ORIGINAL PAPER

OCCURRENCE, COMPOSITION PROFILES AND POTENTIAL RISK ASSESSMENT OF POLYCHLORINATED BIPHENYLS (PCBs) IN TRANSFORMER OILS

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Abstract. *One of the most common problems encountered in everyday life is the pollution of the environment. Therefore, regardless of the nature of the daily activities of the population, this destructive effect of the natural balance finds its origin perhaps in the lack of capacity over time to treat the problem of waste (industrial and domestic) as it should. Although important steps have been taken in this regard, and prevention and control rules have been imposed in most countries of the world, these steps must be supported by all socio-economic / political components through clear measures, anchored to global developments. There are among these generic sources of pollution, which are referred to above, processes that requires high power equipment (generators, transformers, etc.) or hydraulic who which also requires thermal insulation fluids. The aim of this study is to investigate the polychlorinated biphenyls (PCBs) presence in the dielectric oil of power transformers. In this matter, fifteen samples were collected from transformer oils, in the summer of the year 2017, and analyzed from a physicochemical point of view. A more specific aim was to corroborate the findings regarding the amount of chlorine in oil samples with the age of the power transformer using statistical methods. The cluster's analysis indicated that the investigated samples are clustered into four major groups, providing useful information for further studies on PCBs mobility in the environment.*

Keywords: *transformer oil; PCB; viscosity; chlorine content; statistical analysis.*

1. INTRODUCTION

Polychlorinated biphenyls (PCBs) are an important group of pollutants, similar in many aspects to dioxins [1, 2]. Polychlorinated biphenyls belong to a broad class of chlorinated aromatic hydrocarbons with a biphenyl structure (i.e., two phenyl rings $(C_6H_5)_2$ and general formula $C_{12}H_{(10-n)}Cl_n$) and at least one hydrogen atom substituted by a chlorine atom (Fig. 1) [3]. Molecules with the same degree of chlorination are isomers; therefore, ten possible isomer groups of PCBs are revealed (Table 1) [1]. Depending on the position and the number of chlorine atoms that replace the hydrogen atoms in the structure, the molecule may have different chemical properties, with a huge chemical stability [1] as well. In this respect, there are 209 different molecules (Table 1), called congeners, of which only about 130 may be found in commercial products [4].

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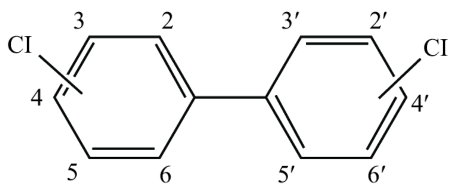

Figure 1. Chemical structure of PCBs.

PCBs were discovered in a wide application field such in the hydraulic and thermal insulation fluids used for electrical transformers and capacitors [5]. PCBs do not easily degrade in the environment, being lipophilic, and tend to bioaccumulate in adipose tissue of humans. Thus, PCBs have been demonstrated as being human carcinogens [6], and causing also a variety of noncancer health effects such as diseases on the reproductive function [7], immune system [8, 9], nervous system [10-12], endocrine system [13-15], or even loss of biological diversity [5].

Table 1. Composition of PCB isomer groups.

PCB isomer group	Chemical formula	Molecular weight	Chlorine percent	Isomers numbers
Monochlorobiphenyl	$C1_2H_9Cl$	188.65	18.79	3
Dichlorobiphenyl	$C1_2H_8Cl_2$	233.10	31.77	12
Trichlorobiphenyl	$C1_2H_7Cl_3$	257.54	41.30	24
Tetrachlorobiphenyl	$C1_2H_6Cl_4$	291.99	48.65	42
Pentachlorobiphenyl	$C1_2H_5Cl_5$	326.43	54.30	46
Hexachlorobiphenyl	$C1_2H_4Cl_6$	360.88	58.93	42
Heptachlorobiphenyl	$C1_2H_3Cl_7$	395.32	62.77	24
Octachlorobiphenyl	$C1_2H_2Cl_8$	429.77	65.98	12
Nonachlorobiphenyl	$C1_2HCl_9$	464.21	68.73	3
Decachlorobiphenyl	$C1_2Cl_{10}$	498.66	71.10	1
Total isomers				209

