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Use of topological indices of polychlorinated biphenyls in structure–retention relationships

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Abstract

Topological parameters (Balaban index and electrotopological index) were used as structural parameters in the structure–retention relationships (SRRs). These relationships were found to be statistically valid and useful in the prediction of retention data of polychlorinated biphenyls (PCBs) despite of the temperature program used in the gas chromatographic experiment.

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1. Introduction

The complexity of the polychlorinated biphenyl (PCBs) chromatographic separation problem is defined by the number of possible congeners (209) and their chemical and physical similarities. There are a few papers that describe structure–retention relationships (SRRs) models for PCBs. Most often “coelution database” [1] or “calculation algorithm” [2] is proposed as a tool for solving the problem of identification of the congeners in chromatogram. The SRR model was proposed by Sabljic [3–5] using classical topological indices. He proposed valence molecular connectivity indices (MCIs)–retention index (*I*) relationships for various compounds, i.e. chlorobenzenes,

polychlorinated dibenzodioxins (PCDDs), polychlorinated dibenzofurans (PCDFs).

We have tried to apply relationship proposed by Sabljic [5] in SRR for polychlorinated biphenyls assuming that their structure is similar to above mentioned:

$$I = -1580.2 + 457.5^1\chi + 14.8\text{Cl}_M + 19.9\text{Cl}_{OM} - 35.4\text{Cl}_{13} - 12.9\text{Cl}_{14} + 22.1\text{Cl}_{SEQ3} + 15.3\text{ClOCl} \quad (1)$$

where $^1\chi$ is the first order MCI; Cl_M the number of *meta*-chlorine substituents; Cl_{OM} the number of pairs of *ortho/meta*-chlorine substituents; Cl_{13} the number of pairs of chlorine substituents in positions 1 and 3; Cl_{14} the number of pairs of chlorine substituents in positions 1 and 4; Cl_{SEQ3} the number of sequences of three consecutive chlorine substituents and ClOCl the simultaneous presence of chlorine substituents in either positions 1 and 9 or 4 and 6.

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Results calculated according to the above equation were not satisfactory. Relative error varied from 1.05 to 59%. The structure differences between PCBs and PCDDs/PCDFs are too significant to use the above equation (derived for PCDDs and OCDFs) for calculation of retention parameters for PCBs. In fact, first, second and third order molecular connectivity indices [3–5] exhibit too low discrimination power for whole polychlorinated biphenyls group. Moreover, the additional elements indicate the “hand-made” character of this multilinear correlation. The good idea is to use only topological indices in structure–retention or structure activity relationships. Topological indices can be divided depending on way of their calculation. Connectivity indices and valence molecular indices are based on adjacency matrix while Wiener index, Rouvray index and Balaban index are based on distance matrix ([6–8,9,10] including references cited therein).

Usability of topological indices as a structural parameters depends mainly on its degeneracy level and discrimination power as well. The discrimination power is sufficient only if indices values diversification is significant for each isomer. The indices are degenerated when their values for two or more various isomers are equal. The applicability of classical topological indices, i.e. based on neighbourhood or distance matrix, in the description of structure of PCBs—their discrimination power and degeneracy level—was discussed earlier [11,12]. In this group of classical topological indices only Balaban index exhibits sufficient discrimination power for polychlorinated biphenyls.

Molecular connectivity indices were used by Gankin et al. [13] to evaluate quantitative structure–retention relationships for PCBs. They applied multilinear regression to achieve satisfactory quality of the model. As it was shown by Kaliszán [14] the use of such a model is not always justified as the resulting regression coefficients are often intercorrelated. It seems to be fruitful to increase the discrimination power of structural parameter (topological index) instead of constructing excessively expanded models. Moreover, the use in the model of such elements as, e.g. $({}^3\chi^v)({}^4\chi^v)$ is vague and not justified. Gankin et al. [13] have shown that even their models exhibit limited predicting ability. Very often several congeners have been assigned for one chromatographic peak (see Table 2 in [13]).

Looking for indices (or better structural parameters) which are able to differentiate all 209 PCBs *E*-state parameter and then based on that electrotopological index TI^E was calculated.

