FLAVOUR AND FRAGRANCE JOURNAL, VOL. 4, 221–224 (1988)

Study of Different Parameters Influencing the Composition of Hydrodistilled Sweet Fennel Oil*

Stavros T. Katsiotis

Division of Pharmaceutical Technology, School of Pharmacy, Aristotelion University, P.O. Box 19589, GR-54006 Thessaloniki, Greece

Different parameters applied in the hydrodistillation of an essential oil not only influence the percentage yield of the oil but also the concentration of each of its constituents. In the present study the simultaneous influence of three parameters, i.e. degree of comminution, distillation period and distillation rate, on the composition of sweet fennel oil was investigated, using the Latin square method for the experimental programme. The percentage composition of the oil samples was determined by GLC. The results showed for each oil constituent the optimum combination of the parameters studied. The degree of comminution influenced the oil composition to a much larger extent than the other two parameters.

KEY WORDS Essential oil *Foeniculum vulgare* Mill. ssp. *capillaceum* (Gilib.) Holmboe var. *dulce* Mill. Sweet fennel Distillation parameters

INTRODUCTION

The essential oil of sweet fennel, which is used in cosmetics, pharmaceuticals, perfumery and as a food additive, is isolated from the plant material by hydrodistillation. Improvements of the yield and quality of the oil are of economic importance.

The aim of the present study was to elaborate standard procedures for hydrodistillation of the oil.[1,2] In order to determine the optimum conditions for the isolation of the oil, the simultaneous influence of three parameters, viz. degree of comminution of the plant material, distillation period and distillation rate, on the composition of the oil was studied. Oil samples obtained under different conditions were analysed by GLC to detect possible variations in the concentrations of the various oil constituents. Such variations can be important for the application of the oil, since they may affect the oil quality.

EXPERIMENTAL

Material

Fruits of *Foeniculum vulgare* Mill. ssp. *capillaceum* (Gilib.) Holmboe var. *dulce* Mill. were collected on the Halkidiki peninsula near Thessaloniki in June 1985. The plant material was identified by the Department of Systematic Botany of the Aristotelion University, Thessaloniki.

The fruits were submitted to hydrodistillation using the apparatus of the European Pharmacopoeia;[3] 20 g samples of fruits were added to 600 ml of deionized water.[4] The other parameters are given in the next paragraphs. The oils obtained were dried over anhydrous magnesium sulphate and then stored at 8–10° C.

Three parameters influencing the isolation of the oil were investigated: the degree of comminution of the fruits (A); the distillation period (B); the distillation rate (C). For each of them, four values were selected.

Table 1. Matrix of the various distillation parameters applied in the hydrodistillation of sweet fennel oil and arranged according to the Latin square method (for details, see the section 'Experimental')

1. $A_1B_1C_1$	5. $A_2B_1C_4$	9. $A_3B_1C_2$	13. $A_4B_1C_3$
2. $A_1B_2C_3$	6. $A_2B_2C_2$	10. $A_3B_2C_4$	14. $A_4B_2C_1$
3. $A_1B_3C_4$	7. $A_2B_3C_1$	11. $A_3B_3C_3$	15. $A_4B_3C_2$
4. $A_1B_4C_2$	8. $A_2B_4C_3$	12. $A_3B_4C_1$	16. $A_4B_4C_4$

*Based on a lecture originally given at the 17th Workshop on Essential Oils, Bad Bevensen, FRG, September 1986.

0882-5734/89/040221-04$05.00

Received 11 July 1988
Accepted (revised) 16 June 1989

Table 2. Percentage composition of sweet fennel oil samples obtained by hydrodistillation using 16 different parameter combinations (see Table 1). The percentages are average values of three GLC analyses

Component	Parameter combination															
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
α-Pinene	0.11	0.23	0.38	0.45	0.22	0.22	0.19	0.19	0.13	0.13	0.16	0.15	0.04	0.02	0.04	0.05
Camphene	0.09	0.17	0.23	0.26	0.16	0.15	0.16	0.13	0.10	0.10	0.11	0.10	0.03	0.02	0.04	0.04
β-Pinene	0.02	0.05	0.07	0.07	0.03	0.05	0.03	0.03	0.03	0.03	0.04	0.02	0.01	0.01	0.02	0.02
Limonene	1.63	3.25	4.45	4.75	3.54	3.42	3.60	3.08	2.60	2.52	2.80	3.97	1.08	1.03	1.48	1.63
γ-Terpinene	0.08	0.18	0.23	0.26	0.25	0.22	0.25	0.13	0.16	0.16	0.15	0.13	0.09	0.09	0.12	0.13
Terpinolene	0.24	0.36	0.47	0.44	0.33	0.33	0.30	0.27	0.26	0.26	0.23	0.23	0.11	0.11	0.14	0.16
Fenchone	2.03	2.57	2.68	2.60	2.23	2.12	2.10	1.86	1.82	1.60	1.52	1.58	0.93	0.83	0.93	0.94
Linalol	0.09	0.13	0.08	0.12	0.12	0.11	0.09	0.11	0.06	0.08	0.09	0.15	0.07	0.08	0.08	0.07
Estragole	4.16	4.68	4.39	4.45	4.42	4.32	4.22	4.18	4.31	4.01	3.74	4.11	3.69	3.45	3.66	3.75
cis-Anethole	0.30	0.30	0.40	0.23	0.25	0.16	0.29	0.29	0.24	0.26	0.23	0.25	0.21	0.28	0.25	0.27
trans-Anethole	89.70	86.10	83.63	84.65	86.42	88.31	86.83	87.98	88.75	88.85	88.96	89.05	92.45	92.47	91.97	91.02
p-Anisaldehyde	1.23	1.59	2.58	1.38	1.58	1.33	0.90	1.29	0.91	1.60	1.39	0.90	0.93	0.78	0.95	1.68

