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Physicochemical Determinants and Functional Properties of the Senses of Irritation and Smell

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Abstract

Airborne volatile organic compounds (VOCs) are prime suspects in indoor air related illnesses (e.g., sick building syndrome). Frequent symptoms in those cases involve sensory responses of odor, nasal pungency (irritation), and eye irritation. We separated the trigeminal (pungent) from the olfactory (odor) response of the nose to a common set of substances by measuring nasal detection thresholds in subjects lacking a functional sense of smell (i.e., anosmics) and in matched-subjects with normal olfaction (i.e., normosmics). Eye irritation thresholds for the same compounds were also measured. Stimuli comprised homologous series of alcohols, acetates, ketones, and alkylbenzenes. Physicochemical properties change orderly and systematically in such series allowing to relate those properties with the observed sensory responses. In all series and for the three sensory modalities, thresholds decreased with carbon chain length. For nasal pungency — but not for odor — thresholds across chemical series bore a uniform linear relationship with simple physicochemical properties (e.g., saturated vapor concentration at room temperature). Eye irritation thresholds fell remarkably close to those of nasal pungency. Mixtures of VOCs can reach odor, pungency, and eye irritation thresholds when all components are below their individual thresholds.

Introduction

Sensory irritation of eyes, nose, and throat is a frequent symptom in occupants of spaces with indoor air quality problems (1). In most cases, the responsible agent(s) elude specification, but volatile organic compounds (VOCs) emitted from indoor materials and present at relatively low levels are prime suspects (2-4).

Detection of airborne chemicals by humans relies on two sensory channels: olfaction (5) and the so-called common chemical sense (CCS) or chemical irritation sense (6). Odor sensations are carried by the olfactory nerve (cranial nerve I). They originate in specialized structures: the olfactory neurons located in the olfactory epithelium that covers a relatively small patch in the upper back portion of the nasal cavity. These bipolar neurons send a dendrite to the epithelium surface and an axon that joins other axons from neighboring cells to form the olfactory nerve. The nerve penetrates through perforations in the ethmoid bone (cribriform plate) and makes the first synapse of the pathway in olfactory bulb structures called glomeruli. The dendrite on the other end of each neuron reaches the epithelium surface and ends in an olfactory knob from which a number of cilia protrude and are immersed in the mucus bathing the surface. These cilia are most likely the sites of olfactory transduction (7, 8). Chapter   in this book describes in detail the neurobiology of smell (Greer, 1994).

Unlike smell, the CCS lacks specific receptor structures — CCS reception relies on free nerve endings — and is widely distributed since there is general chemical sensitivity in all body mucosae (conjunctival, nasal, oral, upper and lower respiratory tract, genital, and anal). Of special relevance to sensory irritation generated by indoor air are two face mucosae: conjunctival and nasal. Common chemical sensations from the eyes and the nose are mediated by free nerve endings from the trigeminal nerve (cranial nerve V). We like to group together under the term "**nasal pungency**" an array of responses elicited in the nose by CCS stimulation. These pungent sensations include: stinging, prickliness, tingling, irritation, burning, piquancy, and freshness, among others. For simplicity, we will refer to CCS responses in the conjunctiva as **eye irritation** although most of the terms just mentioned can be applied to the eye and thus, we can also talk of an ocular pungency.

Sensory responses measured and compounds tested

It would be convenient to be able to separate the odor from the pungent response of the nose to airborne chemicals. Unfortunately, almost all chemicals evoke responses from both sensory systems. In general, at relatively low concentrations only odor is apparent, but as concentration increases pungency joins in. In this way, nasal pungency thresholds would have to be measured, in most cases, against a strong odorous background. This makes such thresholds heavily dependent on the individual criterion of when to call an odor sensation barely pungent.

In order to overcome the problem, we measured nasal pungency thresholds in persons lacking a functional sense of smell (i.e., anosmics) for whom odor did not interfere. Odor thresholds, on the other hand, were measured on subjects with normal olfaction (i.e., normosmics) matched to the anosmics by age, gender, and smoking-status (variables known to affect chemosensory sensitivity) (9, 10).

The approach was complemented by a thoughtful selection of the chemical stimuli. Our goal was to establish relationships and trends between chemical structure and potency of the compound as odorant or irritant, using substances relevant to indoor air. For these reasons we selected homologous series of relatively nonreactive chemicals. Physicochemical properties in such series change orderly and systematically, allowing us to track down how these gradual changes reflect themselves in the sensory responses. The choice of relatively nonreactive chemicals was based on the fact that, in the great majority of cases of indoor air quality complaints, chemical analysis fails to reveal the presence of any overt irritant (e.g., formaldehyde) of the type that will react readily with mucosal tissue. Given these considerations, the substances tested included: n-alcohols (11), acetates (12), branched alcohols and acetates, ketones (13), and alkylbenzenes (14). The compounds were delivered in vapor phase from squeeze bottles adapted to stimulate one nostril (15) or one eye (12). Concentration in the headspace of the bottles was measured by gas chromatography.

Nasal pungency and odor thresholds

Figure 1 shows the average pungency and odor thresholds — measured in anosmics and normosmics, respectively — obtained for normal homologous alcohols from methanol to 1-octanol. Also shown are the thresholds for some secondary alcohols: 2-propanol, 2-butanol (or sec-butyl alcohol), and 4-heptanol; and one tertiary alcohol: 2-methyl-2-propanol (or tert-butyl alcohol).

Insert Figure 1 about here

Within the n-alcohols, both nasal thresholds tend to decline with carbon chain length albeit not at the same rate, a feature that will be seen repeatedly in all series tested. Odor thresholds do not reach a plateau (as will be seen with the following chemical series), but ability to elicit pungency starts to fade with 1-octanol where the anosmics failed to reach threshold criterion in 25 % of instances. Interestingly, switching the alcohols' chemical functional group (HO^-) from a primary to a secondary carbon (e.g., 1-heptanol to 4-heptanol) or even to a tertiary carbon (e.g., 1-butanol vs. tert-butyl alcohol) raises both odor and pungency thresholds.

