

A Novel Technology to Study the Emission of Fragrance from the Skin

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This article challenges the common belief that people perceive a fragrance by the relative volatility of its components. According to this belief, a fragrance is described as having a top note, a middle note and a bottom note. A new technology has now been developed to prove that this is not so. A fragrance actually is perceived by the diffusion of molecules. This diffusion is an inherent property of the compounds and is independent of their molecular weight, boiling point and odor threshold or odor value.

By means of this technology we have shown for the first time that, depending upon the fragrance, various skin may or may not have a significant effect on the emission of fragrance molecules. In this article, both the technology and the results will be discussed in detail.

The Aura

When the sun is totally eclipsed by the moon, as it is in the accompanying photo (Figure 1) taken from *National Geographic* magazine, the surrounding glow is called the "aura." Similarly, if we consider a drop of fragrance, the molecules surrounding the drop form an aura of that particular fragrance.

It is a common belief that one smells a fragrance layer by layer. For example, one first smells the most volatile components, called the *top note*. Then one smells the components with boiling points in the middle range; the *middle note*. Finally one perceives the components

with the highest boiling point; the *bottom note*.

In reality, this is not the case. When a drop of fragrance is placed on the skin, several selected molecules from the lowest boiling to the highest boiling—irrespective of their molecular weights, boiling points and vapor pressures—form an "aura," eventually hit our nose, and give our first impression of that particular fragrance. The composition of this aura is dependent on a characteristic property of each fragrance molecule known as its *diffusivity*.

Diffusivity

What is diffusivity? Diffusivity is the inherent property of a compound to emit its molecules into the air. One compound is said to be more diffusive than another if its molecules tend to pass into the air to a greater extent than those of the other compound. This is called *relative diffusivity*. Diffusivity is independent of boiling point, molecular weight, odor threshold or odor value.

This phenomenon is true not only for a fragrance placed on the skin but also for the fragrance of a living flower, which forms an aura that we smell when we sniff the flower. This aura is actually due to the aroma molecules that are constantly being produced by the living flower and are coming off the surface of the petals.

SPME Technology

This new diffusion technology was developed by IFF and trademarked Aura of Aroma.^a It evolved naturally from IFF's Living Flower^b technology. Now IFF has developed a novel technology to capture the aroma molecules surrounding the flower petals without touching the flower or any other part of the plant. This process is called solid-phase micro-extraction (SPME). To our knowledge, no one has ever before applied

Figure 1. Aura from a solar eclipse

This article is adapted from a presentation given at the Flavours and Fragrances Conference of the Royal Society of Chemistry at the University of Warwick, England, April 30–May 2, 1997.

^a Aura of Aroma is a registered trade name of International Flavors & Fragrances Inc., New York, NY

^b Living Flower is a registered trade name of International Flavors & Fragrances Inc., New York, NY

Figure 2. SPME needle in sampling position near a *Dendrobium superbum* orchid flower

this technique to the analysis of the aroma of living flowers.

The SPME needle,^c which is a 2–3 mm solid glass fiber coated with a high-boiling liquid adsorbent, is placed in close proximity to a flower, without touching it, and is kept there for a period of 30–60 minutes depending on the odor strength of the blossom. The aroma molecules around the petals are adsorbed onto the fiber. Then with GC/MS, we analyze the fiber to determine the aroma profile of that particular flower.

Figure 2 shows the *Dendrobium superbum* orchid, now growing in our greenhouse. This orchid comes from New Guinea and is unique in the world because of its raspberry-like fruity odor.

The data in the column labeled "Living orchid fragrance" in Table I shows the composition of the live *Dendrobium superbum* orchid obtained with classic liquid extraction technology. This composition represents more than 98% of the flower's profile. When we reconstituted this fragrance we discovered it lacked the diffusivity of the live *Dendrobium* orchid.

