finally succeeded. The long-established view that the transition metal to carbon bond is weak is now untenable and must be discarded. We can expect other types of transition metal alkyls to be made in due course and can hope that in addition to their own intrinsic interest some of them may find uses in catalytic or other syntheses. The use of titanium and zirconium alkyls in alkene polymerization (*13*) and the use of alumina treated with hexamethyltungsten for alkene metathesis (*17*) give good grounds for optimism.

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Physicochemical Correlates of Olfactory Quality

A series of physicochemical variables are weighted mathematically to predict olfactory quality.

Susan S. Schiffman

A model does not exist in olfaction which strictly relates quantitative measures of olfactory quality with quantitative physicochemical measures. Although each of the more notable olfactory theories makes useful suggestions, none provides a basis for developing a complete model which could accurately order all olfactory stimuli.

Olfactory Theories

Amoore (*1*) suggested that the shape and size of a molecule are the crucial physicochemical parameters for stimulation in olfaction. He proposed receptor sites which relate to molecular stimuli in a "lock and key" fashion. Molecules of similar size and shape are expected to have similar odor quality. In testing this hypothesis, Amoore and Venstrom (*2*) found significant correlations between odor quality and a hand-calculated index of molecular size and shape for five classes of odors (ethereal, camphoraceous, musky, floral, and minty). Amoore (*3*) found a correlation of 0.90 between odor quality and a computer-generated molecular shape index when 25 substances were compared with benzaldehyde (almond odor).

Wright (*4*) has challenged Amoore's results, indicating that it is inappropriate to represent anything as complicated as a three-dimensional molecular shape by an index consisting of a single number. Many different three-dimensional profiles could share the same molecular shape index. Wright's theory (*5*) is that the mechanism for stimulation of olfactory receptors is low energy molecular vibrations. Molecules with similar vibrational frequency patterns are expected to have similar odor quality. In testing this hypothesis Wright and Robson (*6*) determined that a pattern of frequencies in the far infrared spectra correlated highly with the bitter almond odor. Wright and Brand (*7*) found a relationship between pheromone activity and molecular vibration in insects.

Dravnieks and Laffort (*8*) have related four factors concerned with intermolecular interaction forces (an apolar factor, a proton receptor factor, an electron factor, and a proton donor factor) to both quantitative and qualitative odor discrimination in human beings. Dravnieks (*9*) has also devised a chemical building block model which relates the hedonic qualities of olfactory stimuli to a series of physicochemical variables.

Other theories concerning the process of olfactory stimulation have also been proposed: Beets' theory of profile functional groups (*10*); Henning's emphasis on the position of functional groups in relation to the rest of the molecule (*11*); and Mozell's suggestion that the olfactory epithelium is similar to a gas chromatograph (*12*).

In spite of these efforts, the ability to predict olfactory quality from quantitative physicochemical measures for a wide range of olfactory stimuli has yet to be achieved. The purpose of this article is to demonstrate a procedure which, when extended to a wide range of stimuli, can provide us with means for predicting olfactory quality from physicochemical parameters.

Multidimensional Scaling

The recently developed methodology of multidimensional scaling can aid in deepening our understanding of the relationship between psychological dimensions in olfaction and physicochemical variables. Multidimensional scaling procedures can order olfactory sensations in spaces to reveal relationships and distances among the olfactory stimuli based on experimental mea-

