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Research Article

Determination of Solvents in Ballpoint Pen Ink by Gas Chromatograhy Mass Spectrometry (GC-MS)

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Abstract

Forensic Document examination is a specialized area that focuses on examining the alterations that have been made to legal documents, as well as identifying their contents. The primary inquiry that forensic document experts are called upon to answer by the legal system is the date when a document was created. To determine this, specialists in forensic document examination have conducted research on how dyes, resins, and solvents present in ink change over time. They have attempted to create several techniques and protocols for identifying these changes. One of these approaches is solvent analysis, which examines how ink ages over time. However, the aging process of ink is affected by various factors such as temperature, light, and humidity, which makes it quite challenging to determine the exact date of a document. Recent research indicates that phenoxyethanol is useful in determining the age of ink since it undergoes time-dependent changes. The purpose of study is to develop a single method that analyze phenoxyethanol (PE), phenoxyetoxyethanol (PEE), Proplylene Glycol (PG) and etoxyethanol (EE) with Gas Chromatography Mass Spectrometry (GC-MS).In this study, a method that allows analysis of solvents in blue ballpoint ink that are phenoxyethanol (PE), phenoxyetoxyethanol (PEE), Proplylene Glycol (PG) and etoxyethanol (EE) with Gas Chromatography Mass Spectrometry (GC-MS) for use of forensic science laboratories has been develop and validated. Also age curve of phenoxyethanol (PE) was drawn up to 280 days and age curve of phenoxyetoxyethanol (PEE) was drawn up to 230 days by using developed method. Further studies in ink aging analysis can be extended by adding different color of ballpoint pens, different types of pens such as gel pens or analysing different solvents in ink composition. Thus, justice will be served by clarifying more document forgery cases.

Keywords: Forensic science; GC-MS; Ink aging; Questioned document; Solvents

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Introduction

One of the most frequently committed offences is document forgery. This type of crime is the subject of forensic document examination and is detected by forensic document experts. The use of methods to determine the time of writing of suspicious writings that are subject of the case in service of judicial system system not only ensures that justice is served, but also reduces the workload of the judiciary. In order to clarify this type of crime, the judicial system asks two main questions to the experts. The first one is who created the document and the second one is when it was created. While there are many optic techniques for answer to first question, the number of both analytical methods and experts for answer to second question is very low. The studies on age determination have been carried out since 1903 [1,2]. The basis of age determination studies is to analyse and interpret the time-dependent changes of the substances in the ink structure by analytical techniques. These changes are degradation of dyes or pigments, polymerisation of resins and evaporation of solvents.

Inks in ballpoint pens generally consist of a mixture of dye-based colourants, resins and solvents that are insoluble in water but soluble in organic solvents or oils. According to Weyerman and Bügler, solvents are 50%, colourants and pigments 25% and resins 25% of the ink. Other components are in very small proportions, e.g. surfactants, corrosion inhibitors, thinning agents, etc. [3]. Solvents are used to lighten the colour of ink and to ease the transition from ink to paper. Oil-based solvent, originally used as a solvent for ballpoint pens, has a higher density than water and therefore has a low boiling point and vapour pressure, so it remains in the pen chamber for a long time and is used. Glycol-based solvents keep the ink in the cartridge fluid, but allow the ink to dry quickly after application to the paper. Initially olefins, castor oil or mineral oils were used as solvents. Today, chemicals such as phenoxyethanol, phenoxyethoxyethanol, propylene glycol, benzyl alcohol, butylene glycol, etc. are preferred as ink solvents. Chemical structure of these solvents are shown (Figure 1).

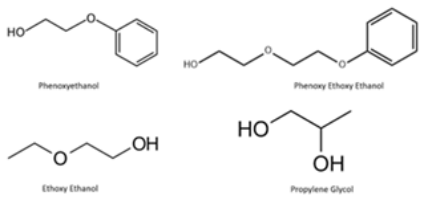

Figure 1: Chemical structure of solvents in ballpoint pen ink.

Avcı et al. found the equation in (Figure 2) which explains the change of evaporation of volatile solvents from non-volatile solvents with respect to time developed at the end of 1990s [4].

$$y(t) = m_o + m_f e^{-(t/t_f)^{1/2}} + m_s e^{-(t/t_s)^{1/2}}$$

Figure 2: The equation that was found by Avcı et al.