When we used the SPME technique to obtain the live orchid's aura (the column labeled "Aura" in Table I), we found that the headspace consists primarily of highly diffusive molecules, and the nature of the composition is independent of the molecules' molecular weight, boiling point

Figure 3. SPME needle in sampling position for fragrance samples on skin

and odor strength. For example, benzyl acetone, with the lowest molecular weight, did not increase at all in the headspace. On the other hand, linalool, which has a higher molecular weight than benzyl acetone, increased dramatically—more than 15 times. Benzyl acetate increased 25 times, while 2-tridecanone, a so-called middle note component based on its molecular weight and boiling point, increased 275 times. Oxyphenolon, another middle note component and the compound responsible for the characteristic raspberry odor, decreased substantially in the aura, while compounds of lower volatility, such as 2-pentadecanone and ethyl myristate, still were substantially present in the aura. When we recreated this formula and added it back to the living orchid fragrance at a level of 10%, the fragrance became very diffusive.

Oil vs. Aura: Compositions Compared

The next step was to find out what happens when a fragrance is applied to the skin.

Using the SPME technique: We have developed a very simple yet elegant technique in which a 10-microliter sample of a perfume is applied to a clean inner forearm. Immediately a small glass globe is placed over it and sealed by contact with the skin (Figure 3). The SPME needle is inserted through a septum in the top of the vessel and positioned so that the fiber tip is approximately 1 cm above the sample area. The needle is kept in place for a period of 30–60 minutes; following which, it is immediately analyzed by GC/MS to determine the composition of the fragrance aura.

In this connection, we would like to mention that, to our knowledge, no one has ever used the SPME method to study the release of fragrance from skin. We did this work at least three years ago and disclosed it in 1996.¹

Shalimar: The first fragrance we studied was the classic fragrance Shalimar^d created by Guerlain in 1925.

Table II shows that limonene, the most volatile component, which constitutes 30% of the fragrance oil, is present at only 20% in the aura, whereas linalool, which is only 1.7% in the oil, is eight times greater in the aura. Similarly, linalyl acetate also doubled in the aura. Ethyl vanillin, a high-boiling chemical used for the first time in Shalimar to

Table I. Composition of fragrance via liquid extraction vs. composition of aura via SPME for a live *Dendrobium superbum* orchid flower

Compound		Mol. wt.	Living orchid fragrance (%)	Aura (%)
benzyl acetone	top note	148	0.02	0.03
benzyl acetate		150	0.20	5.30
linalool		154	2.20	34.10
oxyphenolon	middle note	164	11.90	1.70
2-tridecanone		198	0.02	5.50
2-pentadecanone	bottom note	226	69.00	33.50
ethyl myristate		256	14.80	8.50

^c SPME fibers and ancillary equipment from SUPELCO, Bellefonte, PA

^d Shalimar is a registered trade name of Guerlain SA, Paris, France

the extent of 0.2% in the oil, was dramatically higher (eight times greater) in the aura. Similarly, methyl ionone, another high-boiling compound used for the first time in Shalimar, doubled in the aura. Most interesting is the musk xylol, the highest-boiling compound, which is found only in trace amounts in the oil but was 0.3% of the aura.

This shows that the aura of a fragrance is composed not only of highly volatile molecules, but also of both low-boiling and higher-boiling compounds that are highly diffusive.

Amarige: Next, we studied a very successful 1990s fragrance, Amarige,^e introduced by Givenchy.

Table III shows that top note constituents linalool, benzyl acetate and styralyl acetate were greater in the aura than the oil. At the same time, medium-boiling components such as cashmeran, which could not even be detected in the fragrance oil, showed up in the aura to the extent of 0.5%. Similarly, other high-boiling compounds, such as Bacdanol,^f Cedramber^g and iso E Super,^h were twice as great in the aura.

So-called bottom note compounds included Ambroxⁱ and muskalactone (cyclopentadecanolide), both among the highest boiling of fragrance chemicals. They showed their presence in the aura in appreciable amounts, indicating that they are very diffusive molecules that play important roles in the first impression of Amarige.

A unisex fragrance: Next, we studied a highly successful unisex commercial fragrance (Table IV).

As we expected, ethyl linalool and linalyl acetate show greater percentages in the aura than in the oil due to their high diffusivity. But, interestingly, middle-boiling-range compounds such as floralozone, cyclogalbaniff and β -ionone all were a factor of three greater in the aura.

High-boiling compounds such as Polysantol,^j iso E Super and Ambrox were present in the aura in appreciable quantities due to their diffusivity. Once again, note the poor diffusivity of some materials.

A feminine fragrance: The next perfume we studied was a very successful modern feminine fragrance also introduced in the 1990s (Table V).