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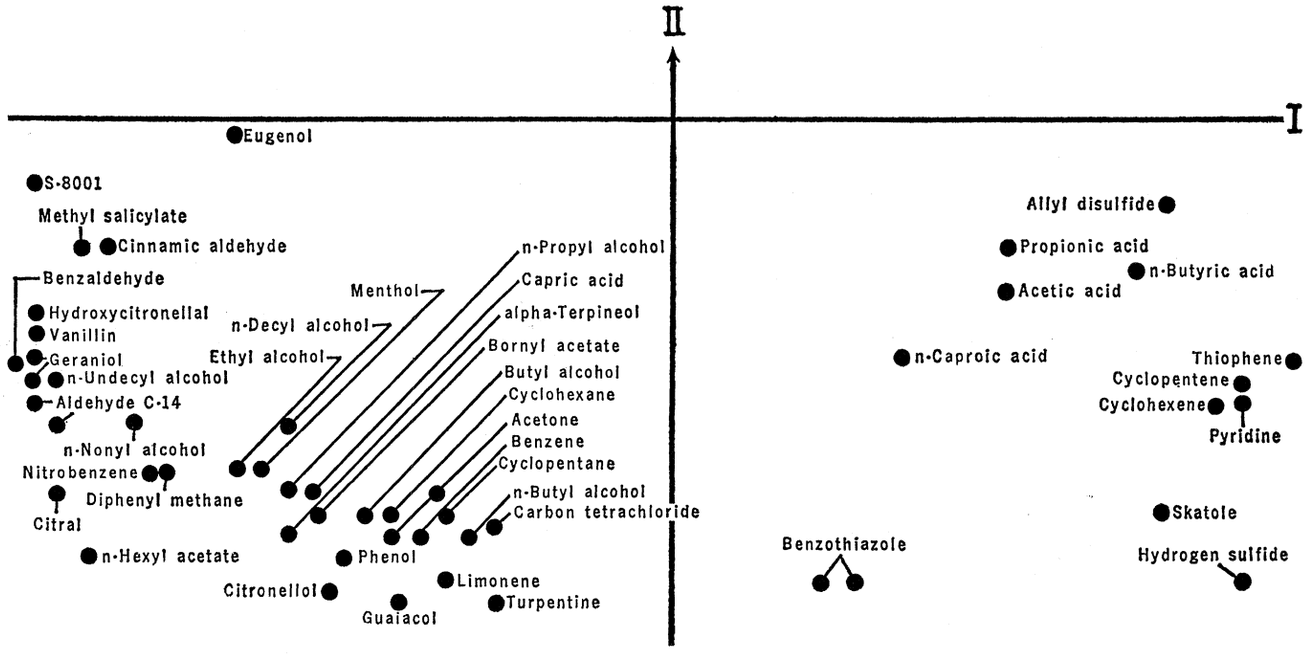

Fig. 1. Two-dimensional solution representing Wright and Michels' psychophysical olfactory data (*17*) for 50 stimuli of which 5 are duplications. The solution was obtained by Guttman's method (*18*). Substances found by Wright and Michels to be highly correlated with each other on the basis of olfactory quality are proximately located in this space. The more pleasant stimuli are located in the subset on the left; the unpleasant stimuli are in the subset on the right.

sures. The input for multidimensional scaling procedures consists of measures of similarity. For example, if two substances are judged by human subjects to have similar olfactory quality, they will be placed near each other in a multidimensional quality space. Stimuli judged to be dissimilar will be located distant from one another.

Multidimensional scaling techniques have been successfully applied in both color vision and gustation. Multidimensional scaling of both psychophysical data on similarities between colors (*13*), and spectral absorption data for single cones in the goldfish retina (*14*), produced a color circle. In this laboratory (*15*), we found a three-dimensional solution revealing five gustatory groups (sweet, sour, salty, bitter, and alkaline) when nonmetric multidimensional scaling techniques were applied to psychophysical gustatory data. We also found that this multidimensional space could be described by the molecular weight, *p*H, and hedonic qualities of the stimuli. The arrangement of the gustatory stimuli in our model, based on psychophysical measures, is similar to the ordering of the stimuli based on neural measures achieved by Doetsch and Erickson (*16*) who used multidimensional scaling techniques.

There are two sets of olfactory psychophysical data to which multidimensional scaling techniques may be applied. Wright and Michels (*17*) compared 50 olfactory stimuli (5 of these were duplications) with 9 odorant standards which ranged widely in quality. The 50 odorants were then correlated across the standards with the assumption that odorants having similar smell quality would be highly correlated. The 50 by 50 correlation matrix was factor analyzed by Wright and Michels, and eight factors were obtained. This correlation matrix was reanalyzed according to Guttman's general nonmetric multidimensional scaling technique (*18*). The results (*19*) from multidimensional scaling are shown in Figs. 1, 2, and 3.

Figure 1 demonstrates that Wright and Michels' data can be represented by a two-dimensional solution (accounting for 91 percent of the data). It divides the stimuli roughly into two groups; the larger subset on the left is affectively more pleasant than the one on the right. Döving (*20*) achieved a solution similar to that in Fig. 1 for Wright and Michels' data using Kruskal's multidimensional scaling routine (*21*).

