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It has often been said that variety is the spice of life. It can also be said that in the world of spice we have a grand variety. Defining the limits of spice can be a trying task. We use all parts of a wide variety of plants including the wood or bark, leaves, flowers, fruits and seeds.

It can be said in a general sense that anything we add for flavor or fragrance value could be construed as a spice. This study examines many of the more common spices that one would find on a good supermarket spice rack as well as some that are relatively obscure.

#### **Use of Spices in Fragrances**

While most people are aware of how and where spices are used, little is known about where and how they are used in perfumes or fragrances. There is scarcely a fragrance on the market that does not contain some sort of spice note. The most commonly used spice in fragrance and flavor is vanilla or its chemical surrogate 4 hydroxy 3 methoxybenzaldehyde, commonly known as vanillin.

The importance and widespread use of vanilla along with chemicals like vanillin, ethyl vanillin, coumarin, heliotropine and anisic aldehyde, (which are all components of vanilla), cannot be overstated. It is difficult to identify fragrances of any type that do not employ these materials to some degree.

Fragrances like Shalimar, Emeraude, Obsession, Oscar de la Renta, Gloria Vanderbilt, Passion, Bal a Versailles, and Lou-Lou employ vanilla as part of

their main theme. Many other fragrances are dependent on vanilla or a vanilla chemical as a part of a balsamic foundation.

Some are built on the foundation of spices like clove and cinnamon. Examples of these would be Youth Dew, Opium, Cinnabar, Canoe, Brut, Pierre Cardin and of course Old Spice. Less obvious examples which employ spice notes are L'air du Temps, Norrell, L'Origan, Paco Rabanne, Halston and Aramis. The list is endless.

Almost all fragrances use some type of spice note to brighten up the composition. They are all used in the same manner as they are used for cooking. They are not usually the dominant character but blend with the main theme of the preparation and add life to the flavor.

Like an artist or musician who searches for just the right color or sound, we examine all the spices so we are familiar with their character and store them in our memory for judicious use at the proper moment.

Many florals whose odors are well known would not have the same sparkle without chemicals which we normally regard as spicy. Carnation and lilac are two that are quite spicy, the responsible constituent for the odor of carnation is eugenol. Eugenol, most normally associated with clove, is found in many florals including jasmin, ylang, rose, tuberose, gardenia and many others.

Group I: Spices with Single Components.						
		Characterizing				
Spice	%	Component	Character			
Allspice	80	Eugenol	Spice			
Almond	95	Benzaldehyde	Nutty/Almond			
Anise	90	Anethole	Anise/Licorice			
Armoise	50	Thujone isomers	Minty/Woody			
Badiane	80	Anethole	Licorice			
Balsamite	80	1-Carvone	Minty			
Basil	84	Methyl Chavicol	Licorice/Medicinal			
Caraway	50	d-Carvone	Grassy/Caraway			
Cinnamon Bark	71	Cinnamic Aldehyde	Cinnamon/Spicy			
Cinnamon Leaf	80	Eugenol	Clove/Spice			
Clove	80	Eugenol	Clove/Spice			
Estragon	80	Methyl Chavicol	Licorice/Medicinal			
*Leek	—	Dipropyl Disulfide	Onion/Garlic			
	—	Allyl Methyl,				
	-	+ Methyl Propyl Sulfides <sup>1</sup>				
Mustard	80	Allvl Isothiocvariate	Mustard/Pungent			
Peppermint	50	Menthol + Menthone 20%	Mint			
Perilla	50	Perilla Aldehyde	Nutty			
Spearmint	60	1-Carvone	Mint			
Wintergreen	95	Methyl Salicylate	Mint			
Vanilla	—	Vanillin	Vanilla/Powder			

\*While leek may have many components, the most important odor components are the sulfides. Other terpenes and classes of chemicals play only a very minor role in the aroma profile.<sup>1</sup>

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The spicy odor of lilac is due to cinnamic aldehyde and cinnamic alcohol. These notes, blended with other floral constituents like phenyl ethyl alcohol, geraniol and citronellol create a pleasant lilac floral composition where the spicy character is not perceived as cinnamon, but as lilac.

#### **Characterization of Various Spices**

The variety of spice notes found throughout nature is enormous. We are in a continuing process, trying to categorize, memorize and understand these natural wonders we call spice. This is no small undertaking and the following pages attempt to bring some order and understanding to our current knowledge of spice oils and their character. I will begin by grouping the spices that consist mainly of a single character donating component.

Although the spices listed in Group I predominantly consist of a single component, they are, of course, not entirely responsible for the full rich character of the total essential oil. For example, while cinnamon contains about 80% cinnamic aldehyde, the total oil is quite different. This is due to the variety and character of the other constituents, most of them only in

trace quantitites, that make a significant contribution to the overall odor and performance.

