ESTIMATION OF CRITICAL PROPERTIES FROM THE NUMBER OF CARBON ATOMS IN HOMOLOGOUS SERIES

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Received November 26, 1993
Accepted March 14, 1994

Equations correlating the normal boiling points (T_B), critical temperatures (T_C) and critical pressures (P_C) with the respective carbon numbers (n) were developed for the straight-chain homologous series H–$(CH_2)_n$–Y, where Y denotes CH_3, OH, CH=O, $OCOCH_3$ and COOH and n varies from 5 to 18. The estimates based on these equations were found to reproduce experimental data within the average errors less than 0.27% for T_B and T_C, and 1.45% for P_C thus allowing a prediction of new data.

A knowledge of the normal boiling points (T_B) and critical properties (T_C, P_C and V_C) of pure substances is important in many thermodynamic and transport property calculations[1]. In spite of the fact that several compilations of critical properties have appeared[1–5], reliable experimental data are still lacking for many substances. This is particularly true for higher molecular weights materials that decompose or associate before attaining their critical states. Values for oxygen compounds, e.g. for 1-alkanols, acetates and alkanals are especially scarce. Further, various compilations of boiling points usually quote the boiling point of a high-boiling compound at a reduced pressure, while for theoretical calculations knowledge of the normal T_B is required.

The methods proposed for the estimation of normal boiling points[1,6] usually require, besides molecular structure, some extra parameters, like critical temperature, molar refraction, ionization potential, etc. It is very unlikely that these parameters will be known for a compound when its boiling point is not known. Although some of these parameters can be estimated through group contribution methods, the propagation of error through such a route would be fairly high. The only method which is based on the molecular structure only was proposed by Joback[7]. This method uses the equation $T_{BJ} = 198 + \Sigma\delta_i$, where T_{BJ} is in Kelvins and the contributions δ_i of the various groups are listed in Reid et al.[1].

A number of methods have been proposed to predict the critical properties[1,3,8–10] when experimental data are not available. These methods are generally based on the Eq. (*1*) according to which the critical property Y_C (or some function of this property) of a compound m is expressed as a linear sum of terms incorporating some functions of

the carbon number n and the normal boiling point T_B, as well as contributions for various atoms of groups of atoms δ_i.

$$Y_C(m) = a_1 + a_2 f(n) + a_3 f(T_B) + a_4 \sum \delta_i \quad . \tag{1}$$

The relationship (*1*) involves two extreme approaches. One uses the last (two) term(s) by setting the coefficients $a_1 = a_2$ (or even a_3) = 0 (e.g. the known group contribution methods of Ambrose[3], Fedors[11] and Somayajulu[4,12]), while the other makes use of an empirical equation which requires only the number of carbon atoms n ($a_3 = a_4 = 0$); the last approach is intended to apply only to homologous series of compounds and may be represented by procedures developed by Kreglewski[13] (Eq. (*2*)) and Fisher[14] (Eq. (*3*)).

$$Y_C = Y^{\infty} - \exp(a_1 + a_2 n^{2/3}) \tag{2}$$

$$Y_C = a_1 + \frac{a_2}{n + a_3} \quad . \tag{3}$$

According to the Eq. (*2*), it is assumed that the property Y_C (i.e. T_C, P_C or even T_B) is compatible with the corresponding limiting property Y^{∞} and that all infinite-length homologues have the same composition [$(CH_2)_n$] and hence the same properties. Various limiting values for an infinitely long alkyl chain, ranging from 960 K to 1 190 K (refs[4,15]) for T_C^{∞} and from 0 bar* to 8.4 bar[13,16] for P_C^{∞}, have been recommended. Recently, Eq. (*3*) was suggested[14] to correlate some physicochemical properties of homologous fatty acids. All three parameters (a_1, a_2 and a_3) were optimized to give the best fit. But, such an equation is not generally suitable since it would tend to fit the data scatter and not give a valid reflection of random experimental uncertainty. Moreover, from a statistical point of view, a relatively large data base would be needed to optimize three parameters.

The principal objective of the present investigation is (i) to demonstrate that Eq. (*3*) can be modified by setting a_1 equal to the fixed limiting property so as to reduce the number of adjustable parameters at a minimum while maintaining both the flexibility and accuracy of the function, and (ii) to present a new empirical approach for the estimation of physical properties of the pheromone-like homologous series H–$(CH_2)_n$–Y, where Y denotes CH_3, OH, CH=O, $OCOCH_3$ and COOH, and n varies from to 5 to 18.

