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DISTINGUISHING BRANDS OF A-4 PAPER THROUGH ELEMENTAL ANALYSIS USING LASER-INDUCED BREAKDOWN SPECTROSCOPY AND PRINCIPAL COMPONENT ANALYSIS

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Abstract

In the realm of forensic science and materials analysis, understanding the composition and characteristics of everyday materials can unveil a plethora of information. This study leverages the precision and efficiency of Laser-Induced Plasma Spectroscopy (LIBS) to dive deep into the elemental makeup of A-4 paper across different brands, offering applications beyond academic research. The results can offer a valuable reference in forensic investigations where distinguishing between paper types could provide key evidence in document authentication or fraud cases. Furthermore, the method provides insights for quality control in paper manufacturing by identifying unique elemental markers that can verify the origin and consistency of paper products. This innovative research employed the ECCO®² brand Laser-Induced Breakdown Spectroscopy (LIBS) device, a state-of-the-art spectrometric system. The device was used to analyze samples from seven distinct A4 paper brands: Mopak®, Office International Copier Bond®, Double A®, Excellent Copy Paper®, Gold®, Master®, and Vera®. Controlled via a computer system and specialized ECCO®² software, the ECCO®² facilitated a comprehensive elemental analysis of these paper samples, focusing on the presence of specific inorganic materials such as glass, inks, and toners that hold forensic value. The study meticulously selected 11 elements (K, Sr, Si, Na, Ba, Mg, Ca, C, Al, Fe, and Ti) for elemental data analysis, based on their relevance and potential to reveal the composition and source of the materials. These elements were chosen due to their prevalence in common additives, fillers, and bleaching agents used in paper manufacturing, making them effective markers for differentiating paper brands based on elemental composition. The LIBS technique, a non-contact and rapid analytical method, allowed for the precise detection of these elements without the need for sample preparation, showcasing its practicality in forensic and analytical applications.

Through Principal Component Analysis (PCA), 76.65% discrimination was initially achieved among the A4 paper samples. Notably, when three elements (Al, Fe, and Ti) were excluded, discrimination increased to 83.78%. Further analyses using Principal Component Analysis (PCA) on peak height and peak area data from A4 paper samples yielded discrimination rates of 80.916% and 85.947%, respectively. In this context, peak height refers to the maximum intensity of each elemental emission line, while peak area represents the integrated area under each spectral peak. These distinct spectral metrics capture different aspects of the elemental composition of A4 paper samples, enhancing PCA's ability to effectively distinguish between paper brands. Finally, when PCA was performed using six elemental pairs identified by the ECCO®^{2} device as optimal for A4 paper differentiation, the discrimination rate reached 97.313%, with variation values calculated at approximately 80%. These results underscore the LIBS method's reliability and precision in distinguishing different paper types, affirming its valuable application for forensic and quality control purposes.

Keywords: LIBS, A-4 office paper, elemental analysis, PCA

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1. Introduction

Document review is an important forensic discipline, and the legal system regularly requires the knowledge and skills of a scientist when documents under question are involved in criminal or civil matters. Among the many aspects of scientific review of documents, elemental analysis can provide useful results. Verification and identification of the source of written or printed documents can be applied to a variety of documents of forensic value : ransom notes, altered wills, insurance fraud, counterfeit money, travel and identity documents, business and personal checks, legal malpractice, money order, prescription labels, traveler's checks, medical records and financial documents [1], [2]. When a fraudulent page entry is suspected in a multi-page document, the forensic examiner usually tries to answer some basic questions. These questions include: Is the document an original or a copy? Was more than one technology used to prepare the document? Is there any evidence that the text has been changed or added? Does the page have obvious imperfections that could indicate machine customization? What type of ink or toner product was used, and are there any unique identifiers or variations in printing techniques? These aspects help establish the document's authenticity. [3] Various analytical methods can be applied to distinguish document elements using their physical and optical properties. Elemental composition can improve the discrimination and identification of samples. These methods can be relatively simple or more complex to implement, and their respective specificities and sensitivities also vary. Improvements in quality control during the modern papermaking process limit the variability in the physical and optical properties of such materials, thereby reducing the ability to distinguish samples from different sources [4], [5], [6]. Several studies have been conducted on evaluating different instrumental methods for classifying and distinguishing paper documents using chemical composition data. Applied methods include Flame Atomic Absorption Spectroscopy (FAAS), X-ray Fluorescence Spectroscopy (XRF), X-ray photoelectron spectroscopy, Neutron Activation Analysis (NAA), Scanning Electron Microscopy (SEM-EDX) with Energy Dissipative X-ray Analysis and inductive Combined plasma-based methods are available [5], [7], [8]. However, few studies have included laser-induced plasma spectroscopy (LIBS) for the detection of trace elements in paper and ink samples [5], [7], [8]. Despite the potential benefits of LIBS over other basic analysis techniques – simple, fast, simultaneous, multi-element detection without sample preparation (direct on paper), minimal sample destruction, and reduced instrument cost and maintenance – it is a routine forensic technique for basic profiling of interrogated documents. it is not yet widely used [9]. The forensic analysis of paper, a seemingly mundane material, has taken a significant leap forward with the advent of advanced spectroscopic techniques and statistical methods.

Recent literature showcases the remarkable efficacy of these methods in distinguishing between A-4 paper samples, underlining the potential of elemental analysis in forensic science and beyond. Techniques such as Multivariate Curve Resolution-Alternating Least Squares (MCR-ALS), Laser-Induced Breakdown Spectroscopy (LIBS), Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS), and various statistical analyses have demonstrated exceptional accuracy rates in paper identification, pushing the boundaries of what's possible in material discrimination. The implications of these findings are far-reaching. In forensic document examination, the ability to accurately differentiate between papers can be crucial in authenticating documents or identifying forgeries. Beyond forensic applications, these techniques offer valuable tools for quality control in the paper manufacturing industry, ensuring consistency and adherence to specifications. Moreover, the environmental aspect of paper production and recycling can benefit from these analytical methods. By precisely identifying the composition of paper, manufacturers can optimize raw materials usage, improve recycling processes, and reduce waste, contributing to more sustainable practices. The purpose of this study is; To develop fast, reliable, and sensitive methods for detecting fake documents by using spectroscopic fingerprint information from A-4 paper samples. In this context, the steps are focused on optimizing the measurement parameters, determining the appropriate data analysis technique, and supporting the obtained results with PCA, which is the reference analysis technique.

2. Material and method

The ECCO®² spectrometer used in this study consists of four distinct parts, each equipped with an optical sensor capable of detecting radiation within a specified wavelength range. Before analysis, each of the four sensors was calibrated. While the main unit of the ECCO®² system was pre-calibrated during manufacturing, additional calibration was performed using Standard Glass (NIST 1411), which is supplied with the equipment. This calibration process ensured accuracy in detecting emission lines, and the resulting ECCO®² spectrum, showing characteristic emission lines at specific wavelengths with the Standard Glass, is illustrated in Fig 1. For the analysis, the paper samples were handled with latex gloves to prevent contamination and placed in the chamber of the device. The MOPAK brand was analyzed first, with 10 laser shots fired at each sample, after which the corresponding ECCO®² spectrum for MOPAK was generated and recorded using the ECCO®² software on the PC. This procedure was repeated for each paper sample, and all ECCO®² spectra were systematically documented.