The clear concentration difference between pungency and odor thresholds does not result from an artifact of averaging, as the individual thresholds from each anosmic and normosmic demonstrate (Figure 2). The groups show no overlap of threshold values. Note that: (i) the spread of individual thresholds among anosmics is much smaller that among normosmics (something that will be repeated in all series); and (ii) there seems to be a general factor of odor sensitivity: the normosmic most sensitive (i.e., having the lowest threshold) to one particular n-alcohol tends to be the most sensitive to the other homologous compounds (the same holds for the least sensitive normosmic).

Insert Figure 2 about here

Figure 3 depicts pungency and odor thresholds for homologous acetates ranging from methyl to dodecyl acetate and for secondary and tertiary butyl acetate. Similarly to the outcome with the alcohols, both thresholds decline as the series progresses, there is no overlap between anosmics and normosmics (see SD bars), and odor threshold variability is higher than that for pungency. In addition, odor thresholds tend to plateau after butyl acetate, while pungency thresholds tend to fade after heptyl/octyl acetate since only one of the four anosmics reliably detected pungency form decyl and dodecyl acetate. The branched butyl acetates (sec and tert) did not show a systematic increase or decrease in their thresholds compared to the unbranched member.

Insert Figure 3 about here

Thresholds for the ketones series are shown in Figure 4. As seen with unbranched alcohols and acetates, pungency and odor thresholds for the ketones decline with carbon chain length, and in this case plateau after 2-heptanone.

Insert Figure 4 about here

Figure 5 presents the results for the alkylbenzenes. Many features observed in the previous chemical families are repeated here (e.g., decline of both thresholds with carbon chain length, greater variability in odor thresholds, no overlap between anosmics and normosmics) but, in the alkylbenzenes, both the fading of pungency and the plateau of odor thresholds occur relatively early in the series (members above propyl benzene).

Insert Figure 5 about here

In order to get a general sense of how our odor thresholds compare with those reported in the literature, we looked at a recent compilation, standardization, and averaging of human olfactory thresholds (16). There is a high correlation (r = 0.90) between our odor thresholds and the average values from the compilation for the 25 substances common to both sets (Figure 6). From the lowest to the highest value, our thresholds cover a larger span, approximately six orders of magnitude rather than their four orders, and, thereby, offer better resolution among compounds. Averaging across studies undoubtedly accounts for much of the apparent constriction in range in the compiled data. Our thresholds, which represent the point of 100% detection, understandably often fall above those in the compilation, which presumably represent principally the points of 50% or 75% detection.

Insert Figure 6 about here

When both nasal thresholds (pungency and odor) for the total 42 VOCs studied so far are plotted as a function of a simple physicochemical property, e.g., saturated vapor concentration at room temperature (23 °C), a picture such as that shown in Figure 7 emerges. Pungency thresholds taken as a whole — irrespective of molecular size or chemical functional group — exhibit a linear relationship with saturated vapor (r = 0.97), having a slope of 1.02. Pungency for individual homologous series conforms to the general picture (slopes: 0.90 for the alcohols, 1.07 for the acetates, 1.06 for the ketones, 1.17 for the alkylbenzenes, and 0.95 for miscellaneous substances). The linear correlation with slope close to 1.00 suggests that when a certain uniform percentage of vapor saturation is achieved (approximately 32 %), nasal pungency would occur in the anosmics if it where to be evoked at all.

Insert Figure 7 about here

On the other hand, odor thresholds as a whole depicted more substance-to-substance scatter than pungency thresholds. They generally failed to show a linear relationship with saturated vapor, displaying systematic deviations, or, even if the odor thresholds of an individual series approximated a linear relationship, its slope departed from unity. The odor thresholds for no individual homologous series exhibited as strong a correlation with vapor saturation as did the pungency thresholds for all series and miscellaneous compounds grouped together. For odor, the best correlation occurred with the ketones, where r = 0.95, and the worst occurred with the acetates, where r = 0.87. The slopes of the relationships for odor thresholds commonly departed from unity: the alcohols had a slope of 1.62; the acetates, 0.83; the ketones, 1.64; the alkylbenzenes, 0.68; and the miscellaneous chemicals, 1.44.

Eye irritation thresholds

As mentioned in the Introduction, common chemical sensitivity in the face includes the conjunctiva. Eye irritation is another symptom often mentioned by occupants of spaces with indoor air quality complaints. Also, from a more basic point of view, we are interested in exploring the relative sensitivity of the CCS in the nasal and ocular mucosae, both served by branches of the same trigeminal nerve but with different mucus and epithelial layers.

A modification in the squeeze bottles cap allowed us to produce ocular stimulation with vapors (12). We, then, measured eye irritation thresholds for selected members of all homologous series previously studied, using an analogous procedure to that employed before (i.e., two-alternative forced choice with an ascending concentration approach) (11). The results showed that eye irritation thresholds were remarkably close to the nasal pungency thresholds measured before in anosmics (Figure 8). This opens the possibility of learning about the nasal pungency-eliciting potential of airborne VOCs (irrespective of odor) by simply testing them in the eyes.

Insert Figure 8 about here

Thresholds for mixtures of substances

In the real world we are almost invariably faced with chemical stimulation from mixtures of compounds. Nevertheless, few investigations have addressed the issue of how odor thresholds for individual substances compare to odor thresholds for mixtures of them (17-20). None addressed that issue for CCS thresholds (nasal pungency, eye irritation).

We designed an experiment where we prepared five mixtures of varying complexity: two three-component mixtures, two six-component mixtures, and one nine-component mixture. The constituents of such mixtures were selected compounds from the series studied before. The reason behind having two mixtures with three constituents and two with six was that one of each pair contained relatively high vapor pressure-highly water soluble substances while the other member of the pair contained relatively low vapor pressure-highly lipid soluble substances. This was done in an attempt to explore trends between the sensory thresholds in mixtures and general physicochemical properties.