This unique feminine note depends on two very volatile chemicals (aldehyde AA and methyl phenyl acetate), sev-

^e Amarige is a registered trade name of Parfums Givenchy Lavallois-Perret, France

^{f,g,h} Bacdanol, Cedramber and iso E Super are registered trade names of International Flavors & Fragrances Inc., New York, NY

ⁱ Ambrox is a registered trade name of Firmenich SA, Geneva, Switzerland

^j Polysantol is a registered trade name of Firmenich SA, Geneva, Switzerland

Table II. Shalimar composition: oil vs. aura on skin

Component		Oil (%)	Aura on skin after 60 min (%)
limonene	top note	30.0	20.4
linalool		1.7	17.9
linalyl acetate		9.9	21.6
ethyl vanillin	middle note	0.2	1.6
coumarin		1.7	7.8
methyl ionone		1.1	2.1
musk xylol	bottom note	trace	0.3

eral highly diffusive middle-boiling compounds (ethyl linalool, diphenyl ether, cyclogalbaniff and methyl ionone) and several high-boiling yet very diffusive materials (iso E Super, Ambrox and cyclopentadecanolide).

Women's fragrance: Finally, we studied a very sensuous and highly successful women's fragrance, recently launched in both Europe and the U.S.

Table VI shows why this fragrance is so diffusive. The aura contains three extremely diffusive chemicals that cannot even be detected in direct analysis of the perfume oil; namely, lolitol, passionfruit compound and methyl octin carbonate. In addition, the aura contains greater amounts of several other very diffusive molecules, including Givescione,^k Floralazone,^l α - and β -damascone, cis-jasmone and undecavertol. It also contains the already discussed ethyl linalool, linalyl acetate, methyl ionone and cyclopentadecanolide.

^k Givescione is a registered trade name of Givaudan-Roure SA, Vernier, Switzerland

^l Floralazone is a registered trade name of International Flavors & Fragrances Inc., New York, NY

Table III. Amarige composition: oil vs. aura on skin

Component		Oil (%)	Aura on skin after 60 min (%)
linalool	top note	1.7	17.9
benzyl acetate		4.9	22.7
styrallyl acetate		1.2	9.7
cashmeran	middle note	-	0.5
Bacdanol		0.2	0.5
Hedione		29.9	4.9
Cedramber		1.5	4.9
iso E Super	bottom note	7.1	12.1
Ambrox		0.2	0.1
benzyl salicylate		32.5	1.1
muskalactone		0.9	0.4

Table IV. Unisex fragrance composition: oil vs. aura on skin

Component	Oil (%)	Aura on skin after 60 min (%)	Increase or decrease in aura (factor)
ethyl linalool	0.7	2.8	4.00
linalyl acetate	10.4	36.0	3.00
Floralazone	0.1	0.3	3.00
Cyclogalbaniff	0.2	0.7	3.00
dihydro myrcenol	5.8	10.6	2.00
linalool	7.7	11.5	1.50
limonene	4.5	1.4	0.30
β -ionone	2.5	6.2	3.00
polysantol	0.2	0.4	2.00
iso E Super	4.8	4.8	-
Ambrox	0.7	0.4	0.50
Hedione	25.9	5.0	0.20
Galaxolide	5.5	0.6	0.10
Tonalid	3.3	0.1	0.03

Conclusions: From these studies, we have proved beyond a doubt that the first impression of a fragrance is not due only to the highly volatile so-called top note chemicals, as once believed. Instead, it consists of a combination of highly volatile, middle-boiling molecules, as well as high-molecular-weight, high-boiling compounds possessing a quality called high diffusivity.

Table VII summarizes our finding with regard to the aroma molecules, from lowest-boiling to highest-boiling, that appear simultaneously in the aura of any fragrance in which they are present. Relatively higher-molecular-weight sulfur compounds such as 8-para-menthane thiol and passionfruit compound play key roles in many fragrances at very low levels, not only due to their extremely low odor thresholds but also because of their very high diffusivities.

At the same time, one sees in the aura of fragrances various so-called middle note compounds (ethyl vanillin, the ionones, cashmeran and Cedramber). One also sees bottom note

compounds (sesquiterpenic materials such as cedrol; Polysantol; very-high-boiling amber and musk compounds such as Ambrox and cyclopentadecanolide). These compounds constitute the real first impression of any fragrance.