In terms of data analysis, the initial focus was on identifying a set of emission lines, elements, and/or isotopes, depending on the analytical method and sample type. From there, a series of elemental ratios were generated to facilitate comparison. To analyze the laser-induced breakdown spectroscopy (LIBS) data, the Principal Component Analysis (PCA) method, one of the multivariate data analysis techniques, was applied to assess the discriminatory power between the paper samples. PCA helped in reducing the number of variables, grouping the samples, and determining the distinguishing features of each brand. For this purpose, we used Paleontological Statistics Software (PAST Version 4.03 for Windows 10; Hammer, Ø., Harper, D.A.T., and Ryan, P.D. 2001). The PCA method allowed for the effective separation of the paper brands based on their elemental composition, ensuring they could be distinguished from one another with high accuracy.

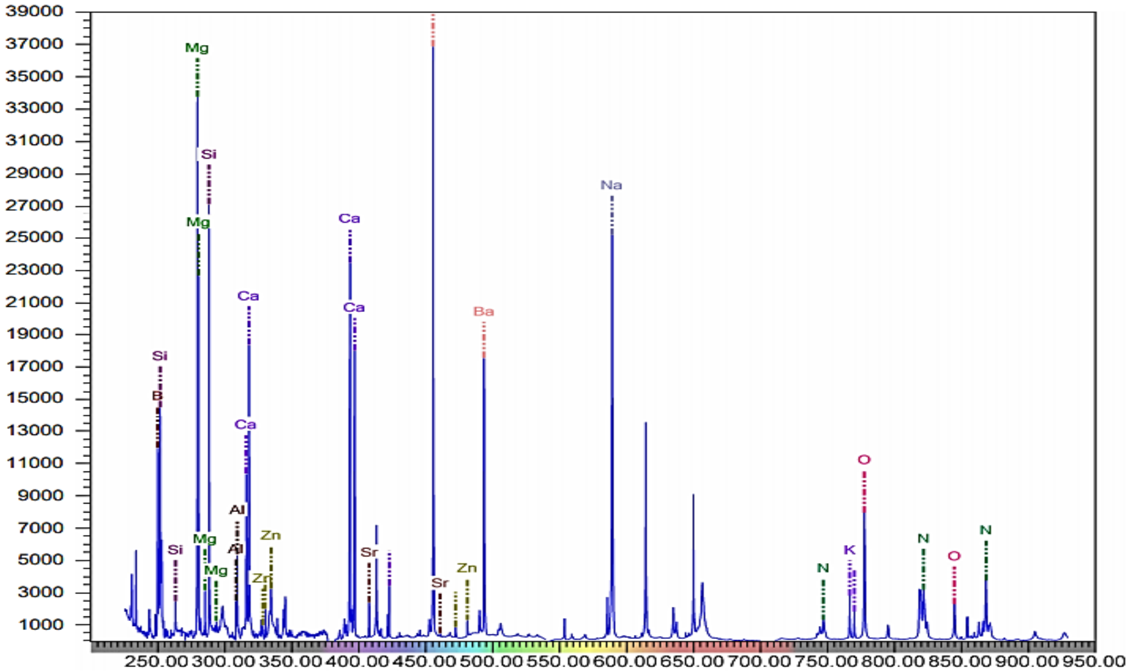

Figure 1. ECCO®² calibration spectrum for standard glass (NIST 1411).

Table 1. Examined papers with origin and year of production.

A-4 PAPER BRAND	ORIGIN	YEAR OF PRODUCTION
MOPAK®	Kemalpaşa, Izmir, Türkiye	2019
Office International Copier Bond®	Sefaköy, Istanbul, Türkiye	2018
Double A®	Thailand	2018
Excellent Copy Paper®	Mindanao, Philippines	2017
Gold®	Selangor,Malaysia	2019
Master®	People's Republic of China	2019
Vera®	Brazil	2019

3. The main result

3.1 Analysis of A-4 paper samples belonging to different brands

The target samples were placed inside the ECCO®² system and 10 laser shots were fired at it. Afterward, the obtained spectrum was found in Fig.2, Fig.3, Fig.4, Fig.5, Fig.6, Fig.7, and Fig.8. In this study, the basic elemental concentrations of white office papers, which are frequently encountered in forensic document analysis, were examined using the LIBS method. Thus, an attempt was made to distinguish the papers from each other.

The results indicate that the initial discrimination rate for A4 paper samples produced at different times and locations was 76.65%. This rate improved by 7.138%, reaching 83.788%, when a common elemental subset was used. Efforts to enhance sample distinguishability included analyzing values based on peak height and peak area. Analysis using peak height yielded a discrimination rate of 80.916%, while peak area analysis achieved a higher rate of

85.947%. The highest discrimination power, 97.313%, was obtained by analyzing elemental pairs, demonstrating the effectiveness of this approach in distinguishing between samples.

In this study, the homogeneity and variation of the elemental composition within the paper were not examined, as it was reported by Trejos et al. that no significant difference was observed for certain areas within a single paper as a result of the analysis conducted with LIBS [5].

The MCR-ALS approach achieved a paper identification rate of 96.3%, showcasing the strength of this method in decomposing complex data matrices without prior information about the composition or number of components [12]. The combination of LIBS and LA-ICP-MS provided an exemplary discrimination rate of 98% between samples, highlighting the complementary strengths of these techniques in elemental analysis. While LIBS offers rapid, in situ analysis with minimal sample preparation, LA-ICP-MS brings a high level of sensitivity and specificity to elemental quantification [5]. LIBS Analysis of 17 Paper Samples yielded a discriminatory power of 98.5%, proving its efficacy in handling a broad range of samples with high accuracy [9]. Ternary plot, PCA, and Student's t-test methods reported discrimination rates ranging from 99.4 to 100%, depending on the applied method [15]. These statistical analyses enhance the discrimination power by effectively handling the complexity of the data obtained from spectroscopic measurements. In this study, the highest discrimination rate of elemental pairs obtained by the PCA method was 97.313%.