The Stockholm Convention stipulates that all existing PCBs stock/waste and all PCB-contaminated equipment to be disposed of in an environmentally safe manner (without danger to human health and the environment) by the year of 2025 [16]. The most important categories of waste are used substances or articles which either contain or are contaminated with PCBs (polychlorinated biphenyls) and/or PCTs (polychlorinated terphenyls) and/or PBBs (polybrominated diphenyls), and heavy metal compounds. Regarding these RoHS (Restriction of Hazardous Substances), Directive 2002/95/EC (known as RoHS-Recast) restrict the use of six hazardous materials found in electrical/electronic transformers/products (Table 2) [17, 18]. Even though PCBs are no longer manufactured or used in industrial activities, they continue to be encountered as contaminants of concern at hazardous waste sites and in the environment.

In Romania, the oil from transformers containing PCBs at concentrations of 50 parts per million (ppm) or greater ($\geq$ 50 ppm) is regulated for disposal under the Decision no. 173/2000 for the regulation [19] of the special arrangements for the management and control of polychlorinated biphenyls and other similar compounds (Table 1). Despite that, according to Romanian regulations, it is still authorized the use of intact and non-leaking PCB-containing dielectrics in transformers in order to ensure the useful life of the types of equipment.

Table 2. Admitted concentrations of RoHS [17, 19].

Substances	Allowed Limit [% weight]
Pb	< 0.1 (1000 ppm)
Hg	< 0.1 (1000 ppm)
Cd	< 0.01 (100 ppm)
Cr(VI)	< 0.1 (1000 ppm)
Polybrominated diphenyls (PBBs)	< 0.1 (1000 ppm)
Polybrominated diphenyl ethers (PBDEs)	< 0.1 (1000 ppm)
Polychlorinated biphenyls (PCBs)	Amount < 50 ppm (according to HGR 173/2000)

To overcome these problems through PCBs use, it is very important to shift the world attention towards eco-friendly alternatives, which not only increase the performances of electrical transformers sustainably but also protect human health and the environment [20]. In this respect, the current investigations were performed in order to highlight the amount of chlorine, as well as to establish the physicochemical properties of lubricants as conditions for environment and life quality. Statistical analysis revealed that is a strong relationship between chlorine content in PCBs and the age of power transformers, providing useful information about emission sources and PCBs mobility in the environment.

2. MATERIALS AND METHODS

2.1. MATERIALS. SAMPLING

All chemicals and reagents were purchased from commercial suppliers and were of the highest available purity. Dielectric oil samples were collected from fifteen power transformers, in the summer of the year 2017, establishing in the reporting sheet a series of characteristics (Table 3) related to transformer performance (i.e., age, total weight, rated power, cooling type, cooling medium, oil weight).

Table 3. The characteristics of power transformers.

Code	Year	Total weight [kg]	Oil weight [kg]	Rated power [kVA]	Cooling type*	Cooling medium
S1	1985	1500/Al	405	400	ONAN	Air
S2	1985	1650/Al	384	400	ONAN	Air
S3	1978	1780/Al	434	400	ONAN	Air
S4	1988	1700/Al	434	400	ONAN	Air
S5	1982	1650/Al	384	400	ONAN	Air
S6	1986	1780/Al	434	400	ONAN	Air
S7	1986	1780/Al	434	400	ONAN	Air
S8	2000	760/Al	170	400	ONAN	Air
S9	1989	1430	434	400	ONAN	Air
S10	1986	1780/Al	434	400	ONAN	Air
S11	1985	2500/Al	365	400	ONAN	Air
S12	1988	1780/Al	404	400	ONAN	Air
S13	1982	1780Al	434	400	ONAN	Air
S14	1982	1780Al	434	400	ONAN	Air
S15	1983	1780/Al	431	400	ONAN	Air

**ONAN (previous designations OA) - Oil Natural Air Natural*

The samples were collected with the consent of the economic agent, according to practical guide published by Wells and Hess [21]. This was achieved to provide a better understanding of the management of PCBs in the old transformer's oils.