Based on our previously measured average thresholds, we prepared the mixtures so they would be odor balanced, i.e., all components would be present at the same multiple or submultiple of their particular odor threshold. Odor (in normosmics), nasal pungency (in anosmics), and eye irritation thresholds were measured not only for the mixtures but also for the single compounds. At present we are exploring different ways to process the data. One of the problems we have encountered is that the new individual subject odor thresholds measured in this study depart from the odor threshold for the "average" subject on which the balanced mixtures were prepared. Thus, for each particular subject, the components in the mixtures were not necessarily odor balanced as intended (and described above).

A preliminary look at the results indicates that as mixtures get more complex (higher number of components) the concentration of each individual constituent necessary for a sensory threshold (odor, pungency, or eye irritation) to be elicited from the mixture as a whole becomes lower. Figure 9 depicts the average sensory threshold for each of nine substances. The nine sections of the graph correspond to the nine compounds. In each section, the first value (e.g., 1-propanol) represents the threshold for the substance alone. The following values represent the concentration at which the substance was present in mixtures of increasing complexity (e.g., in mixture A, in C, in E), when the mixture as a whole achieved threshold. A and B are the three-component mixtures, C and D are the six-component mixtures, and E is the nine-component mixture. Note that, for every chemical, the concentration at which the chemical was present when the stimulus reached threshold is always lower in mixtures vs. when single, and also tends to be even lower if the mixture is more complex (i.e., if it has a higher number of components). This trend holds for all three sensations: odor, pungency, and eye irritation.

Insert Figure 9 about here

A comparison of the results obtained with mixtures of equal number of substances but of contrasting general physicochemical properties (i.e., mixture A vs. mixture B, and C vs. D) suggest that for the attributes nasal pungency and eye irritation — but not for odor — mixtures of substances with higher lipid solubility drive down the threshold for the mixture to a larger extent than those with lower lipid solubility.

Conclusions and needs for future research

Odor thresholds are always below (most times well below) nasal pungency and eye irritation thresholds. A common feature found in all series studied is the decline in all three sensory thresholds with increasing carbon chain length. The decline in odor thresholds tends to be steeper than that in pungency thresholds, at least for the first few members of each series, so, up to that point, the gap between odor and pungency grows larger as the series progresses. It should be pointed out that substances not usually regarded as irritants (e.g., 1-heptanol, 2-nonanone), not only can be detected by anosmics (i.e., they have pungency) but their pungency threshold is lower than that of more typical irritants (i.e., methanol, acetone).

In general, the decline in odor thresholds across each series reaches a plateau. The exception are the alcohols where, up to 1-octanol, no tendency to plateau is observed in the odor thresholds. On the other hand, pungency thresholds across the series do not plateau, but a member is reached were the ability to evoke pungency is lost for one or more of the anosmics tested (1-octanol, octyl acetate, propylbenzene). Interestingly, the anosmics describe the quality of the pungency evoked by the first members of each series as "sharp" or "biting", as opposed to that of the last members capable of being detected whose pungency is more "dull" or "pastel". This raises the question for future investigations of the discriminative capability of the human CCS, in the absence of olfaction, to distinguish among chemical vapors.

One issue that needs to be explored further due to its direct implications on indoor air is that of the sensory thresholds for mixtures of substances. Up to what extent can sensory thresholds for mixtures be driven down by adding more components? Are there certain compounds that add their sensory effects better than others? Is there a sensory channel that integrates the signals from different chemicals more completely than the others? What implications might this have regarding the mechanisms of the reception process(es) by which these airborne substances are perceived?

There are also at least two additional important parameters that need to be better understood. One of them is the effect of the presentation procedure on the sensory thresholds obtained. All the results described here were obtained with squeeze bottles. Our recent work indicates that the absolute value of the thresholds — although most likely <u>not</u> the relative value among compounds — changes if the stimuli are presented through face masks fed from small chambers, or in environmental chambers. Results show that thresholds are lower from face masks, and even lower in whole-body chambers. An important question is whether there is a constant factor that would allow comparison of thresholds measured with these different procedures (at least within related chemicals) or if the relationship among the procedures is very different from one chemical to another and cannot be generalized.

The second parameter — very relevant to indoor air quality — is the time-course characteristics of the various sensory impressions from airborne chemicals. All the results described here represent very short-term exposure responses (typically one or two sniffs). It is well known that odor sensations tend to diminish with time (olfactory adaptation) while common chemical sensations can build-up for 30 or more minutes (temporal integration or summation) before adaptation starts to be produced (21, 22). Unveiling the temporal properties of all these sensations is central to understanding the adverse sensory reactions produced in indoor spaces.

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Figure Legends

Figure 1. Average nasal pungency thresholds measured in anosmics (filled symbols) and odor thresholds measured in normosmics (empty symbols) for homologous n-alcohols, and secondary and tertiary alcohols. Only n-members of the series are joined by a line. Each point in the anosmic group represents the mean of at least 36 thresholds; each point in the normosmic group represents the mean of 48 thresholds. Bars indicate standard deviation.

Figure 2. Individual thresholds measured in anosmics and normosmics for n-alcohols from methanol to 1-octanol (carbon chain length 1 to 8, respectively). Each point represents the median of 12 thresholds measured in one subject.

Figure 3. Average nasal pungency thresholds (filled symbols), and average odor thresholds (empty symbols) as a function of carbon chain length for homologous n-acetates, and secondary and tertiary butyl acetate. Nasal pungency threshold values for decyl and dodecyl acetate are not connected to the rest by lines since they represent the result from only one anosmic; the other three anosmics did not reliably detect these two stimuli. Bars indicate standard deviation.

Figure 4. Nasal pungency thresholds and odor thresholds for homologous ketones. Each point represents a geometric mean across subjects, and bars indicate standard deviations.

Figure 5. Nasal pungency thresholds and odor thresholds for alkylbenzenes. Each point in the pungency or odor function represents the average of 48 thresholds measured in four subjects. (The standard deviation on pungency thresholds is small enough to be covered by the symbol.)

Figure 6. Comparison between our odor thresholds (11-13), sorted in descending order, and those compiled, standardized, and averaged by Devos et al. (16).

