Comparison of Elemental Analysis for Different Kind of Papers by Using Energy Dispersive X-Ray Spectrometer

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In this study, comparative analyses of different 27 brand papers widely used in Turkey as ashes of paper in the form of pellets. The white paper and photocopied paper have been analyzed by using the energy dispersive X-ray spectrometer system. The analyses indicated the elemental concentration values for specimens in parts per million. Besides, Pearson correlation relations were calculated with statistical package for the social sciences. When the obtained values in this study are examined carefully, it is evident that the changing content of paper is considerable. The aim of this study is to compare the original paper with photocopied paper and ashes of paper in terms of elemental concentration. It is believed that the obtained results in this study will contribute to the data base on the paper elemental characterization.

Keywords: EDXRF, Paper analysis, Elemental analysis.

INTRODUCTION

Since the beginning of history, our ancestors have made use of different types of materials, including stone, ivory, metals, textiles, *etc* to draw signs and share information. Although many materials have been employed during the history of human kind, the most commonly used graphic supplies have been papyrus, parchment and paper [1]. When one speaks about paper, the first thing that comes to mind is that it is for carrying information, not for wrapping and packaging [2]. Nowadays, paper is the most common information carrier as it used to be. Pulp is diluted to at least 99 % with water and a mineral filler and water soluble substances such as optical brighteners and polyvinyl alcohol are added in the production of paper. Silicates, sulphates, carbonates, oxides and sulphides are used as mineral fillers depending on the paper application [3,4].

Rozic *et al.* [2] have made elemental analyses for ashes of office paper by using EDXRF spectrometry. Artificial aging processes in modern paper have been researched with X-ray spectrometry technique by Manso *et al.* [5]. Lartigue *et al.* [6] have analyzed some of cigarette paper with the help of EDXRF. The elemental characterization from papyrus to paper has been investigated with X-ray fluorescence spectrometry by Manso *et al.* [7]. Manso *et al.* [8] have compared the elemental content between modern and ancient paper by EDXRF. The characterization of paper which belongs to 18th and 19th century

documents has been examined using X-ray fluorescence spectrometry by Manso *et al.* [9] and Van Es *et al.* [10] have surveyed the discrimination of document paper by XRF, LA-ICP-MS and IRMS using multivariate statistical technique.

The raw material of paper is cellulose since it is made from trees. However, various elements are added by the manufacturer. The most important stuff in all of the additives is the polyelectrolytes which make better the fineness and filler retention, provide the drainage of the paper suspension more efficient and at the same time add specific qualifications [11]. For the improvement of surface properties of paper, picture colour and quality, polymers soluble in water predominantly starch or soluble cellulose products are added in paper mass in a separate process, or paper mass is coated with these substances. The pigment in coating layer could be clay platelets, calcium carbonate, titanium dioxide, calcium sulphate, talc, or synthetic pigments [4]. Thus, the paper has become a doped. The main problem starts at this stage. There is no standard for paper additives used in the production process. In addition to this, paper manufacturers keep the proportions of additives used as a secret. This situation makes the paper manufacturing process an enigma for the users. The lack of standardization in papermaking has led us to perform this study. This present study will contribute to the database on paper's elemental characterization and provide efficient paper identification.

The office paper was analyzed by using energy dispersive X-ray fluorescence technique (EDXRF) in this study. The main advantage of this technique is that it is non-destructive, which makes it a convenient tool for elemental analysis of paper. The application of this technique preserves the integrity of the paper and allows the characterization of each type of paper [12,13]. The aim of this study is to compare the white paper, photocopied paper and ashes of paper in terms of elemental concentration.

EXPERIMENTAL

Twenty seven different brands of paper weighing 80 g/m² and paper size A4 210 × 297 mm in each three groups as original, photocopied paper and ashes of paper, were collected and then analyzed with the energy dispersive X-ray fluorescence (EDXRF) system.

