through a plot thresher, and carefully cleaned to remove foreign materials.

Oil extraction. Oils were extracted by vigorous shaking of triplicate seed samples (each 5 g) in stainless-steel tubes with four steel balls and 30 mL of hexane/isopropanol (3:1, vol/vol) for 1 h as described in detail elsewhere (18).

HPLC analysis of oil-soluble lignans and g-tocopherol. The extracted oil was dissolved in *n*-hexane (*ca.* 100 mg/10 mL) in triplicate and analyzed by an HPLC system equipped with a Bischoff HPLC pump (Bischoff Analysentechnik und -geräte GmbH, Leonberg, Germany), an Agilent 1100 series fluorescence detector (Agilent Technologies, Waldbronn, Germany), and Chromeleon software (Version 4.12, Gynkotek HPLC; Mahwah, NJ) for data collection and evaluation. Sesame lignans and γ-tocopherol were separated using an Alltima SI 5U silica column (4.6 × 250 mm; Alltech Associates Inc., Deerfield, IL) and hexane/1,4-dioxane (94:4, vol/vol) as mobile phase. The fluorescence detector was operated at an excitation wavelength of 296 nm and an emission wavelength of 324 nm. The concentration of γ-tocopherol was quantified against authentic γ-tocopherol used as external standard. The coefficient of variation (CV) for triplicate determinations of sesamin, sesamolin and γ-tocopherol was not higher than 6%.

The limit of detection was determined as 5 mg/100 g oil for sesamin and 10 mg/100 g oil for sesamolin using standard solutions and a signal-to-noise ratio of 3. The concentration of sesamin (15 mg/100 g oil) and sesamolin (35 mg/100 g oil) having a peak height 10 times higher than noise was considered as the limit of quantification. Linearity was confirmed between quantification limit and highest concentration of sesamin and sesamolin used in standard solutions (75 μg/mL).

Statistics. ANOVA and regression were performed using Minitab 14 software (Minitab, State College, PA), and significant differences between sample means were determined using Tukey's test.

RESULTS AND DISCUSSION

The oil content of the seeds ranged from 32.8 to 50.6% (mean ± SD, 44.3 ± 3.1%). This range was wider but the average was lower than those reported by Tashiro *et al*. (10) and Yermanos *et al*. (12). There was a significant difference in the oil content between black (mean ± SD, 41 ± 3.5%) and white (mean ± SD, 45 ± 2.7%) seeds ($P < 0.001$) in agreement with Tashiro *et al*. (10). γ-Tocopherol was the only tocopherol detected in this study and ranged from 34 to 75 mg/100 g oil (mean ± SD, 47 ± 7 mg/g oil), which is consistent with previous reports (9). Black seeds contained higher γ-tocopherol levels (mean ± SD, 54 ± 9.8 mg/g oil) than white seeds (mean ± SD, 46 ± 5.0 mg/g oil) ($P < 0.001$).

Sesamin and sesamolin were analyzed in the oil extracts by HPLC (Fig. 1), and the contents of sesaminol and sesamolinol were calculated from their glucosides, as analyzed before (5,6). The levels of lignans in the oils from 65 seeds are presented in Table 1. Sesamin content ranged from 7 to 712 mg/100 g seed (mean ± SD, 163 ± 141 mg/100 g) and sesamolin content ranged from 21 to 297 mg/100 g seed (mean ± SD, 101 ± 58 mg/100 g). The analysis of data indicated a wider range of sesamin content than that reported by Tashiro *et al*. (10) but Hemalatha and Ghafoorunissa (11) reported higher sesamin and sesamolin contents than what we found in this study. The low sesamin and sesamolin contents found in this study were not observed by previous investigators (9–14). Statistical analysis revealed no significant difference between sesamin and sesamolin contents of white and black seeds in contrast with what Tashiro *et al.* had reported (10).

The contents of sesamin and sesamolin in seeds were positively and significantly correlated ($R^2 = 0.69$, $P < 0.001$) as were the contents of the glucosylated lignans, sesaminol and sesamolinol ($R^2 = 0.53$, $P < 0.001$) (Fig. 2). The correlation between sesamin and sesamolin was stronger for black ($R^2 = 0.77$) than for the white seeds ($R^2 = 0.66$). The oil-soluble lignans, sesamin and sesamolin, correlated negatively with the glucosylated lignans, sesaminol and sesamolinol (Fig. 2). It is likely that the positive correlation between sesamin and sesamolin is related to similar pathways in their biosynthesis and that the negative correlation with the glucosylated lignans is related to the activity of some enzyme(s) in the biosynthetic pathway. The few studies on the biosynthesis of sesamin and sesamolin do not provide information about pathways to the glucosylated lignans (19). There was no significant correlation between oil content and the contents of sesamin and sesamolin, indicating the oil might be just a dispersing medium.