E-state parameter was proposed by Kier et al. [15] as a molecular description of electrotopological state of atom in molecule and was mostly calculated for one characteristic atom in molecule, i.e. for nitrogen atom in pyrazine (1,4-diazine).

$$E\text{-state} = \frac{I_i - I_j}{r^2} \quad (2)$$

where I_i, I_j are the internal state parameters $((\delta^v + 1)/\delta)$; r the graphic distance between atoms “*i*” and “*j*” including “*i*” and “*j*” atoms.

The idea of Kier et al. [15] have been used later to characterize PCBs’ structures [16]. Heinzen and Yunes [17] applied electrotopological state parameter (calling it electrotopological state index, *S*) to describe the structure of linear alkylbenzene isomers. However, they calculated values of this parameter for the *arbitrary* chosen “... C7 position of the linear alkyl chain”. Variation of values of this topological index for different vertices of topological graph has been discussed elsewhere [11,12,18]. Full description of the molecule is possible with the matrix of *E*-parameter values. Any selection of “characteristic” vertex of the graph might be the source of significant error.

Because it is impossible to find one, very characteristic atom for all 209 PCBs, *E*-state parameter for each atom (graph bound) in each of 209 PCBs molecules was calculated and then used to calculate electrotopological TI^E index [19]:

$$\text{TI}^E = \frac{q}{\mu + 1} \sum_{k_i-j} (E\text{-state}_i \times E\text{-state}_j)^{-1/2} \quad (3)$$

where q and μ are the numbers of bounds and of cycles in the molecule, respectively, and k the edge.

2. Experimental

2.1. Materials and methods

2.1.1. Determination of retention parameters of PCBs

Standard solution (A) of 47 individual congeners in isooctane was prepared by mixing of three standard solutions CLB1A, CLB1B and CLB1C as well

as the standard solution (B) of 17 congeners in isooc-tane (Mallinckrodt Nanograde). Certified solutions of PCBs (CLB1) were purchased from National Research Council of Canada, Marine Analytical Chemistry Standard Program (Toronto, Canada). Individual polichlorinated biphenyls, included in solution B were purchased from Dr. Ehrenstorfer, Germany.

Standard solution of alkanes (C₈–C₄₀) TRHP Standard (Florida), was obtained from Ultra Scientific, USA.

The analyses of PCB and alkane mixtures were carried out with fused silica capillary column PE-5MS, Perkin-Elmer, USA (60 m × 0.25 mm, 0.25 μm i.d.; 5% diphenyl–95% dimethyl siloxane) installed in Autosystem XL chromatograph (Perkin-Elmer, USA) equipped with autosampler, split–splitless injector and low-resolution mass spectrometry (LRMS) detector. Helium was used as the carrier gas at the flow rate 21.2 cm³/min. The injector was operated in the splitless mode at 250 °C.

The electron impact ionization MS source, 70 eV nominal was employed, with the source set at 300 °C. Mass spectra data acquisition was initiated directly after sample injection. The dwell time was set at 100 ms and the multiplier voltage at 450. The mass spectrum was scanned from 100 to 650 u once every second. Experiments were repeated three times to calculate relative retention time (RRT) and values of standard deviation varied from 0.0004 to 0.0093.

All the analyzed compounds were eluted during linear increase of temperature. The following temperature profiles were used in our experiments: Program I—0.5 min at 80 °C; 25 °C/min until 140 °C and 4 °C/min until 300 °C, holding for 10 min and Program II [1] of the column was 2 min at 75 °C; 15 °C/min until 150 °C and 1.2 °C/min until 300 °C, holding for 8 min. The retention times were measured with accuracy of 0.01 min by using a Turbomass Data System, Perkin-Elmer.

Following Frame [1], relative retention times for all peaks on chromatograms for PCBs mixture A were calculated against the sum of the retention times (*t_R* values) of PCBs 52 and 180. RRTs for all peaks on the alkanes mixture chromatogram were calculated against the sum of *t_R* values of alkanes C₁₈ and C₂₆.

Relative retention indices (RRI) of examined PCB congeners were calculated according to the modified van der Dool and Kratz equation [20] for elution with

a programmed temperature:

$$\text{RRI} = 100c_z + 200 \frac{x_i - x_z}{x_{z+2} - x_z} \quad (4)$$

where *c_z* is the number of carbon atoms in *n*-alkane eluting before given PCB congener while *x_i*, *x_z* and *x_{z+2}* refer to RRT of the congener and *n*-alkanes having *z* and *z* + 2 atoms, respectively.