Sampling: One A4 sheet of each paper sample was used for all the analytical procedures. Twenty-seven different brands of white paper did not undergo any processing. Photocopy paper was obtained by pulling double-sided copies of the original text. Samples were cut diagonally from top left to bottom right for white paper as shown in Fig. 1 and for photocopied paper specimens were obtained by cutting in the way as shown in Fig. 2. Each circle has a diameter of 37 mm. Due to the heterogeneity of the paper and in order to evaluate the variation in composition within a sheet, three areas of the paper were sampled. Ashes of paper are prepared as the paper was combusted on the glass and finally all of the samples were shaped as pellets as shown in Fig. 3.

Fig. 1. Locations of white paper samples were cut

X-ray fluorescence instrumentation and method: An EDXRF spectrometer (Epsilon5, PANalytical, Almelo, the Netherlands) was used to determine the metal content of paper sample. This system is a three-dimensional geometry and optical polarized anode X-ray tube 600 Gd, 100 kV generator for up to 15 high resolution of secondary and polarized target consists of PAN-32 detector. The XY axis is able to receive up

Fig. 2. Locations of photocopied paper samples were cut

Fig. 3. Pellet forms of ashes of paper

to 133 samples with sample changer, measurements can be carried out for solid and liquid samples in a vacuum and helium environment. Secondary sources of EDXRF system used in this study are Al₂O₃, CeO₂, Ag, Mo, Zr, Ge, Fe, Ti and Al in pellet form as same thicknesses at the different densities. The thickness and densities of pellets of these secondary sources are shown in Table-1. Each sample was analyzed by EDXRF system; the averages of the three measured values of the same paper were calculated.

Statistically evaluation of data: The statistical data evaluation was done with statistical package for social sciences (SPSS) program and Pearson correlation was used to examine the properties of the correlations among the elements.

TABLE-1
SECONDARY SOURCES OF PRESENT SYSTEM
USED IN PELLET FORM AS SAME THICKNESS
AT DIFFERENT DENSITIES

Secondary source	Thickness (μm)	Density (g/cm^3)
Al_2O_3	600	3.94
CeO_2	600	6.84
Ag	600	10.5
Mo	600	10.2
Zr	600	6.49
Ge	600	5.35
Fe	600	7.86
Ti	600	4.52
Al	600	2.70

RESULTS AND DISCUSSION

The elemental contents of Mg, Al, Si, S, Cl, K, Sc, Ti, Mn and Fe are illustrated in Table-2, Tables 3 and 4 for white, photocopied and ashes of paper, respectively. When looking at these tables, it is seen that the highest concentrations of elements in the white, photocopied and ashes of paper are Al, Si, S; Mg, Al, Si; Si, Fe, Al; respectively. As, Br, Zr, I, Pr, Yb, Hf, Au and Pu are not illustrated in any table due to very low elemental concentrations. These elements are thought to originate from the measurement system and the sampling process. Besides, each of the spectra for white, photocopied and ashes of paper is plotted in Figs. 4-6. If the correlation level is positive, the concentration values of the two elements are directly proportional. When the correlation level is zero, there is no relationship between the concentration values of the two elements. If there is negative correlation level among

Fig. 4. EDXRF spectra of white paper

any two elements, the concentration values of the elements are inversely proportional. The correlation levels and sensitivities for each set of paper are shown in Tables 5-7.

Table-5 showed that there is a high positive linear relationship between S-Al, Ca-Al, Fe-Mn and Pb-Al and it is seen that there is a very high positive linear relationship between Zn and Fe for white papers. If considering Table-6, a high linear correlation was detected between Fe-Mn and Zn-Mn. A statistically significant high positive linear relationship was determined between Zn-Fe for photocopied paper. When considered Table-7, there were no data about correlational relationship at very high level for ashes of paper.