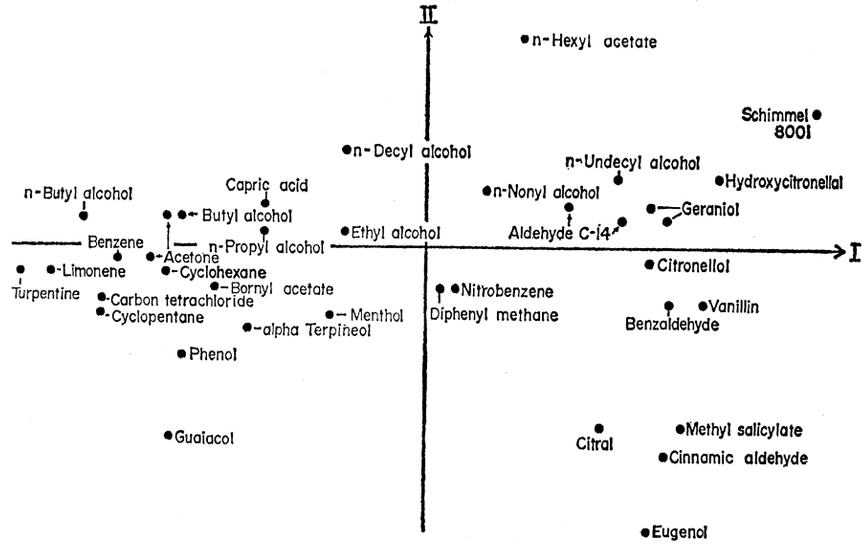

Fig. 2. Two-dimensional arrangement achieved by Guttman's method (*18*) upon reanalysis of the affectively more pleasant stimuli in Fig. 1.

Because multidimensional scaling procedures aim for minimum dimensionality, the case in which there are only two major clusters of stimuli can present a problem. These procedures tend to drive the clusters apart; as a consequence, there can be some loss in the internal relationships within each of the groups. For this reason the two groups were reanalyzed separately to regain any lost internal relationships. Figure 2 illustrates the reanalysis of the more pleasant stimuli in the subset on the left in Fig. 1, while Fig. 3 shows the reanalysis of the more unpleasant stimuli in the right-hand subset. It can be seen by comparing Figs. 2 and 3 that only slight distortion occurred in Fig. 1. Rotations and inversions from Fig. 1 to Figs. 2 and 3 are not meaningful.

Another psychophysical study was done by Woskow (*22*) in which 25 olfactory stimuli were used; many of these stimuli were identical with those used by Wright and Michels. Woskow compared each of the 25 stimuli with one another and thus achieved a measure of similarity for all possible pairs, In his analysis, Woskow used an early multidimensional scaling procedure in which it is assumed that the data are metric (there is no reason to make this assumption), and he found that three dimensions account for 80 percent of the variance. Woskow's data were reanalyzed by the more recently developed nonmetric technique of Guttman. Figure 4 shows the two-dimensional space resulting from this reanalysis which accounts for 84 percent of the variance. Since the reanalysis accounts for more variance than Woskow's metric solution, and because the relationships among the stimuli are not lost in compressing them from three to two dimensions, the two-dimensional space in Fig. 4 must be considered the appropriate solution.

The two-dimensional solutions achieved by application of Guttman's multidimensional scaling procedure to both Wright and Michels' data and Woskow's data are somewhat similar. The spaces in Figs. 1 and 4 both consist of two groups, a more pleasant group (in both) and a more unpleasant group (in both). The stimuli used in common by Woskow and by Wright and Michels (for example, vanillin, benzaldehyde, methyl salicylate, eugenol, and ethanol on the left, and skatole, butyric acid, acetic acid, and pyridine on the right) fall at closely analogous points in Figs. 1 and 4.

The similarity of these two solutions, each obtained from different numbers of stimuli and by different psychophysical techniques, leads me to conclude that a two-dimensional space adequately describes the relationships among a wide range of olfactory stimuli.