2.1.2. Determination of topological indices

Values of Balaban index (*I_B*) were calculated according to equation proposed by Balaban [21]:

$$I_B = \frac{q}{\mu + 1} \sum_k (s_i \cdot s_j)^{-1/2} \quad (5)$$

where *k* is the edge, *μ* the number of cycle in graph and, *s_i* the sum of all elements in all distance matrix vectors:

$$s_i = \sum_j d_{ij} \quad (6)$$

Usability of this index is connected with the construction of the relationship between the structure and physical, chemical, and biological properties of molecules.

Values of *E*-state parameter were calculated according to Eq. (2) while those of *TI^E* by using the Eq. (3).

Regression equations were produced by using internal MS Excel algorithms. Both RRT or RRI were used as retention parameters while *I_B* or *TI^E* as structural descriptors in the appropriate equations.

3. Results and discussion

Structure–retention relationships were constructed with the use of topological indices having high discrimination power. It was shown [12] that for PCBs the Balaban index as a classical index and electrotopological index *TI^E* exhibit the largest discrimination power and the lowest degeneracy level. Therefore their use in SRR is justified. Retention times and retention indices (Program I) for tested PCBs as well as topological indices for these compounds are presented in Table 1. The use of both structural descriptors (*I_B* and *TI^E*) leads to statistically valid and simple, linear regressions (Table 2, only one exemplary linear relationship is presented in Fig. 1). In this case we have used

Table 1
Balaban's indices, *E*-state parameters, relative retention indices and relative retention times for exemplary PCBs (Programs I and II)

Name	PCB (IUAPC no.)	Balaban index	TI ^E	RRI (I)	RRT (I)	RRI (II)	RRT (II)
4,4'-Dichlorobiphenyl	15	2.5232	49.3968	1780.28	0.3449	1788.97	0.2615
2,2',5-Trichlorobiphenyl	18	2.8574	67.4976	1767.22	0.3398	1755.35	0.2553
2,2',3,5'-Tetrachlorobiphenyl	44	2.9456	90.2818	1958.75	0.4161	1966.17	0.362
2,2',5,5'-Tetrachlorobiphenyl	52	2.9323	84.8408	1922	0.4014	1930.08	0.3413
2,3,4,4'-Tetrachlorobiphenyl	60	2.9885	94.9668	2077.12	0.4621	2082.61	0.4314
3,3',4,4'-Tetrachlorobiphenyl	77	2.7335	86.6506	2165.23	0.4958	2171.81	0.4855
2,2',4,5,5'-Pentachlorobiphenyl	101	2.9878	114.0025	2088.89	0.4666	2098.27	0.4409
2,3,3',4,4'-Pentachlorobiphenyl	105	2.8983	123.5879	2266.2	0.5328	2273.52	0.5463
2,3',4,4',5-Pentachlorobiphenyl	118	2.8150	113.3983	2213.13	0.5138	2223.44	0.5165
2,2',3,3',4,4'-Hexachlorobiphenyl	128	3.0531	179.4381	2367.6	0.5691	2375.38	0.6066
2,2',3,4,4',5-Hexachlorobiphenyl	137	3.0668	186.3503	2294.41	0.543	2304.22	0.5644
2,2',3,4',5',6-Hexachlorobiphenyl	149	3.1364	162.1625	2203.07	0.5102	2213.66	0.5108
2,2',4,4',5,6'-Hexachlorobiphenyl	154	3.0873	151.7957	2120	0.4925	2166.53	0.4823
2,3,3',4,5,5'-Hexachlorobiphenyl	159	3.0597	181.4923	2348.32	0.5622	2360.2	0.5976
2,2',3,3',4,4',5-Heptachlorobiphenyl	170	3.1557	269.2653	2514.84	0.619	2526.14	0.6919
2,2',3,4,4',5,5'-Heptachlorobiphenyl	180	3.1464	249.0992	2453.67	0.5986	2466.9	0.6587
2,2',3,4,5,5',6-Heptachlorobiphenyl	185	3.2886	366.8152	2376.82	0.5724	2386.34	0.6131
2,3,3',4,4',5,6'-Heptachlorobiphenyl	191	3.1454	238.9301	2463.57	0.6019	2476.18	0.664
2,2',3,3',5,5',6,6'-Octachlorobiphenyl	202	3.4326	407.2144	2406	0.5827	2417.31	0.6309
2,2',3,4,4',5,5',6-Octachlorobiphenyl	203	3.3085	514.9130	2542.13	0.6281	2555.4	0.7083
2,3,3',4,4',5,5',6-Octachlorobiphenyl	205	3.2600	479.2373	2663.55	0.6671	2678.33	0.7745
2,2',3,3',4,4',5,6,6'-Nonachlorobiphenyl	207	3.4516	1211.8552	2620.97	0.6539	2634.6	0.7515
2,2',3,3',4,5,5',6,6'-Nonachlorobiphenyl	208	3.4957	1443.8898	2600.65	0.6476	2614.45	0.7409
2,2',3,3',4,4',5,5',6,6'-Decachlorobiphenyl	209	3.5491	11855.8585	2801.37	0.7098	2817.33	0.8471

