

The Future of Natural Essential Oils in the Flavor and Fragrance Industry

By Richard C. Pisano, Citrus & Allied Essences, Ltd., Floral Park, New York

The production and use of essential oils is an ancient industry which has its roots in the human desire for better tasting foods and a beautifully smelling environment. Early use of herbs, flowers, fruits and spices added variety to an otherwise bland and uninteresting diet and encouraged the consumption of food products that might otherwise have been wasted. To use these odoriferous materials in religious ceremonies and to enhance the beauty and attractiveness of men and women of early times is well documented.

By the middle ages, simple methods for producing essential oils were developed in the Middle East and these techniques soon spread to the countries of southern Europe, particularly Italy, Bulgaria and France.

During the early part of the eighteenth century, the essential oil industry began to develop in southern France, particularly in the region of Provence. About the same time, an Italian by the name of Paul Feminis doing business in the German city of Cologne, introduced a new and fragrant mixture which became the sensation of the day. Based primarily on neroli, lemon, bergamot and lavender, it was called L'Eau Admirable.

About seventy-five years later a descendant of Feminis, Jean Marie Farina, modified and improved the formula introducing his product under the name Eau D'Cologne. Farina had more success than he anticipated and many perfume houses copied his formula. To the present day, eau d'cologne survives. The contribution of its creator to the art of perfumery was the popularization of a bouquet rather than of a perfume identified with the fragrance of a single flower.

The total use of essential oils as fragrance raw materials has not declined in modern times. In recent articles titled Perfume: Evolution of Its

Techniques,¹ Arcadio Boix Camps has stated, "Perfumery has evolved and is evolving, but the evolution does not mean in any way a break with the past." He goes on to say that "Perfumery of today is no better or worse than it was before. The science continues and those who work in this field continue to strive. We are enriched by the experience that the perfumers of the past have handed down to us. The mystery that they have enclosed in their fantastic perfumes is like a part of them that will never die. By assimilating the experience of the great classics and by their mastery of the basic new elements at their disposal, new generations of perfumers create this present state and provide examples for the future. We cannot be concerned here why it evolves. All the arts evolve and perfumery cannot be any different."

No one associated with our industry should or can deny the magnificent contribution that advances in organic chemistry have made to the science and art of perfumery. The use of synthetics is not a recent development. Early in the nineteenth century, William Henry Perkins, a brilliant British chemist, synthesized coumarin. The years that followed brought a richness of chemical invention that laid the basis for modern perfumery. The synthesizing of vanillin by Professor Ferdinand Tiemann at the University of Berlin was accomplished in 1876, soon to be followed by the synthesis of indole and heliotropine accomplished by the famous Dr. Baeyer.

By the turn of the century, research chemists in Europe were busily analyzing the complicated chemical structures of the substances found in essential oils, terpenes and their oxygenated derivatives. The goal was to create synthetic citral, geraniol and terpineol—not from lemongrass, citronella or pine oils but rather from coal, petroleum, water and air.

Table I. United States Imports for Specific Essential Oils (in pounds)

	<u>1950</u>	<u>1960</u>	<u>1970</u>	<u>1984</u>
Citronella oil	2,652,258	2,132,407	2,742,329	1,860,269
Clove oil	290,960	453,019	1,010,786	1,783,686
Lemongrass oil	589,745	1,497,362	365,979	185,300
Sandalwood oil	9,711	54,339	117,588	71,728
Vetiver oil	41,060	55,502	174,720	167,874
Geranium oil	Unavailable	143,306	129,962	134,270
Patchouli oil	Unavailable	124,428	315,888	725,846

Many chemists had a hand in this task but three of them made outstanding contributions. First was Merling with the synthesis of isoprene starting from acetone. Then in 1901 Ipatieff working with isoprene created methyl heptenone found in lemongrass and other essential oils. In 1919 the great Swiss chemist Ruzicka treating methyl heptenone with acetylene obtained a chemical which on reduction was linalool.

It was not, however, until the late 1950s that synthetic citral, geraniol, nerol and linalool derived from beta pinene became an economically viable alternative to lemongrass, citronella and bois de rose oils.

In the 1960s the acetylene/acetone process, mainly developed as a route to produce vitamin A and subsequently vitamin E, brought with it a number of perfumery chemicals, particularly citral, linalool and the ionones.