* 1 bar = 0.1 MPa.

The experimental property data needed (T_B, T_C and P_C) were taken mainly from compilations[1,5], but for some oxygen containing compounds the critical constant values reported in more recent sources[16–18] were preferred. The critical constants recommended by Ambrose (1992) on behalf of IUPAC (as quoted in ref.[19]) were used for alkane standard set.

For the acetate and aldehyde homologous series, where the experimental critical data are limited to those members having up to five carbon atoms only, these data have also been included in the correlations, the missing data in each series being substituted by reliable estimates based on a recently developed[4] group contribution method.

A commercial statistical package (Statgraphic Plus, Release 7.0) was used for nonlinear regression procedures and graphical analyses.

RESULTS AND DISCUSSION

Equation (*3*) forms the central point of our analysis. Prior to the analysis it may be useful to point out that the evaluation of the physicochemical properties according to this model is generally based on the extent to which the properties have characteristics normally found in members of homologous series, i.e. (i) with increasing chain-length the properties increase or decrease in a regular fashion, (ii) the properties are correlatable, by least squares equations, both with chain-length measured by the number of carbon atoms and with the corresponding properties of alkanes, and (iii) the properties are compatible with the limiting properties.

Our interest in keeping the number of parameters to be estimated at a minimum prompted us to investigate a possibility of setting the a_1 parameter in Eq. (*3*) equal to the corresponding limiting value Y^∞. From a theoretical point of view this idea seems to be reasonable as the derivative of the second term on the right side of Eq. (*3*) with respect to n goes to zero for n equal to infinity. A similar approach was previously applied with a remarkable success[20] on density calculations. Since a variety of literature limiting values is available both for the T_B^∞, T_C^∞ and P_C^∞, the immediate problem was to decide which values of limiting properties to adopt. To select the limiting temperature value suitable for the C_6 to C_{18} carbon span region investigated in this work, a nonlinear regression analysis was applied on the T_B and T_C experimental data of thirteen alkanes, C_6 to C_{18} inclusive. The general procedure used in parametrizing a_1, a_2 and a_3 was to minimize the square of the deviations of a set of calculated properties from their experimental values. By analyzing the $T_B = f(n)$ and $T_C = f(n)$ functions separately, two slightly different estimates resulted, viz. $a_1 = T_B^\infty = 1\ 114$ K and $a_1 = T_C^\infty = 1\ 095$ K. Taking into account the small difference (19 K) between the two estimates as well as generally accepted assumption that the limiting value for the normal boiling point is equal to the limiting value for the critical temperature, an average value $T_B^\infty = T_C^\infty = 1\ 105$ K was selected as the most suitable limiting value for our study. Figure 1 compares mean square errors (MSE) of the $T_B = f(n)$ and $T_C = f(n)$ functions fitted by Eq. (*3*) on

alkane data using both the selected value a_1 = 1 105 K and several other limiting values, 1 021 (ref.[12]), 1 072 (ref.[19]), 1 078 (ref.[13]), 1 143.8 (ref.[16]) and 1 190 (ref.[15]) K suggested by other authors. It is shown that the $T_C = f(n)$ dependence is slightly more sensitive to a correct selection of the limiting value, while there is some toleration (MSE < 1) for both functions in the limiting temperature region of about 1 070 – 1 140 K. It is worth to mention that the value of 1 105 K agree very well with the literature values derived recently[16,19] from other models. However, the value of Teja (1 143.8 K, ref.[16]) is about 4% too high, while that of Tsonopoulos (1 072 K, ref.[19]) is about 3% too low in relation to our value.

With the value of $a_1 = T_B^\infty = T_C^\infty$ = 1 105 K thus predetermined and with the corresponding value of P_C^∞ = 1.01325 bar, Eqs (*4*) – (*6*) immediately follow from the general equation (*3*):

$$T_B = 1\,105 + \frac{a_B}{n + b_B} \quad \text{[K]} \tag{4}$$

$$T_C = 1\,105 + \frac{a_T}{n + b_T} \quad \text{[K]} \tag{5}$$

$$P_C = 1.01325 + \frac{a_P}{n + b_P} \quad \text{[bar]} \ . \tag{6}$$

These two-parameter equations predict an increase in boiling point or critical temperature and decrease in critical pressure until the values attain a limiting value as n tends to infinity.

