Indian Essential Oils.*

By J. J. Sudborough.

In a recent article on Imperial Co-operation the statement is made that "It is a sound doctrine of political economy that the ideal condition of a given industry is that the country where the raw materials occur should, in the main, and in the absence of antagonistic influences, be also the country in which the finished article is manufactured from the locally produced raw material." If this is true then India should be the country in which the indigenous raw materials for essential oils and perfumes are manufactured into the final marketable products.

This has not been the case in the past.

Table I is a list taken from the London Market of important essential oils which are produced wholly or in part from natural products grown in India and Ceylon.

The only oils in this list which have been actually distilled in India or Ceylon for many years are the last four, namely lemon-grass, citronella, ginger-grass, and palmarosa. The reason why an exception was made in the case of these oils was that the grass would not stand export, as the yield of oil would diminish and the freight would be prohibitive.

Even in the case of these oils the industry is not all that could be desired. The distillation in many cases is crude and very little attention has been paid to the most economic and efficient methods of manufacture Recently Mr. Pearson has suggested using steam for the distillation of palmarosa and ginger-grass oils, as both the yield and quality of the oils are improved.

Another serious defect which has been pointed out by all authorities dealing with essential oils is the tendency for the Indian distiller or seller to adulterate his products. This sophistication is repeatedly met with in connection with such oils as citronella, lemon-grass, palmarosa and cinnamon.

*A paper based on the opening address of the discussion on Essential Oils at the Lahore meeting of the Indian Science Congress, January 1918.

TABLE I.

No.	Oil.	Plant or tree.	Part used for distillation.	Yield of oil per cent.
1	*Anise oil	Pimpinella ansium	Fruit	1·5-6·0
2	†Cardamom oil	Elettaria cardamomum	Seeds	2-7
3	Cinnamon oil	Cinnamomum zeylanicum	Bark	0·5-1·0
4	Cinnamon leaf oil	Do	Leaves	1 0-1·8
5	Coriander	Coriandrum sativium	Fruit	0·15-0·8
6	E. Indian Dill oil	Peucedanum graveolens	Fruit	3-4
7	Ginger oil	Zingiber officinale	Rhizome	2-3
8	Sandalwood oil	Santalum album	Heart wood	2·5-6·0
9	Thymol	Carum copticum	Fruit	2 3-4·0 of oil containing 40-50% of thymol
10	Vetivert oil	Vetiveria zizanoides	Root	0.4-0·9
11	Lemon-grass oil	Cymbopogon citratus	Grass	
12	Palmarosa oil	Do martini motia	Grass Heads	0·3
13	Ginger-grass oil	Do do sofia	Grass Heads	0 5-1·0
14	‡Citronella oil	Do nardus	Grass	0·5-1·0

*Quite different from Star anise oil of China which is sold at about one-fourth the price of European anise oil.

†The oil from certain varieties is the only oil which is official.

‡The citronella oil from Java is superior to Ceylon oil and fetches a higher price.

TABLE II.

Exports of Raw Materials.

No.	Article.		1912—13. Quantity.	1912—13. Value. £	1913—14 Quantity.	1913—14 Value. £	1914—15. Quantity.	1914—15. Value. £	1915—16. Quantity.	1915—16. Value. £
1.	Ajwan seed ...	cwt.*	21650.	6135.	9784.	2983.	7368.	2736.	†13062.	4871.
2.	Aniseed	cwt.	2478	1978.	1129.	931.	835.	977.	594.	615.
3.	Cardamoms ...	lbs.	266971.	37765.	373401.	49994.	413135.	54369.	482764.	49597.
4.	Cinnamon	lbs.	39650.	1018.	38170.	1015.	39711.	869.	54147.	1144.
5.	Coriander seed ...	cwt.	84587.	35477.	95533.	39099.	84058.	46327.	90104	70953.
6.	Cummin seed ...	cwt.	20130.	30682.	19026.	29338.	13554	25698.	26308	12889.
7.	Cummin seed, black ...	cwt.	1738	1870.	1313	1157.	1411.	985.	2050.	1857.
8.	Dil or Sawa seed ...	cwt.	1920	1294.	2090	1489.	1945.	1351.	3380.	2342.
9.	Ginger	lbs.	9950660.	158425.	9214471.	122661.	7529188.	87291.	6289699.	71351.
10.	Sandalwood ...	lbs.	...	101529.	..	128626.	...	35918.	...	103796.
11.	Citronella oil ...	lbs.	1384628.	...	1586005.	...	1425050.	...	‡1674692.	...
12.	Lemon grass oil ...	gls.	32262	47416.	47522.	67955.	27796.	37914.	31700.	30102.