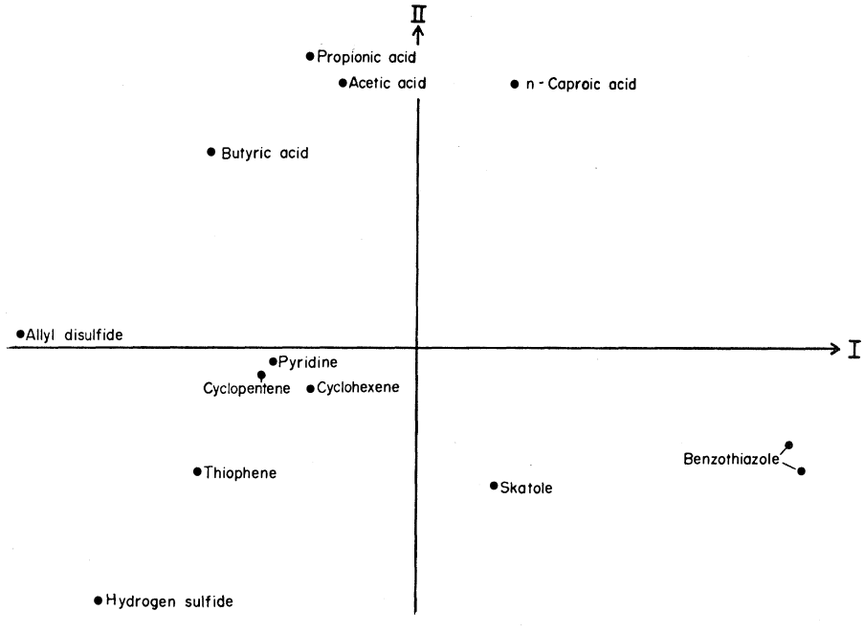

Fig. 3. Two-dimensional arrangement achieved by Guttman's method (*18*) upon reanalysis of the unpleasant stimuli in Fig. 1.

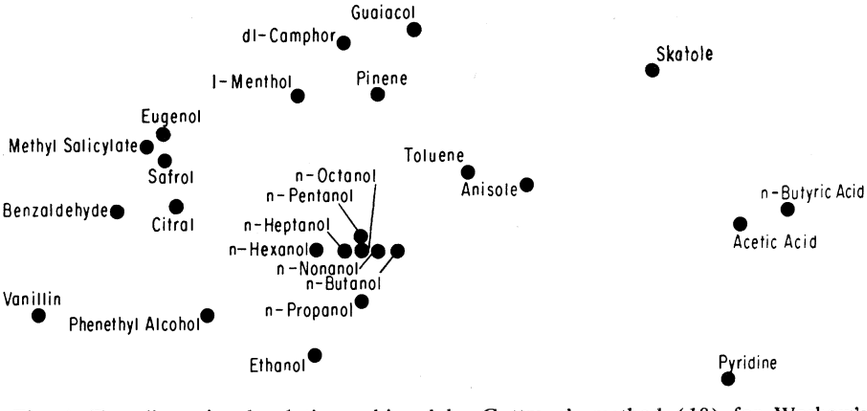

Fig. 4. Two-dimensional solution achieved by Guttman's method (*18*) for Woskow's psychophysical olfactory data (*22*). This arrangement is similar to that in Fig. 1.

Olfactory Dimensions

The arrangements of olfactory stimuli based on measures of similarity (Figs. 1 to 4) raise the obvious question of what psychological and physicochemical parameters describe these spaces. Because Wright and Michels (*17*) employed more stimuli in their experiment than Woskow (*22*), I used the spaces in Figs. 2 and 3 to search first for psychological dimensions and then for physicochemical correlates of olfactory quality. (Figures 2 and 3 were used rather than Fig. 1 simply for visual ease in examining the trends.)

Psychological dimensions. To search for the psychological dimensions, the range of stimuli in Figs. 2 and 3 were examined for the smell qualities traditionally assigned to them in Moncrieff's *The Chemical Senses* (*23*) or in *The Merck Index* (*24*). According to these sources, the quality of the compounds shown in Fig. 2 changes from right to left, from a flowery, fruity, or generally quite pleasant odor to one which is more spiritous and resinous. From the top to the bottom there is a slight tendency toward an increase in sharpness or spiciness in the quality. The descriptions of the quality of the com-