Figure 7. Pungency and odor thresholds for homologous alcohols, acetates, ketones, alkylbenzenes, and miscellaneous chemicals, depicted as a function of their saturated vapor concentration at room temperature. In decreasing order of saturated vapor concentrations, the alcohols include methanol, ethanol, 2-propanol, tert-butyl alcohol, 1-propanol, 2-butanol, 1-butanol, 1-pentanol, 4-heptanol, 1-hexanol, 1-heptanol, and 1-octanol; the acetates include methyl, ethyl, propyl, tert-butyl, butyl, sec-butyl, pentyl, hexyl, heptyl, octyl, decyl, and dodecyl acetate; the ketones include acetone, 2-pentanone, 2-heptanone, and 2-nonanone; the alkylbenzenes include toluene through octyl benzene; and the miscellaneous substances include 1-octyne, 1-octene, pyridine, chlorobenzene, menthol, and ß-phenyl ethyl alcohol. The line representing pungency has a slope of 1.02 and an $r = 0.967$. The saturated vapor identity line is shown for reference.

Figure 8. Comparison of eye irritation thresholds (triangles) and nasal pungency thresholds (squares) for selected members of

homologous series of alcohols, ketones, alkylbenzenes, and acetates. Bars indicate standard deviations.

Figure 9. Thresholds (ppm ± SD) for nasal pungency (filled squares), eye irritation (triangles), and odor (empty squares). The nine sections of the graph correspond to nine substances. Each section lists, first, the threshold for the substance by itself (e.g., 1-propanol), then, consecutively, the level at which that substance was present when the threshold was achieved for mixtures of increasing complexity (e.g., 1-propanol in mixture A (3 components) when A achieved threshold, 1-propanol in mixture C (6 components) when C achieved threshold, 1-propanol in mixture E (9 components) when E achieved threshold).

FIGURE 1

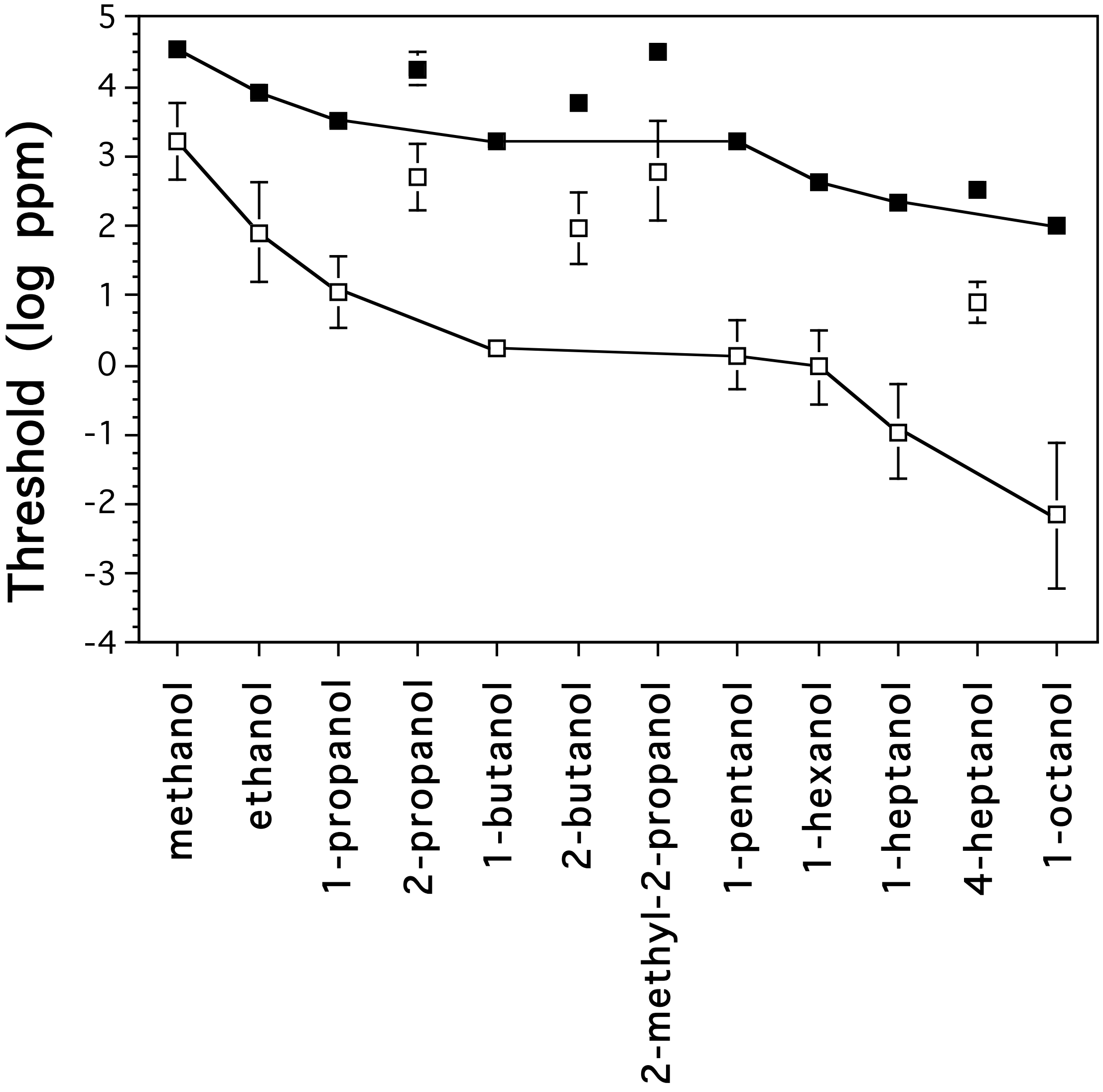

Threshold (log ppm)
5
4
3
2
1
0
-1
-2
-3
-4
methanol
ethanol
1-propanol
2-propanol
1-butanol
2-butanol
2-methyl-2-propanol
1-pentanol
1-hexanol
1-heptanol
4-heptanol
1-octanol