The amount of Palmarosa and Ginger grass oils produced each year averages 150,000lbs: about 100,000lbs. are exported.

*Only 772 cwt. to British Empire.

† 3882 cwt. to British Empire.

‡ 506000 lbs. to British Empire.

As recently as May 1917 the following statement was made in the Perfumery and Essential Oil Record in an article on citronella oil. "Java citronella oil is always pure—we have never met a sample of direct import which there was any reason to believe was tampered with—whilst, we regret to say, Ceylon citronella oil is almost invariably adulterated. It is true that there is a certain small quantity of Ceylon Oil, usually designated "Estate Oil", which is a really pure oil, but the enormous majority of exports of this oil from Ceylon are deliberately adulterated with petroleum."

The same complaint used to be made with reference to the Chinese Cassia oil shipped from Hong Kong The Chamber of Commerce of that port took up the matter and requested the Hong Kong Government to undertake the sampling and analysis of all shipments of cassia oil and the sealing of each original case with a Government mark. This has not been enforced, partly owing to the war, but the enunciation of the regulations has led to the improvement in the quality of the shipments.

The necessity for similar regulations in India and Ceylon dealing with the shipment of oils distilled in the country is obvious to anyone connected with the Essential Oil trade. The regulation should include the following oils :—lemon grass, palmarosa, ginger-grass, citronella, cinnamon, sandalwood and any oils subsequently prepared in bulk in the country.

In addition to the oils just mentioned large scale experiments have been made on the distillation of sandalwood oil, and of thymol from ajwan seed.

Both these have been carried out under scientific direction and the distillations have resulted in a considerable financial return to those who undertook the manufacture.

In the case of sandalwood oil the two factories in the Mysore State aim at turning out 25,000 lbs. of high grade oil per month.

There are several factories making appreciable quantities of thymol, but in several cases the quality is not equal to that of the product turned out by German factories in pre-war time.

It is highly probable that some of the other oils mentioned in Table I could be distilled profitably in India, although in most of these cases the conditions are more complex.

With both sandalwood oil and thymol the conditions are simplified by the fact that India practically possesses a monoply of the raw materials from which the oils are obtained viz., the sandalwood tree (*Santalum album*) and the ajwan fruit (*Carum copticum*). No other country at present grows timber which yields an oil comparable with East Indian sandalwood oil. The so-called West Indian sandalwood oil obtained from *Amyris balsamfera* is physically and chemically quite different from the East Indian oil. It does not contain santalol, the chief constituent of the East Indian oil, and can never be a substitute for the latter but is often used for adulterating it.

The W-Australian oil is derived from *Santalum cygnorum* and is inferior to the East India oil. Its alcohol content is about 70% as compared with 92 to 96% for East Indian oil and it is not certain that the alcohols present are santalols.

In the case of thymol attempts are being made to grow *Carum copticum* in other countries and so far these experiments have met with success in the Seychelles. In America attempts are being made to obtain the chemical from other natural sources. The plant *Monarda punctata* has been found to yield 0·25 per cent. of oil, 66 per cent. of which consists of thymol.

Of the oils enumerated in Table I it is probable that Vetivert, Cardamom and East Indian Dill oils could be distilled successfully in this country from the financial standpoint, as in these cases India is one of the chief countries in which the raw materials are grown. The question of the distillation of cinnamon, cinnamon leaf, ginger, cummin, fennel and coriander is not so simple, as these raw materials can be obtained from other parts of the world *e. g.*

Cinnamon — from Ceylon, where it is cultivated, and Seychelles. Very little distillation is carried out in Ceylon, the chips are exported, *e. g.* in 1911 2,644,598 lbs. and distilled in Europe. This oil is similar to the Chinese cassia oil, but has a much more delicate odour and fetches a higher price. The oil which is distilled in Ceylon and exported is largely adulterated with cinnamon leaf oil or cassia oil.