In his 2012 paper, Cantu showed that, based on this equation, if the non-volatile solvent in Avcı's model is replaced by the cellulose of the paper and the volatile solvent is replaced by phenoxyethanol, the solvent of the ink, the ink aging curve obtained in practice is in accordance with the mathematical equation in the model [5]. (Figure 3) shows the ink aging curve of Cantu.

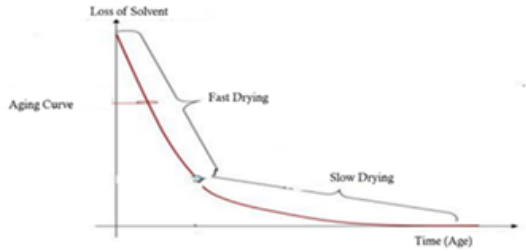

Figure 3: The ink aging curve of Cantu.

In order to determine the creation time of the document, the dyestuffs [6-11] and resins [12-15] in the ink were also analysed. Since the time difference required for dyestuff and resin analyses is greater than the time difference required for solvent analysis, it is used in the analysis of documents 4 years or older [2]. Solvent analysis is used in the chronological order of inks used on the same document at different times within the same year. Also, result of literature review, it was determined that there are more studies on the time-dependent change of phenoxyethanol [16-25]. However recent studies focuses ink aging analysis by using different analytical instruments and different componenets in ink structure such as dyes, solvents and resins [26-28].

This study aims to develops a method for analyse the detection of phenoxyethanol, phenoxyethoxyethanol, ethoxyethanol and propylene glycol solvents using GC-MS. In addition to drawing the age curve of phenoxyethanol solvent, it was aimed to draw the age curves of phenoxyethoxyethanol, ethoxyethanol and propylene glycol solvents in the structure of ballpoint pen ink to increase the discrimination power of suspicious writings in suspicious documents.

Material and Methods

Materials

Chemicals used as referece were pure phenoxyethanol (PE), ethoxyethanol (EE), Propylene Glycol (PG) purchased from Sigma-Aldrich,Germany and phenoxyethoxyethanol (PEE) purchased from TCI, Japan. 10 blue ballpoint pens of different brands and models (Table 1) were collected from the local markets. Blue ballpoint pens were applied on standart white A4 office paper from CopierBond (80 g/m^2, Türkiye). Extraction of solvents from ballpoint pens that applied on A4 paper were made with dichloromethane (DCM, Merck, Germany) containing internal standart 1,3-benzodioxol-5-methanol (IS, Sigma-Aldrich,Germany). Extraction was made in 1.5 ml glass vials (Agilent, USA).

Brand and Model of Blue Ballpoint Pens
Bic Round Stic
Faber Castell 1425
Gestetner
Mikro M-25
Office Time
Pensan Büro
Pensan My Tech
Pensan Ofis
Scrikss F108
Uni-Ball Laknock

Table 1: Brand and Model of Blue Ballpoint Pens.

Instrumentation

For better dissolution samples were vortexed VTX-3000 L from Mixer Uzusio (LMS, Japan) and kept in water bath BM-302 from Nüve (Nüve, Türkiye). Analysis of the solvents was made on a Gas Chromatography-Mass Spektrometer 7820A/5977E MSD from Agilent (Agilent Technologies, USA). Separation was carried out on a HP 5MS capillary column from Agilent (Agilent Technologies, USA). The column was 30 m long and had an internal diameter of 0.25 mm and film thickness of 0.25 µm. The chromatographic elution was temperature programmed as follows: at 40^{0}C for 2 min, then from 40^{0}C to 200^{0}C at a rate of 10 ^{0}C/min, and finally at 400^{0}C for 2 min. Totaly analysis takes 11 minutes. The carrier gas was helium with a constant flow of 1 ml/min. For the chromatographic separation, a solvent delay of 5 min. was chosen. The sample was injected in the splitless mode and the injector temperature was maintained at 250^{0}C. The MS part of the GC/MS was a highly sensitive quadrupole instrument with a mass range up to 1000 u. For qualitative analysis, the instrument was used in the selected ion monitoring (SIM) mode. 12 ions were selected and monitored, corresponding to the masses: 43, 45, 59, 65, 72, 77, 93, 94, 135, 138, 152 and 182 u.