FIGURE 2

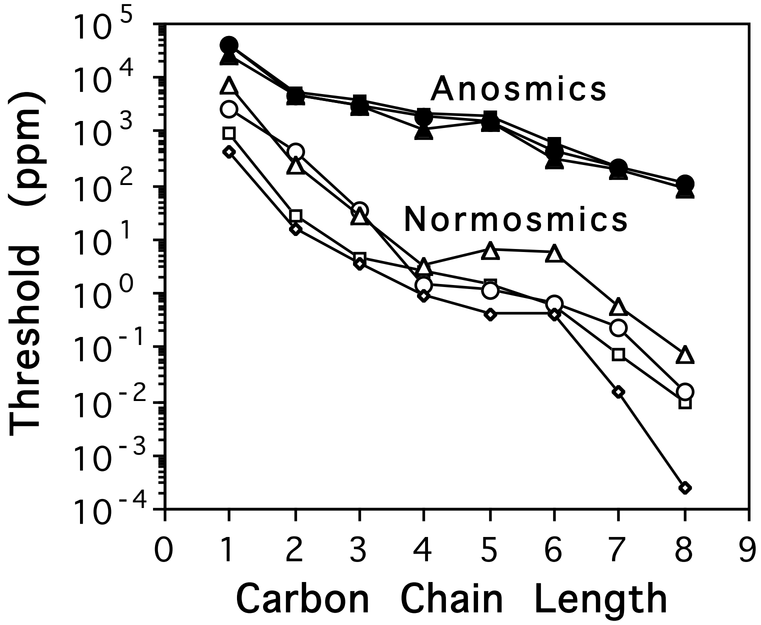

Anosmics
Normosmics
Threshold (ppm)
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
0
1
2
3
4
5
6
7
8
9
Carbon Chain Length

FIGURE 3

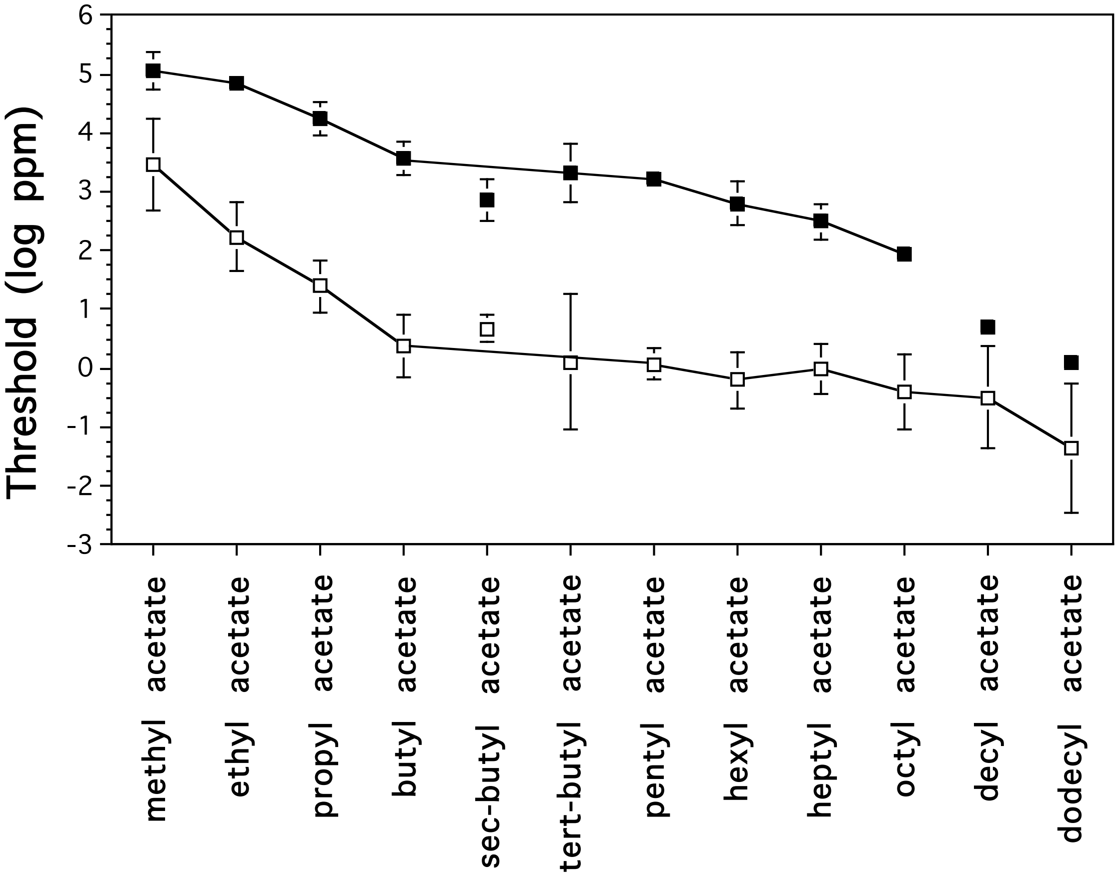

Threshold (log ppm)
6
5
4
3
2
1
0
-1
-2
-3
methyl acetate
ethyl acetate
propyl acetate
butyl acetate
sec-butyl acetate
tert-butyl acetate
pentyl acetate
hexyl acetate
heptyl acetate
octyl acetate
decyl acetate
dodecyl acetate