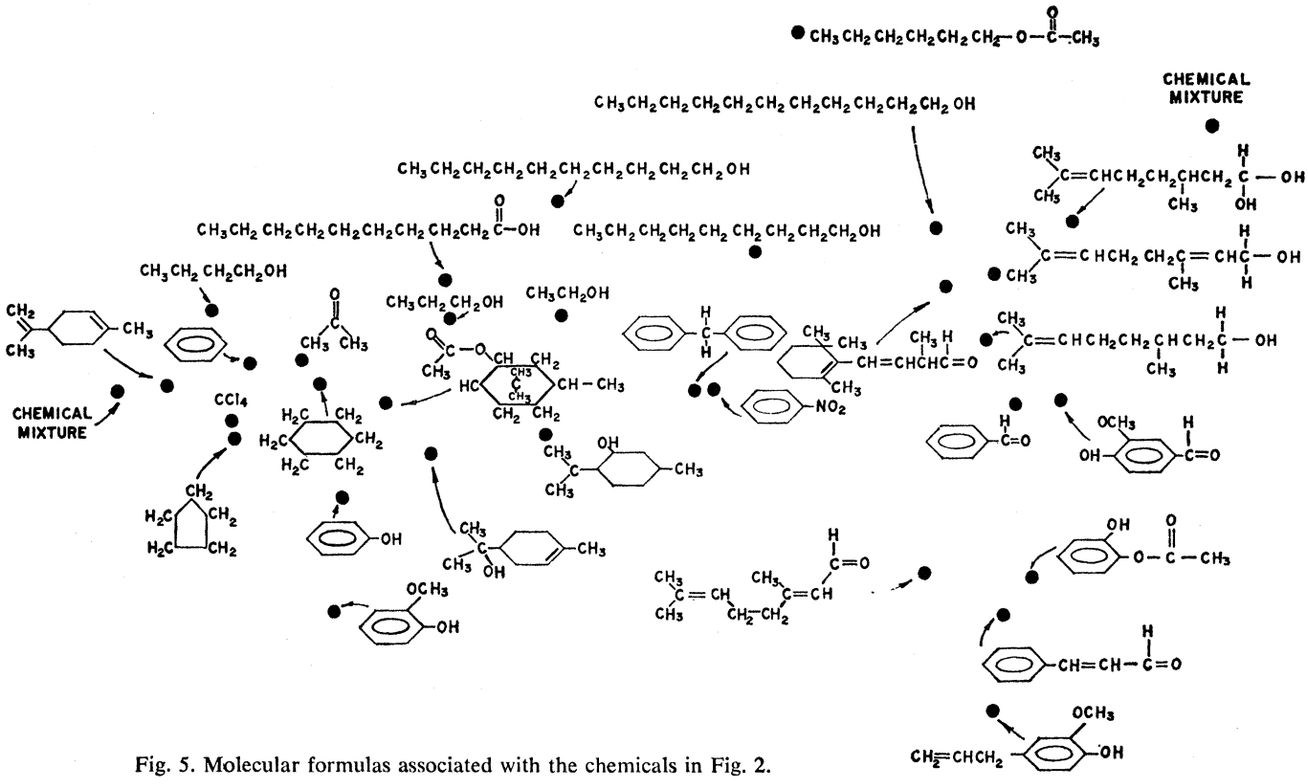

Fig. 5. Molecular formulas associated with the chemicals in Fig. 2.

pounds shown in Fig. 3 are more nebulous and it is difficult to get as good a picture of the change in quality throughout the space. For example, propionic acid is merely described as a "strong, penetrating" odor; pyridine is considered "rank, unpleasant, and burnt," and cyclopentene is classified as "unpleasant and pungent."

Although generalizations must be tempered by the fact that only a few representative stimuli for each chemical class were used in these spaces, overall it is concluded that the use of adjectives alone to order olfactory stimuli (a procedure employed previously by many classifiers) cannot provide a stable, clear, and unequivocal ordering of olfactory quality. There are no clear psychological groups or classes of stimuli, merely trends. The positioning of the olfactory stimuli at specific points in a space by multidimensional scaling techniques gives a much more stable ordering of smell quality based on quantitative measures than the assignment of adjectives, especially for unpleasant stimuli.

Physicochemical dimensions. To search for the physicochemical correlates to smell quality, I began by examining the molecular formulas of the olfactory stimuli. Figures 5 and 6 demonstrate the molecular structures of the stimuli in Figs. 2 and 3, respectively. The molecular formulas were written in such a way as to describe approximately the size and shape of the substances. Although there are certainly some trends in the spaces, especially with regard to cyclic molecules, it would be impossible to conclude from even this limited sample of chemicals that the stereochemical properties alone determine olfactory quality. Each of the molecules in Figs. 5 and 6 was built to scale with models, and Amoore's theory (*1*) was not confirmed.