2.2. METHODS

The analysis of PCBs and other chlorinated organic compounds in the matrices of transformer oil samples was performed by non-specific tests using L2000DXT PCB/Chloride analyzer (Dexsil, Hamden, CT, USA). This device is a versatile and easy-to-use for analyzing samples of dielectric oil, water, soil, and contaminated dust. The range of measured values is 2 - 2000 mg/kg, for oil samples. The L2000DX analyzer is programmed with conversion factors for the main PCBs and most chlorinated substances. The method can apply results correction factors, taking into account the extraction efficiency, the degree of dilution and the control sample. The analysis of each oil sample is done very quickly in about 5 minutes. After calibration, a blank test was performed in order to determine the background level, which is important for the accuracy of the measurements in the range of low concentrations. The preparation steps involve the extraction of the chlorinated organic substances from the oil samples and its treatment with a sodium-based reagent, in order to convert the chlorinated organic substances into chlorides. Subsequently, the chloride level was measured by the electrochemical method [22, 23].

The lower and upper limit of oil samples inflammability were performed using an automates closed cup flash point test system from Tanaka Scientific Limited Corporation, Japan (i.e., APM-7 automated Pensky-Martens Closed Cup tester, PMCC) [24].

Water content determination from oil samples were performed with Karl Fischer coulometric titrator (Mettler-Toledo Ltd., Melbourne, Australia). Engler viscometer (Globecore GmbH, Oldenburg-Eversten, Deutschland) was used for measuring the viscosity of oil samples.

Statistical Package for the Social Science (SPSS) version 20.0 software for MS Windows [25-29] was used to interpretation the analytical results and to investigate the correlations between parameters.

3. RESULTS AND DISCUSSION

The measurements performed on the provided samples have led to the results systematized in Table 4. The analysis of the results presented in Table 4 shows that the density of oils varies between 0.8700 and 0.9082 g/cm^3, and the viscosity of samples varies from 1.14 to 1.29 °E. Non-specific tests were standardized for Arochlor 1242 (i.e., PCB blank sample), with a chlorine content of 42%. Consequently, analysis of samples with more chlorinated PCB isomers (for example, Arochlor 1260, with a chlorine content of 60%) shows a higher result than the actual PCB concentration. Non-specific tests are designed to indicate the presence of chlorine without error.

The density of the oil samples, determined at 20°C, shows values in the range of 0.870 g/cm^3 (S8) and 0.908 g/cm^3 (S10), with an average value of 0.884 g/cm^3 and standard deviation of 0.011 g/cm^3. The recorded values for flash point are similar for all samples: 137-141°C, with an average value of 138.667±1.543°C. Regarding the viscosity of the samples, two parameters were reported: oil viscosity (expressed in °E) and viscosity index. The oil viscosity ranged from 1.17°E (S7) to 1.34°E (S1), with the median value of 1.23±0.05°E. The total PCB concentration in oil samples ranged from 1.17 mg/kg (S8) to 10.30 mg/kg (S2) with an average value of 4.501 ppm and standard deviation of 2.557 ppm.

Table 4. Analysis values for transformers which operated after 1975 and worked in the normal parameters.