FIGURE 4

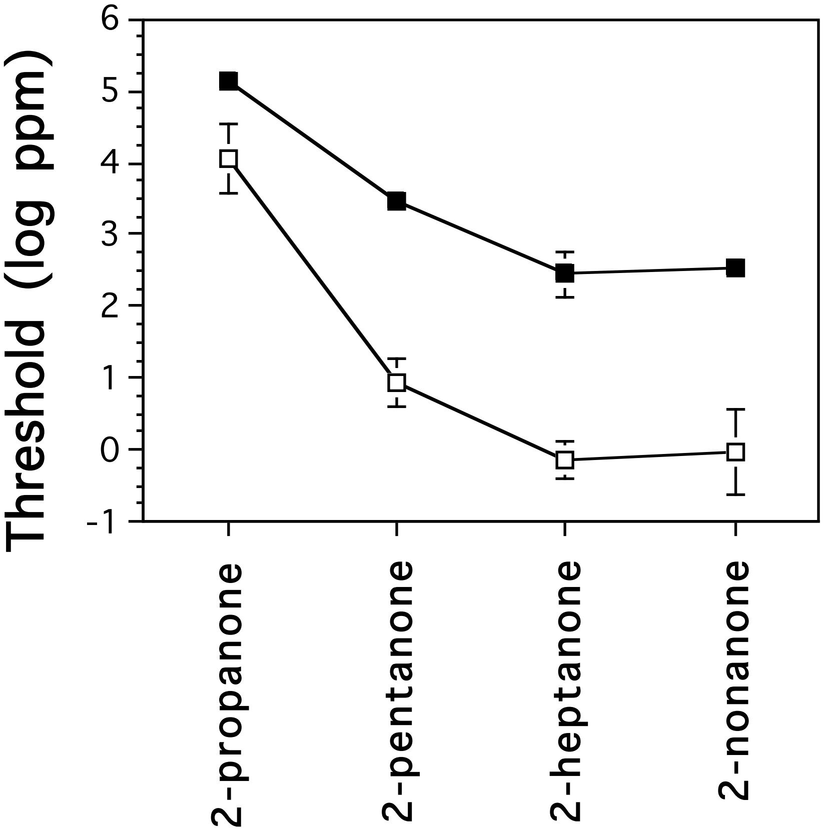

Threshold (log ppm)
6
5
4
3
2
1
0
-1
2-propanone
2-pentanone
2-heptanone
2-nonanone

FIGURE 5

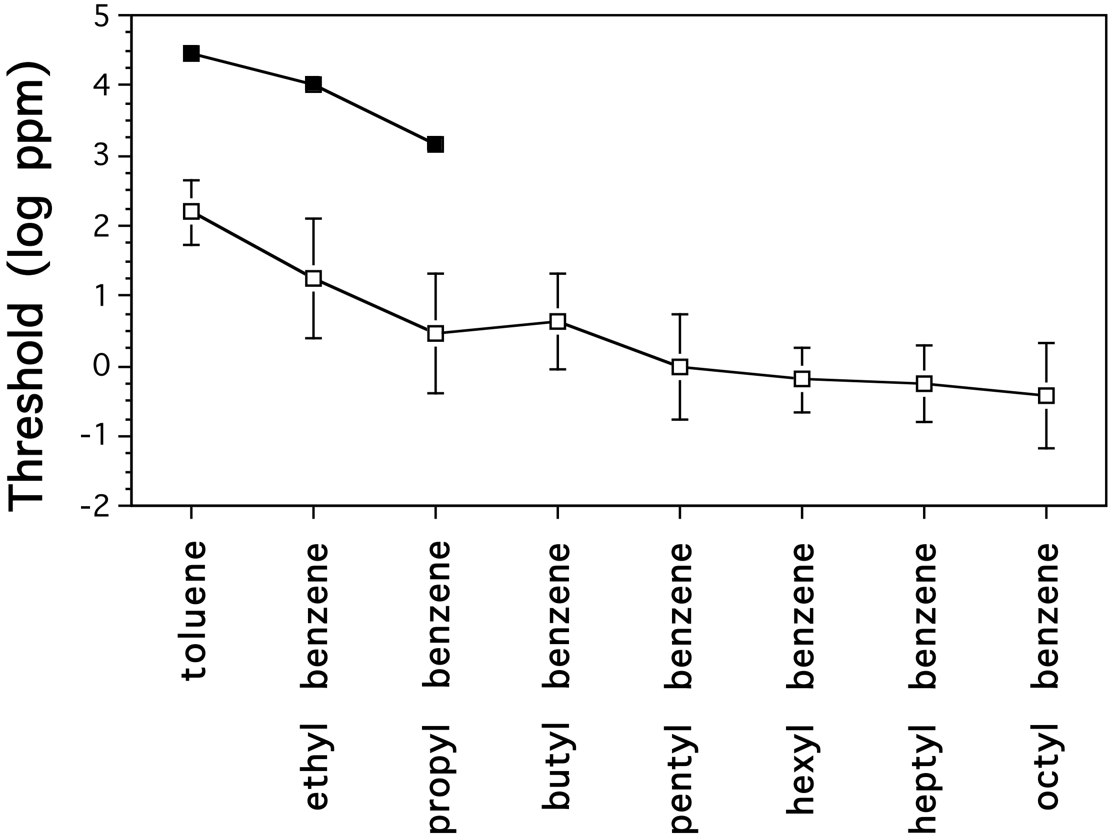

Threshold (log ppm)
5
4
3
2
1
0
-1
-2
toluene
ethyl benzene
propyl benzene
butyl benzene
pentyl benzene
hexyl benzene
heptyl benzene
octyl benzene