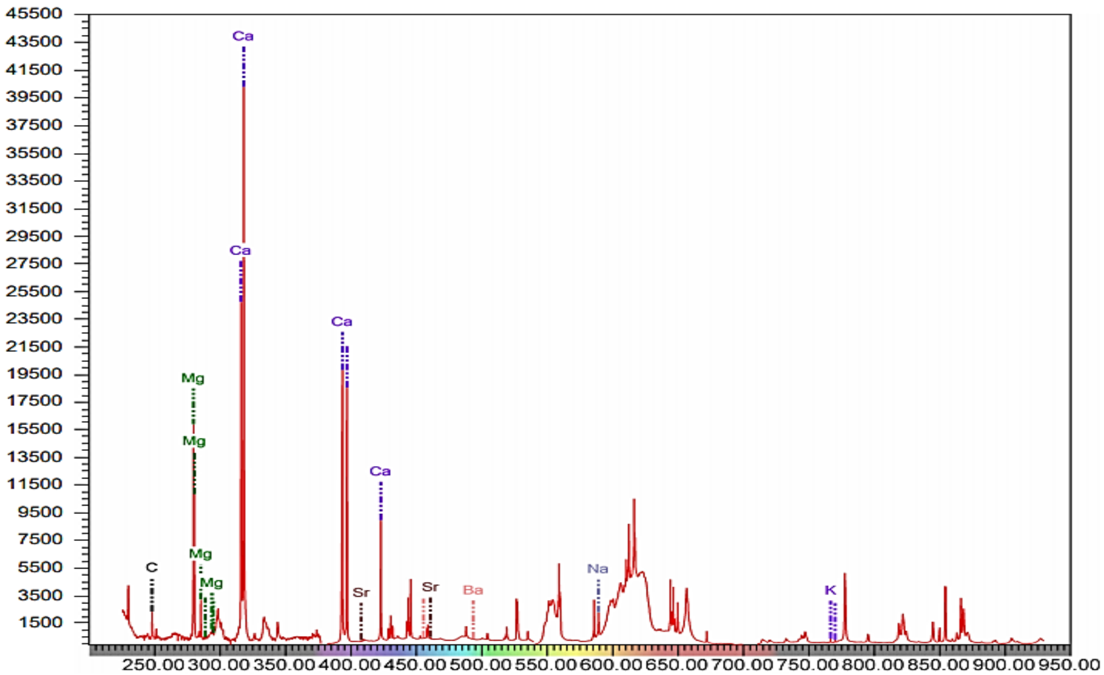

Figure 2. Spectrum of A4 paper sample from MOPAK brand.

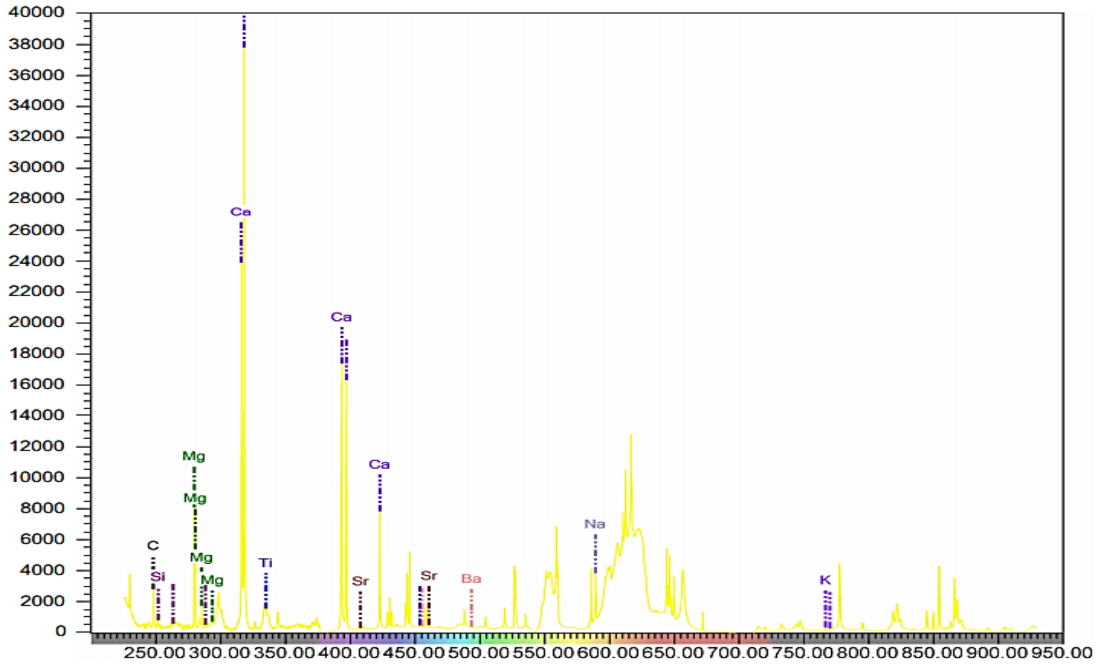

Figure 3. Spectrum of A4 paper sample from OFFICE INTERNATIONAL COPIER BOND brand.

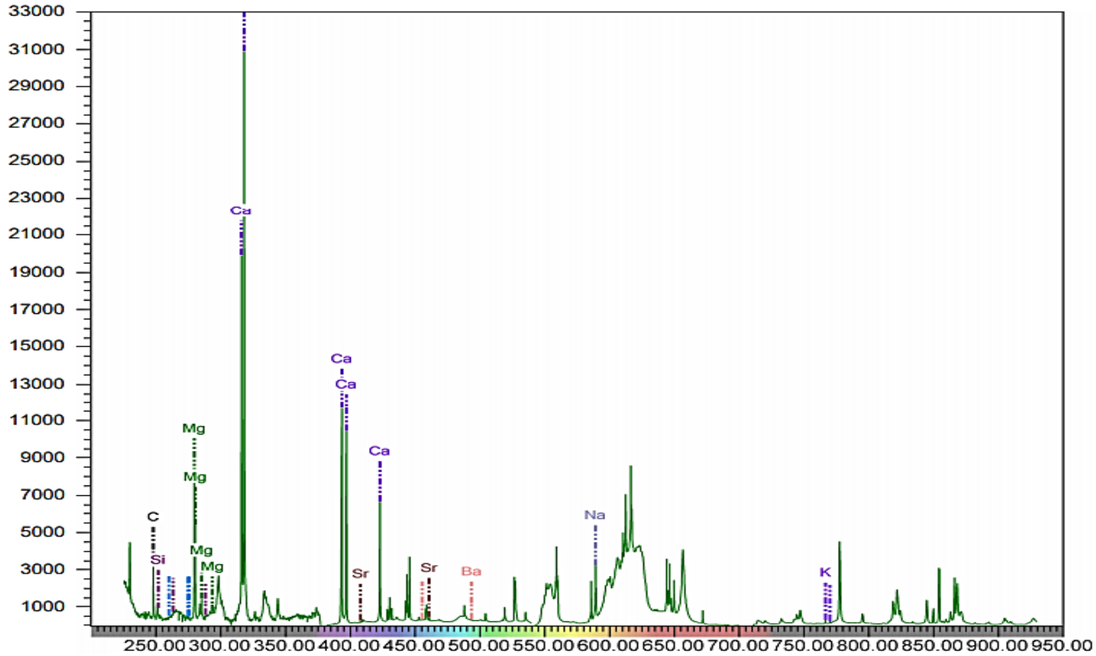

Figure 4. Spectrum of A4 paper sample from DOUBLE A brand.