On the other hand, many compounds used throughout the fragrance industry in ton quantities are relatively non-diffusive and play little role in the aura of a fragrance.

One could easily imagine that a creative perfumer, selecting from compounds mentioned here, could easily create a characteristic fragrance that is both long-lasting and highly diffusive.

Effect of Skin on Emission of Fragrance

It is commonly believed that the smell of a fragrance differs greatly from skin to skin. Therefore, we studied whether the fragrance composition changes depending upon the skin to which it is applied. Of course, we used SPME technology to study this interesting phenomenon. Now, the question of what kind of skin should be studied?

A number of investigators using Caucasian skin have stressed the importance of the skin's fat and moisture con-

Table V. Feminine fragrance composition: oil vs. aura on skin

Component	Oil (%)	Aura on skin after 60 min (%)	Increase or decrease in aura (factor)
Aldehyde AA	0.05	0.80	16.00
methyl phenyl acetate	0.02	0.20	10.00
ethyl linalool	5.00	30.00	6.00
diphenyl ether	0.01	0.04	4.00
Cyclogalbaniff	0.20	0.90	4.00
methyl ionone	2.50	9.50	4.00
ethyl acetoacetate	1.80	3.40	2.00
iso E Super	2.60	2.10	0.81
Ambrox	0.20	0.10	0.50
Hedione	18.00	2.40	0.13
cyclopentadecanolide	4.80	1.20	0.25
Galaxolide	14.00	0.90	0.07

Table VI. Recent women's fragrance composition: oil vs. aura on skin

Component	Oil (%)	Aura on skin after 60 min (%)	Increase or decrease in aura (factor)
Lolitol	-	0.10	-
passionfruit compound	-	0.01	-
methyl octin carbonate	-	0.10	-
Givescone	0.10	1.20	12.00
Floralazone	0.01	0.10	10.00
α -damascone	0.10	0.50	5.00
β -damascone	0.04	0.20	5.00
ethyl linalool	1.40	6.00	4.00
undecavertol	0.30	1.00	3.00
linalyl acetate	2.00	7.80	3.00
cis-jasmone	0.10	0.20	2.00
methyl ionone	2.00	4.60	2.00
cyclopentadecanolide	0.90	0.20	0.25
Galaxolide	8.00	1.00	0.13

Table VII. Summary of diffusive fragrance molecules

Fragrance molecule	Mol. wt.
<i>Extremely diffusive</i>	
passionfruit compound	160
methyl octin carbonate	168
8-para-methane thiol	170
<i>Diffusive top note</i>	
Aldehyde AA	138
Lolitol	144
linalool	154
dihydro myrcenol	156
styrallyl acetate	164
cis-jasmone	164
ethyl linalool	168
diphenyl ether	170
linalyl acetate	196
<i>Diffusive middle note</i>	
coumarin	146
ethyl vanillin	166
Floralazone	190
α - and β -damascone	192
ionone	192
methyl ionone	206
Cyclogalbaniff	198
Cashmeran	206
Cedramber	236
<i>Diffusive bottom note</i>	
cedrene	204
cedrol	222
α - and β -Santalol	220
Polysantol	222
patchouli alcohol	222
iso E Super	234
Ambrox	236
cyclopentadecanolide	240

tent. We think these are important considerations. But, we selected the skin from a global perspective. In other words, we have selected people from different parts of the world:

- A professional Indian woman in her mid-40s (Figure 4a).
- A Jamaican woman in her mid-40s who works in a clerical position (Figure 4b).
- A light-skinned Caucasian working woman in her mid-30s (Figure 4c).
- A managerial Caucasian woman in her mid-50s (Figure 4d).

Each of these women was first placed on a bland diet and asked to maintain her normal working conditions without too much physical activity. The women also were asked to clean their skin by washing with non-fragranced soap. We performed a series of tests on these four women using two different fragrances.

A unisex fragrance: The first perfume we selected for our skin-effect studies was the unisex fragrance whose aura was discussed earlier. Since this was a unisex fragrance, we also studied the skin effect of five different male lab workers between 20 and 55. One was Jamaican, one was Indian and the rest were Caucasian.