Code	Aspect/Color/ Impurities	Density at 20°C [g/cm^3]	Flash point [°C]	Water content [g]	Oil viscosity [°E] / Viscosity Index (V.I.)	PCBs concentration depending on chlorine content [ppm]
S1	Clear/Orange/ Lack of coal	0.8934	137	17.67	1.34 / 111	6.45
S2	Clear/Orange/ Lack of coal	0.8812	139	10.38	1.28 / 98	5.15
S3	Clear/Orange/ Lack of coal	0.8845	137	12.16	1.21 / 99	10.3
S4	Clear/Yellow - citron/Lack of coal	0.8700	140	8.820	1.19 / 87	2.97
S5	Clear/Orange/ Lack of coal	0.8801	139	14.38	1.23 / 96	6.02
S6	Clear/Orange/ Lack of coal	0.8800	137	15.15	1.22 / 99	5.71
S7	Clear/Orange/ Lack of coal	0.8819	138	14.22	1.17 / 101	3.30
S8	Clear/Yellow - citron/Lack of coal	0.8700	141	8.563	1.19 / 87	1.17
S9	Clear/Yellow - citron/Lack of coal	0.8723	141	8.614	1.20 / 88	1.37
S10	Clear/Orange/ Lack of coal	0.9082	137	16.23	1.28 / 99	6.65
S11	Clear/Orange/ Lack of coal	0.8912	139	10.38	1.32 / 95	6.90
S12	Clear/Yellow - citron/Lack of coal	0.8726	141	8.609	1.21 / 88	1.22
S13	Clear/Orange/ Lack of coal	0.8893	138	15.45	1.26 / 103	4.70
S14	Clear/Orange/ Lack of coal	0.8934	139	12.34	1.28 / 97	3.39
S15	Clear/Orange/ Lack of coal	0.8914	137	14.87	1.26 / 98	4.17

The obtained results highlight that all samples have PCBs content higher than the reference dose (RfD) established for oral exposure to Aroclor 1016 ($7\cdot10^{-5}$ mg/kg/day) and Aroclor 1254 ($2\cdot10^{-5}$ mg/kg/day) [30, 31]. The oral R_fD is based on several studies led on PCBs effects on monkey: hairline hyperpigmentation, nail changes (prominent beds, elevated nails), decreased birth weight, possible neurologic impairment (deficits in discrimination-reversal learning and delayed spatial alternation), reduction of I_gG and I_gM antibodies, and human infants: deficits in visual recognition and short-term memory [30, 31].

However, the Integrated Risk Information System (IRIS) of U.S. Environmental Protection Agency, National Center for Environmental Assessment, does not offer the reference concentration (RfC) for inhalation exposure to PCBs [32]. The inhalation exposure is more probable than swallowing.

PCB concentrations were highest in older transformers and decreased as transformers were installed earlier (Fig. 2).