The wide variation in lignan contents seems to exceed the natural variation due to agroclimatic conditions and is most probably related to genetic factors influencing the biosynthetic enzymes. The effect of capsule type on the lignan contents was considered in Table 1. Sesame seeds with shattering capsules are the variety most used today and are mainly grown in Asian and African countries. The plants are harvested by hand before drying of the capsules to prevent the loss of seeds on the field following capsule shattering. Seed samples from plants with semishattering ($n = 7$) and nondehiscent ($n = 29$) capsules were

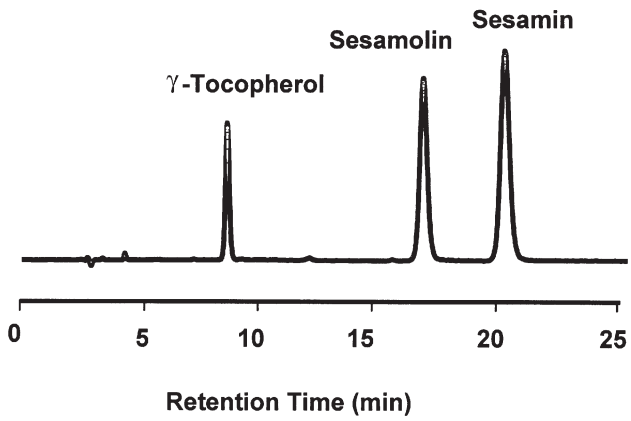

FIG. 1. A typical normal-phase HPLC chromatogram of sesame oil analysis using a fluorescence detector (excitation 296 nm and emission 324 nm).

TABLE 1
Variation in the Contents of Oil, Sesamin, Sesamolin , Sesaminol, and Sesamolinol in 65 Different Sesame Seed Samples (mg/100 g seed)

Color	Oil %	Sesamin	Sesamolin	Sesaminol	Sesamolinol	Total
			Plants with shattering capsules (n = 29)			
White	41	247	99	82	20	447
White	43	37	34	324	53	449
White	47	249	159	208	42	658
White	49	415	187	61	8	670
White	44	232	99	196	33	560
White	48	712	297	139	11	1159
White	46	204	208	226	28	667
White	42	472	201	212	30	915
White	48	167	106	361	49	683
White	47	41	29	302	110	481
White	47	209	50	299	49	607
White	47	166	122	276	31	596
White	45	194	172	273	34	673
Whitish	43	187	122	310	53	671
Whitish	44	48	52	363	66	529
Yellow	43	186	77	141	34	439
Yellow	49	166	99	356	59	680
Yellow	44	178	193	326	41	738
Brown	44	208	78	149	34	469
Grey	43	543	272	32	ND[a]	846
Black	43	153	131	262	24	570
Black	33	295	136	16	ND	447
Black	42	59	65	120	14	259
Black	45	29	65	130	34	258
Black	45	184	147	140	21	493
Black	43	13	57	437	101	607
Black	42	105	126	344	58	634
Black	37	54	105	101	15	275
Black	43	210	146	239	45	640
Range	**33–49**	**13–712**	**29–297**	**16–437**	**ND–110**	**258–1159**
Mean ± SD	**44 ± 3**	**206 ± 159**	**125 ± 67**	**222 ± 112**	**38 ± 26**	**590 ± 190**
			Plants with semishattering capsules (n = 7)			
White	46	37	101	355	68	561
White	48	129	128	244	45	546
Whitish	46	115	134	281	52	582
Whitish	42	12	44	425	97	577
Whitish	41	126	113	212	48	499
Whitish	51	227	157	258	33	675
Whitish	51	380	160	110	18	668
Range	**41–51**	**12–380**	**44–160**	**110–425**	**18–97**	**499–675**
Mean ± SD	**46 ± 4**	**146 ± 124**	**119 ± 40**	**269 ± 101**	**52 ± 25**	**587 ± 64**
			Plants with nondehiscent capsules (n = 29)			
White	45	266	110	206	37	619
White	41	148	21	324	124	617
White	44	30	40	720	45	835
White	42	13	33	501	90	637
White	44	80	64	491	74	709
White	46	25	37	475	73	611
White	43	60	82	482	70	695
White	41	18	59	473	87	637
White	42	37	66	501	72	676
White	47	149	66	474	84	772
White	44	84	67	436	107	693
White	44	122	74	373	77	647
White	44	52	76	440	75	644
White	44	100	43	580	100	823
White	46	114	53	442	64	673
White	43	331	118	268	48	765
White	48	349	125	134	9	617
White	46	236	115	329	35	715
White	45	151	135	435	16	736
White	47	334	136	273	43	785
Whitish	45	90	70	631	71	862
Whitish	47	11	31	442	41	526
Whitish	38	7	53	340	44	445
Whitish	45	201	80	433	104	817
Whitish	46	110	66	585	82	844
Yellow	40	37	30	310	57	433
Black	42	26	36	500	111	674
Black	42	17	49	480	102	648
Range	**38–48**	**7–390**	**21–167**	**134–720**	**9–124**	**433–862**
Mean ± SD	**44 ± 2**	**124 ± 115**	**73 ± 37**	**424 ± 131**	**68 ± 30**	**688 ± 108**
			All sesame seeds (n = 65)			
Range	**33–51**	**7–712**	**21–297**	**16–720**	**ND–124**	**224–1148**
Mean ± SD	**44 ± 3.1**	**163 ± 141**	**101 ± 58**	**153 ± 317**	**31 ± 53**	**581 ± 153**