Characterization of any complex (or simple) composition is a difficult task. It is analgous to a piece of music. All the parts can be heard simultaneously, but no one part is obscured by any of the other parts. It is a question of balance and blend, all done very tastefully. It is an analytical task to dissect the composition and understand how it was composed originally.

In order to determine how the effect is achieved, we begin by examining the individual component parts until we find a component that is individually responsible for the character of the whole. For example, eugenol is quite characteristic of clove. Unfortunately, this is not always the case. Even after we discover several components, it is not always clear how they contribute to the overall composition. It often becomes necessary to reconstruct the composition, component by component, to determine how they interact.

We have seen analysts focus on threshold to emphasize the importance of trace ingredients. While threshold values are important for comparison purposes, the same values may not be valid and hence, not as useful in a composition. We can be certain that the physical properties or evaporation rate of one component affects the properties of another. It would be more accurate to assume that factors of polarity, solubility, and evaporation rate all play a role in an aroma profile. With these things in mind we will examine the composition of coriander oil in detail.

Coriander oil consists primarily of 80% linalool. For those who appreciate the subtleties and intricacies of this complicated spice, it is not necessary to indicate that linalool is not the characterizing agent of this essential oil.

Table I examines the composition of this interesting spice by separating the characteristic elements and grouping them by chemical class.<sup>2</sup>

The odor characters of Coriander oil are: Citrus, (lemon-like), Floral, Fruity, Spicy, Minty, Woody and Waxy.

It is the combination of these characters and their specificity to coriander that we keep in mind as we match their components to the coriander character. Table II illustrates the components, and their particular contribution to the character of coriander.

It is often the case that a chemical may contribute more than a single character. This is the case with citronellol. It contributes both to the floral and to the waxy character.

Next, we will examine the oils that exhibit a slightly more complex character. The spices that have at least two or more ingredients that characterize the flavor and aroma are listed in Group II.

As in the previous group, the components in Group II only give the shell of the character of the whole oil. In fact when we blend two or three of the constituents the difference is enormous. However, in almost all cases, when we look at the blends of components

Table I. Constituents of coriander oil grouped by chemical class						
19 Hydrocarbons $\alpha$ -Pinene Camphene $\beta$ -Pinene Sabinene Myrcene 3-Carene $\alpha$ -Thujene $\alpha$ -Phellandrene Limonene $\alpha$ -Terpinene $\beta$ -Phellandrene $\gamma$ -Terpinene p-Cymene Heptadecane Octadecane Octadecane A-isopropenyl-1-methylbenzei $\beta$ -Careno	3 Thiazoles   Thiazole, 2,4,5-trimethyl   Thiazole, 2-isopropyl, 4-methyl   Thiazole, 2-isobutyl, 4,5-dimethyl   2 (Ep) Oxides   β-Caryophyllene epoxide   Linalool oxide   10 Alcohols   Dodecanol   Citronellol   Geraniol   Nerol   Linalool   α-Terpineol   Decanol   Terpinene-ol-4   Eiemol	9 Bases Pyrazine, 2,3,5 trimethyl Pyrazine, 2-methyl 6-ethyl Pyrazine, 2-ethyl 5-methyl Pyrazine, 2-ethyl 3.6-dimethyl Pyrazine, 2-ethyl 3,6-dimethyl Pyrazine, 2-ethyl 3,5-dimethyl Pyridine, 2-butyl Pyridine, 2-pentyl Pyridine, 2-acetyl <b>10 Furans</b> Furan, 2-pentyl Furan, 2-hexyl Furan, 2-hexyl 5-methyl Furan, 3-(3-pentenyl-4-methyl) Furan, tetrahydro 2-methyl- 2-vinyl-5-lsopropenyl Furan, tetrahydro 2-methyl-	Carbonyis 7 Aidehydes (E)2-Decenal Undecanai Dodecanal (E)2-Dodecenal 2-Tridecenal Geranial Decanal 1 Ketone Camphor 2 Acids Acetic Decanoic 4 Esters Geranyi acetate Linalyi acetate			
3 Phenois 2-Isopropyl-5-methyl phenol (Thymol) 1-Methoxy-4-propenyl benzene (Anethole) 1-Allyl-5-methoxy-3,4-methylenedioxy benzene (Myristicin)		2-vinyl-5-isopropanol (Lilai alcohol) Furan, tetrahydro 2-methyl- 2-vinyl-5-isopropanoic acid (Davana acid Furan-2-one, 5-methyl 5-vinyl (Lavender lac Furan-2-ol, 5-methyl 5-vinyl	isoamyi 3-metnyi Butanoate ) ctone)			

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in Table II, the basic character of the spice is recognizable.

In the next section we will look at those spices which have a very complex character. It is often the case that even after all the constituents have been recombined the character is only approximate. In some cases the final composition is not even recognizable.