the retention data collected with the use of temperature Program I. However, significantly higher values of correlation coefficient and lower values of relative error of estimation were found when electrotopological index is used as structural parameter. This effect is probably caused by higher discrimination power and lower degeneracy of TI^E in comparison to I_B. The selection of retention parameter also influences the quality of SRR. Somewhat statistically better relationships (higher *r* values and lower relative errors) were found for RRI than for RRT.

However, the assessment of relative errors of estimation by using of experimental data applied earlier in the evaluation of the given regression is always controversial. Therefore, we evaluated the appropriate regressions by using only limited number of experimental data. Their quality was than checked by the calculation of relative errors of estimation for congeners not applied in the evaluation of given relationship. We have used two groups of congeners as the basis of our calculations: (i) regression equation was evaluated for 30 congeners, randomly selected

Table 2
Retention parameters–topological indices relationships (Program I)

Relationship	Regression parameters		Correlation coefficient, <i>r</i>	Relative error (%)	Significance coefficient, <i>t</i>	Base for calculations, <i>n</i>
	<i>a</i>	<i>b</i>				
RRT = <i>a</i> × I _B + <i>b</i>	0.3393	−0.5180	0.7774	10.35	49.43	42
RRI = <i>a</i> × I _B + <i>b</i>	937.3102	−627.4920	0.7840	6.26	50.53	42
RRT = <i>a</i> × 1/log TI ^E + <i>b</i>	−1.2810	1.0981	−0.9167	6.40	91.75	42
RRI = <i>a</i> × 1/log TI ^E + <i>b</i>	−3535.5048	3836.7740	−0.9249	3.70	97.33	42