Ginger — from China, Japan and Jamaica. Sierra Leone and East Africa.

Anise seed — from Egypt, Bulgaria, Chili, but mainly Russia and Asia Minor.

Cummin — from Egypt, Sicily, Malta, China.

Fennel — from Mediterranean littoral, Asia Minor, Persia, Japan and especially Galicia and Roumania.

Coriander — from Russia and Thuringia.

As a factor of first importance in considering the question of distillation of some of these oils is the yield of oil from E. Indian raw materials as compared with materials from other sources. For example coriander seed is exported from India but is never used for distillation as the yield of oil is only 0·15 to 0·2% compared with 0·8 to 1·0% for seed grown in Russia or Moravia. Similarly E. Indian aniseed distilled in the Institute laboratories gave a 0·88% yield of oil as compared with 2·4 to 3·2 for Russian seed and 2·7 to 3·5% for Italian seed. The oil from the Indian seed also appears to contain less anethole, the important odorous constituent of anise oil. It is therefore highly improbable that the distillation of Indian grown aniseed would pay.

The oil obtained from Indian grown dill seed and known as East Indian dill oil differs appreciably in composition from European dill oil, but is a product which is quoted on the London market. The E. Indian oil has a high specific gravity, contains less carvone than the European oil, and is further characterised by containing an appreciable quantity of a complex phenolic ether known as dill apiol. The price of the E. Indian oil is only about 2/3rds that of the European oil, but the yield from the seeds is good.

The composition of vetivert oil varies appreciably. The two well known types of oil are the Reunion and the Java, both apparently being obtained from the roots of *Vetiveria zizanoides*.

The oil which is distilled in Reunion has a specific gravity 0·980 to 0·995 whereas Java oil has a sp. gr. 1·01 to 1·03.

The cause of the difference is stated to be that the Reunion oil is obtained from fresh roots whereas the Java oil is obtained from the dried roots, and further that the methods of distillation are somewhat different.

The Reunion oil fetches only about half the price that the Java oil does.

Experiments made at Dehra Dun and also in Bangalore indicate that the oil obtained from Indian cuscus roots approximates in properties to the Java type.

Extraction of the Oil.

Before the distillation of any oil is undertaken on a commercial scale a considerable amount of preliminary work is necessary.

In India it is more or less assumed that all that is necessary is to put the raw material in an iron or copper vessel with water and then to heat over a free fire or perhaps by a closed steam coil. It must not be assumed that a method which gives the best result with one oil will necessarily give the best result with a different oil.

Questions which have to be decided are :—

1. Water distillation or steam distillation. In most cases the method of blowing steam through the mass is preferable to boiling the material with water, this is true of sandalwood, ajwan seed and many others, but in a few cases *e. g.* rose leaves the water distillation gives better results. As examples of the difference obtained by water and steam distillation may be cited the cases of Lavender, Palmarosa and Sandalwood oils.

In the case of lavender it has been shown that the percentage of oil and also the ester content of the oil are increased by using steam distillation instead of the older water distillation. These results are clearly shown from the following numbers :—

	Yield per cent.		Esters per cent	
	(1)	(2)	(1)	(2)
Steam	0·81	0·82.	50·9	53·7.
Water	0·71	075.	44·0	43·6.

which prove that in the old method a loss of 12 to 20 per cent. of the total esters resulted. In the case of palmarosa Mr. Pearson claims that an increased yield of oil of about 10% can be obtained by substituting steam for water distillation. In the case of sandalwood oil, the amount of steam necessary to distil a ton of oil is considerably diminished by using steam in place of water distillation.

2. The previous treatment of the material to be distilled, the extent to which it should be bruised, crushed or disintegrated.

In most cases the finer the material the greater the extent to which the oil cells are broken and the more readily is the oil volatilised. On the other hand the finer the material the greater the tendency of the steam to blow tunnels, the formation of which is disastrous, if good results are to be obtained.

3. The extent to which the material should be dried before distillation. In the case of lavender it is found that previous drying leads to a slight loss of oil but as this loss is mainly due to the more volatile terpenes, the quality of the oil from the dried flowers is rather better than that from the fresh.

In some plants the oil does not exist free but in combination with glucose and in such cases previous decomposition of the glucoside by a fermentation process is necessary, *e. g.* essential oil of mustard.