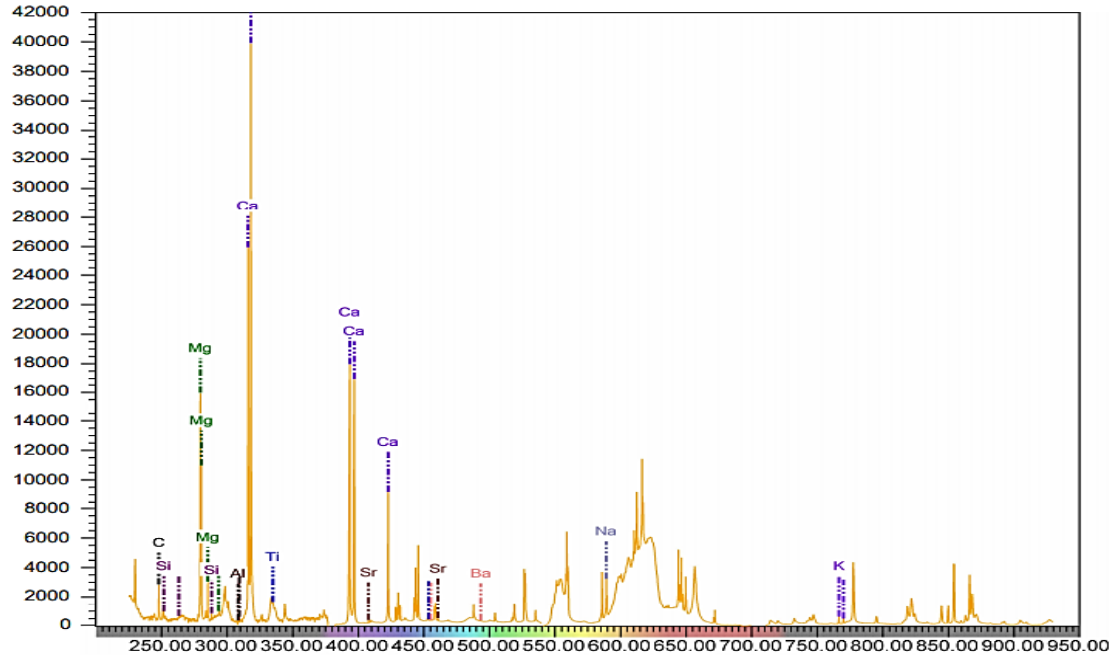

Figure 5. Spectrum of A4 paper sample from EXCELLENT COPY PAPER brand.

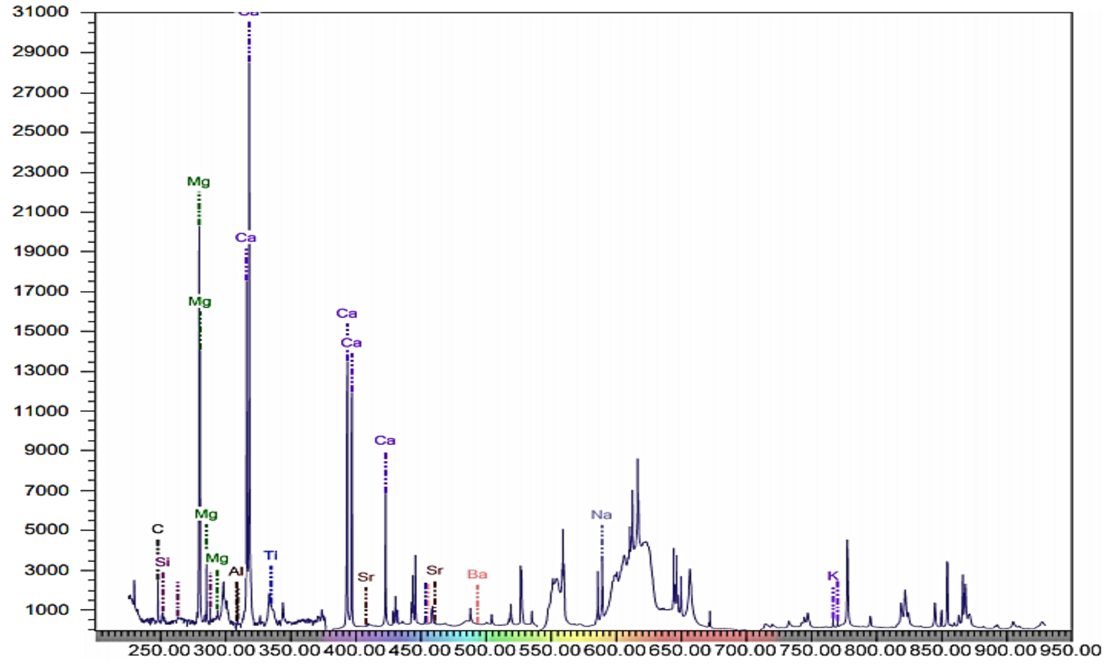

Figure 6. Spectrum of A4 paper sample from GOLD brand (ECCO®²).

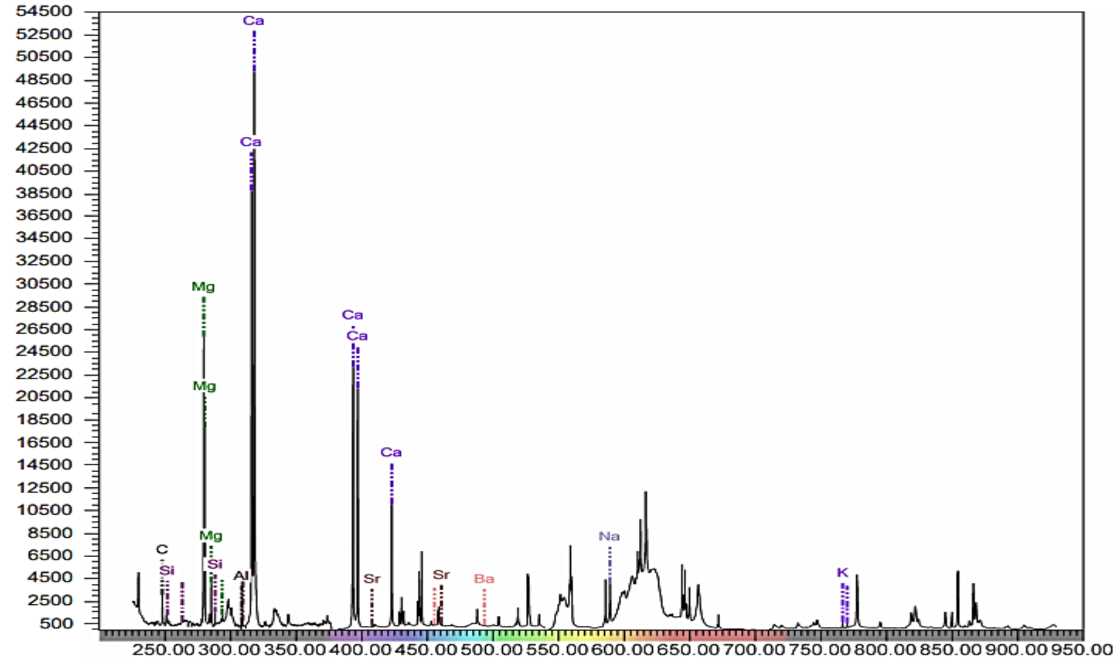

Figure 7. Spectrum of A4 paper sample from MASTER brand.