We applied two drops of the fragrance to the forearm skin of each subject, then collected the auras for one hour

Figure 4. Female subjects in tests of fragrance emission from various skins: a) Indian mid-50s; b) Jamaican mid-50s; c) Caucasian mid-30s; d) Caucasian mid-50s

starting immediately after application. This experiment was repeated twice. Table VIII shows the composition of fragrance from each skin.

One can easily see that among the so-called highly volatile components there is essentially no change from skin to skin. Among the women, for instance, the limonene composition is in the 9–13% range; linalool, the second major component, is in the 14–15% range; the major constituent, linalyl acetate, basically did not change at all. Similarly, none of the minor components changed appreciably.

The composition of the auras for the middle and bottom notes showed basically no change from skin to skin. The slight changes observed in the case of β -ionone, coumarin and Tonalid[™] for one of the Caucasian women (PAM) is considered within the range of experimental error.

When we compare the men's results with those of the women, we see that there were no significant differences from men to women for the components of the unisex fragrance. It is obvious that different skin types do not significantly affect the emission of this particular fragrance.

A women's fragrance: The next perfume we studied for

[™] Tonalid is a registered trade name of PFW Limited, Perivale, Greenford, Middlesex, UK

the effect of skin on the emission of fragrance was a women's fragrance that was introduced in the U.S. in 1994. In this case, we included one young male with the four women.

The auras were collected from the five different subjects for one hour immediately after application of the fragrance. As one can see from Table IX, there was no significant change in the composition of the initial auras from the skin of all of these individuals.

Since this particular fragrance lasts longer on the skin than the unisex fragrance, we also collected the auras 45 minutes after applying the fragrance. Table X shows that after 45 minutes none of the skins retained any of the lower-boiling components. However, for the higher-boilers, there are some significant differences.

Conclusions: We studied two fragrances that are of totally different composition. In the initial auras of the fragrances we observed basically no changes based on skin differences. However, when evaluated approximately one hour after application, one of the fragrances (the women's fragrance) showed some differences in the composition of the higher-boiling constituents of the auras.

Therefore, we may assume that skin may have some effect, depending on the composition of the fragrance. To

Table VIII. Unisex fragrance: comparison of emission (%) from selected skins
(Applied two drops to skin and immediately collected for 1 hour)

Component	Men					Women			
	BK	RW	JK	DA	RM	PAM	CB	CP	SM
cis-3-hexenol	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
limonene	9.4	11.0	9.4	10.9	10.5	13.3	10.1	9.3	10.7
dihydro myrcenol	8.6	8.3	7.7	8.3	7.8	9.3	9.2	10.3	9.5
cis-3-hexyl methyl carbonate	0.2	0.3	0.2	0.3	0.2	0.3	0.3	0.3	0.3
linalool	13.4	13.3	12.3	13.5	12.5	13.9	13.8	15.4	14.1
benzyl acetate	0.3	0.4	0.3	0.4	0.3	0.4	0.4	0.4	0.4
4-terpinenol	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
ethyl linalool	1.9	1.8	1.9	1.9	1.8	1.8	1.8	2.0	1.8
C ₁₀ aldehyde	-	-	-	-	-	-	-	0.1	-
citronellol	0.6	0.5	0.6	0.5	0.5	0.4	0.4	0.4	0.4
carveol	0.4	0.5	0.5	0.4	0.5	0.3	0.5	1.0	0.5
allyl amyl glycolate	0.2	0.3	0.2	0.3	0.3	0.2	0.2	0.3	0.2
citral	0.2	0.2	0.2	0.2	0.2	0.1	0.2	0.2	0.2
linalyl acetate	31.5	32.7	34.8	33.7	32.9	32.0	31.3	31.0	30.9
α -terpineol acetate	0.5	0.4	0.5	0.4	0.5	0.3	0.3	0.3	0.3
neryl acetate	1.0	1.0	1.0	0.9	1.0	1.1	1.0	1.0	1.0
geranyl acetate	1.5	1.5	1.5	1.4	1.5	1.7	1.6	1.6	1.6
cis-jasmone	0.2	0.2	0.1	0.2	0.1	0.1	0.2	0.2	0.1
coumarin	0.1	0.1	0.1	0.1	0.1	0.0	0.1	0.1	0.1
Cyclogalbaniff	0.3	0.3	0.3	0.3	0.3	0.1	0.3	0.3	0.2
Floralozone	0.1	0.1	0.2	0.1	0.2	0.1	0.2	0.2	0.2
caryophyllene	0.7	0.6	0.8	0.6	0.6	0.7	0.6	0.6	0.6
β -ionone	4.3	3.5	4.4	3.4	4.1	1.9	4.2	3.6	4.1
Polysantol	0.2	0.2	0.2	0.2	0.2	0.1	0.2	0.2	0.2
Hedione	2.1	1.5	1.9	1.5	2.1	0.1	0.5	0.3	0.6
Lilial	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	1.6
Helional	0.4	0.3	0.3	0.2	0.4	0.5	1.4	1.2	1.2
iso E Super	3.0	2.3	2.9	2.3	2.8	1.1	2.3	1.7	2.0
Ambrox	0.2	0.2	0.2	0.2	0.2	0.1	0.2	0.1	0.1
Galaxolide	0.8	0.6	0.6	0.6	0.7	0.1	0.2	0.2	0.2
Tonalid	0.1	0.1	0.1	0.1	0.1	0.0	0.1	0.1	0.1