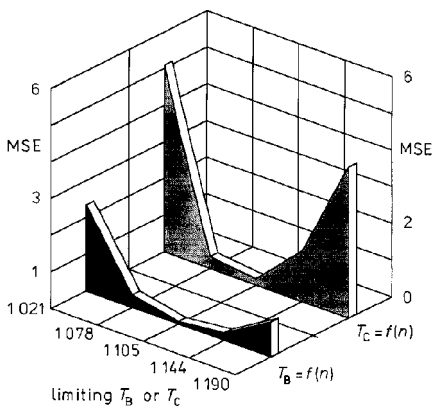

FIG. 1
Mean square errors of Eq. (*3*), $Y_i = T_B$ or T_C, fitted on alkane (C_5 to C_{18}) experimental data. Influence of selected limiting property values adopted for a_1 parameter

Equation (*4*) was tested by using available experimental normal boiling points for 44 compounds belonging to five homologous series. The series included alkanes, 1-alkanols, alkyl acetates, alkanals and carboxylic acids ranging in carbon number from 5 to 18. At least five data points (alkanals) were used for correlations in each series. The boiling points were calculated by a nonlinear, least squares fit of the input data. The results of the calculation are included in Table I along with the corresponding correlation coefficients. A comparison of the calculated and experimental values (see Appendix) shows a good agreement, generally within 0.3%. However, the invariably nonlinear $T_B = f(n)$ relationships found in this work markedly contrast with an inherent linearity assumed by Joback[7] in his group contribution method. Thus, we should be cautious in using T_B estimates based on Joback's method.

Regression equations and associated statistical data that relate T_C and P_C data of the homologous series to n according to the Eqs (*5*) and (*6*) are shown in Tables II and III. The different classes of compounds appear to be equally well correlated (in all cases $r^2 > 0.995$). The close juxtapositions of data points and calculated lines (Figs 2

TABLE I
Coefficients of Eq. (*4*) for correlation of boiling points

Series	n	a_B	b_B	r^2
Alkanes	5 – 18	–19 073.4	20	0.9999
Alkanols	5 – 12, 17, 18	–22 457.3	27.34	0.9993
Alkyl acetates	5 – 8, 10, 12, 14	–21 966.2	27.27	0.9993
Alkanals	3 – 8	–21 193.3	25.08	0.9991
Carboxylic acids	4 – 9, 11, 13, 15, 17	–20 560.6	27.73	0.9988

TABLE II
Coefficients of Eq. (*5*) for correlation of critical temperatures

Series	n	a_T	b_T	r^2
Alkanes	5 – 17	–10 616.8	12.76	0.9999
Alkanols	5 – 12	–10 720.8	15.71	0.9996
Alkyl acetates	(5 – 17)[a]	–13 254.9	21.33	0.9999
Alkanals	(5 – 17)[a]	–12 515.9	18.57	0.9999
Carboxylic acids	4 – 9	–10 612.9	18.95	0.9973

[a] Input data based on the group additivity method of Somayajulu[4].

and 3) seem to give assurance that the equations in Tables II and III could be used to predict T_C and P_C satisfactorily from the corresponding carbon numbers for all classes of compounds investigated. Two points should be made, however, regarding the input data used for the correlations: (i) as recommended in the original work[21], "smoothed" experimental P_C data were used for alkanoic acids; this allowed to improve a rather modest correlation ($r^2 = 0.976$) following from the use of not-smoothed data (inclusive an apparently erroneous value, 27.0 bar, for octanoic acid); (ii) since either the T_C or P_C experimental data were available for the alkyl acetate and alkanal homologous series ($n > 5$), estimated values based on the group additivity method of Somayajulu[4] had

TABLE III
Coefficients of Eq. (*6*) for correlation of critical pressures

Series	*n*	a_P	b_P	r^2
Alkanes	5 – 17	245.57	3.408	0.9981
Alkanols	5 – 12	268.23	2.002	0.9953
Alkyl acetates	(5 – 17)[a]	245.8	4.151	0.9998
Alkanals	(5 – 17)[a]	249.64	2.858	0.9998
Carboxylic acids	4 – 9	275.73	3.879	0.9994

[a] See note in Table II.