[a]ND, not detected. The detection limit for sesamolinol was 3 mg/100 g seed, and the quantification limit was 9 mg/100 g seed.

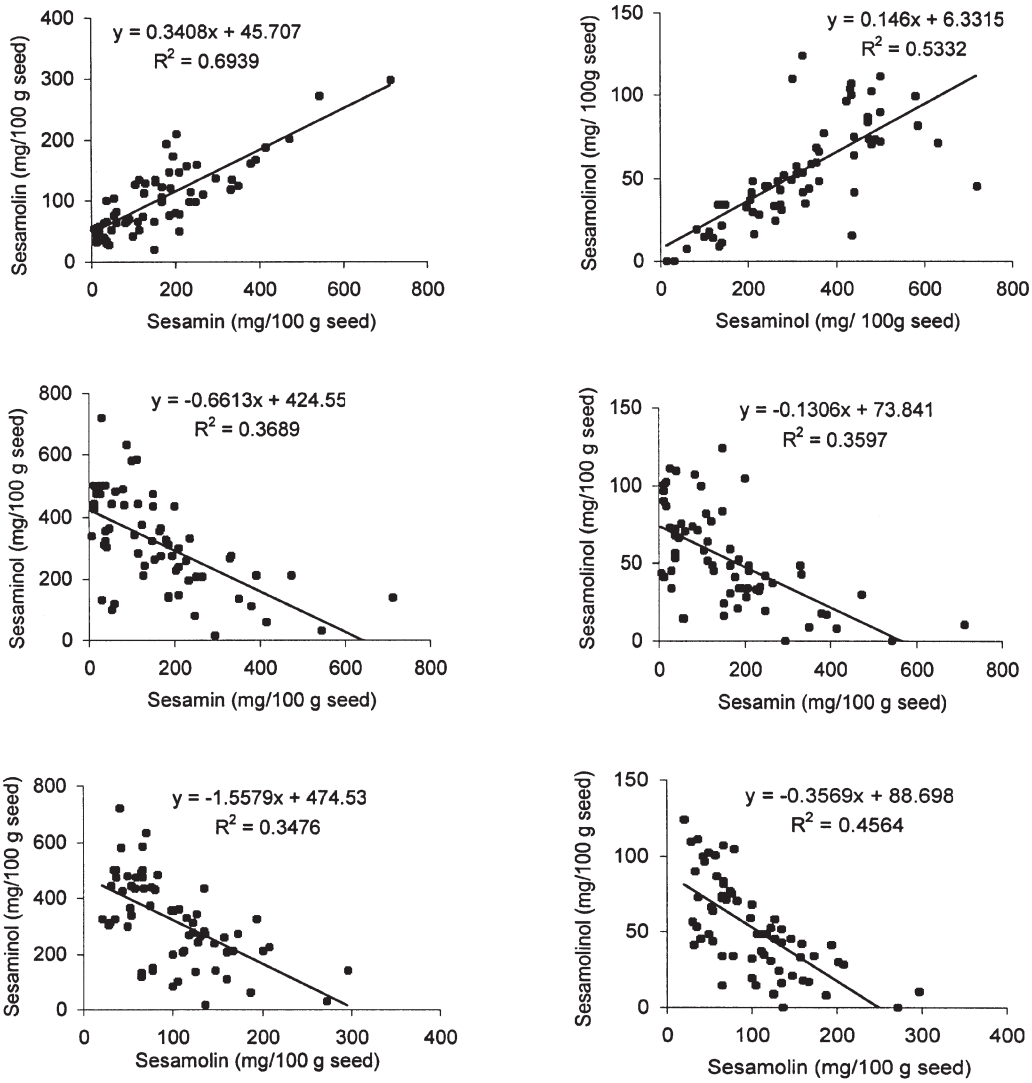

FIG. 2. The correlation between sesamin, and sesamolin and sesaminol contents in the 65 sesame seed samples analyzed ($P < 0.001$) (mg/100 g seed).

included, for the first time, in this study. These plants can be mechanically harvested on the field, thus providing economical advantages over the traditional, capsule-shattering plants. ANOVA showed that there is no difference between the sesamin content in seeds from plants with shattering and with nondehiscent capsules, but the content of sesamolin was significantly higher in seeds from plants with shattering capsules (mean ± SD, 125 ± 67 mg/100 g) than in seeds from nondehiscent capsules (mean ± SD, 73 ± 37 mg/100 g, $P < 0.001$). Conversely, the contents of sesaminol and sesamolinol were higher in the seeds from plants with nondehiscent capsules (mean ± SD: 424 ± 131 and 68 ± 30 mg/100 g seed, respectively) than their contents in the seeds from plants with shattering capsules (mean ± SD: 222 ± 112 and 38 ± 26 mg/100 g seed), respectively ($P < 0.001$). The total lignan content, i.e., the sum of sesamin, sesamolin, sesaminol and sesamolinol, was higher in seeds from plants with nondehiscent (mean ± SD: 688 ± 10 mg/100 g seed) than the seeds from plants with shattering capsules (mean ± SD: 590 ± 19 mg/100 g seed, $P < 0.05$). These differences should be investigated in the breeding of traditional capsule-shattering sesame plants with semishattering and nondehiscent capsules. These different capsule-type plants might also be useful for studies pertinent to understanding the pathways/enzymes involved in sesame lignan biosynthesis.