Additional related processes using isobutylene/acetone, isoprene/acetone and alpha pinene have also proved commercially successful. The development and introduction of many unique synthetics such as hedione, sandela, lillial and vertofix all have significantly influenced modern perfumery. The list of important new aroma chemicals is unending and provides a variety of aromas to stimulate and excite the creative abilities of today's perfumers.

With all the progress in the field of chemistry, has the essential oil industry dropped by the wayside, a victim of technological advance? The answer is emphatically no!

For example, let us evaluate some statistics covering thirty-five years which clearly demonstrate the continuing demand for essential oils (see Table I). In the United States citronella oil continues to be important, not as a starting material for the manufacture of aroma chemicals, but for its odor value. Lemongrass oil has ceased to be an important source of citral, having been replaced by litsea cubeba and citrals of synthetic origin. Finally the modern decline in the use of sandalwood oil is most likely due to the success of synthetics which partially simulate the aroma of this

important oil. Table II shows amounts and values for a selection of forty-two essential oils recently imported by the United States.

The import statistics* covering selected essential oils for the United Kingdom, France, the Federal Republic of Germany, the Netherlands and Japan from 1975 to 1984 may be seen in Tables III-VII.

It is interesting to note that the demand for citronella oil in Europe and Japan was greatest during 1975 and 1976 and then declined until 1982 when use again began to increase. The imports of geranium and vetiver oils have been relatively uniform during this period while imports for lemon and orange oils have increased very dramatically.

Natural essential oils, because of their complex and unique structure which adds richness to their aroma, continue to play an important role in perfumery. However, the majority of raw materials used in fragrance compounds are and will continue to be synthetic. The demand for fragrance compounds worldwide has increased very significantly during the last twenty years. Although the need for essential oils also has increased, it has not done so proportionately with the demand for fragrances.

Flavor Materials

Certainly among all the natural flavor materials, essential oils and oleoresins are the most important (see Table VIII). Isolated directly from plants by means of extraction, distillation or mechanical expression, essential oils have been used in household extracts since their introduction over 150 years ago. Almond, anise, cinnamon, lemon and vanilla have been the most popular.

Beginning nearly one-hundred years ago, the dynamic growth of the flavor industry is due to the shift from home processed to factory pro-

* These statistics were provided courtesy of the Tropical Development and Research Institute, London, England.

Future of Essential Oils

duced foods and beverages. Citrus oils, including orange, lemon, lime, grapefruit and mandarin/tangerine, spice oils, particularly cassia, clove, ginger and nutmeg; mint oils, especially peppermint (*mentha piperita*), cornmint (*dementholized mentha arvensis*), and spearmint all find increasing use in beverages, convenience foods,

confectionery, packaged meats and pharmaceuticals.

One group of essential oils which I describe as specialty materials have become increasingly important to the creative flavorist. These include floral and related absolutes such as beeswax, boronia, cassis, jasmin and orris, and some small

Table II. United States: Imports of Specified Essential Oils, 1983 and 1984

Essential Oils	1983		1984	
	Quantities (in pounds)	Value (in U.S. \$)	Quantities (in pounds)	Value (in U.S. \$)
Almond oil bitter	207,491	367,624	231,347	433,514
Anise oil	43,789	186,798	107,413	533,140
Bergamot oil	69,063	917,759	65,760	852,701
Bois de rose	141,018	1,096,972	239,984	1,654,909
Camphor oil	177,357	193,639	128,207	170,546
Caraway oil	22,791	401,302	12,619	220,367
Cassia oil	203,268	6,788,710	215,227	6,893,836
Cedarleaf oil	9,897	147,938	21,445	366,478
Cedarwood	316,127	848,073	516,007	1,311,100
Cinnamon oil	84,892	314,665	72,620	241,135
Citronella oil	1,461,906	2,763,368	1,860,269	3,280,836
Citrus	2,204	39,617	35,117	317,066
Clove oil	864,583	1,534,148	1,793,686	3,440,192
Cornmint	649,329	2,883,558	633,137	2,994,522
Eucalyptus oil	669,284	1,428,091	846,875	1,908,983
Geranium oil	119,780	2,444,633	134,270	3,018,737
Grapefruit oil	96,867	52,199	37,358	84,204
Lavender and spike lavender	233,178	1,683,802	200,511	1,242,995
Lemon oil	1,645,116	7,940,964	1,363,347	7,418,361
Lemongrass	234,627	795,588	185,300	726,202
Lime oil	1,354,277	15,894,892	1,188,383	14,329,394
Nutmeg oil	206,171	1,364,993	233,480	1,859,243
Onion and garlic	13,623	840,721	16,913	797,905
Orange flower oil	14,046	303,409	1,403	278,380
Orange oil	9,638,104	1,786,960	6,979,381	2,828,242
Origanum oil	5,292	67,245	10,262	145,605
Orris oil	1,138	276,280	980	226,966
Palmarosa oil	29,071	467,603	37,853	617,758
Patchouli oil	749,716	10,673,942	725,846	9,047,027
Peppermint from <i>mentha piperita</i>	17,856	145,380	13,609	223,291
Petitgrain oil	343,156	1,738,823	211,918	1,354,377
Pine needle oil	61,422	451,149	188,231	695,270
Pine oil	640,614	265,120	891,988	326,459
Rose oil	40,415	3,425,168	63,508	3,431,493
Rosemary oil	136,987	616,653	179,719	679,913
Sandalwood oil	52,500	1,980,258	71,728	2,885,592
Sassafras	472,334	756,541	1,082,387	2,780,569
Spearmint	57,393	330,063	166,093	901,973
Thyme oil	31,720	291,749	18,614	182,717
Vetivert oil	179,213	3,196,270	167,874	3,346,614
Ylang ylang	71,343	1,345,466	82,503	1,724,925