Changes in a molecule which do not alter its size and shape appreciably have a profound impact on smell quality. For example, if one compares the triangular relationship among benzene, cycloheptane, and cyclopentane on the more pleasant side of the space (Fig. 5) with the triangular relationship among pyridine, cyclohexene, and cyclopentene on the more unpleasant side of the space (Fig. 6), one finds that the close relationships among the three

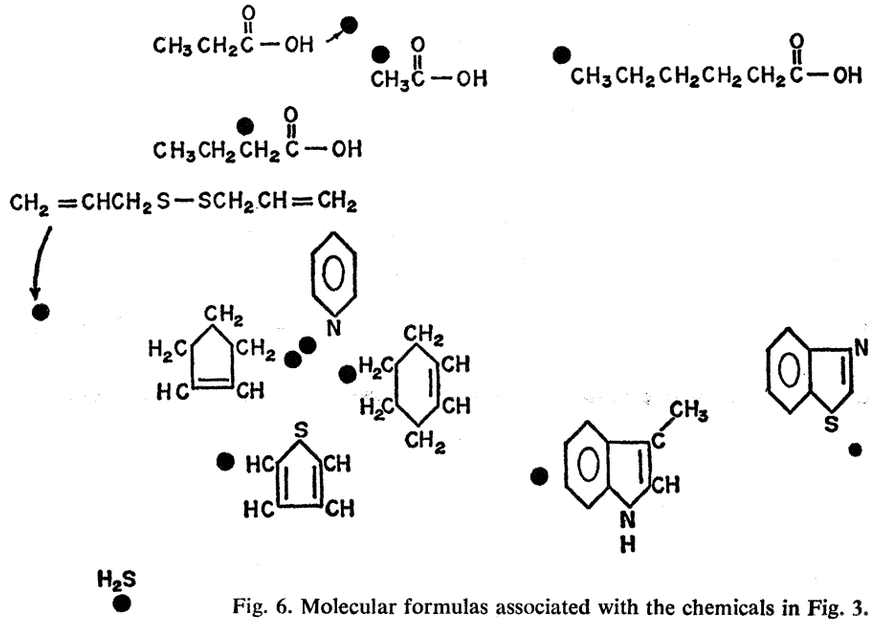

Fig. 6. Molecular formulas associated with the chemicals in Fig. 3.

Table 1. Weights that were applied to standard scores for physicochemical variables to achieve the regenerated space in Fig. 7. Means and variances for these variables are also shown. Functional groups were coded by their number in a molecule; thus, benzaldehyde was coded "1" and the mean number of aldehyde groups for all the molecules in Fig. 7 is 0.10. Cyclic compounds were coded "1" while noncyclic compounds were coded "0."

Physicochemical variable	Mean	Variance	Weight
Molecular weight	116.57	1788.64	6.24
Number of double bonds	0.74	0.55	0.51
Phenol	0.13	0.11	2.33
Aldehyde	0.10	0.09	3.21
Ester	0.05	0.05	0.24
Alcohol	0.26	0.19	2.54
Carboxylic acid	0.13	0.11	5.50
Sulfur	0.08	0.07	3.44
Nitrogen	0.08	0.07	3.15
Benzene	0.33	0.27	−0.14
Halogen	0.03	0.02	−0.34
Ketone	0.03	0.02	−0.19
Cyclic	0.31	0.21	4.56
Mean Raman intensity			
Below 175 cm^{-1}	0.51	3.14	0.01
176–250 cm^{-1}	2.36	9.30	3.57
251–325 cm^{-1}	1.65	7.10	−0.75
326–400 cm^{-1}	1.56	5.74	3.81
401–475 cm^{-1}	2.10	7.23	1.65
476–550 cm^{-1}	1.54	5.22	−3.63
551–625 cm^{-1}	2.07	7.09	−0.69
626–700 cm^{-1}	1.07	5.14	−1.16
701–775 cm^{-1}	2.36	11.01	0.07
776–850 cm^{-1}	4.36	13.84	3.04
851–925 cm^{-1}	3.44	15.77	0.24
926–1000 cm^{-1}	2.06	8.29	0.36