4. The size of the stills. For a given charge is it advisable to have a short wide still or a tall narrow one? Preliminary experiments will soon determine which type gives the greater oil to steam ratio, a ratio which is of fundamental importance in an essential oil distillery.

5. The most economic steam velocity. In most cases by passing steam through the material with a high velocity the oil is removed more quickly than if a lower steam velocity is used. It will be found, however, that the oil to steam ratio is low owing to the steam not being saturated with the vapour of the oil, and in most cases the aim of the distiller is to keep this ratio as near its maximum as possible. A high steam velocity has the further disadvantage with finely pulverised material that it increases the tendency to tunnel formation.

6. The type of the condensers to be used. Various types of patent double surface, worm and tubular condensers can be used. The most efficient for the purpose has to be selected and this becomes a question of considerable importance when the amount of cooling water is limited or when the price for the same is high.

The material of which the condenser can be made is also a point which is frequently of high importance. In some cases, *e. g.* thymol, bare copper is best, with other oils, *e. g.* sandalwood, bare copper affects the colour of the oil with results of a disastrous nature. Experiments will settle whether bare copper, tin, lead or aluminium is the best. If copper condensers with a tin coating are used then it will be found that the tin is gradually worn

away and has to be replaced and it is essential to have the condensers so constructed that the parts can be readily removed and retinned.

Many of these details have been worked out in particular cases in America or Europe, but as the results are not available in each case the best conditions must be determined experimentally in this country.

Cultivation of the oil-bearing plants.

There is also another aspect of the question which must receive considerable attention, namely the proper cultivation of the materials, woods, seeds, flowers, roots, etc.—from which the essential oils are obtained. The questions of climate, soil and manure as affecting both the yield and also the composition of the essential oil, and lastly the question of plant diseases and pests need far more attention than they have received in the past.

The variation in composition of an oil from a plant grown under different conditions has been noted in several cases. At one time it was thought that the essential oils known respectively as ginger-grass and palmarosa were derived from the same variety of grass grown under different conditions, but it is now known that they are derived from two different varieties of the same species. Norris in America has drawn attention to one or two remarkable alterations in the composition of plant products with change in environments. Cases of change not quite so noticeable have been observed in Europe in the case of lavender and peppermint. Both these are grown at Mitcham in England and give oils of the finest quality both as regards flavour and odour. Most attempts to cultivate the Mitcham peppermint in other countries have ended in the production of an oil quite different from Mitcham in odour or flavour. Recently oil of the Mitcham type has been produced in Italy.

The effect of suitable manuring is shown by figures based on experiments by Irk on peppermint plants.

	Oil in kilos per hectare.*
1. With no manure	10·01.
2. Farmyard manure	16·40.
3. Sodium nitrate	18·76.
4. Superphosphate and Sodium nitrate ...	14·16.
5. Superphosphate, Kainite and Sodium nitrate	15·84.
6. Superphosphate, Kainite, Sodium nitrate and farmyard manure	29·83.

*Hectare=2·47 acres.

The researches of Dr. Charabot on the influence of certain manures on the composition of peppermint oil are of considerable interest. He has been able to show that the addition of sodium chloride or nitrate to the soil always increases the percentage of esters in the oil as compared with an oil from plants grown without such additions. In this way the flavour and odour of the oil can be controlled to a certain extent by the addition or non-addition of such salts as nitrate and chloride of sodium to the soil.

In the case of lavender excellent results have been obtained by Professor Zacetiarewitz of Avignon by the use of a special manure consisting of sodium nitrate 20%, potassium chloride 20% and superphosphate 60%. The use of 500 kilos per hectare of this manure, increased the yield of oil from 12 to 28 kilos per hectare.

The variation of the oil with the part of the plant distilled and also with the age of the plant :—

One of the best examples of the former is the Ceylon cinnamon tree *Cinnamonum zeylanicum*, from which three distinct types of oil can be obtained.

(a) Cinnamon oil from the bark consists mainly of cinnamaldehyde with a little engenol

(b) Cinnamon leaf oil from the leaves contains 70-90% of engenol.

(c) Oil from the root contains camphor.

The two former are oils of commercial importance, the bark oil realising a much better price than the leaf oil with which it is frequently adulterated.