Sample Preparation

Lines were drawn on paper with blue ballpoint pens and the help of a ruler by the same person every month to eliminate the press difference. Lines were about 0.5 mm wide, 20 cm long and 5 cm intervals (the reason for this distance is that there is no interference due to the horizontal and vertical diffusion of the solvents in the ink on the paper surface). In order to prevent solvent contamination in created documents, they were placed one by one in transparent files and filed. They were stored at a temperature of 23-25^{o}C and a humidity range of 45-55%. These values were measured with a temperature-humidity meter. To determine calibration curves, pure solvents were dissolved in dichloromethane at concentrations of 0.1, 0.2, 0.3, 0.4, 0.5 and 0.6 mg/ml with 0.05 mg/ml internal standard concentration. Then validation of the method was performed. After method validation samples of 1 mm x 5 mm, 20 rectangles were taken from aged documents at the time of first drawing and then every month. The samples were placed in 1.5 ml vials with DCM and internal standard. To increase dissolution samples were vortexed for 5 min. and kept in a water bath at 80^{o}C for an hour. Then samples analysed by GC-MS.

Results

For specificity and selectivity parameters, the internal standard was added on the blank paper sample and analyzed by GC-MS to detect whether there was interference from the paper at the retention times of the solvents. Then 0.1 mg/ml mixture solution was prepared and analyzed. The chromatogram of the blank paper is shown in (Figure 4) and the chromatogram of the mixture is shown in (Figure 5). As a result, it was determined that retention time and specific ions of solvents. It was shown in (Table 2).

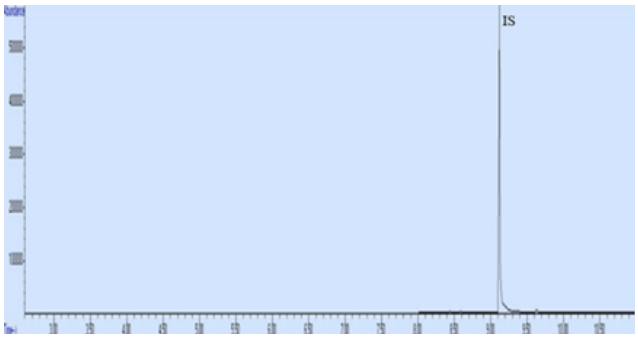

Figure 4: Chromatogram of blank paper sample containing internal standard.

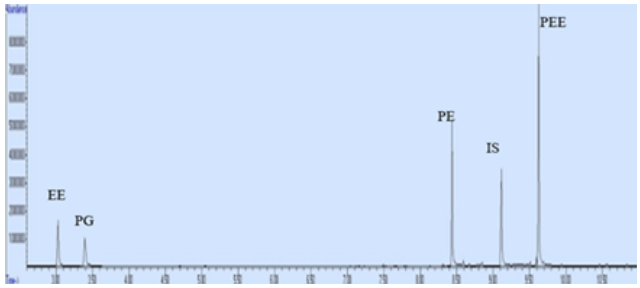

Figure 5: Chromatogram of 0.1 mg/ml mixture of solvents containing internal standard.

Analytes	EE	PG	PE	IS	PEE
Retention Time (min)	3.026	3.391	8.447	9.115	9.626
Specific Ions (m/z)	59, 72, 45, 43	45	94, 138, 77	152, 135, 93, 65	94, 45, 182, 77

Table 2: Retention times and specific ions of analytes.

For Limit Of Detection (LOD) and Limit Of Quantitation (LOQ) was obtained by preaparing phenoxyethanol, phenoxyethoxyethanol, propylene glycol and ethoxyethanol mixture solutions at the concentrations of 0.1 mg/ml, 0.05 mg/ml and 0.025 mg/ml and analyzed in GC-MS. Result of this, LOD and LOQ values were determined as 0.025 mg/ml. Result was shown in (Figure 6). Mixture solutions of 0.1, 0.3 and 0.5 mg/ml were prepared by the same analyst on the same day and given to the system in 3 consecutive repetitions. The mean peak area, Standard Deviation (SD) and Relative Standard Deviation (%RSD) values were calculated and shown in (Table 3).