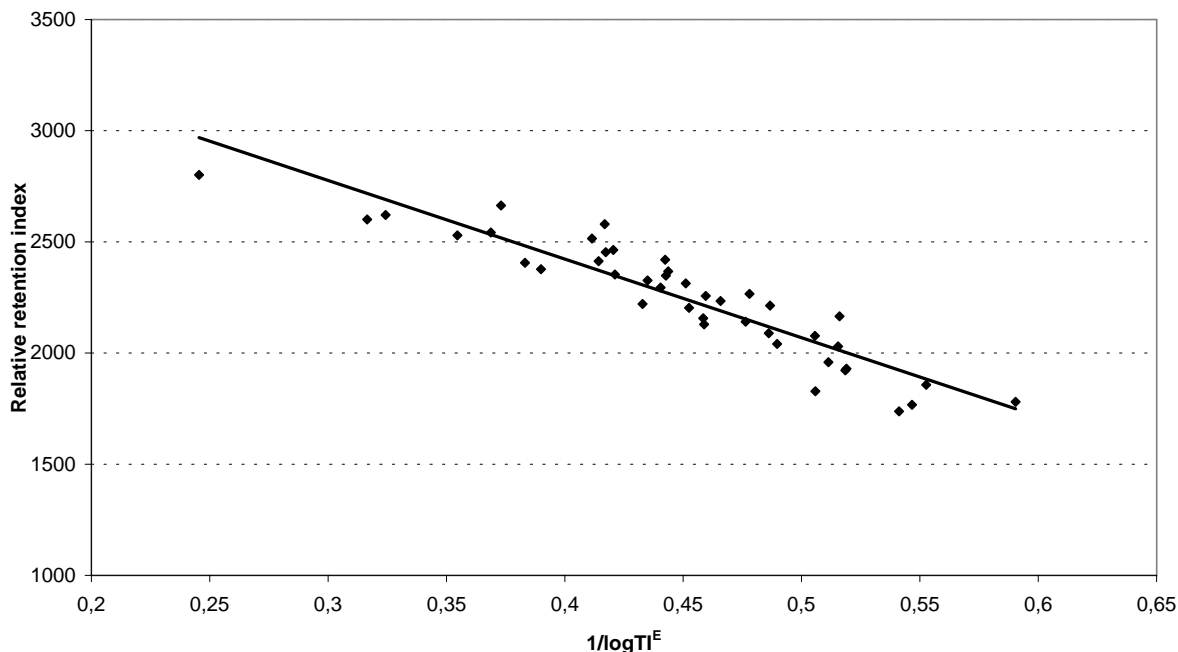


Fig. 1. Relative retention index–electrotopological index TI^E relationship (Program I).

(15, 28, 30, 44, 52, 54, 60, 74, 77, 86, 101, 114, 118, 121, 129, 137, 143, 153, 156, 159, 171, 180, 183, 191, 199, 202, 203, 207, 208, 209); the applicability of this equation was further checked by calculation of relative errors (and average error) for the group of the following 12 congeners: 18, 49, 87, 105, 128, 138, 149, 154, 170, 185, 189, 205; (ii) regression equation was determined for only six congeners 18, 52, 101, 143, 185, 203 (for these congeners the best correlation between RRT and the number of chlorine atoms has been found, $R = 0.9998$). Relative errors were calculated for 36 congeners. Results obtained according to such calculations are presented in Table 3.

Relations between the regression quality found for two examined structural descriptors and two different retention parameters are the same as for the data presented in Table 2 and discussed earlier.

The decrease of the number of experimental data leads to the increase of the values of the respective correlation coefficients (Table 3). However, this effect is accompanied by the increase of average relative error of the determination of retention parameters for twelve and thirty six selected PCBs. For example, values of correlation coefficient for $RRI = f(TI^E)$

regression (Program I) increase from 0.9249 to 0.9390 and 0.9905 when the number of experimental data decreases from 42 to 30 and 6, respectively. At the same time, values of average relative error (Tables 2 and 3) change from 3.7% ($n = 42$) to 3.42% ($n = 30$) and 1.57% ($n = 6$), respectively, when calculated for all compounds taken into account during evaluation of the relationship. However, determination error for the “test congeners”, i.e. not used in relationship determination *increases* to 4.47 and 4.91%, respectively. The best statistical characteristics were found for RRI and TI^E relationships. The quality of regressions always decreased when I_B was used in place of TI^E or RRT instead of RRI.

Various temperature programs are often used in the chromatographic analysis of PCBs. We have applied two such programs (see Section 2).

We conclude, after analysis of regressions, that the retention data obtained in temperature Program I, are generally similar to those from Program II (especially for RRI versus TI^E relationships). We like to point it out as it means that topological parameters may be successfully used in the prediction of PCBs retention data despite the temperature program applied in the

Table 3

Influence of number of experimental data used in the evaluation of regression equation on the relative error of estimation of retention parameters (Program I)