The alteration of the nature of the oil with the age of the plant is exhibited in a marked degree in the case of the orange.

From orange flowers the oil of neroli is extracted, the very small undeveloped fruits yield oil of petit grain and the peel of the ripe fruit yields orange oil.

It is essential to study the physiological aspect of plant changes and to determine experimentally what period in the life history of the plant should be selected for the extraction of the oil or perfume.

In this connection may be mentioned the fact that both lemon-grass and ginger-grass give two types of oil

(*a*) Soluble in 70% alcohol

(*b*) Insoluble in 70% alcohol

The soluble variety always fetches a higher price than the unsoluble, in the case of ginger-grass oil the difference is as much as one shilling per pound.

The cause of this is not known at present but may be due to the age of the grass when distilled.

New oils and new sources of oils.

So far we have dealt with the production, from Indian grown materials, of oils which already find a sale on the London market. There are other aspects worth consideration viz. (1) attempts to grow in India Essential oil producing raw materials not already grown to any appreciable extent and the extraction of oils from these. (2) The production of essential oils or perfumes from materials present in the country but at present not used on the manufacturing scale. (3) The production of artificial perfumes from natural products grown in India.

1. As examples of the growth of essential oil bearing materials more or less new to India may be cited the following :—

(*a*) Species of Eucalyptus In the Nilgiris *E. globulus*—the common blue gum—is readily grown and small scale distilleries are run at a profit. Many other species give oils which fetch a much higher price on the market. A few of these are (*1*) *E. citriodora* which gives an oil rich in citral and comparable with lemon grass oil and (2) *E. Macarthuri* an oil rich in geraniol and geranyl acetate. These trees grow in Australia, but it is stated that the distillation of the oils does not pay on account of the high cost of collection and heavy labour charges. It is possible that their cultivation and distillation in certain parts of India, where labour is much cheaper than in Australia, might be profitable.

(*b*) ***Liquidamber orientalis***, the tree which gives the oleoresin known as liquid-storax, grows in Asia Minor and, according to E. M. Holmes, might be grown in Cashmere and Punjab.

(*c*) Balsam of Tolu and of Peru. The trees from which these resins are obtained might grow in Ceylon.

(*d* The plant *Piper cubeba* appears to grow in Ceylon and India and its cultivation might be profitable,as the dried berry yields 10—16% of a volatile oil—oil of cubebs—used in medicine.

(*e*) The production of Essential oils from lemon,orange and pummelo has never been carried out in this country. The oils would have to be expressed or extracted and not distilled.

2. As examples of the production of oils from materials grown in the country, but at the present time not used for the manufacture of oils to any appreciable extent, may be mentioned:-

(*a*) The production of a substitute for Copaiba oil from the oleoresin from either *Hardwickia pinnata* or *Dipterocarpus indicus*. A detailed examination of the oils from these two oleoresins indicates that they are practically identical with copaiba oil. Experiments are now being conducted in Calcutta in order to see whether they have the same medicinal value as copaiba oil.

(*b*) The production of cinnamic acid and its derivatives from the oleoresin of *Altingia excelsa* of Burma and Assam.

(*c*) The distillation of oils from Eagle wood (*Aquilaria agallocha* of Burma and Assam), spikenard (*Nardostachys Jatamansi*) of costus oil from the roots of *Saussurea hypolenia*.

(*d*) The production of pine needle oil from certain species of pines on the Himaliyas.

(*e*) The determination of the commercial value of the oil from *Myristica Malabarica* and other species of Myristaceae.

3. Synthetic Oils. A certain number of oils which occur naturally or which were originally obtained from natural sources are now manufactured on a large scale synthetically. Some of the more important of these are oil of Bitter Almonds (=Benzaldehyde). Cinnamon oil (=Cinnamaldehyde), essential oil of mustard(=Allyl thiocyanate) and Wintergreen oil (=Methyl

salicylate). In several cases the synthical products can be manufactured and sold at a lower price than the natural products. Numerous other products which occur in small quantities in plants or which have odours resembling plant oils are now produced synthetically, *e. g.*, esters of benzyl alcohol, phenylethyl alcohol and its esters, ethers of naphthols. The production of such synthetic products in India at the present time is scarcely a commercial proposition, as the starting point for the manufacture of many of these is coal tar, large quantities of which are not available in this country.