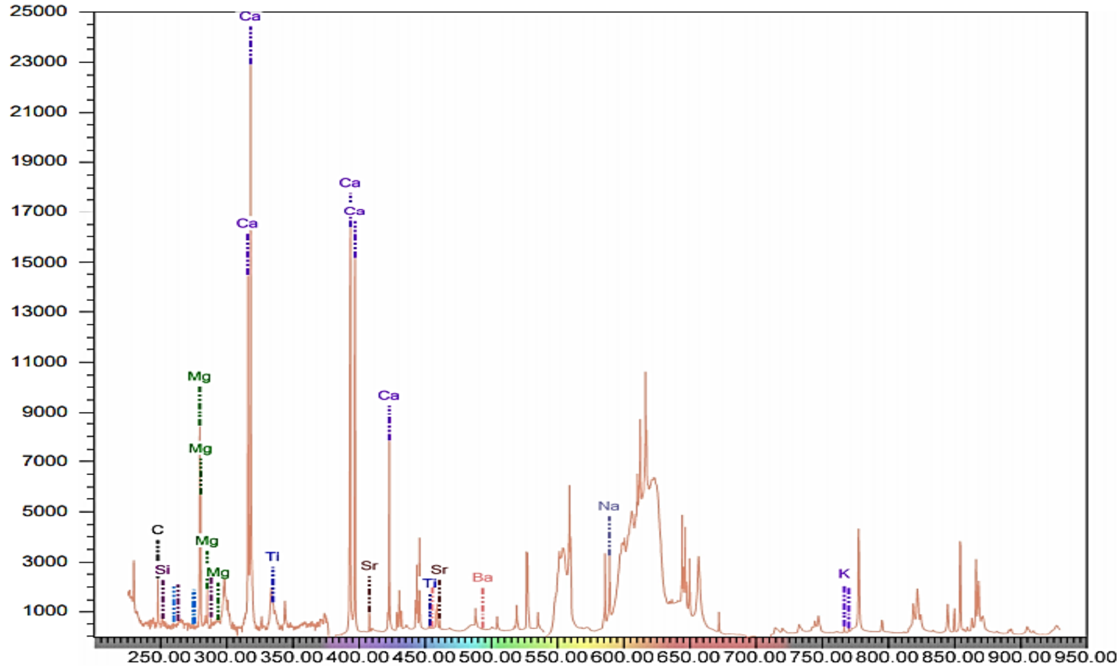

Figure 8. Spectrum of A4 paper sample from VERA brand.

3.2 Evaluation of LIBS ability to discriminate A-4 papers with PCA

The study's most novel approach involved PCA with six elemental pairs presented by the ECCO®² device, pushing the boundaries of conventional analysis. Although specific outcomes of this particular analysis were not detailed, it signifies a promising direction for future research, potentially offering even more refined discrimination capabilities.

Among the chemometric methods, PCA was used to cumulatively evaluate major and minor elemental composition differences between paper samples. Classification of groups of paper samples was carried out by PCA. For this purpose, in the first stage, Match (% Match) data (Table 5) between the spectrum data of the samples and the characteristic spectra of pure elements was used. PCA analysis was performed with these data. In the second stage, the height peak data of all peaks (Table 2) and in the third stage, the peak area values of all peaks (Table 3) were examined with the PCA technique. In the final stage, the comparison rates of the elemental pairs (Table 4), which were presented as the best determinants of A-4 papers, were examined with the PCA technique.

Table 2. Heights of peaks.

EMISSION LINE	CALIB.	MOPAK	BOND	DA	COPY	GOLD	MSTR	VERA
Na (589 nm)	25212.6	2380.9	3865.5	3294	3263.5	3450	4141	3262
Mg (279.55 nm)	35208	16148.3	8337	8342.4	16447.7	20599.5	26511.6	8601.9
Mg (518.36 nm)	388	1012.3	1218.7	742.3	1111.4	1041.5	1310.1	899.3
Al (309.28 nm)	5487.9	407	263.2	381.1	610.7	506.1	738.5	435.6
Si (288.16 nm)	28008.1	553.6	508.5	573.8	829.1	1176.5	1511.6	754.5
Ca (317.93 nm)	18505.2	40286.6	37745.1	30951.7	40104.5	28509.5	49272	22911.7
Ca (422.67 nm)	3363	8862.5	7736	6612.5	9071.8	6794.7	10965.6	7724.7
Ca (430.25 nm)	484.1	2104.6	2289.6	1560.5	2371.8	1720.8	2914.3	1870.1
Ca (442.54 nm)	251.8	1663.7	1883.2	1332.9	1916.7	1374	2493.9	1414.2
Ca (445.59 nm)	620.5	4663.1	5120.1	3656.4	5448.9	3703	6824.8	3877
Ca (558.88 nm)	505.3	5921.6	6912.7	4335	6527.3	5149.4	7591	6111.1
Fe (438.35 nm)	243.4	334.2	306.2	256.1	283.2	235.7	283.2	238.2
Sr (407.77 nm)	2509.2	216.7	182.6	207.3	312.1	185.2	273.8	929.3
Sr (460.73 nm)	381	408.4	355.4	360.3	424.1	350.2	426.4	607.9
C (247.86 nm)	1748.2	2391.9	2816	3204.5	2847.2	2536.5	3085	2360.1
Mn (403.31 nm)	199.4	139.2	136.2	129.5	144.1	141	147.9	141.5

Table 4. Comparison ratios of elemental analysis for A4 office papers (Presented by ECCO®[2]).

RATIO	CALIB.	MOPAK	BOND	DA	COPY	GOLD	MSTR	VERA
$\frac{Mg\ (279.55)}{Ca\ (317.93)}$	1.903	0.401	0.221	0.27	0.41	0.723	0.538	0.375
$\frac{Sr\ (407.77)}{Ca\ (445.59)}$	4.044	0.046	0.036	0.057	0.057	0.05	0.04	0.24
$\frac{Sr\ (460.73)}{Ca\ (445.59)}$	0.614	0.088	0.069	0.099	0.078	0.095	0.062	0.157
$\frac{Na\ (589)}{Ca\ (558.88)}$	49.894	0.402	0.559	0.76	0.5	0.67	0.546	0.534
$\frac{Al\ (309.28)}{Ca\ (317.93)}$	0.297	0.01	0.007	0.012	0.015	0.018	0.015	0.019

Table 3. Peak areas for all peaks.