Note: All values refer to F.O.B. country of origin.
Source: U.S. Department of Commerce.

Table III. United Kingdom: Imports of Selected Essential Oils, 1975-1984

		1975	1976	1977	1978	1979	1980	1981	1982	1983	1984
Vetiver oil	Q	-	22	25	22	21	13	14	10	8	6
	V	-	505	781	750	527	274	219	227	202	175
Citronella oil	Q	-	446	297	195	212	178	306	136	167	214
	V	-	590	574	359	586	592	999	416	481	641
Eucalyptus oil	Q	138	191	241	252	447	217	278	298	372	321
	V	691	339	484	448	755	380	590	751	1,152	1,131
Lemon oil	Q	304	497	597	633	542	481	608	485	541	605
	V	1,922	3,126	4,640	4,717	4,720	7,187	7,721	4,357	4,234	4,504
Orange oil	Q	373	786	719	1,060	1,006	776	813	1,053	1,720	1,781
	V	282	423	551	844	1,012	670	549	704	960	1,666
Geranium oil	Q	12	43	19	31	32	22	23	30	32	33
	V	203	733	454	886	608	392	357	461	667	885
<u>Mentha arvensis</u> oil	Q	283	443	549	417	478	554	840	834	754	1,017
	V	2,374	4,174	5,683	3,871	3,395	4,799	7,429	8,106	10,354	15,754

Q = quantities in tons
 V = values in thousands (Br £)
 Source: Trade Returns

Table IV. France: Imports of Selected Essential Oils, 1975-1984

		1975	1976	1977	1978	1979	1980	1981	1982	1983	1984
Vetiver oil	Q	86	143	118	117	82	86	95	100	94	107
	V	15,632	36,739	36,294	36,330	22,013	18,049	20,356	29,068	31,207	42,070
Citronella oil	Q	707	742	503	250	366	298	347	271	291	300
	V	11,408	9,691	8,662	4,649	8,086	9,767	12,373	10,479	9,467	10,490
Eucalyptus oil	Q	243	1,524	431	444	860	653	626	721	513	709
	V	4,301	6,783	6,940	6,811	9,268	9,618	14,112	18,140	15,799	26,443
Lemon oil	Q	193	413	407	311	376	258	275	395	293	357
	V	8,433	16,549	18,668	18,292	26,003	31,106	28,580	27,241	28,047	27,408
Orange oil	Q	508	1,139	1,162	1,122	1,799	1,289	1,511	1,195	1,745	2,267
	V	3,479	5,674	8,000	7,054	12,159	11,038	12,517	11,360	15,737	26,688
Geranium oil (other than Bourbon)	Q	17	83	59	60	73	51	52	58	53	65
	V	2,204	12,389	12,693	13,668	14,201	8,783	10,499	13,036	14,662	23,532
Geranium oil, Bourbon	Q	13	75	41	43	40	31	35	36	31	24
	V	3,313	20,572	13,319	15,626	15,143	12,438	12,948	15,238	14,505	12,948
Peppermint oil	Q	634	938	750	677	1,040	674	712	911	930	965
	V	42,016	64,382	66,967	53,609	61,120	41,080	57,421	77,305	97,691	114,965

Q = quantities in tons
 V = values in thousands (FF)
 Source: Trade Returns

Table V. Federal Republic of Germany: Imports of Selected Essential Oils, 1975-1984