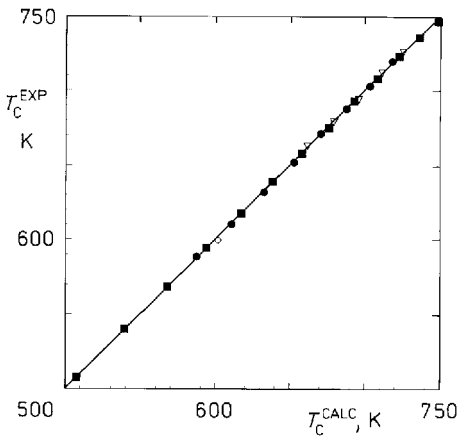

FIG. 2
Comparison of estimated (Eq. (*5*)) T_C with literature experimental values; ■ RH, ● ROH, ✧ ROAc, ∇ RCOOH

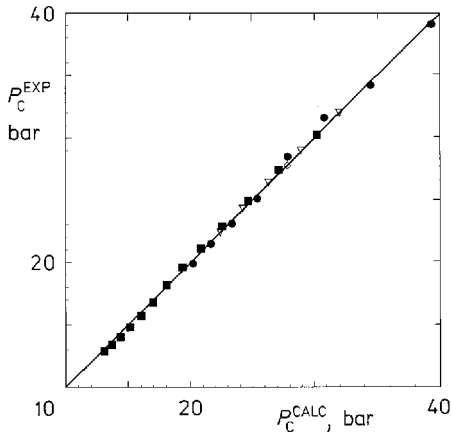

FIG. 3
Comparison of estimated (Eq. (*6*)) P_C with literature experimental values; ■ RH, ● ROH, ✧ ROAc, ∇ RCOOH

to be used as input data to treat the corresponding T_C or $P_C = f(n)$ dependence. Therefore, for these two homologous series, the proposed correlations should be regarded only as a more simple alternative to the above mentioned group additivity method.

Table IV presents the average deviations associated with different classes of compounds and thus assists in a broad sense in assessing the capability of the method to estimate selected physicochemical properties for substances corresponding to their respective classification.

Combining each of the Eqs (*4*) – (*6*) for alkyl homologous series with corresponding equations for alkanes enables the appropriate differences to be expressed by

$$Y_i - Y_i^a = \frac{a_i}{n + b_i} - \frac{a_i^a}{n + b_i^a} \quad , \tag{7}$$

where Y_i represents T_B, T_C or P_C and Y_i^a is that property for the n-alkane with the same number of carbon atoms n; a_i, b_i and a_i^a, b_i^a are fitted constants which are characteristic of the respective homologous series. Note, that a formally similar (and more simple) equation (Eq. (*8*)) has been suggested[16] to describe T_C or $P_C = f(n)$ dependence of 1-alkanols as perturbations of the corresponding properties of alkanes.

$$Y_i - Y_i^a = \frac{A_i}{n + B_i} \quad . \tag{8}$$

TABLE IV
Summary of average deviations according to classes of compounds

Series	Average deviation, % (n-range)		
	T_B	T_C	P_C
Alkanes	0.05 (5 – 18)	0.09 (5 – 17)	1.45 (5 – 17)
Alkanols	0.27 (5 – 12, 17, 18)	0.11 (5 – 12)	1.36 (5 – 12)
Alkyl acetates	0.24 (5 – 8, 10, 12, 14)	–	–
Alkanals	0.25 (5 – 9)	–	–
Carboxylic acids	0.32 (5 – 9, 11, 13, 15, 17)	0.15 (5 – 9)	0.32 (5 – 9)

To make the results based on Eqs (*7*) and (*8*) comparable, we recalculated the coefficients of Eq. (*8*) using the same *n*-range and input data as for Eq. (*7*). This afforded the numerical values of A_T, B_T, A_p and B_p equal to 861.1, 2.303, 19.795 and −1.3771, respectively. Prediction results for the $Y_i - Y_i^a$ differences (ΔY^{CALC}) using the correlation equations (*7*) and (*8*) are compared with the experimental differences (ΔY^{EXP}) in Figs 4 and 5. It may be observed (Fig. 4) that the correlating ability of Eqs (*7*) and (*8*) for ΔT_C (n = 6 to 12) is nearly the same, the absolute difference between ΔT_C^{CALC} and ΔT_C^{EXP} not exceeding 2.5 K. On the other hand, Fig. 5 demonstrates a relatively large scatter of points for the corresponding ΔP_C differences. There are two possible explanations for this type of behaviour. First, and most obvious, the data scatter could be a manifestation of a high uncertainty of experimental P_C data for the alkanol series in general and for heptanol and octanol, in particular. Note, that for these two compounds the newly reported[18] data (31.6 and 28.5 bar) used in this work differ by 1.0 and 0.7 bar from the equivalent data obtained[16] by another technique. From this point of view, even the largest differences (≈0.7 bar) between the ΔP_C^{CALC} and ΔP_C^{EXP} (Fig. 5) seem to lie in the range of experimental errors. Second, this might be a signal of severe problems with the models examined at a very fundamental level (e.g. an unappropriate description of the $T_C = f(n)$ and $P_C = f(n)$ dependences by the same functions). Both explanations can be assessed only via additional (and accurate) experiments.