Relationship	Regression parameters		Correlation coefficient, r	Relative error (%)	Significance coefficient, t	Base for calculations, n
	a	b				
$RRT = a \times I_B + b$	0.3183	-0.4579	0.8031	9.92 11.34	37.74	30 Checking for congeners 12 not used in the evaluation of the regression
	0.5425	-1.1902	0.9728	2.52 15.39	16.81	6 Checking for congeners 36 not used in the evaluation of the regression
$RRI = a \times I_B + b$	881.7302	-467.4170	0.8134	5.86 7.19	39.15	30 Checking for congeners 12 not used in the evaluation of the regression
	1451.4040	-2335.7500	0.9718	2.52 9.23	16.49	6 Checking for congeners 36 not used in the evaluation of the regression
$RRT = a \times 1/\log TI^E + b$	-1.2033	1.0584	-0.9284	6.04 7.17	69.93	30 Checking for congeners 12 not used in the evaluation of the regression
	-1.4860	1.1668	-0.9904	2.64 8.27	28.70	6 Checking for congeners 36 not used in the evaluation of the regression
$RRI = a \times 1/\log TI^E + b$	-3328.8996	3731.0428	-0.9390	3.42 4.47	76.42	30 Checking for congeners 12 not used in the evaluation of the regression
	-3980.2273	3972.5168	-0.9905	1.57 4.91	28.75	6 Checking for congeners 36 not used in the evaluation of the regression

GC experiment. However, the correlation coefficients are much worse especially when I_B and/or RRT are used as structural and retention parameters, respectively.

The similar values of correlation parameters were found for relationships between the topological indices and retention parameters (according to Programs I and II) (Tables 2 and 4). Although those relationships are charged by higher relative error (even 17%), correlation coefficient values are in the range 0.77–0.93.

Comparing calculated correlation parameters and relative errors we can observe that the results are similar for two examined temperature programs, especially for RRI and TI^E relationships (Tables 3 and 5).

The quality of the evaluated relationships is also proved by the proper prediction of the elution order. Correlation between predicted and experimental RRTs for the series of 12 PCBs not involved in the model evaluation process is given in Fig. 2. The calculated elution order is in accordance with data published

Table 4
Retention parameters–topological indices relationships (Program II)

Relationship	Regression parameters		Correlation coefficient, r	Relative error (%)	Significance coefficient, t	Base for calculations, n
	a	b				
$RRT = a \times I_B + b$	0.5483	-1.1463	0.7841	17.98	50.54	42
$RRI = a \times I_B + b$	948.0030	-650.0312	0.7848	6.28	50.66	42
$RRT = a \times 1/\log TI^E + b$	-2.0651	1.4632	-0.9224	10.31	95.53	42
$RRI = a \times 1/\log TI^E + b$	-3576.9081	3864.7625	-0.9248	3.73	97.26	42

Table 5
Influence of number of experimental data used in the evaluation of regression equation on the relative error of estimation of retention parameters (Program II)

Relationship	Regression parameters		Correlation coefficient, r	Relative error (%)	Significance coefficient, t	Base for calculations, n
	a	b				
$RRT = a \times I_B + b$	0.5153	-1.0519	0.8111	17.39 18.92	38.82	30 Checking for congeners 12 not used in the evaluation of the regression
	0.8551	-2.1648	0.9734	6.62 24.38	16.98	6 Checking for congeners 36 not used in the evaluation of the regression
$RRI = a \times I_B + b$	889.4769	-48.2713	0.8131	5.90 7.15	39.11	30 Checking for congeners 12 not used in the evaluation of the regression
	1483.9990	-2430.3539	0.9699	2.75 9.42	15.94	6 Checking for congeners 36 not used in the evaluation of the regression
$RRT = a \times 1/\log TI^E + b$	-1.9437	1.4010	-0.9354	9.92 11.51	74.07	30 Checking for congeners 12 not used in the evaluation of the regression
	-2.3438	1.5513	-0.9915	4.01 12.64	30.51	6 Checking for congeners 36 not used in the evaluation of the regression
$RRI = a \times 1/\log TI^E + b$	-3359.8271	3754.8291	-0.9391	3.43 4.55	76.54	30 Checking for congeners 12 not used in the evaluation of the regression
	-4071.1523	4020.2865	-0.9889	1.78 5.05	26.62	6 Checking for congeners 36 not used in the evaluation of the regression