All of the components in the next group (Group III) play an important role in the character of their respective oils. After each oil I have indicated the com-

Group II: Spices with two or more components.					
Spice	%	Characterizing Component	Character		
Bay	40	Eugenol	Medicinal/Spice		
	10	Chavicol	Medicinal/Spice		
Cassia	70	Cinnamic Aldehyde	Cinnamon/Spice		
	10	o-Methoxy Cinnamic Aldehyde	Cinnamon/Spice		
Cumin	50	Cuminic Aldehyde	Nutty/Fatty		
	10	Dihydro Cuminic Aldehyde	Nutty/Fatty		
Dillseed	40	d-Carvone	Grassy/Dill		
	30	α Phellandrene	Woody/Citrus		
Dillweed	40	d-Carvone	Grassy/Dill		
	30	$\alpha$ Phellandrene	Woody/Citrus		
Fennel	80	Anethole	Licorice		
	10	Fenchone	Minty/Woody		
	3	Methyl Chavicol	Herb/Medicinal		
Marjoram	40	Eucalyptol	Medicinal/Minty		
(Spanish)	15	Linalool	Citrus/Floral		
Oregano (Greek)	65	Carvacrol	Oregano/Medicinal		
	7	Thymol	Spicy/Medicinal		
	6	Para Cymene	Fresh/Spicy		
Sage Spanish	15	Eucalyptol	Medicinal		
(Lavandulifolia)	30	Camphor	Minty/Medicinal		
Sage Dalmation	40	Thujone isomers	Mint/Herbaceous		
(Officinalis)	20	Camphor	Minty/Medicinal		
Thyme	45	Thymol	Spicy/Medicinal		
	18	Para Cymene	Spicy/Terpene		
	2	Carvacrol	Medicinal/Spice		

ponents that have been identified. In every case in Group III the most important components have not yet been discovered.

Each spice has a distinct and unique character that stems from more than just the combination of the identified components. As indicated above, the most important components have not yet been identified for all of these. What might these components be? I feel they are trace ingredients with enormous impact. Compounds like pyrazines, thiazoles, sulfur and nitrogen containing molecules along with unsaturated aldehydes and ketones that exhibit very specific characters.

The fourth section of this examination of spices might be viewed as the most difficult. There is a common denominator in that they all have a characteristic aroma of Black Pepper. While there are differences in constituents, they have a great deal in common. It is most interesting to note that the character contributing component or components have so far remained elusive to many scientists. We will examine these in Group IV.

There are over 120 components identified in Black Pepper. They are composed of the following classes of chemicals. Forty-six hydrocarbons, 31 alcohols, 16 carbonyls, 1 acid, 10 esters, 11 bases, 2 ethers, 1 nitrile, 8 phenols, and 3 oxides.

None of these chemicals is responsible for the extremely powerful and characteristic aroma of black pepper. It is possible to see the character contribution of para cymene and gamma terpinene as well as other unsaturated hydrocarbons to the aroma of black pepper. However, it is not possible to achieve the characteristic aroma of black pepper by mixing all of the above ingredients. This is a clear indication that we have not identified the characterizing component(s) of black pepper.

It can further be seen that there are a great many terpenes and sesquiterpenes in all three of these oils. It has been my experience that many commercial ses-

Table II. An illustration of the components responsible for the character of coriander.*						
Citrus character**	Sabinene, myrcene, $\alpha$ -phellandrene, limonene, $\alpha$ -terpinene, $\beta$ -phellandrene, $\gamma$ -terpinene, p-cymene, ocimene, terpinolene, $\alpha$ -thujene. All 7 aldehydes with particular emphasis on (E) 2-decenal and geranial, acetic acid and linalyl acetate.					
Floral character	Linalool, nerol, geraniol, citronellol and most of the 10 furans listed in Table I.					
Fruity character	Geranyl acetate, linalyl acetate and isoamyl 3 methyl butanoate enhance the citrus character by introducing the sweet fruity flavor.					
Spicy character	2-Isopropyl-5-methyl phenol and 1-methoxy-4-propenyl benzene.					
Minty character	Linglool oxide and camphor.					
Woody character	β Caryophyllene, linally acetate, terpinen-4-ol and myristicin.					
Waxy character	Alcohols C-10 through C-12 and citronellol.					

\*Listed in order of their importance to the aroma profile.

\*\*It should be noted that commercial grades of many chemicals are not sufficiently pure. As a consequence, a clean fresh citrus character cannot be easily achieved synthetically.