The difference between Eqs (*7*) and (*8*) is more apparent when examining the predictions based on $\Delta T_C = f(n)$ and $\Delta P_C = f(n)$ functions for a more extended *n*-range. For

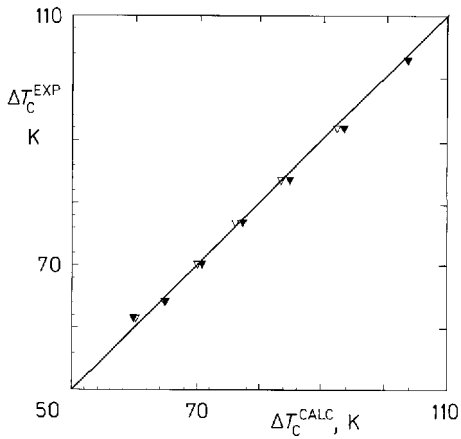

FIG. 4
Comparison of ΔT_C^{EXP} and ΔT_C^{CALC} (Eqs (*7*) and (*8*)). Solid line indicates a 1 : 1 correspondence. ▼ Eq. (*7*); ∇ Eq. (*8*)

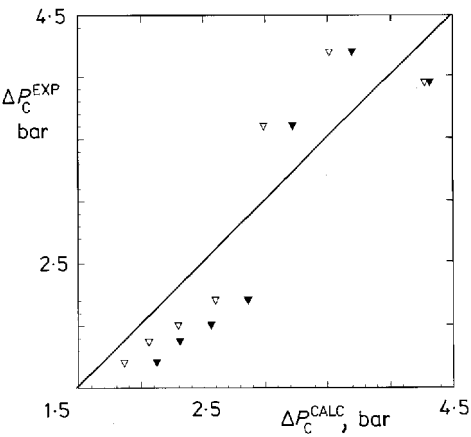

FIG. 5
Comparison of ΔP_C^{EXP} and ΔP_C^{CALC} (Eqs (*7*) and (*8*)). Solid line indicates a 1 : 1 correspondence. ▼ Eq. (*7*); ∇ Eq. (*8*)

APPENDIX

Calculated (Eqs (*4*) – (*6*)) and experimental (in parentheses) values of properties of homologous series $H–(CH_2)_n–Y$

n	Y = CH_3[a]			Y = OH			Y = $OCOCH_3$			Y = CHO			Y = COOH		
	T_B, K	T_C, K	P_C, bar	T_B[b], K	T_C[c], K	P_C[c], bar	T_B[d], K	T_C, K	P_C, bar	T_B[d], K	T_C, K	P_C, bar	T_B[e], K	T_C[f], K	P_C[f], bar
5	342.1	507.3	30.2	410.6	587.2	39.3	424.3	601.6	27.9	400.5	574.0	32.8	476.8	661.9	32.0
	(341.9)	(507.6)	(30.3)	(411.2)	(588.5)	(39.1)	(422.4)	(599.9)[g]	(27.8)[g]	(401.1)	–	–	(478.4)	(663.0)	(32.0)
6	371.4	539.2	27.1	431.4	611.1	34.5	444.7	620.1	25.2	423.1	595.6	29.2	495.5	679.6	28.9
	(371.6)	(540.2)	(27.4)	(430.6)	(610.4)	(34.2)	(444.7)	–	–	(425.9)	–	–	(495.4)	(679.0)	(29.0)
7	398.6	567.8	24.6	451.0	632.8	30.8	464.0	637.2	23.1	444.4	615.5	26.3	513.0	696.0	26.3
	(398.8)	(568.7)	(24.9)	(449.5)	(632.0)	(31.6)	(465.6)	–	–	(444.2)	–	–	(512.0)	(694.0)	(26.4)
8	423.8	593.7	22.5	469.5	652.7	27.8	482.2	653.1	21.2	464.4	634.0	24.0	529.6	711.2	24.2
	(424.0)	(594.6)	(22.9)	(468.4)	(652.2)	(28.5)	(483.2)	–	–	(464.1)	–	–	(527.7)	(712.0)	(24.3)
9	447.3	617.2	20.8	487.0	671.0	25.4	499.3	668.0	19.7	483.2	651.0	22.1	545.2	725.3	22.4
	(447.3)	(617.7)	(21.1)	(486.4)	(671.3)	(25.1)	–	–	–	(482.1)	–	–	(541.9)	(726.0)	(22.3)
10	469.2	638.6	19.3	503.6	687.9	23.4	515.6	682.0	18.4	500.9	666.9	20.4	560.1	738.4	20.9
	(469.1)	(639.0)	(19.6)	(504.3)	(687.7)	(23.1)	(517.2)	–	–	–	–	–	–	–	–
11	489.7	658.2	18.1	519.3	703.5	21.6	531.0	695.1	17.2	517.6	681.7	19.0	574.1	750.6	19.5
	(489.5)	(658.0)	(18.2)	(521.2)	(703.0)	(21.5)	–	–	–	–	–	–	(572.1)	–	–
12	509.0	676.3	17.0	534.1	718.0	20.2	545.6	707.3	16.2	533.5	695.6	17.8	587.5	762.1	18.4
	(508.6)	(675.0)	(16.8)	(537.8)	(719.4)	(19.9)	(543.9)	–	–	–	–	–	–	–	–