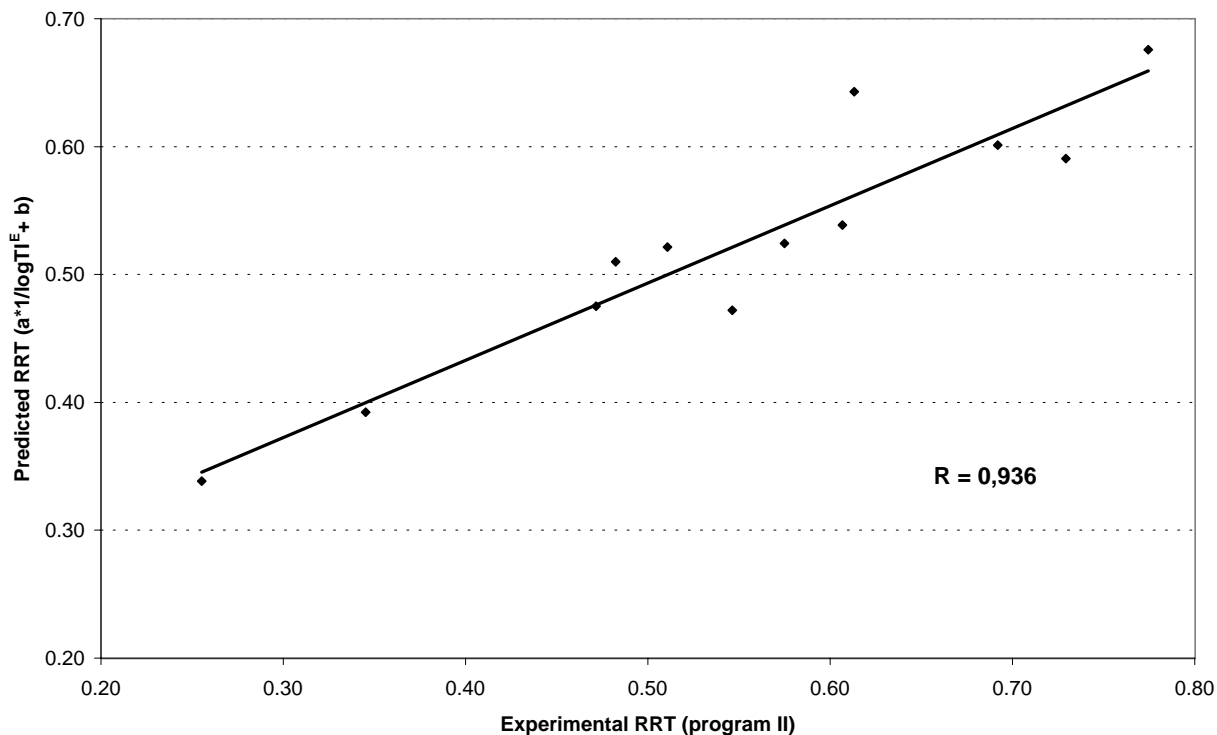


Fig. 2. A plot of RRT-predicted ($a \times 1/\log TI^E + b$) vs. experimental values of RRT for the 12 PCBs not used in the evaluation of the regression (Program II).

earlier by Frame [1] as well as with our experimental results. However, the difference was found for congeners 105, 170 and 189.

4. Conclusion

Electrotopological index TI^E with its highest discrimination power can be used to predict retention times and retention indices for polychlorinated biphenyls. The use of $RRI = f(TI^E)$ allows to determine reliable values of retention parameters for polychlorinated biphenyls with the high correlation significance and low relative error (the lowest average relative error equals to 3.7%). The replacement of TI^E by Balaban index I_B , i.e. by structural descriptor characterized by lower discrimination power and higher degeneracy, caused the decrease of the quality of evaluated SRR. The proposed relationship can be used in correlating the retention data collected in

various temperature programs. Best results were found for Program I. Here, the relative errors are lowest, especially for $RRI = f(TI^E)$ relationship. Prediction error for 12 congeners not used in the evaluation of the relationship is similar to the relative errors found for all congeners. In this case (Program I) for $RRI = f(TI^E)$ relationship the relative error is 4.5%. Prediction error, found for relationship using retention data from Program II, is only slightly higher.

Electrotopological index TI^E exhibits the highest discrimination power. The relative errors for retention parameters- TI^E relationship are lowest and correlation coefficients are highest. Therefore relationships: $RRI = a \times 1/\log TI^E + b$ and $RRT = a \times 1/\log TI^E + b$ (somewhat worse solution) can be used as a tool for the prediction of retention parameters (according to Program I) for polychlorinated biphenyls. Topological parameters may be successfully used in the prediction of PCBs retention data despite the temperature program applied in the GC experiment.

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