		1975	1976	1977	1978	1979	1980	1981	1982	1983	1984
Vetiver oil	Q	5	7	7	5	7	8	5	5	7	7
	V	870	1,000	1,143	875	1,056	1,154	811	436	909	1,046
Citronella oil	Q	231	226	239	111	91	80	54	54	97	105
	V	2,226	1,491	2,050	817	848	1,064	746	723	1,185	1,226
Eucalyptus oil	Q	130	193	596	240	207	282	301	248	285	363
	V	2,087	1,729	4,410	1,897	1,506	2,332	3,029	2,930	3,646	5,088
Lemon oil	Q	170	207	179	175	162	153	177	156	176	211
	V	5,589	5,663	5,883	5,208	5,712	7,802	9,379	6,628	6,369	6,987
Orange oil	Q	865	1,226	1,724	1,503	2,003	1,842	2,037	2,097	2,061	1,928
	V	3,443	3,846	6,243	6,371	8,860	8,903	7,631	6,514	5,883	8,858

Q = quantities in tons
 V = values in thousands (DM)
 Source: Trade Returns

Future of Essential Oils

Table VI. Netherlands: Imports of Selected Essential Oils, 1975-1984

		1975	1976	1977	1978	1979	1980	1981	1982	1983	1984
Vetiver oil	Q	2	8	7	5	4	7	5	3	4	5
	V	312	1,013	1,169	843	525	636	401	376	428	714
Citronella oil	Q	33	237	198	141	145	154	63	86	54	143
	V	312	1,635	1,863	1,195	1,574	2,499	1,569	1,289	832	1,678
Eucalyptus oil	Q	39	94	30	24	89	64	68	69	49	37
	V	396	792	323	229	604	670	856	868	748	677
Lemon oil	Q	38	68	60	70	87	84	106	116	97	74
	V	1,458	2,236	2,876	3,025	4,437	6,932	8,414	6,377	4,095	3,365
Orange oil	Q	299	477	471	477	543	492	399	452	584	485
	V	886	1,199	1,641	2,196	2,960	1,549	1,716	1,748	2,903	2,784

Q = quantities in tons
V = values in thousands (NF)
Source: Trade Returns

Table VII. Japan: Imports of Selected Essential Oils, 1975-1984

		1975	1976	1977	1978	1979	1980	1981	1982	1983	1984
Patchouli oil	Q	10	31	26	32	42	31	30	25	41	30
	V	58	137	134	140	228	269	216	224	360	228
Vetiver oil	Q	3	8	10	11	15	9	8	9	15	9
	V	61	132	168	163	197	123	73	103	155	97
Lemongrass oil	Q	13	28	43	29	44	30	28	44	40	22
	V	26	39	85	47	69	53	50	109	90	50
Citronella oil	Q	762	484	352	166	274	132	29	16	34	33
	V	860	397	355	136	289	257	46	26	40	40
Eucalyptus oil	Q	15	38	49	34	46	31	24	44	39	37
	V	25	38	51	32	45	37	29	65	55	59
Lemon oil	Q	89	145	142	155	213	163	150	159	202	228
	V	480	651	706	705	1,131	1,411	1,033	944	962	1,050
Orange oil	Q	1,755	3,061	2,513	1,789	4,132	3,849	3,409	2,606	4,466	3,067
	V	498	890	832	815	1,356	1,331	1,215	1,256	1,275	1,223
Mint oil (excl. spearmint)	Q	485	463	544	489	689	588	512	718	657	630
	V	3,378	2,241	2,957	2,287	2,512	2,056	1,593	2,832	2,137	2,005
Spearmint oil	Q	85	49	58	118	101	95	68	94	51	84
	V	697	248	448	679	548	576	443	766	394	609
Geranium oil	Q	8	16	19	20	23	21	15	16	18	16
	V	115	193	246	235	321	305	175	191	202	187
Clove oils	Q	71	216	240	92	220	159	123	173	145	171
	V	96	275	300	117	250	175	131	197	164	199
Cassia oil	Q	18	23	14	19	22	16	39	37	42	39
	V	150	155	173	218	285	238	614	591	654	560

Q = quantities in tons
V = values in million yen
Source: Trade Returns

volume essential oils including angelica root, buchu, davana, neroli and tagette. These absolutes and essential oils find increasing use as minor components in natural and artificial flavors. Unlike fragrances, the use of essential oils in flavors has increased very dramatically in recent years. In the United States with its "black and white" differentiation between natural and artificial, the use of synthetic aroma chemicals in flavor formulations require that a flavor be desig-

nated *artificial* and identified as such on the particular food or beverage label. The trend towards naturally obtained flavor chemicals, often derived from essential oils, has been encouraged by food manufacturers who perceive an advantage emblazoning their products *natural*.