APPENDIX
(*Continued*)

n	Y = CH_3[a]			Y = OH			Y = $OCOCH_3$			Y = CHO			Y = COOH		
	T_B, K	T_C, K	P_C, bar	T_B[b], K	T_C[c], K	P_C[c], bar	T_B[d], K	T_C, K	P_C, bar	T_B[d], K	T_C, K	P_C, bar	T_B[e], K	T_C[f], K	P_C[f], bar
13	527.0	692.0	16.0	548.3	731.5	18.9	559.5	718.9	15.3	548.5	708.6	16.8	600.2	772.8	17.3
	(526.7)	(693.0)	(15.7)	–	–	–	–	–	–	–	–	–	(599.4)	–	–
14	544.0	708.3	15.1	561.8	744.1	17.8	572.7	729.9	14.6	562.7	720.7	15.8	612.3	782.9	16.4
	(543.8)	(708.0)	(14.8)	–	–	–	(572.2)[h]	–	–	–	–	–	–	–	–
15	560.1	722.6	14.4	574.6	755.8	16.8	585.3	740.2	13.8	576.2	732.2	15.0	623.8	792.4	15.6
	(560.0)	(723.0)	(14.0)	–	–	–	–	–	–	–	–	–	(624.7)	–	–
16	575.2	735.9	13.7	586.8	766.9	15.9	597.3	750.0	13.2	589.1	743.0	14.3	634.8	801.3	14.9
	(575.5)	(736.0)	(13.4)	–	–	–	–	–	–	–	–	–	–	–	–
17	589.5	748.3	13.0	598.5	777.2	15.1	608.8	759.2	12.6	601.4	753.1	13.6	645.3	809.8	14.2
	(590.0)	(747.0)	(12.9)	(597.0)[i]	–	–	–	–	–	–	–	–	(649.2)	–	–
18	603.1	759.9	12.5	609.7	786.9	14.4	619.8	768.0	12.1	613.1	762.8	13.0	655.4	817.8	13.6
	(603.9)	–	–	(608.0)[i]	–	–	–	–	–	–	–	–	–	–	–

[a] Experimental data taken from ref.[12] (T_B) and ref.[19] (T_C and P_C); [b] experimental data taken from ref.[16]; [c] refs[16,18]; [d] ref.[5]; [e] ref.[23]; [f] ref.[21]; [g] ref.[17]; [h] from the Antoine equation (ref.[22]); [i] ref.[1].

higher n the functions represented by Eqs (*7*) and (*8*) display a slightly different behaviour, with the absolute deviation between them reaching about 4 K and 0.25 bar for $n = 18$. The deviations may be even more significant for higher ($n > 20$) homologous series members. Again, because of the lack of the experimental data for 1-alkanols with $n > 12$, it is yet difficult to draw a conclusion which of the functions more correctly extrapolates the data. The discrepancy between the predicative ability of Eqs (*7*) and (*8*) can be resolved only by new experimental measurements.

To conclude, correlations based on limiting properties concept were developed for the estimation of normal boiling points, critical temperatures and critical pressures for homologous series of compounds related to pheromones. The correlations are valid especially for the $C_5 - C_{18}$ carbon atom range and produce estimates that generally agree with literature values when these are available. One outcome of this development could be that biologically oriented chemists with a rather modest working knowledge of physical chemistry can easily make profitable use of Eqs (*4*) – (*6*) to aid in interpretation of pheromone transport phenomena.

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Translated by the author (B. K.).