EMISSION LINE	**CALIB.**	**MOPAK**	**BOND**	**DA**	**COPY**	**GOLD**	**MSTR**	**VERA**
Na (589)	20070.2	2436.7	3731	2949.6	3100.8	3306.7	3841.5	3264.8
Mg (279.55)	20763.1	11012.1	5195.8	5280	10822.3	14271	17441.2	5841.6
Mg (518.36)	632.8	1203.2	1332.6	864.5	1388.4	1354.3	1646	1160.7
Al (309.28)	3106.8	708.4	514.7	653	672.4	694.8	791	753.5
Si (288.16)	11127.5	909.3	817.3	932.7	1023.9	1069.1	1221.8	959.7
Ca (317.93)	10070.6	24348.3	22828.3	17622.2	22788.7	19865.2	35013.6	15336.7
Ca (422.67)	1567.9	4935.3	4889.9	3940.3	5205.9	4106.3	6352.5	4478.2
Ca (430.25)	509.3	1968.7	2062.1	1438.7	2112.6	1605.1	2489.8	1770
Ca (442.54)	458.9	2249.6	2376.6	1660	2426	1796.3	2866.7	1910.8
Ca (445.59)	646.7	3169.7	3519.1	2388.2	3487.1	2681.4	4247.6	2890.3
Ca (558.88)	609.8	6848.3	8433.3	5067.9	7587.4	6402	8751.1	7700
Fe (438.35)	385.6	699	591.3	533.9	589	500.9	603.3	490
Sr (407.77)	1225.7	261.8	255.6	268.2	297.9	255.7	285.5	526.3
Sr (460.73)	485.8	643.1	542.8	497.5	574.9	505.3	570.8	648.7
C (247.86)	1164.2	1938.5	2084.1	2270.8	2132.2	1772.1	2300.4	1904.6
Mn (403.31)	345.2	243.2	255	233.9	255.6	254	264.2	250.8
$\frac{\boldsymbol{Ca}\ (\mathbf{442.54})}{\boldsymbol{Fe}\ (\mathbf{438.35})}$	1.035	4.979	6.151	5.205	6.769	5.83	8.808	5.936

3.2.1 Evaluation of match (% match) data with PCA method

The role of experimental data, which is considered a component in PCA analysis, is very important in this technique. This technique uses the method of calculating the weights of the effects of the data on the result while revealing the relationship between the data. Therefore, PCA analysis is examined for two cases in this section.

The first case was to examine all values in Table 13 with this technique. The elemental data harvested through LIBS underwent Principal Component Analysis (PCA), a statistical method used to reduce the dimensionality of data while preserving most of the variation in the dataset. This method was instrumental in analyzing the elemental compositions and distinguishing between the A-4 paper samples. Initially, the PCA on all element ratios yielded a 76.65% discrimination rate among the samples. Intriguingly, when the analysis was refined by removing three elements (Al, Fe, and Ti) to create a common elemental subset, the discrimination rate saw a significant increase to 83.78%.

This difference value of 7.138% from the initial situation corresponds to a significant increase due to the small number of samples selected and the fact that A-4 papers are known to be different from each other.

Table 5. Match (% match) values from LIBS analysis of A4 paper samples.

ELEMENTS	MOPAK	BOND	DA	COPY	GOLD	MSTR	VERA
K	94	93	91	91	93	91	90
Sr	93	91	89	92	89	91	91
Si	93	30	21	50	68	55	44
Na	92	70	70	73	71	88	81
Ba	89	50	44	55	49	63	41
Mg	87	77	76	89	91	90	76
Ca	84	81	81	83	72	82	78
C	81	80	85	83	76	80	76
Al	74	0	0	50	32	59	0
Fe	44	0	22	20	0	0	21
Ti	22	21	0	22	21	20	22

3.2.2 Evaluation of data of height peaks by PCA

In this section, the height peak values in Table 1 were examined by the PCA technique. Thus, 80.916% discrimination ability was achieved between paper samples.

The results PCA Score Graph are shown in Figure 9. In addition, score values and summary information for this graph were obtained in Tables 6 and 7.

Table 6. Summary of PCA results using peak height data across all spectral peaks.

BASIC COMPONENT (PC)	EIGENVALUE	% VARIANCE
1	1.07741E08	**80.916**
2	2.34085E07	17.58
3	1.40287E06	1.0536
4	426928	0.32063
5	164061	0.12321
6	7852.67	0.0058976

Table 7. PCA score values from peak height data analysis of all spectral peaks.

SAMPLES OF PAPER	PC 1	PC 2	PC 3	PC 4	PC 5	PC 6
MOPAK	4395.2	-1616.8	-845.93	-1186.5	-312.61	-59.359
BOND	-2004.8	-6722.3	401.13	776.25	-506.73	-25.662
DA	-8014.6	-2702.3	-1592.9	252.47	602.26	-22.734
COPY	4605.9	-1343	214.67	-207.39	69.025	192.24
GOLD	-3050.4	8776.1	-868.77	444.23	-350.81	11.155
MSTR	18160	1729.4	807.66	309.77	335.7	-67.343
VERA	-14091	1879	1884.2	-388.84	163.17	-28.299

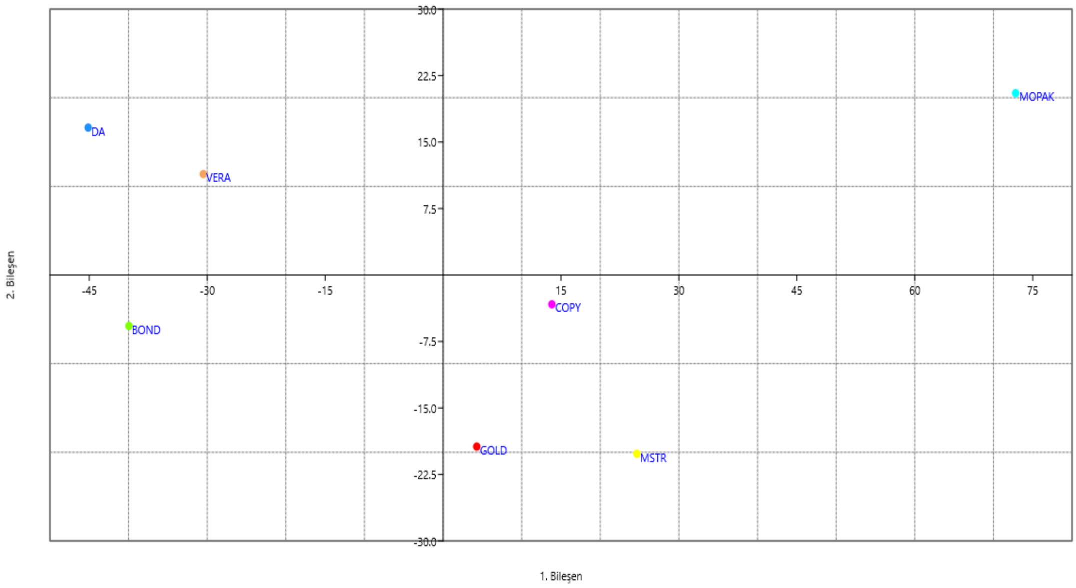

Figure 9. PCA score graph for matching (% match) rates of all elements.

3.2.3 Evaluation of peak area data of Aall peaks with PCA

In this section, the peak area values in Table 2 were examined with the PCA technique. Further analysis of the peak height data across all elements yielded a success rate of 80.916% in distinguishing the samples. This discrimination rate improved significantly when peak area data were analyzed, rising to 85.947%. This progression highlights the critical role of selecting the appropriate data representation in enhancing analytical outcomes.