TABLE-2
ELEMENTAL CONCENTRATIONS OF TWENTY SEVEN WHITE PAPERS IN PARTS PER MILLION

	Al	Si	S	Cl	K	Ca	Ti	Mn	Fe	Co	Ni	Cu	Zn	Sr
W1	7120	—	764	495	—	201943	—	52	166	—	70	26	16	21
W2	7547	—	999	1021	593	183350	—	10	95	—	53	26	21	—
W3	10597	2730	1003	1253	1135	204233	—	12	103	—	42	23	18	11
W4	59737	2970	3376	2273	707	403303	—	61	191	—	—	40	14	28
W5	47010	—	4228	1169	543	340483	—	8	96	—	—	32	23	—
W6	7977	—	946	922	623	175513	—	11	130	—	60	25	23	—
W7	11580	4160	1883	1699	—	197180	56	18	331	—	—	26	17	20
W8	9073	2640	1181	2630	—	163020	—	17	175	—	38	22	27	104
W9	10323	6603	849	691	—	201233	167	27	283	—	159	22	25	36
W10	8980	5540	869	1143	519	153450	238	95	8850	18	—	30	60	48
W11	13250	3550	2610	4410	—	201233	79	21	321	—	—	20	19	19
W12	41540	30190	5565	2025	1824	153647	—	17	153	—	—	33	30	132
W13	12753	3870	2138	5783	—	189893	74	10	348	—	—	26	22	19
W14	9377	2540	1118	519	573	200883	—	10	93	—	—	25	22	14
W15	8307	3580	1693	1412	—	183533	—	11	168	—	—	24	15	12
W16	8683	—	976	419	—	192873	—	8	99	—	97	26	24	—
W17	7260	—	1013	375	—	167960	—	10	99	—	—	69	17	9
W18	10740	3693	1324	1334	—	209487	—	15	173	—	97	28	23	19
W19	10133	4197	1269	924	—	196590	—	18	205	—	—	27	18	17
W20	9560	—	1342	1227	—	193213	—	8	76	—	—	26	23	—
W21	7640	2785	1969	613	516	166793	—	13	98	—	143	27	17	96
W22	12610	3707	1919	4087	701	199747	88	15	340	—	—	22	17	24
W23	11090	—	1399	1114	—	209017	—	8	77	—	77	24	19	—
W24	8810	3645	1710	1161	—	183583	—	16	117	—	54	25	23	161
W25	11223	3573	2247	5693	—	174640	—	8	126	—	—	27	18	—
W26	8660	3055	1635	834	—	180173	—	13	183	—	24	23	24	134
W27	5837	2140	1009	1614	424	126717	—	27	119	—	—	33	20	31

TABLE-3
ELEMENTAL CONCENTRATIONS OF TWENTY SEVEN PHOTOCOPIED PAPERS IN PARTS PER MILLION

	Al	Si	S	Cl	K	Ca	Ti	Mn	Fe	Co	Ni	Cu	Zn	Sr
P1	7507	1291	904	754	698	165257	175	187	18837	38	–	22	86	23
P2	8300	2800	1035	1005	738	18	170	161	19467	27	–	22	92	16
P3	11600	3250	1046	723	–	192780	185	162	19817	27	–	21	88	16
P4	9163	3185	911	734	564	183470	178	204	23093	31	–	24	100	23
P5	9093	2830	1228	541	–	16440	166	152	19253	23	–	24	92	15
P6	10320	2700	887	533	–	182343	162	153	18183	25	–	18	87	16
P7	10623	4813	1729	1548	–	183983	159	142	16920	25	40	23	79	25
P8	7010	3345	1092	2405	–	153290	163	162	19015	25	–	17	90	123
P9	12077	6780	825	793	542	176523	273	160	17507	21	–	22	82	40
P10	11220	6520	875	897	522	183067	302	216	26277	54	–	34	136	63
P11	9733	3750	2413	4407	–	194930	182	151	17470	25	72	26	88	24
P12	8153	3403	1859	501	–	160433	161	153	18567	26	122	19	86	107
P13	13493	4467	2387	4073	–	205073	167	137	16883	20	–	21	82	23
P14	9603	2820	998	571	–	192520	178	154	18887	26	–	26	90	18
P15	10033	3767	1618	1751	–	170803	171	150	18440	26	–	24	85	17
P16	9583	2910	903	485	–	175070	145	142	17497	24	–	26	87	18
P17	6770	2525	1382	662	–	136427	213	170	21277	–	151	24	101	20
P18	10310	3907	1290	1181	–	197657	187	160	19177	28	–	25	92	22
P19	10837	4957	1164	965	–	177000	173	165	19460	27	–	24	90	20
P20	10027	3223	1211	1349	–	172277	147	136	17177	25	–	25	83	15
P21	7953	3585	2080	660	577	159540	175	169	20193	–	–	27	101	112
P22	13177	4410	1862	3773	638	192483	216	162	19533	28	–	24	95	31
P23	9227	3633	2273	589	–	182463	160	160	18517	29	–	23	87	195
P24	11043	–	1294	1288	678	19490	153	152	18723	27	–	23	88	14
P25	11060	3880	2433	5737	–	160727	166	155	19230	32	–	30	92	24
P26	8590	3320	1570	973	–	161590	182	157	19537	32	–	24	95	140
P27	6027	2503	993	1705	491	114593	173	170	18320	–	–	36	95	37