Phenoxyethanol, phenoxyethoxyethanol, propylene glycol and ethoxyethanol solutions at 0.5 mg/ml, 0.3 mg/ml and 0.1 mg/ml concentrations were prepared by different analysts and analyzed 3 times using the same method and system. The method was found to be

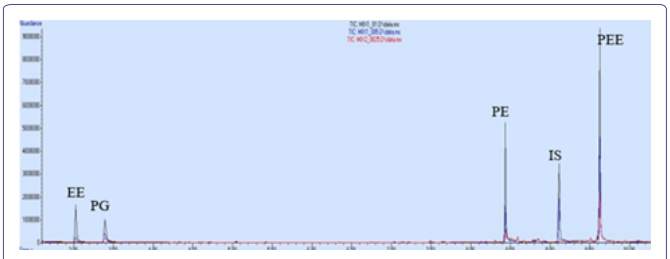

Figure 6: Superimposed chromatograms of 0.1 mg/ml, 0.05 mg/ml and 0.025 mg/ml mixture solutions.

Cons. (mg/ml)	Solvents	1st Analyze	2nd Analyze	3rd Analyze	Mean Area	SD	%RSD
0.1	EE	3281608	3154131	3265159	3233633	69340	2,144
	PG	2228935	2315968	2478337	2341080	126583	5,407
	PE	5166014	4903437	5076309	5048587	133466	2,644
	PEE	1082025	1043091	1121718	1082278	39314	3,633
0.3	EE	7037628	8069216	8251549	7786131	654602	8,407
	PG	10606901	12332362	12223089	11720784	966197	8,243
	PE	13113263	14843649	14028608	13995173	865677	6,186
	PEE	27676662	32728571	27041014	29148749	3116465	10,692
0.5	EE	13375306	12622643	10949090	12315680	1241894	10,084
	PG	22022801	20025194	18134474	20060823	1944408	9,693
	PE	20695200	18147709	19914240	19585716	1305133	6,664
	PEE	41865048	43744745	39622002	41743932	2064038	4,945

Table 3: The mean peak area, Standard Deviation (SD) and Relative Standard Deviation (%RSD) values of Analytes.

reproducible within the laboratory. The chromatogram of 0.1 mg/ml concentration prepared by two different analysts is shown in (Figure 7). F-test was applied for the analyzes made by both analysts. Since the values we obtained were less than 5.79, they were within the 95% confidence interval (α=0.05).

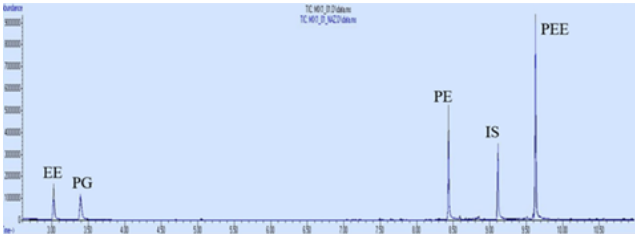

Figure 7: Superimposed chromatograms of 0.1 mg/ml mixture solutions prepared by two different analysts.

For solvent analysis from documents, 14 documents between 0-12 months stored at room temperature, 45-55% humidity and in transparent files were analysed by GC-MS. Relative Peak Area (RPA) values were calculated and RPA-time graph was drawn. RPA was calculated with the formula given below. The chromatogram of fresh ink is shown in (Figure 8).

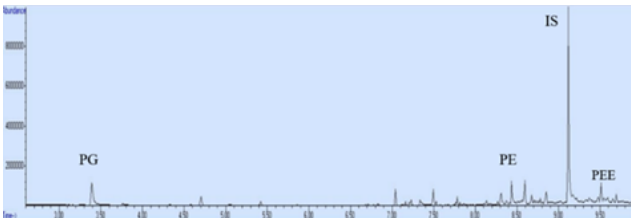

Figure 8: The chromatogram of newly created document.

$$RPA = \frac{\text{Peak Area of Analyte}}{\text{Peak Area of Internal Standard}}$$

The chromatogram of newly created document (t=0 day) is shown in (Figure 9). The chromatogram of 230 days old and 280 days old document are shown in (Figure 9&10) The time dependent changes of the peak areas of PE obtained from GC-MS analysis and RPA values are given in (Table 4). RPA-Time graph was drawn with the obtained data. RPA-Time graph of PE is shown in (Figure 11).