FIGURE 6

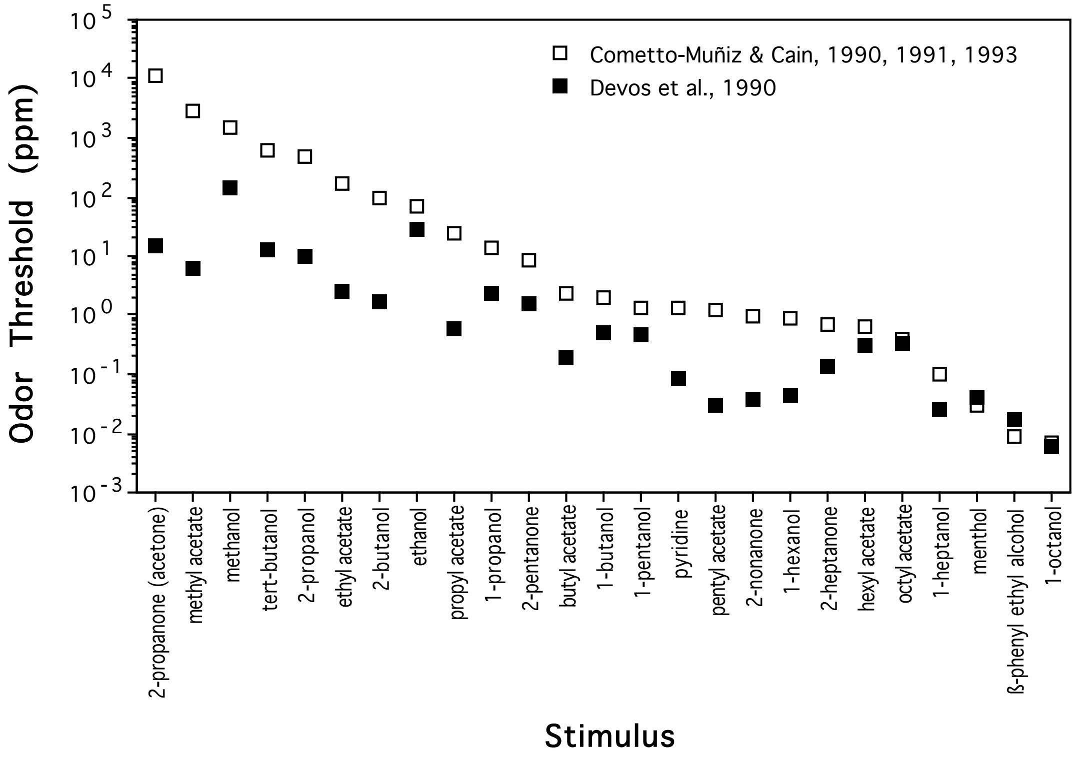

Odor Threshold (ppm)
10^5
10^4
10^3
10^2
10^1
10^0
10^{-1}
10^{-2}
10^{-3}
☐ Cometto-Muñiz & Cain, 1990, 1991, 1993
■ Devos et al., 1990
2-propanone (acetone)
methyl acetate
methanol
tert-butanol
2-propanol
ethyl acetate
2-butanol
ethanol
propyl acetate
1-propanol
2-pentanone
butyl acetate
1-butanol
1-pentanol
pyridine
pentyl acetate
2-nonanone
1-hexanol
2-heptanone
hexyl acetate
octyl acetate
1-heptanol
menthol
ß-phenyl ethyl alcohol
1-octanol
Stimulus

FIGURE 7

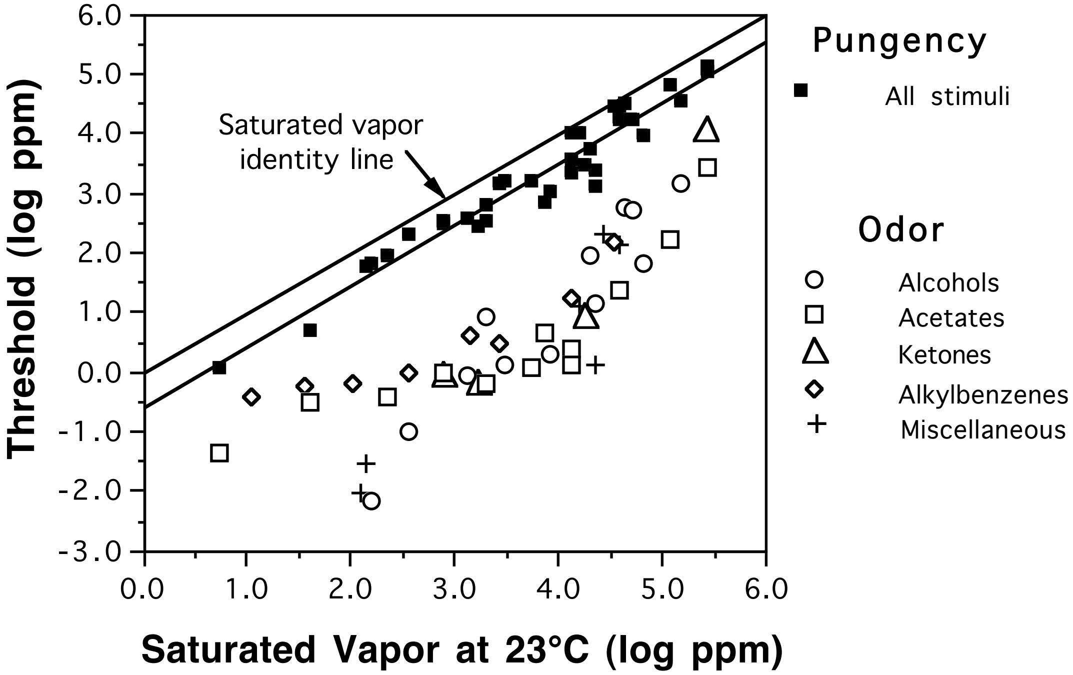

Threshold (log ppm)
6.0
5.0
4.0
3.0
2.0
1.0
0.0
-1.0
-2.0
-3.0
0.0
1.0
2.0
3.0
4.0
5.0
6.0
Saturated Vapor at 23°C (log ppm)
Saturated vapor identity line
Pungency
All stimuli
Odor
Alcohols
Acetates
Ketones
Alkylbenzenes
Miscellaneous