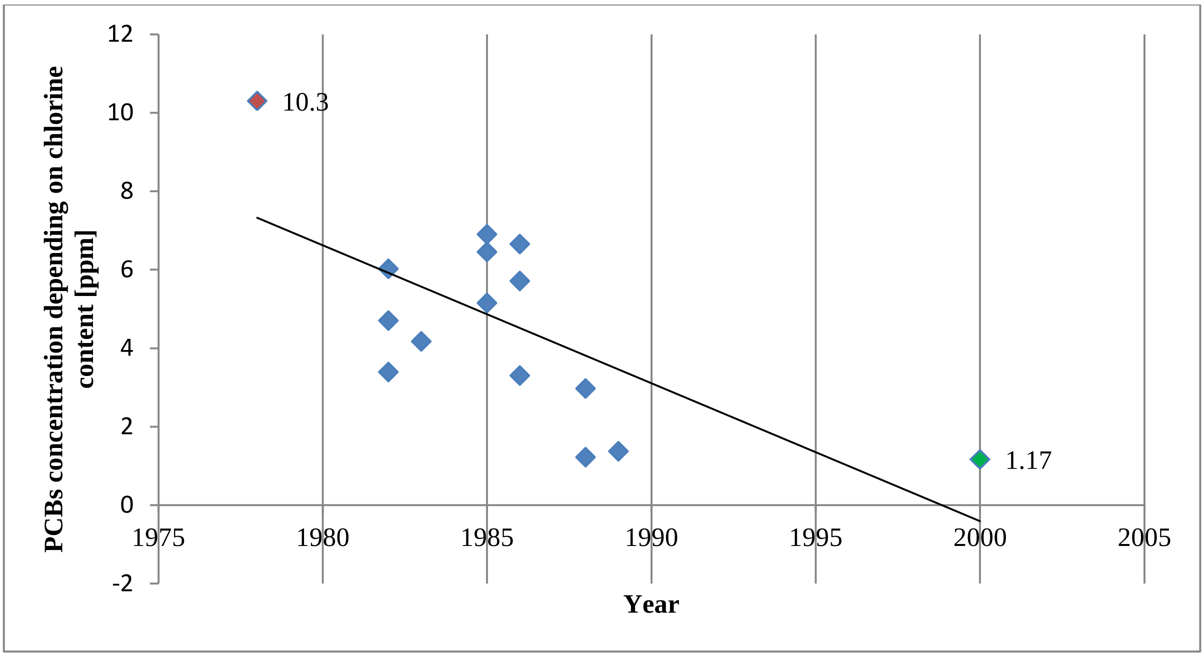

Figure 2. Dynamic analysis of PCBs concentration determined in transformers.

Analysis of variance was used to assess the difference in contamination levels between the studied transformers, while the Pearson correlation, and Sperman`s indicators were used to establish relationships among the PCB content identified and with other oil properties. All statistical analyses were carried out at a significance level of p = 0.05.

Table 5. Pearson correlations of the analysed variables.

		Density	Flash point	Water content	Oil viscosity	PCBs concentration
Density	Pearson Correlation	1	-.721**	.718**	.723**	.560*
	Sig. (2-tailed)		.002	.003	.002	.030
Flash point	Pearson Correlation	-.721**	1	-.871**	-.409	-.753**
	Sig. (2-tailed)	.002		.000	.130	.001
Water content	Pearson Correlation	.718**	-.871**	1	.444	.520*
	Sig. (2-tailed)	.003	.000		.097	.047
Oil viscosity	Pearson Correlation	.723**	-.409	.444	1	.441
	Sig. (2-tailed)	.002	.130	.097		.100
PCBs concentration	Pearson Correlation	.560*	-.753**	.520*	.441	1
	Sig. (2-tailed)	.030	.001	.047	.100	
	N	15	15	15	15	15

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

A moderate correlation relationship can be observed between PCBs concentration and density, water content and oil viscosity and Sig. <0.05, demonstrates a directly proportional relationship. Between the density and water content, or between oil viscosity and density it can be observed a high dependency relationship, sustained by the fact that the value of the Person coefficient is higher than 0.7. The flash point variable is the only characteristic about doesn't affirmed it is in a relation of correlation with other analyzed variables.This can be explained by the fact that the values of this indicator do not depend on the dynamics of the other variables or their evolution. Applying statistical tests on the data collected is a reliable way to characterize the statistical population. Based on the information presented in Table 5, is observed the existence of a moderate or high connection between the analyzed variables.

Sperman's rank correlations also validate the information obtained through the Pearson coefficient as it can see from the information presented in Table 6.

Table 6. Sperman's correlations of the analysed variables.

		Density	Flash point	Water content	Oil viscosity	PCBs concentration
Density	Correlation Coefficient	1.000	-.688**	.720**	.769**	.624*
	Sig. (2-tailed)	.	.005	.002	.001	.013
Flash point	Correlation Coefficient	-.688**	1.000	-.858**	-.407	-.710**
	Sig. (2-tailed)	.005	.	.000	.132	.003
Water content	Correlation Coefficient	.720**	-.858**	1.000	.529*	.599*
	Sig. (2-tailed)	.002	.000	.	.043	.018
Oil viscosity	Correlation Coefficient	.769**	-.407	.529*	1.000	.631*
	Sig. (2-tailed)	.001	.132	.043	.	.012
PCBs concentration	Correlation Coefficient	.624*	-.710**	.599*	.631*	1.000
	Sig. (2-tailed)	.013	.003	.018	.012	.
	N	15	15	15	15	15
Correlation is significant at the 0.01 level (2-tailed).**						
Correlation is significant at the 0.05 level (2-tailed).*						

The value of correlation coefficient ranges between − 1 (indicating perfectly negative correlation) and + 1 (indicating perfectly positive correlation). The sign indicates the direction of the trend (i.e., positive or negative), and the absolute value quantifies the strength of the relationship [33]. Thus, in our case it can observe a strong positive relationship between four of the analyzed variables (PCBs concentration, density, water content and oil viscosity) and a strong negative correlation between flash point and the other analyzed variables.