Flaxseed has been considered to have the highest level of lignans (15–17). Vegetables, cereal brans, legumes, and tea also are good sources, based on the amounts consumed in the diet (15). Since sesame seeds are consumed on a large scale in Asian and African countries, it is important to determine their contribution to dietary lignans. Results from the present study show that sesame seeds contain an average of 0.63% of lignans mainly as sesamin, sesamolin, sesaminol, and sesamolinol with the lowest and the highest lignan contents being 0.26 and 1.16%, respectively. The highest level of secoisolariciresinol,

the major lignan, reported so far is 1.3% (16). Therefore, the analytical data presented in this study indicate that sesame seed should also be considered as a rich source of dietary lignan. However, the variation in the content of lignan in sesame is larger than that in flaxseed (16,17). Moreover, sesame seeds contain lignan both in the oil and the defatted sesame flour whereas the presence of lignan in flaxseed oil has not been reported. Recently, Coulman *et al.* (20) showed that sesame seeds are as rich a source of mammalian lignan precursors as whole flaxseed. It has been shown that sesamin can be converted to the mammalian lignan enterolactone (21). This will be important in research estimating the dietary intake of sesame lignans.

Sesame and flaxseed lignans seem to have different physiological effects. For example, Frank *et al.* (22) showed that secoisolariciresinol diglucoside, the major lignan glucoside in flaxseed, causes an increase in liver cholesterol and a twofold reduction in the levels of α- and γ-tocopherols in rat plasma and liver, in contrast to sesamin, which reduces cholesterol and increases liver and plasma γ-tocopherol (23). All the major lignans and lignan glucosides in sesame seeds contain a methylenedioxy bridge in their molecular structures, which is similar to a functional group found in many medicines (24). The major lignans found in the other seeds, secoisolariciresinol, lariciresinol, pinoresinol and matairesinol, lack this methylenedioxy bridge, which potentially is responsible for the physiological effects of sesame lignans.

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