FIGURE 8

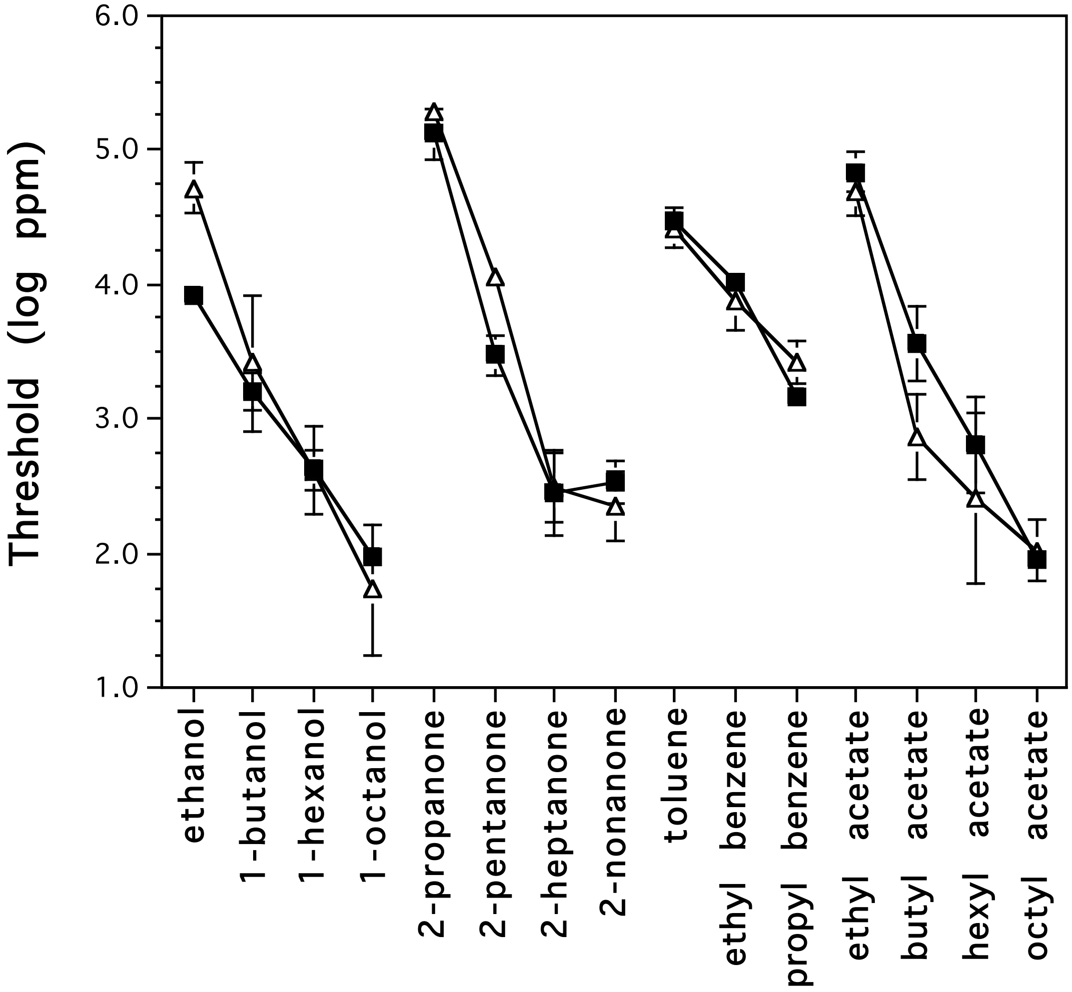
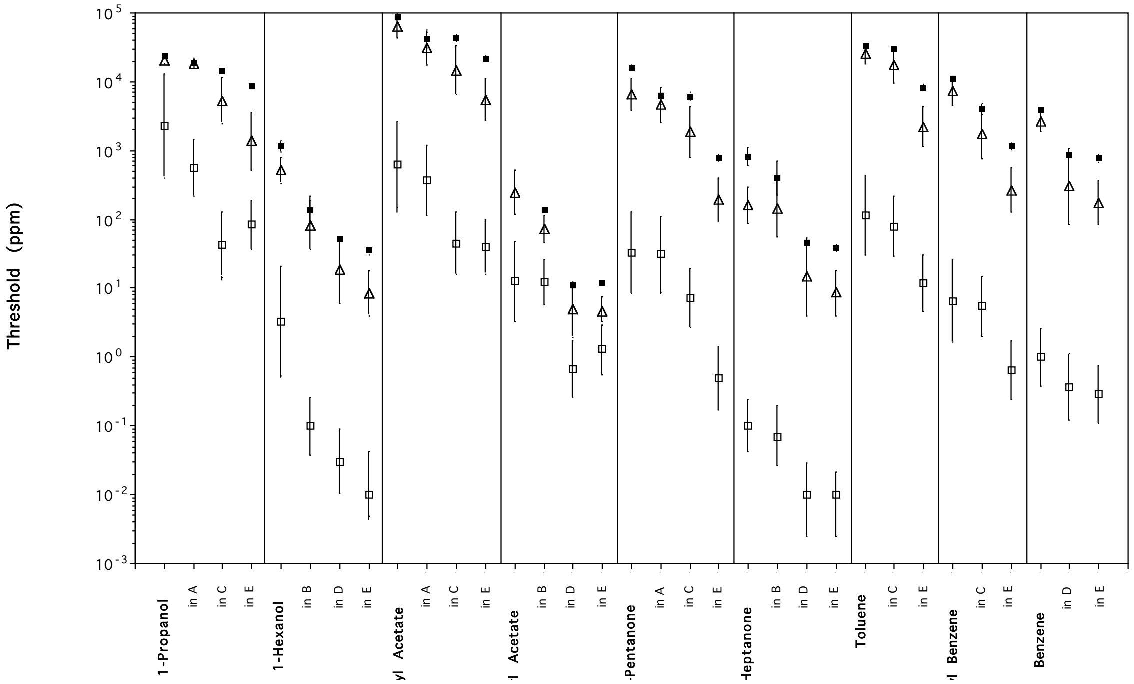

Threshold (log ppm)
6.0
5.0
4.0
3.0
2.0
1.0
ethanol
1-butanol
1-hexanol
1-octanol
2-propanone
2-pentanone
2-heptanone
2-nonanone
toluene
ethyl benzene
propyl benzene
ethyl acetate
butyl acetate
hexyl acetate
octyl acetate

FIGURE 9


Threshold (ppm)
10^5
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
10^-3
1-Propanol
in A
in C
in E
1-Hexanol
in B
in D
in E
yl Acetate
in A
in C
in E
l Acetate
in B
in D
in E
-Pentanone
in A
in C
in E
Heptanone
in B
in D
in E
Toluene
in C
in E
l Benzene
in C
in E
Benzene
in D
in E