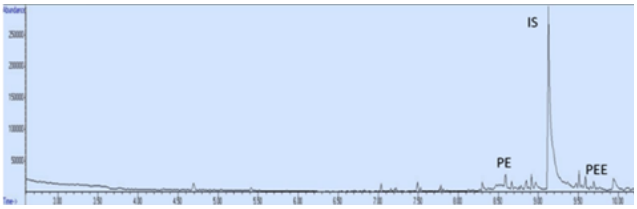

Figure 9: The chromatogram of 230 days old document.

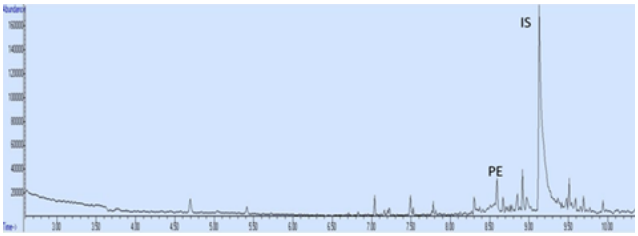

Figure 10: The chromatogram of 280 days old document.

Time (Days)	Area of PE	Area of IS	RPA
0	248842	1075977	0,231
30	121088	711392	0,170
60	102088	860394	0,119
80	209224	2320073	0,090
120	50258	838066	0,060
140	25964	528166	0,049
160	37568	841097	0,045
180	38554	998926	0,039
210	24362	705966	0,035
230	23077	688084	0,034
240	23925	765768	0,031
260	21324	758664	0,028
270	22723	821961	0,028
280	148114	5432583	0,027

Table 4: The time dependent changes of the peak areas of PE (Mean PE values of 10 blue ballpoint pens).

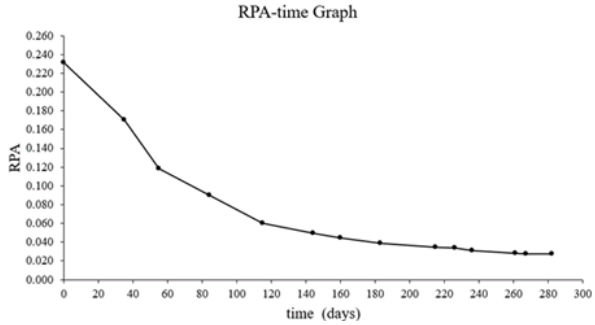

Figure 11: RPA-Time graph of PEE.

Also time dependent changes of peak areas of PEE and RPA values are given in (Table-5). RPA-time graph was drawn with the obtained data. RPA-Time graph of PEE is shown in (Figure 12). RPA value was calculated by the same formula given above.

Time (Days)	Area of PEE	Area of IS	RPA
0	83819	1075977	0,0779
30	40692	711392	0,0572
60	35792	860394	0,0416
80	80043	2320073	0,0345
120	24472	838066	0,0292
140	15053	528166	0,0285
160	23635	841097	0,0281
180	27870	998926	0,0279
210	18990	705966	0,0269
230	18165	688084	0,0264

Table 5: The time dependent changes of the peak areas of PEE (Mean PEE values of 10 blue ballpoint pens).

Discussion

In this study, in addition to the phenoxyethanol solvent known to be in the structure of the blue ballpoint pen, other solvents such as ethoxyethanol, propylene glycol and phenoxyethoxyethanol, which are also known to be in the blue ballpoint pen, were analysed and ink age determination was made with the data obtained. While phenoxyethanol and phenoxyethoxyethanol were detected in all 10 ballpoint pens studied, propylene glycol was found in only one (Faber Castel 1425). However, no ethoxyethanol was found in any of them. Ethoxyethanol is known to be widely used as a solvent in gel pen inks. Since ethoxyethanol solvent is glycol-based like the solvents used in ballpoint pen inks, it was assumed that it could be used. However, it was not found in the pens we studied.