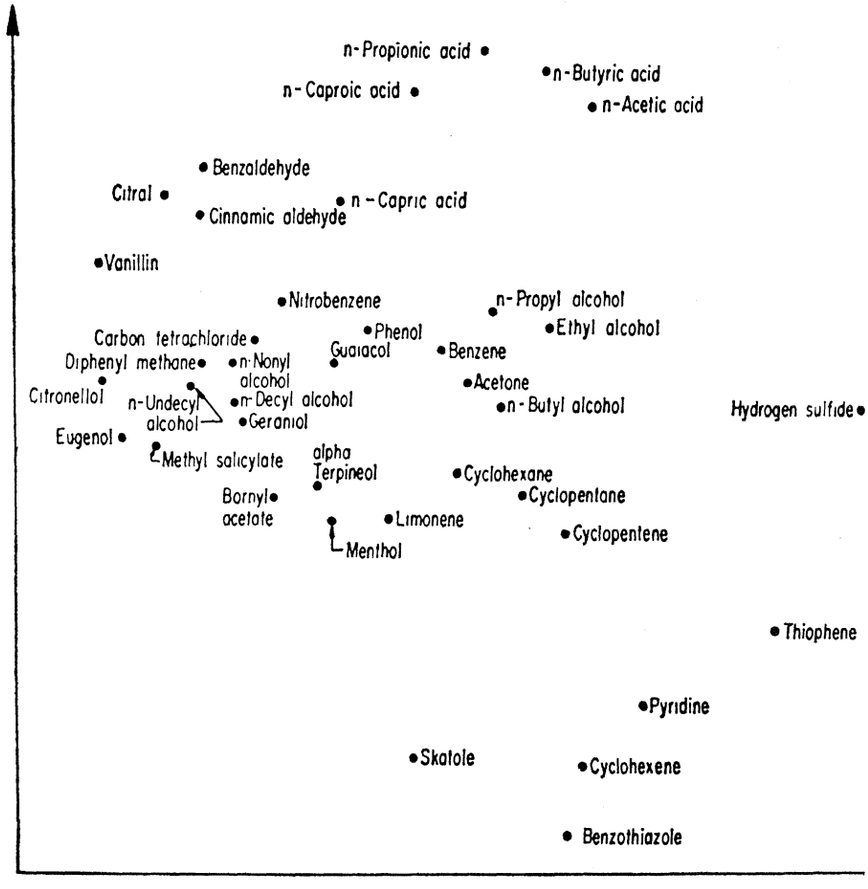

Fig. 7. Space regenerated from weighting the physicochemical variables shown in Table 1 in an attempt to reproduce the psychological space illustrated in Fig. 1.

compounds are retained in both instances, although they are shifted radically in olfactory quality when a nitrogen is substituted for a carbon in the benzene ring in the case of pyridine and double bonds are added in the cases of cyclohexene and cyclopentene.

Next I examined the relationships among the functional groups of the compounds in Figs. 5 and 6. The functional groups seem to be good distinguishing factors for olfactory quality. The aldehydes, esters, alcohols, phenols, ketones, and ethers fall in the more pleasant side of the space (Fig. 5) while the sulfur- and nitrogen-containing compounds (not oxygenated), and the light carboxylic acids, are located on the more unpleasant side of the space (Fig. 6). One heavy, less volatile carboxylic acid fell in the more pleasant space.

There is also order within each of the spaces. For example, in Fig. 5 the aldehydes tend to group together to the right. The alcohols are distributed along the top of the space. In Fig. 6 the carboxylic acids group tightly, separated from the sulfurs and nitrogens. Cyclic compounds tend to group in both Figs. 5 and 6.

Other aspects of the molecules in the spaces were examined as well. There are some trends for molecular weight (and boiling point, with which molecular weight is highly correlated). Among the more pleasant stimuli in Fig. 5, those substances with low molecular weight and low boiling point tend to be located toward the left. For the more unpleasant stimuli, with the exception of allyl disulfide, the same trends hold —the heavier compounds are located toward the right in Fig. 6.

There were no trends for freezing point, solubility in water, dipole moment, and number of double bonds for the stimuli in these spaces. Since all of these stimuli, for which data existed, were soluble in ether, this may imply that fat (ether) solubility is necessary for stimulation to occur. The relative ether solubilities were not used here as factors to discriminate between molecules on their olfactory quality.

The Raman spectra of these molecules from 100 to 1000 reciprocal centimeters were found to contain much information that could be correlated with the pleasantness or unpleasantness of the molecules. Wright and Michels (*17*) divided this range into 12 intervals of 75 cm^{-1} each. I found that when a discriminant function analysis was applied to the mean intensities

of the wave numbers for each of the 12 intervals given by Wright and Michels, statistical weights could be found to classify the stimuli as pleasant or unpleasant with considerable accuracy. No unpleasant stimuli were incorrectly classified as pleasant. Only five pleasant stimuli (geraniol, *n*-butyl alcohol, ethyl alcohol, cyclohexane, and acetone) were incorrectly classified as unpleasant on the basis of their Raman spectra.