The results of the PCA Score Graph are found in Figure 10. Additionally, score values and summary information for this graph are given in Tables 8 and 9.

Table 8. Summary of information from PCA applied to peak area data of all peaks.

PRINCIPAL COMPONENT (PC)	EIGENVALUE	% VARIANCE
1	5.73007E07	**85.947**
2	8.00057E06	12
3	1.14521E06	1.7177
4	157538	0.2363
5	50990.8	0.076482
6	14921.2	0.022381

Table 9. PCA score values from analysis of peak area data for all peaks.

Paper Samples	**PC 1**	**PC 2**	**PC 3**	**PC 4**	**PC 5**	**PC 6**
MOPAK	1999.5	-28.249	-807.94	683.84	-256.79	-60.022
BOND	-2262.4	4381.5	587.82	-319.31	-263.39	71.695
DA	-7014.4	711.58	-1893.2	-244.77	188.08	-15.944
COPY	768.7	-419.86	491.39	342.83	230.33	215.53
GOLD	-17.55	-5223.3	132.05	-373.96	-195.16	35.149
MSTR	14744	868.61	66.772	-209.27	152.27	-87.953
VERA	-8217.3	-290.29	1423.1	120.64	144.65	-158.45

3.2.4 PCA evaluation with comparison ratios of elemental pairs

ECCO has special elemental pairs that enable comparisons between samples for various types: $\frac{Mg\ (279.55)}{Ca\ (317.93)}$, $\frac{Sr\ (407.77)}{Ca\ (445.59)}$, $\frac{Sr\ (460.73)}{Ca\ (445.59)}$, $\frac{Na\ (589)}{Ca\ (558.88)}$, $\frac{Al\ (309.28)}{Ca\ (317.93)}$ ve $\frac{Ca\ (442.54)}{Fe\ (438.35)}$. When PCA analysis was performed with these ratio values calculated separately for each brand sample, a variation of 97.313% was reached. It was observed that this ratio was the highest variation value obtained in the entire study and approached perfect discrimination power (100%). Summary information (Table 10), PCA Score values (Table 11) and graph (Figure 11) of the PCA analysis were found as follows.

Table 10. Summary of information from PCA applied to comparison ratio data of elemental pairs.

PC	EIGENVALUE	% VARIANCE
1	1.63711	**97.313**
2	0.0261581	1.5549
3	0.0131203	0.7799
4	0.00591653	0.35169
5	5.11513E-06	0.00030405
6	1.39654E-06	8.3013E-05

Table 11. PCA score values from analysis of comparison ratio data for elemental pairs.

PAPER SAMPLE	PC 1	PC 2	PC 3	PC 4	PC 5	PC 6
MOPAK	-1.2581	0.0062074	-0.17738	-0.060869	0.0019825	-0.00073031
BOND	-0.095863	-0.19284	0.024164	-0.050817	-0.0038044	-0.00075217
DA	-1.0418	-0.079747	0.1967	-0.010305	0.0025362	0.00013099
COPY	0.52948	-0.038841	-0.054121	-0.022703	-0.00021048	0.0025332
GOLD	-0.39826	0.33113	0.051693	-0.010797	-0.0017904	-7.6368E-05
MSTR	2.5714	0.017626	0.0072034	-0.012767	0.001586	-0.00089126
VERA	-0.30687	-0.043535	-0.048263	0.16826	-0.00029942	-0.00021406

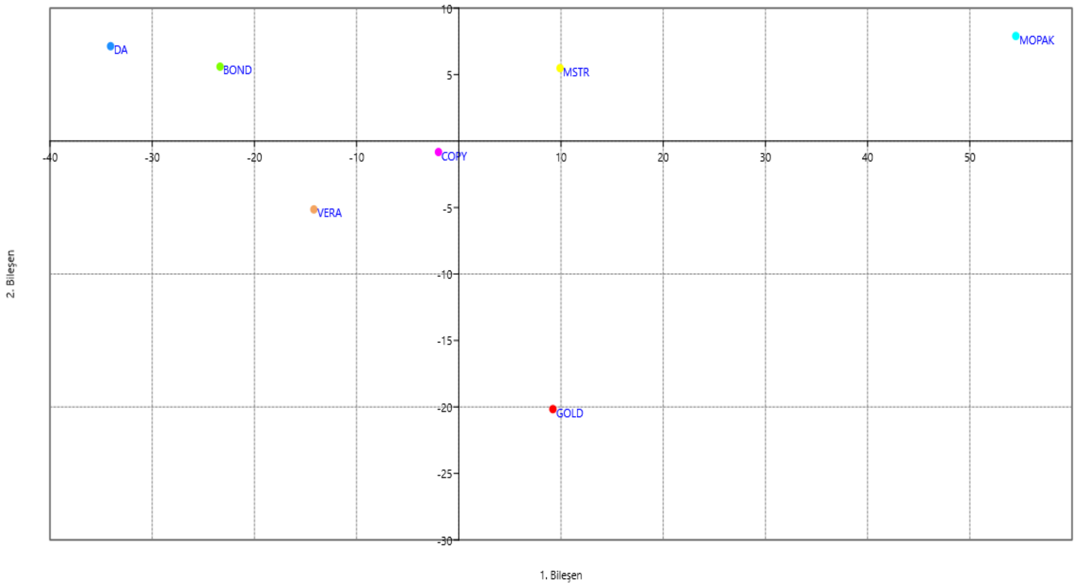

Figure 10. PCA score graph for matching (% match) rates of selected elements (K, Sr, Si, Na, Ba, Mg, Ca, C) excluding Al, Fe, and Ti.

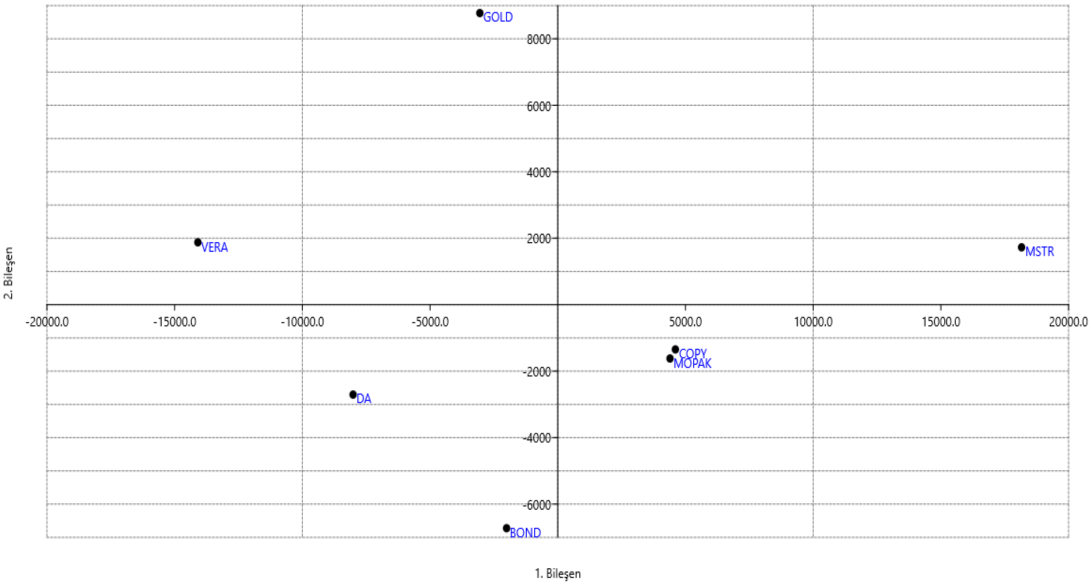

Figure 11. PCA score graph from PCA analysis of peak height values for all peaks.