This trend towards the natural has also begun in Western Europe and Japan as a reaction to governmental regulations and to the impact of consumerism. Dr. Jan Stofberg, in a recently

published article titled *The Flavor of Food—How Natural is Natural*,² has concluded that the simple, subjective and emotional distinction between natural and artificial flavorings is not justified. He urges consumer education regarding the true value and benefit of food and its ingredients. As the standard of living improves within the developing nations of Africa, Asia, Latin America and the Middle East, the demand for processed food and beverages and their need for natural and artificial flavors will grow significantly.

Safety of Flavor Raw Materials

Beginning in the middle 1950s, the Food and Drug Administration of the United States and other national health authorities in Europe and Japan became interested in the safety of flavor raw materials. A number of essential oils and aroma chemicals were banned from use due to their presumed toxicity. Most notable among these were oil sassafras, due to its safrole content, and tonka bean vanilla extract for its coumarin content.

Concern on the part of the United States flavor industry prompted the Flavor and Extract Man-

ufacturers Association (FEMA) to create in 1959 an expert panel composed of independently recognized experts to review the safety of essential oils and aroma chemicals for their intended use in flavors. The comprehensive list of these materials, known as the GRAS list (Generally Recognized As Safe), was compiled. This list is added to periodically and GRAS List #14 will be published in the near future. The guiding forces behind the GRAS concept are Dr. Richard Hall and Dr. Bernard Oser. Obviously, the safety of new essential oils and aroma chemicals will have to be reviewed by the expert panel prior to their use in flavors within the United States. It should be noted that the findings of the FEMA Expert Panel are recognized and respected not only in the United States but in many other parts of the world.

Internationally, the flavor and fragrance industry has recognized the increasing legal and toxicological requirements imposed by various governments. In 1966 the industry established the International Organization of the Flavor Industry (IOFI) and in 1973 a sister association, the International Fragrance Association (IFRA), was created. Both these organizations are based in Geneva, Switzerland and through their member

national associations maintain contacts with concerned government agencies. IOFI and IFRA have also established guidelines concerning the safety aspects of using essential oils and aroma chemicals.

Quality Evaluation

Quality and authenticity has become an increasingly important factor pertaining to the use of essential oils. Ultra violet and infrared spectrometry, gas chromatography and GC/MS are used extensively for evaluating the quality of essential oils. Recently, isotopic analysis has been employed to determine the authenticity of essential oils and other flavor ingredients. Through carbon C₁₄ analysis, the addition of nature identical synthetics can easily be detected. Most flavorists and perfumers prefer to use pure essential oils, and continued adulteration of these materials not only violates national laws pertaining to fraud, but also brings discredit to our industry. The availability of blended and reconstituted essential oils is important to perfumers and flavorists; however, these materials should be identified and labeled accordingly.

In summary, the essential oil industry worldwide remains healthy, although at present there may be over-production for some specific

essential oils. Continued significant growth in use, however, will be related to those materials that are unique or that are extracted from crops that can be cultivated easily. The future will depend on highly sophisticated and efficient agricultural techniques like those utilized for the production of mint oils in the United States, for citrus oils in North and South America, and for lavender and lavandin as in Bulgaria and France.

IFEAT will continue to encourage the production and utilization of essential oils and looks forward to cooperating with essential oils producers, dealers and users, and with our sister organizations, IOFI and IFRA.

Acknowledgement

This paper was presented at the IFEAT/Pharmachim Flavor & Fragrance Summit Conference in Varna, Bulgaria, in October 1985.

References

Address correspondence to Richard C. Pisano, Citrus & Allied Essences, Ltd., Floral Park, NY 11001.

1. Arcadio Boix Camps, *Perfumery: Evolution of Its Techniques—Part I*, *Perfum. Flavorist* **10**(3)1, June/July 1985; *Part II*, *op.cit.* **10**(4) 15, August/September 1985; *Part III*, *op.cit.* **11**(3)9; June/July 1986.
2. Jan Stoffberg, *Consumption Ratio for Flavor Materials*, *Perfum. Flavorist*, **11**(1)5, February/March 1985