A New Approach

Because no physicochemical variable, when considered individually, can be used to account totally for the ordering of the olfactory stimuli in Figs. 1, 2, and 3, a series of variables were weighted to determine whether a group of physicochemical variables, when considered together, could account for the ordering. A recently developed technique (*25*) was used to weight a series of physicochemical variables in such a way that the distances and thus the spatial arrangements among the stimuli in Fig. 1 could be regenerated.

The purpose of this procedure is to maximize the configurational similarity of the psychologically determined space with a space generated by physicochemical parameters. The procedure attempts to minimize the error by least squares criteria between the proximities $\mathbf{P}$ in Fig. 1 based on subjective measures and some proximity measures $\hat{\mathbf{P}}$ based on weighted physicochemical parameters. The basic matrix equations are:

$$\mathbf{P} = \hat{\mathbf{P}} + \mathbf{E} \qquad (1)$$

$$\hat{\mathbf{P}} = D\mathbf{Q} \qquad (2)$$

$$\mathbf{P} = D\mathbf{Q} + \mathbf{E} \qquad (3)$$

where $\mathbf{P}$ is an $(n)(n-1)/2$ column vector whose elements p_{ij} represent all the interstimulus distances between stimulus i and stimulus j and where n is the total number of stimuli; $\hat{\mathbf{P}}$ is an $(n)(n-1)/2$ column vector representing the proximity measures based on weighted physicochemical parameters; D is an $[(n)(n-1)/2]$ by k scalar distance matrix whose elements $d^2_{(ij)k}$ are the squared differences between stimulus i and stimulus j for each physicochemical parameter k; $\mathbf{Q}$ is a k element column vector of weights for the k physicochemical parameters; and $\mathbf{E}$ is an $(n)(n-1)/2$ column vector representing the error between the subjective proximities and the proximities based on physicochemical measures.

The error to be minimized is

$$\frac{\partial \mathbf{E}'\mathbf{E}}{\partial \mathbf{Q}_k} = 0 \qquad (4)$$

leading to the least squares solution

$$\mathbf{Q} = (D'D)^{-1} D'\mathbf{P} \qquad (5)$$

Thirty-nine stimuli for which Raman intensities, molecular weights, number of double bonds, functional groups, and cyclic structures were known were ordered in a multidimensional space, interstimulus distances found by weighting these physicochemical variables being used to reproduce the psychophysical space in Fig. 1. The two-dimensional regenerated space is shown in Fig. 7. The more pleasant stimuli are located toward the left and the more unpleasant stimuli are distributed in a semicircle toward the right. The correlation coefficient between interstimulus distances found from weighting the physicochemical variables with the interstimulus distances in Fig. 1 (original distances) is + 0.76. The two-dimensional solution achieved by Guttman's routine (*18*) in scaling the regenerated distances accounts for 84 percent of the variance.

The weights that were applied to standard scores for each of the physicochemical variables to achieve the space in Fig. 7, as well as the means and variance for these physicochemical variables, are shown in Table 1. Functional groups were coded according to their number in a particular molecule; for example, benzaldehyde has one aldehyde group and the mean number of aldehyde groups for all the molecules in the space in Fig. 7 is 0.10. Cyclic compounds were coded "1" and noncyclic compounds, "0."

The variables in Table 1 used for prediction are not highly correlated with each other. When variables were highly correlated (for example, molecular weight, boiling point, number of atoms in the molecule, and molecular size), only one of these variables (here, molecular weight) was used in the weighting procedure.

In interpreting the meaning of the weights, it is important to bear in mind that only a limited number of stimuli were scaled in Fig. 7. The meaning of the weights can be understood by examining the impact of the weights on the series of differences between each pair of stimuli on each physicochemical variable. As an example, the differences between the z scores (standard scores) for the molecular weights of benzene and citronellol are found; this difference is then weighted by 6.24, as shown in Table 1. This large weight will expand the difference between these two stimuli more than a small weight, indicating the importance in discrimination of physicochemical variables with large weights.

It appears that this methodology might be of considerable use in finding the appropriate physicochemical variables for olfaction because it strictly relates quantitative measures of olfactory quality with quantitative physicochemical measures. When all the appropriate physicochemical variables for spaces based on a very large sampling of olfactory stimuli are found, the correlation between the psychological space and regenerated space based on physicochemical variables may approach "1"; and from knowledge of these variables we may be able to discover the underlying receptor mechanism.

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