The production of certain oils or perfumes from plant products which are indigenous to India is a question that deserves further consideration. It is quite possible that it would pay to manufacture in India some of the following :—

(*a*) Ionone, the artificial perfume of violets, from citral obtained from Indian lemon grass oil.

(*b*) Geraniol and such esters as geranyl formate, acetate and propionate from palmarosa oil.

(*c*) Rhodinol and citronellol.

(*d*) Halogen cinnamenes from storax cinnamic acid.

(*e*) Aubepinene, the foundation of hawthorn perfumes, from the anethole of aniseed.

(*f*) Heliotropine from the safrole of camphor oil.

(*g*) Santalol and its esters from sandalwood oil.

In the preceding discussion it has been assumed that the only method of obtaining essential oils from natural products is by a process of water or steam distillation.

Up to the present this is the method almost universally adopted for obtaining oils from woody tissues, seeds, fruits and rhizones, but it is probable that some method of extraction with a volatile solvent, such as petroleum ether or carbon tetrachloride, might be cheaper in the cases of oils which volatilise very slowly with steam. Certain woods require as much as 100 hours steam distillation in order completely to remove the oil from the tissue. A process of extraction of the same quantity of the finely divided material by a solvent would require probably 10—20 hours.

Precautions for avoiding loss of solvent would be necessary and further purification of the extracted oil would have to be resorted to in most cases.

Perfumes.

In addition to the essential oils already mentioned, many of which are made use of in perfumery, there are a number of perfumes obtained from flowers.

In India the usual method is to distil the flowers with water and collect the perfume in sandalwood oil.

The method of water or steam distillation can be used with advantage with very few flowers. Experiments have shown in many cases that the yield of perfume is decreased and the delicate odour partially destroyed by high temperature treatment. This is true of jasmine and all oils from species of citrus and also holds good for rose oil to a certain extent.

Some years ago Cassia pomade (obtained from the flowers of *Acacia farnesiana*) was made in appreciable quantities in India and exported, but as the price of the pomade fell its production was discontinued.

Modern methods of extracting the finest odours are used but little in this country.

The modern enfleurage method of extraction by which moist air is drawn through layers of the flowers placed on 5 or 6 trays and is then made to pass over 18 glass plates coated with a suitable fat is largely used in France. A similar method could be used in this country by substituting a vegetable for an animal fat. The solid fat used must be specially purified and should be odourless. It is possible that the solid fat from *Vateria Indica* could be used for this purpose.

Hot or cold maceration of the flowers with a vegetable oil, such as sesame oil, is sometimes used and even mineral oil fractions such as paraffin can be employed.

In most cases an alcoholic solution of the perfume is made by repeatedly shaking the impregnated fat, oil or paraffin with alcohol.

A method of extracting perfumes from flowers which has been largely developed in Europe during recent years, but which so far has not been tried in India, is by the solvent action of a low boiling petroleum spirit. The same solvent can be used for extracting several batches of flowers and, on removal of the solvent by distillation, the so called "Concretes" are obtained and these, when treated chemically to remove resins, colouring matters &c. yield the valuable "Absolutes".

This method should prove successful in India, but can only be done on a large scale and with great care owing to the inflamable nature of the solvent.

The future of the essential oil and perfumery trade will be one of keen competition.

In pre-war times the main manufacture was conducted in S. France and Germany. At the present time the central European nations, France, N. America, Canada and Australia are devoting much thought and energy to the better cultivation of natural products capable of giving odoriferous or medicinal volatile oils. As examples of this are the experimental drug stations and farms started by Austria and Hungary, the attempts of Australia to introduce certain Eucalyptus oils in place of lemon-grass oil and palma-rosa oils, and the attempt in the U. S. A. to compete with S. India in growing lemon grass in Florida, and with Sicily in producing lemon oil in California.

This quickening of the activities of the different nationalities in the essential oil industry can only mean that in the future those with the best natural advantages and most scientific methods of cultivating the raw materials and of distilling and extracting the active oils will meet with the greatest financial success.

The use of some of the cheaper oils, *e. g.* lemon-grass oil, as germicides and insecticides and the use of Cinnamon oil in medicine may increase the demands for these oils so that the market will be able to take larger quantities than in pre-war times without any considerable fall in selling price.

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