CP-Sil 8 CB capillary column is generally used in GC-MS ink analysis, but the column we used is HP 5MS column. Due to the

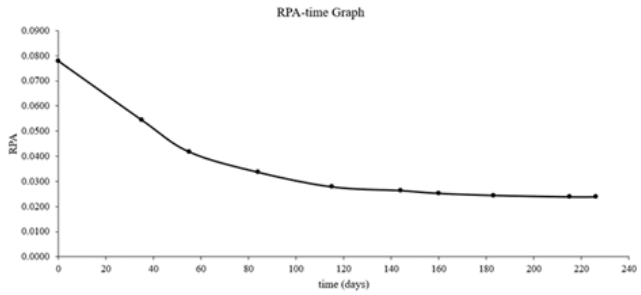

Figure 12: RPA-Time graph of PEE.

unsuitability of the column and the inaccessibility of methods such as thermal desorber or solid-phase micro extraction, no results were obtained from the analysis of a small amount of sample. Accordingly, sample size was increased and 10 cm was taken. Also the reason for not using another solvent other than DCM is that the common solvent in which EE, PG, PE and PEE can be dissolved is chloroform. Chloroform was not preferred because it is toxic and very volatile. In addition, solvents such as acetonitrile, acetone, ethanol and methanol were tried but no significant data were obtained.

The analysis of age curves for phenoxyethanol and phenoxyethoxyethanol revealed that experimental changes persisted for up to 280 and 230 days, respectively, with faster evaporation rates observed on the paper surface during the initial 60-day period that decreased over time due to environmental factors and solvent depletion.

In order to minimise the margin of error in age determination reporting, experts prefer to use time interval instead of direct time. Time interval interpretation for phenoxyethanol if RPA> 0.23, the document is new, if 0.12<RPA<0.23, the document is 0-60 days old, if 0.06< RPA< 0.12, the document is 60-120 days old, if 0.04< RPA< 0.06, the document is 120-180 days old, if 0.03< RPA< 0.04, the document is 180-280 days old and if RPA< 0.03, no time interval can be given for the document. Time interval interpretation for phenoxethoxyethanol if RPA>0.078, the document is new, if If 0.042< RPA< 0.078, the document is 0-60 days old, if 0.029<RPA<0.042, the document is 60-120 days old, if 0.028<RPA< 0.029, the document is 120-180 days old, if 0,026<RPA< 0,028, the document is 180-280 days old and if RPA< 0.026, no time interval can be given for the document.

Conclusion

In this study, it was aimed to determine the date of creation of the document by analysing the solvents of ethoxyethanol, propylene glycol, phenoxyethanol and phenoxyethoxyethanol in the structure of blue ballpoint pens of different brands and models collected from the market by GC-MS. Since phenoxyethanol and phenoxyethoxyethanol are more abundant in the structure of ballpoint pen ink in the archive samples studied and no solvent was detected in documents older than 280 days. In general, ethoxyethanol could not be detected among the other solvents other than phenoxyethanol in the ink age analysis, while propylene glycol was detected in fresh ink and phenoxyethoxyethanol could be detected up to 230 days.

The reason for investigating other solvents is to confirm the accuracy of the analysis result by having another supporting element in the analysis of the document whose creation time is unknown. In order for the ink age analysis studies to be more comprehensive, the number and types of samples (black, red and green colour) should be increased, appropriate column and method should be selected, thermal desorber, solid phase micro extraction etc. methods should be used. The effect of environmental factors (temperature, light, humidity, etc.) on the aging of the ink on the document, the effect of paper type on ink analysis, the results of the studies to be carried out to determine the ink type by elemental analysis methods can better serve the judicial system. In conclusion, the method developed in this study allows the age determination of 0-12 months in documents created with blue ballpoint pen containing phenoxyethanol and phenoxyethoxyethanol.

Another result of this study is that with the developing technology, chromatographic methods that damage the document in ink aging analysis studies have started to be switched to spectroscopic methods that do not damage the document [29-34].

Ethics approval and consent to participate

Not applicable

Consent for publication

All the authors consent to publish this research paper.

Availability of data and material

Istanbul University-Cerrahpaşa Institute of Forensic Sciences and Legal Medicine Toxicology Laboratory, Istanbul, Turkey

Competing interests

The authors declare that they have no competing interests.

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