Table IX. Longer-lasting women's fragrance: comparison of emission (%) from selected skin (applied 2 drops to skin and immediately collected for 1 hour)

Component		SM	CP	PAM	CB	BK
dihydro myrcenol	top note	3.4	2.8	3.3	2.8	2.1
phenyl ethyl alcohol		5.9	7.4	4.2	4.8	4.6
benzyl acetate		20.5	20.3	20.3	24.9	18.1
ethyl linalool		1.9	1.8	1.8	1.7	1.6
citronellol		6.4	6.6	5.2	3.4	5.7
phenyl ethyl acetate		1.2	1.5	0.9	1.1	1.1
linalyl acetate		0.9	0.8	1.1	1.3	0.9
dimethyl octyl acetate		2.1	2.1	2.5	3.1	2.3
citronellyl acetate		11.2	11.4	14.1	14.2	12.7
geranyl acetate	middle note	12.7	13.1	15.8	14.0	14.6
γ -methyl ionone		15.0	15.0	16.5	13.4	18.1
Lilial		0.8	0.8	0.6	0.5	1.0
Baccanol		0.2	0.2	-	0.3	0.3
iso E Supe	bottom note	12.2	11.2	12.9	10.7	12.5
cyclopentadecanolide		-	-	-	-	-
Galaxolide		-	-	-	-	-

SM = 45-year-old Indian woman; CP = 47-year-old Jamaican woman; PAM = 36-year-old Caucasian woman; CB = 53-year-old Caucasian woman; BK = 20-year-old Caucasian man

Table X. Longer-lasting women's fragrance: comparison of emission (%) from selected skin (applied 2 drops to skin, waited 45 minutes, then collected for 1 hour)

Component		SM	CP	PAM	CB	BK
dihydro myrcenol	top note	-	-	-	-	-
phenyl ethyl alcohol		0.6	-	0.5	-	-
benzyl acetate		0.5	-	-	-	-
ethyl linalool		-	-	-	-	-
citronello		-	-	-	-	1.1
phenyl ethyl acetate		-	-	-	-	-
linalyl acetate		-	-	-	-	-
dimethyl octyl acetate		0.3	-	0.4	-	-
citronellyl acetate		4.0	-	5.2	-	1.6
geranyl acetate	middle note	9.3	5.2	10.8	2.4	3.9
γ -methyl ionon		22.7	17.6	24.5	11.5	15.4
Lilial		2.8	3.7	2.8	3.4	2.9
Baccanol		1.5	-	1.5	2.2	2.3
iso E Supe	bottom note	38.6	53.2	35.6	56.2	48.1
cyclopentadecanolide		-	1.7	0.8	1.9	1.5
Galaxolide		-	-	0.8	2.4	1.9

SM = 45-year-old Indian woman; CP = 47-year-old Jamaican woman; PAM = 36-year-old Caucasian woman; CB = 53-year-old Caucasian woman; BK = 20-year-old Caucasian man

really establish the effect of skin, obviously we must do more studies on various fragrances.

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