TABLE-4
ELEMENTAL CONCENTRATIONS OF TWENTY SEVEN ASHES OF PAPERS IN PARTS PER MILLION

	Mg	Al	Si	S	Cl	Ca	Ti	Mn	Fe	Cu	Zn	Sr
A1	11417	31017	–	1286	–	421207	–	107	339	18	–	140
A2	14737	32567	9583	–	–	411310	–	97	323	22	–	138
A3	9103	35410	8245	–	–	432753	–	27	191	17	–	69
A4	7493	29353	7760	1642	–	440337	–	24	219	17	10	71
A5	10243	36530	9820	2350	–	413957	–	–	160	20	–	55
A6	9683	39967	–	–	–	406557	–	23	165	21	–	63
A7	9990	35971	17560	3150	1760	407930	–	35	805	21	13	168
A8	10660	36200	16203	2627	3163	420853	–	41	392	13	11	1314
A9	8943	37990	22443	1535	720	392663	252	54	700	19	14	334
A10	7917	18513	7870	1299	964	237987	–	29	216	32	–	120
A11	7960	31513	10403	3417	4560	387723	–	32	651	21	–	117
A12	8353	30710	12483	3193	801	402113	–	27	336	11	–	1056
A13	8893	34527	13533	2850	6210	377397	64	18	767	20	–	111
A14	6360	28380	–	–	–	404713	–	19	304	19	–	64
A15	8910	34930	14210	3470	1352	406250	–	24	413	22	13	87
A16	8830	34253	8565	–	–	426420	–	19	218	30	9	89
A17	13997	33543	9775	1947	–	42490	–	19	238	17	–	80
A18	9593	33287	13847	2240	1149	439923	–	38	434	20	12	118
A19	11657	40477	18510	2623	1057	427400	–	39	525	20	14	102
A20	7510	32950	–	2155	956	418013	–	19	193	14	–	69
A21	12673	37990	15943	4223	1003	434127	–	34	276	14	13	1124
A22	8317	36587	13260	3350	5043	406887	85	30	861	18	21	194
A23	6030	33017	–	1952	675	438593	–	–	154	13	9	62
A24	8060	31860	15187	3180	1494	418207	–	36	354	12	11	1864
A25	5753	35520	14130	3653	7567	399420	–	17	381	23	9	100
A26	10017	32580	15093	2677	1067	429527	–	24	410	10	9	1283
A27	15853	35430	16013	2837	1747	394563	–	92	376	32	15	332