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	Hydrocarbons	Alcohol	Aldehydes	Ketone	Acid	Ester	Base	Sulfur	Misc.	Total number of
									com	ponents
Ambrette Seed <sup>3</sup> Ambrettolide and hydrocarbons	9	10	9	2	3	10	_	_	1	44
Angelica Root <sup>4</sup> Hydrocarbons and ketones	15	2		2	<u></u>	1	_	_	3	23
Calamus⁵ Furfural, 5-methyl-2 -furfural, methyl eugend	24 DI	12	—	16	_	1	-	_	3	56
Cardamon <sup>6</sup> Eucalyptol and terpinyl acetate and aldehydes	33	28	18	7	3	24	-	-	7	113
Carrot Seed <sup>7</sup> Hydrocarbons, γ- decalactone and coumarin	35	14	7	5	-	4	-	_	9	74
Celery Seed <sup>8</sup> Hydrocarbons, 3-n-buty phthalide and related phthalides	14 1	1	_	<b>—</b> .	_	_		_	2	17
Coriander <sup>9</sup> See Table I	30	45	22	19	26	18	15	3	25	203
Elemi <sup>10</sup> Hydrocarbons particu- larly α-phellandrene	93	6	-		—	_	_		1	100
Ginger <sup>11</sup> Aldehydes especially aldehydes C-3 through C-8 and citral	48	25	13	10	3	7	—	3	12	121
Nutmeg <sup>12</sup> Eugenol, safrole and phenols along with hydrocarbons	26	20	3	2	8	9	_	_	14	82
Parsely <sup>13</sup> Hydrocarbons, cis-3- hexenol, trans-2- hexenal, cis-3-hexenyl acetate and sulfur compounds. (Methylthio methane, carbondisulfid methyl dithiomethane)	33 - e,	5	3	1	-	2	_	3	11	58

Group III: Spices with complex components.

quiterpenes or sequiterpene products have somewhat of a black pepper aroma. Although weak, these products are almost always complex mixtures and it is always difficult, if not impossible, to determine where this characteristic aroma is coming from. It is probable that the characteristic aroma of black pepper has something to do with the terpenes/sesquiterpenes or related compounds. They may be oxygenated or contain a sulfur or nitrogen moiety or both.

When a class separation is done on black pepper and passed through a silica gel column there is a strong and characteristic aroma of black pepper that elutes at the point where the hydrocarbons end and eucalyptol begins.<sup>18</sup>

#### Conclusions

As can be seen from the preceding information, perfumers have an impressive library from which to draw and create new combinations of spice blends. There is an additional aspect to consider, as in cooking, there are many combinations of spices that may be used in a single fragrance theme. These odor types

Group IV: Spices with extremely complex components.					
Component	(Bark) Cascarilla (Ref. 14)	(Fruit) Cubeb (Ref. 15)	Black Pepper (Ref. 16)	(False Pepper) Schinus Molle (Ref. 17)	
α-Pinene	5.1	2.2	9.0	2.7	
α-Thujone	2.8		—	_	
α-Thujene	Tr	Tr	1.8	0.2	
Camphene	1.0		0.1	1.2	
3-Pinene	0.9	—	10.4	0.4	
Sabinene	—	4.6	19.4	0.4	
6-3 Carene	_		5.4	Tr	
a-Phellandrene	—	—	1.7	5.3	
a-Terpinene			0.3	Tr	
8-Phellandrene	—		4.0	7.2	
+ 1,8 Cineol	The second s	0.7		—	
Myrcene	3.6	_	2.0	5.3	
Limonene	2.3	_	17.5	8.4	
oara Cymene	13.0	—	1.3	11.5	
y-Terpinene	0.9	—	0.5	0.8	
linalool	2.7	—	0.5	0.3	
Terpinene-4-ol	0.6		1.0	0.5	
2-Terpineol	—	2.2	_	1.0	
I-Borneol	2.5		_	_	
Eugenol	0.2	—	—	—	
3-Caryophyllene	9.0	3.7	14.7	0.3	
lumulene	—	4.9	0.2	3.0	
6-Cadinene	3.7	8.8	0.5	6.8	
3-Bisabolene	_	1.5	2.0	—	
Copaene	_	0.4	0.5	0.4	
Cubebol	_	10.0	0.1	2.4	
Nerolidol	—	3.5	_	—	
β-Cubebene	—	11.0	_	—	
Allo aromadendrene	—	4.2	—	—	
Calamenene	_	3.7		0.3	

may not necessarily be related. Vanilla can be a main theme and can be accented with basil, thyme or any combination of spices. There are no creative boundaries, only paths.

Further, there are many qualities of each of the preceding oils mentioned. In the clove family for example, we have Clove Bud, Clove Stem and Clove Leaf. All of these are available with varying qualities

#### References

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due to geographic origin as well as different types of extraction techniques.

To understand many uses of spice and the effects that can be achieved requires many lifetimes of work and study. Regrettably, I only have one lifetime to donate to this fascinating and exciting work but it has proved to be an extremely rewarding and enjoyable experience.

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