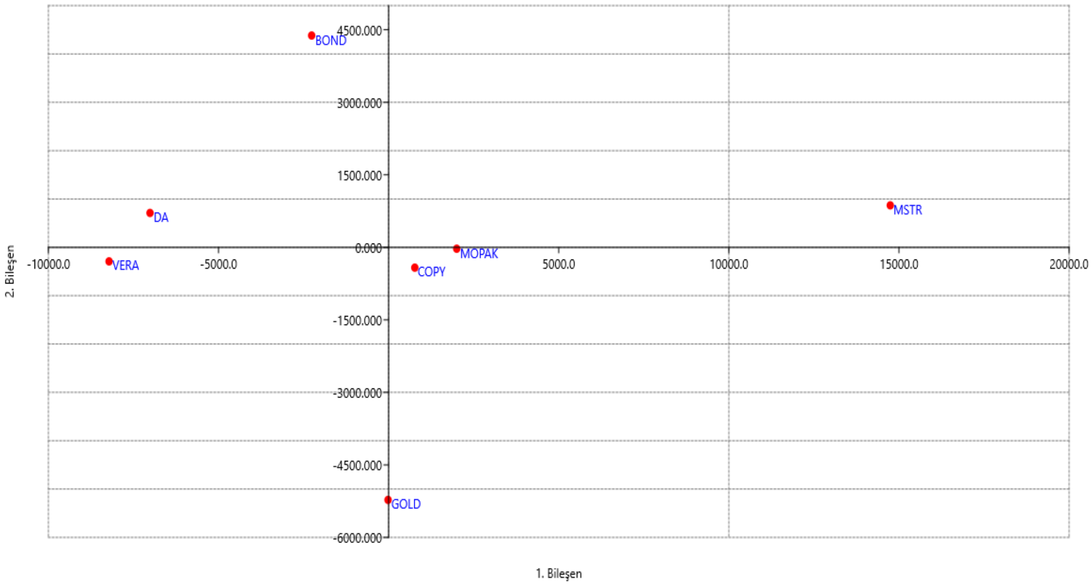

Figure 12. PCA score graph from PCA analysis of peak area values for all peaks.

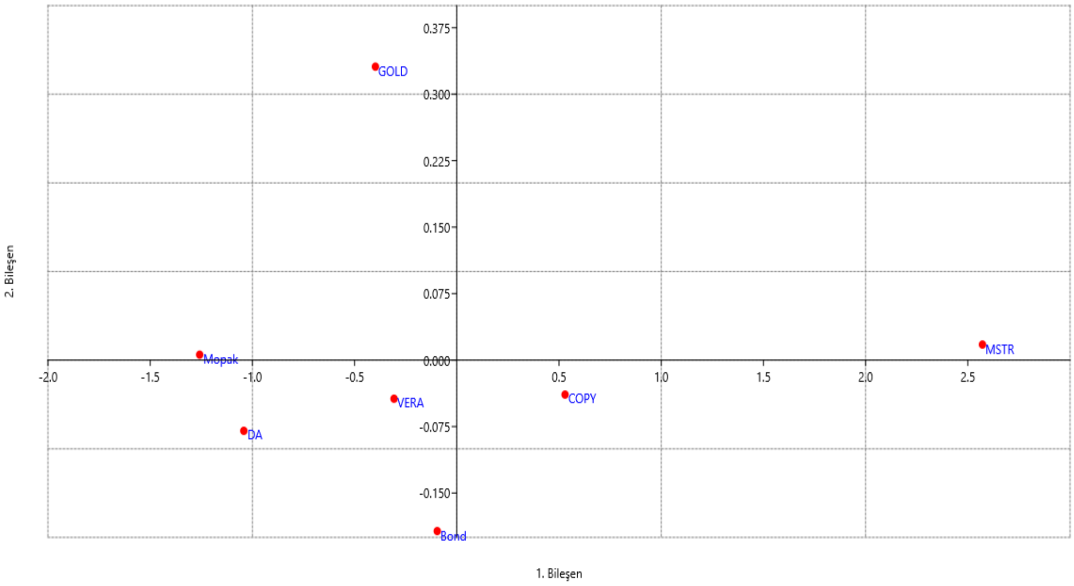

Figure 13. PCA score graph from PCA analysis of comparison ratio data for elemental pairs.

Since it is the first study conducted at a scientific level in our country to show that documents can be distinguished using the LIBS method in the field of forensic document examination, it is anticipated that it will provide a pioneer for other studies to be conducted in this field. It is thought that it can provide an important opinion to all officials serving in the establishment of justice, especially forensic document examination experts and assistants.

4. Conclusion

The ECCO®² LIBS system used in this study has proven to be a highly sensitive and rapid technique for multi-element analysis, making it an attractive method for forensic document examination. This study supports the adoption of LIBS in forensic laboratories for paper analysis due to its unique advantages: simple, fast, multi-element detection with minimal sample destruction and without the need for preparation, along with lower maintenance and operational costs compared to traditional techniques. Although LIBS is not yet routine for forensic profiling of documents, wider adoption could expedite case resolutions involving questioned documents and reduce forensic experts' workload.

This research not only demonstrates LIBS's versatility and effectiveness for analyzing paper but also opens new opportunities for forensic investigations and quality control in the paper industry. The ability to discriminate between paper brands based on their elemental composition has significant implications for forensic document examination, recycling processes, and paper manufacturing, enhancing product consistency and quality.

Further understanding of paper's elemental composition could also support sustainable manufacturing practices and recycling methods. By identifying elemental makeup, manufacturers can optimize raw materials and improve recycling efficiency, contributing to environmental sustainability.

As we move forward, the integration of advanced spectroscopy techniques like LIBS with sophisticated data analysis methods such as PCA will undoubtedly revolutionize our approach to material characterization. This study not only highlights the potential of such technologies in forensic and analytical sciences but also sets the stage for future innovations in material science. The promising results pave the way for further research and development, refining these techniques and exploring applications in other fields of forensic science, materials science, and environmental studies. As technology advances, the integration of machine learning and artificial intelligence with spectroscopic and statistical methods could offer even more sophisticated analysis capabilities, opening new avenues for investigation and innovation.

These advancements in paper analysis reflect a broader trend in forensic and analytical science towards more accurate, efficient, and versatile methodologies. As we continue to explore the potential of these techniques, the future of material analysis appears promising, with significant implications for science, industry, and environmental sustainability.

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