TABLE-5
CORRELATION OF EACH ELEMENT FOR WHITE PAPER

Pearson correlation	Al	Si	S	Cl	Ca	Mn	Fe	Cu	Zn	Sr	Hf	Pb
Al	1											
Si	0.378	1										
S	0.821	0.656	1									
	-	-										
Cl	0.121	0.143	0.327	1								
	0.547	0.478	0.096									
Ca	0.790	-0.189	0.414	0.012	1							
	-	0.345	0.032	0.952								
Mn	0.208	0.085	-0.063	-0.089	0.168	1						
	0.297	0.673	0.753	0.661	0.402							
Fe	-0.075	0.080	-0.151	-0.050	-0.162	0.770	1					
	0.711	0.691	0.452	0.806	0.418	-						
Cu	0.255	0.016	0.122	-0.188	0.134	0.108	0.025	1				
	0.199	0.937	0.543	0.347	0.504	0.590	0.903					
Zn	-0.044	0.273	-0.046	-0.115	-0.265	0.592	0.892	-0.049	1			
	0.827	0.168	0.821	0.569	0.181	0.001	-	0.808				
Sr	0.064	0.492	0.274	-0.079	-0.243	0.084	0.053	-0.102	0.259	1		
	0.751	0.009	0.166	0.696	0.223	0.679	0.792	0.611	0.191			
Hf	0.294	0.452	0.500	-0.012	-0.105	-0.262	-0.178	0.263	0.025	0.269	1	
	0.137	0.018	0.008	0.954	0.602	0.187	0.373	0.185	0.902	0.175		
Pb	0.718	0.450	0.684	-0.035	0.420	-0.085	-0.215	0.232	0.061	0.210	0.533	1
	-	0.019	-	0.862	0.029	0.672	0.282	0.245	0.761	0.293	0.004	

TABLE-6
CORRELATION OF EACH ELEMENT FOR THE PHOTOCOPIED PAPER

Pearson correlation	Al	Si	S	Cl	Ca	Ti	Mn	Fe	Co	Cu	Zn	Sr	Hf	Pb
Al	1													
	-													
Si	0.474	1												
	0.013													
S	0.212	0.125	1											
	0.288	0.535												
Cl	0.372	0.171	0.650	1										
	0.056	0.395	-											
Ca	0.318	0.499	0.205	0.184	1									
	0.105	0.008	0.305	0.359										
Ti	0.265	0.646	-0.23	-0.05	0.199	1								
	0.181	-	0.245	0.793	0.319									
Mn	-0.215	0.150	-0.369	-0.24	0.056	0.594	1							
	0.281	0.454	0.058	0.222	0.782	0.001								
Fe	-0.090	0.189	-0.277	-0.22	-0.023	0.596	0.879	1						
	0.657	0.344	0.161	0.269	0.911	0.001	-							
Co	0.390	0.226	-0.147	0.046	0.150	0.255	0.319	0.347	1					
	0.044	0.256	0.464	0.818	0.454	0.199	0.105	0.076						
Cu	-0.085	0.178	0.019	0.173	-0.004	0.307	0.338	0.357	-0.06	1				
	0.674	0.374	0.926	0.388	0.983	0.119	0.085	0.067	0.787					
Zn	-0.104	0.253	-0.187	-0.11	-0.023	0.642	0.782	0.921	0.265	0.561	1			
	0.604	0.203	0.349	0.579	0.908	-	-	0.182	0.002					
Sr	-0.313	0.126	0.329	-0.15	0.139	0.006	0.116	0.097	0.020	-0.113	0.141	1		
	0.111	0.533	0.094	0.455	0.490	0.976	0.566	0.631	0.919	0.574	0.483			
Hf	-0.206	-0.130	0.371	0.354	-0.014	-0.26	-0.259	-0.216	-0.33	0.616	-0.02	0.146	1	
	0.302	0.517	0.057	0.070	0.945	0.200	0.192	0.279	0.089	0.001	0.910	0.469		
Pb	-0.027	0.035	0.439	0.447	0.137	0.003	-0.160	-0.182	-0.11	0.258	-0.04	-0.110	0.286	1
	0.893	0.861	0.022	0.020	0.494	0.987	0.426	0.363	0.583	0.195	0.846	0.583	0.147	