Hierarchical cluster analysis was used to classify the variations in Σ15 PCB concentrations of the investigated samples by using the rescaled distance cluster combine and average linkage methods. The clustering of the sampling is summarized in the dendrogram presented in Figs. 3-4.

The cluster analysis indicated that the investigated samples are clustered into four major groups. The first group of samples (S8, S9 and S12), are characterized by a very low levels of PCBs (1.17–1.37 ppm). For the second group (S1, S5, S6, S10, and S11) the PCBs concentrations are the highest and ranges from 5.71 to 6.90 ppm. The third group (S4, S7, and S14), reached values of PCBs concentration between 2.97 and 3.39 ppm, while, the last group (S2, S13, and S15) showed values between 4.17 and 5.15 ppm. The sample S3 does not fit into any groups, reaching the highest value of the concentration of PCBs (10.3 ppm).

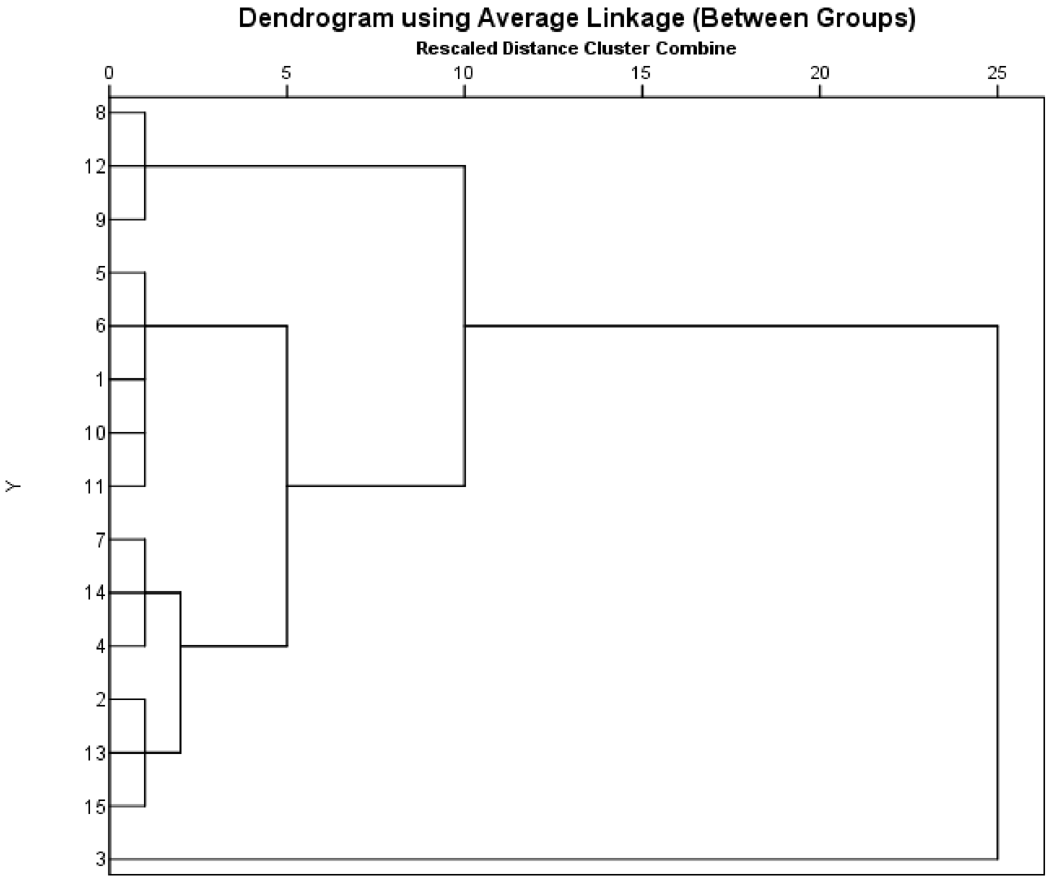

Figure 3. Hierarchical cluster analysis showing the dendrogram of 15 oil samples.