Table 3. Statistical analysis of the percentages determined for *trans*-anethole

Source of variance	Degree of freedom	Sums of squares	Mean of squares	F^a
Comminution	3	84.02	28.01	17.96
Distillation period	3	4.84	1.61	1.04
Distillation rate	3	10.10	3.37	2.16
Error	6	9.36	1.56	

[a]Fisher value.

Parameter A—comminution: fruits submitted to distillation without previous comminution (A_1); comminution of the fruits until all particles passed through a 1.6 mm sieve and more than 40% through a 0.355 mm sieve (A_2); comminution until all particles passed through a 0.63 mm sieve and more than 40% through a 0.22 mm sieve (A_3); comminution until all particles passed through a 0.25 mm sieve and more than 40% through a 0.1 mm sieve (A_4).

Parameter B—distillation period: 90 min (B_1); 150 min (B_2); 210 min (B_3); 270 min (B_4)

Parameter C—distillation rate: 1.5 ml/min (C_1); 3 ml/min (C_2); 4 ml/min (C_3); 5 ml/min (C_4).

The total number of parameter combinations was 16: they were arranged according to the Latin square method, as shown in Table 1.

Gas-Liquid Chromatography

The various oil samples obtained were analysed by GLC, using a gas chromatograph Hewlett-Packard model 7620A, connected with an integrator Hewlett-Packard model 3370B. GC conditions: stainless steel column, 12 ft × 1/8 in; stationary phase, 10% FFAP on Chromosorb 80–100 mesh; injection temperature, 250° C; oven temperature programmed, 60–200° C at 4° C/min; carrier gas, nitrogen; flow rate, 35 ml/min. The percentage composition of the oil samples was determined in triplicate. The relative standard deviation was between ± 0.05% and ± 2.45%. The data were processed statistically by an Amstrad CPC 6128 in BASIC.

RESULTS AND DISCUSSION

GLC analysis of the sweet fennel oil samples obtained by hydrodistillation using 16 different parameter combinations, showed that the percentage composition of the samples varied significantly (Table 2). The percentage of *trans*-anethole varied from 83.63% (combination 3) to 92.47% (combination 14). The relative amounts of estragole, fenchone, and *p*-anisaldehyde varied to a larger extent, while the percentages of the minor components showed relative variations up to 1800%.

Most of the components were found in larger, relative, amounts when the fruits were not comminuted (combinations 1–4), whereas *trans*-anethole was most abundant after the maximum comminution tested. The parameter combination 4($A_1B_4C_2$) yielded the largest relative amounts for most of the constituents, except for *trans*-anethole. Also the amount of *cis*-anethole, an undesirable component, was relatively small under these conditions.

The period and the rate of distillation influenced the percentage composition of the samples as well, as can easily be seen from Table 2 by comparing the data obtained for a certain degree of comminution (combinations 1–4, 5–8, 9–12 and 13–16).

Table 4. Optimum conditions to obtain the highest concentration of certain components of sweet fennel oil, based on the Latin square method

Component	Parameter combination
α-Pinene	$A_1B_4C_2$
Camphene	$A_1B_3C_2$
β-Pinene	$A_1B_3C_2$
Limonene	$A_1B_4C_2$
γ-Terpinene	$A_2B_3C_2$
Terpinolene	$A_1B_3C_4$
Fenchone	$A_1B_3C_2$
Linalol	$A_2B_4C_3$
Estragole	$A_1B_1C_2$
trans-Anethole	$A_4B_1C_1$
p-Anisaldehyde	$A_1B_3C_4$

Elaboration of the experimental data showed that the degree of comminution was the most important parameter; as an example, the statistical analysis of the percentages determined for *trans*-anethole is given in Table 3. It is clear that the highest variance was found for the comminution. Also for the other constituents this parameter was the most important one. Furthermore it was found that the influence of the distillation rate was less significant than that of the distillation period, at least for the distillation rate values investigated.

The parameter combinations which should yield the highest relative amounts of certain oil components are given in Table 4. These combinations were established by statistical analysis using the Latin square method for the experimental programme.

REFERENCES

1. N. G. Iconomou, S. Katsiotis and G. Ktistis, *Pharm. Acta. Helv.*, 57, 196 (1982).
2. S. Katsiotis, G. Ktistis and N. G. Iconomou, *Pharm. Acta. Helv.*, 60, 228 (1985)
3. *European Pharmacopoeia*, Vol. 3, pp. 68–71, Maisonneuve, Saint-Ruffine (1975).
4. S. Katsiotis, *Fitoterapia*, 59, 65 (1988).