TABLE-7
CORRELATION OF EACH ELEMENT FOR THE ASHES OF PAPER

Pearson correlation	Mg	Al	Si	S	Cl	Ca	Sc	Mn	Fe	Cu	Zn	Br	Sr	I	Yb	Hf
Mg	1															
Al	0.296	1														
Si	0.134		1													
S	0.285	0.359		1												
	0.150	0.066														
Cl	-0.015	0.200	0.570		1											
	0.942	0.318	0.002													
Ca	-0.280	0.135	0.345	0.579		1										
	0.157	0.502	0.078	0.002												
Sc	-0.258	0.304	0.025	0.044	0.050		1									
	0.194	0.123	0.900	0.827	0.805											
Mn	-0.276	0.120	0.107	0.160	0.331	0.319		1								
	0.164	0.550	0.597	0.424	0.092	0.105										
Fe	0.633	-0.009	0.122	-0.135	-0.13	0.099	0.033		1							
	-	0.966	0.546	0.501	0.498	0.624	0.872									
Cu	-0.016	0.259	0.591	0.452	0.597	0.090	0.197	0.154		1						
	0.938	0.192	0.001	0.018	0.001	0.654	0.324	0.443								
Zn	0.177	-0.205	0.018	-0.253	0.073	-0.18	-0.11	0.227	0.023		1					
	0.377	0.305	0.930	0.204	0.719	0.367	0.561	0.255	0.908							
Br	0.058	0.407	0.633	0.484	0.235	0.310	0.302	0.070	0.446	-0.01		1				
	0.774	0.035	-	0.011	0.238	0.116	0.125	0.728	0.020	0.980						
Sr	0.068	0.194	0.192	0.294	0.258	0.050	0.202	0.111	0.382	0.260	0.311		1			
	0.735	0.332	0.338	0.137	0.194	0.804	0.312	0.583	0.049	0.190	0.114					
I	0.085	0.018	0.399	0.402	0.026	0.161	-0.12	0.062	0.008	-0.54	0.264	-0.07		1		
	0.673	0.929	0.039	0.038	0.898	0.421	0.554	0.759	0.970	0.004	0.183	0.731				
Yb	-0.329	-0.085	0.001	-0.197	0.221	0.144	-0.09	-0.11	0.033	-0.23	-0.14	-0.34	0.243		1	
	0.094	0.674	0.995	0.326	0.267	0.475	0.627	0.578	0.870	0.259	0.479	0.085	0.222			
Hf	-0.128	-0.102	-0.08	0.078	0.316	0.083	-0.08	0.259	0.317	0.073	-0.04	0.320	0.118	0.299		1
	0.526	0.613	0.710	0.701	0.108	0.681	0.692	0.192	0.108	0.717	0.830	0.104	0.558	0.130		
	0.032	-0.306	-0.13	-0.28	-0.09	-0.54	-0.45	-0.28	-0.17	0.504	-0.25	-0.09	-0.22	0.076	0.022	1
	0.873	0.120	0.518	0.150	0.673	0.004	0.018	0.164	0.395	0.007	0.209	0.664	0.264	0.707	0.914

Fig. 5. EDXRF spectra of photocopied paper

Fig. 6. EDXRF spectra of ashes of samples

Due to the lack of production standard of paper, many chemical elements, doped with different effects to the final properties of the produced paper, shall be added during its manufacturing. As the manufacturing process of paper is strictly protected by each manufacturer, it is difficult to say where certain elements come from but the researchers can deal with this problem as the identification of the content elements should be analyzed.

Unlike white papers, manganese and titanium were detected on photocopied papers additionally, magnesium, scandium and strontium were determined on ashes of papers. It is thought that the contents of photocopied paper differ from the content of white paper because of the photocopying remnant. The content in ash samples is believed to be caused by the burning of papers.

The aim of this study is to compare the white paper, photocopied paper and ashes of paper in terms of elemental

concentration. Besides, it is believed that the results obtained in this study will contribute to the data base on the paper elemental characterization.

Energy dispersive X-ray fluorescence analytical tool seems to be a good elemental analysis technique for analyzing the different types of the paper due to the advantage of being non-destructive.

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