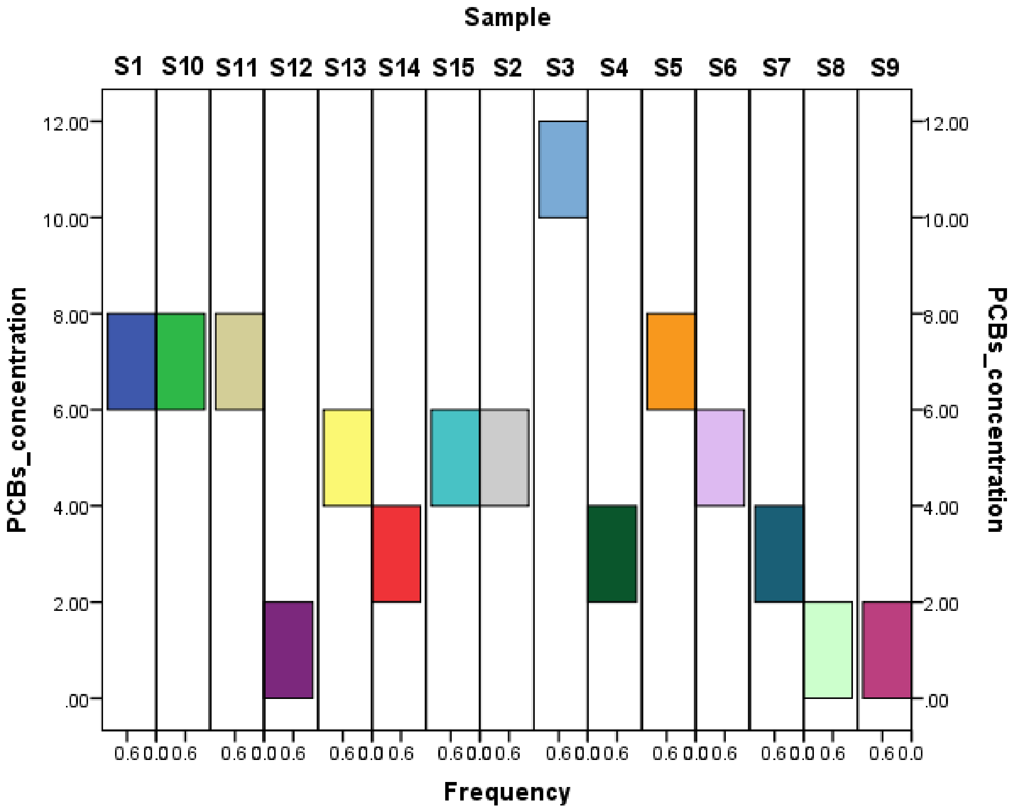

Figure 4. Samples classification about PCBs concentration.

As it may be seen in Fig. 4, apart from the major classes resulting from the dendrogram, a single sample S110 has a much higher concentration of PCB, which cannot be included in any of the groups presented above.

4. CONCLUSIONS

The oil used as dielectric for transformers losing the physicochemical properties during operation. In this study, fifteen oil samples collected from power transformers were analyzed from several point of view, such as aspect (all samples were clear), color (from yellow to orange), impurities (all samples present lack of coal), density at 20°C (values ranged from 0.870 to 0.903 g/cm^3); flash point (137-141°C), water content (8.563-17.670 g/L), oil viscosity (1.17-1.34°E) and PCBs concentration depending on chlorine content (1.17-10.30 ppm).

The PCBs content is proportional with the age of the power transformer and the cluster analysis indicated that the investigated samples are clustered into four major groups.

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