Jan Andrasko,[1] *Ph.D.*

HPLC Analysis of Ballpoint Pen Inks Stored at Different Light Conditions

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ABSTRACT: A method for comparison of ink entries on documents stored in different light conditions is presented. Various blue inks were exposed to light, both daylight and artificial light from fluorescent tubes. Inks were then extracted from the document and analyzed by HPLC (high performance liquid chromatography). Significant changes in composition were noted on exposure to light. These changes were followed by using ternary diagrams constructed for dyes generally present in blue-colored inks—Crystal Violet, Methyl Violet, and Tetramethyl Para Rosaniline. Also, the amount of the various compounds formed by decomposition of these dyes on exposure to light was measured and employed for comparison of inks. An example of the use of the proposed method in casework is given.

KEYWORDS: forensic science, inks, HPLC, ternary diagrams, effect of light, document examination

It is a common task for forensic document examiners to investigate if two or more ink entries on one or more documents have been written with the same type of ink. Several techniques are available for such an examination, including optical techniques, thin layer chromatography (TLC), microspectrophotometry, proton induced X-ray emission (1), X-ray microanalysis (2), Fourier transform infrared spectrophotometry (FTIR), high performance liquid chromatography (HPLC) (3–6), capillary electrophoresis and related techniques (1,7,8). A historical review of several methods for writing-ink differentiation has been written by Harris (9). Roux et al. recently investigated the evidential value of ballpoint pen inks examination (10).

The most common type of ink in these investigations is ballpoint pen ink. It has been stated (11) that at least 80% of all evidence requiring ink analysis contains ballpoint pen ink. If the ink entries occur on the same document, the investigation is straightforward. A more complicated situation appears when the ink entries occur on different documents or on different sides of the same document (e.g., on the front and back). Generally, storage of these documents can be different in terms of humidity, temperature, and exposure to light. Particularly, storage with different amounts of light exposure might cause significant differences in the optical properties (color shade, IR luminescence etc.) of the inks, as well as differences in chemical composition. The knowledge of the effect of light on the composition and on the optical properties of inks is essential for correct conclusions in forensic casework. To our knowledge, only one study has been devoted to monitor changes in dye composition of ballpoint pen inks after light exposure (12).

In this study, HPLC analysis with diode array detection was employed to monitor changes in the chemical composition of ballpoint pen inks after exposure to light and under normal aging conditions. Blue ballpoint pen inks on ordinary writing paper were studied. Changes in dye composition have been evaluated quantitatively and a way to compare ink entries stored in different light conditions has been proposed.

Materials and Methods

Examined Inks

Blue colored ballpoint pen inks, manufactured by Ballograf Bic AB (Göteborg, Sweden) were examined. The manufacturer supplied the information about the code system used (year and month of production). Many different production lots from this manufacturer were analyzed in our laboratory, thus inks with various compositions suitable for this investigation could be chosen. Ink entries in the form of asterisks on ordinary writing paper (KONTORAB Copy, A4 80 g paper for photocopy machines) were examined. Each asterisk corresponded to about 1 cm ink in length.

A series of inks was exposed to daylight inside a laboratory, close to the windows, but not exposed to sunshine. Another series of inks was exposed to light from a fluorescent tube from a short distance (about 10 cm). Normally aged ink was taken from an ordinary notebook, where all the text was intentionally written by the same pen. The text was up to three years old and the notebook stored unopened and, to our knowledge, not unnecessarily exposed to light.

Extraction of Inks

Single asterisks (or a single letter from the notebook) were cut out from the writing paper and placed inside 2 mL glass vials. Approximately 0.2 mL methanol (HPLC grade) was added and the ink material extracted for 30 min at room temperature followed by heating the vial content up to boiling point for 1 to 2 min. The methanol extract was then transferred to a 200 μL conical glass inset and evaporated to dryness by a stream of nitrogen. The dry residue was dissolved in 20 μL methanol. During the whole extraction procedure, the ink and the extract were kept in darkness or protected from the exposure to intense light. Aliquots of 20 μL were taken for analysis. It means that the whole sample was taken for one analysis. Methanol has been used as extracting solvent as the analytical method used, HPLC, employs the mobile phase, which is much weaker in comparison with those used for TLC.

[1] Associate professor, SKL, National Laboratory of Forensic Science, S-58194 Linköping, Sweden.

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Pyridine and other strong solvents should remove more colorant from the ink entry, but these extra components will not be detected by HPLC analysis.

HPLC Analysis

Ink analyses were run on a Hewlett Packard series II Liquid Chromatograph connected to the HP's HPLC^{3D} ChemStation. The instrument was equipped with an auto-sampler, an auto-injector and a diode array detector from Hewlett Packard. HPLC separations were performed by using a 25 cm 5 μm TSKgel ODS-120T (4.6 mm ID, Tosohaas Bioseparation Specialists) stainless steel column. The mobile phase consisted of two solvents. Solvent A was a mixture of 20% acetonitrile and 80% water containing 10 mM $KClO_4$, pH adjusted to approximately 3 with hydrochloric acid. Solvent B was 100% acetonitrile. The gradient was linear from solvent A to solvent B in 20 min at a flow rate of 1.0 mL/min and operated at room temperature. The diode array detector was programed to record chromatograms at 540 ± 60 nm, 254 ± 10 nm, 355 ± 10 nm, 220 ± 10 nm, and 440 ± 20 nm. Full spectra were acquired on all significant peaks from 190 to 600 nm.

Some analyses were run on a Varian Model 5000 Liquid Chromatograph equipped with a variable wavelength detector (model 100). The detector wavelength was set to 540 nm. The HPLC column used was a 20 cm 5 μm Nucleosil C_{18} with a 4.6 mm inside diameter. The mobile phase used was the same as above. The signals from the detector were stored with an Omega data system from Perkin Elmer.

Results and Discussion

Effect of Light on Composition of Inks

Ballpoint pen inks consist of several different constituents, such as various dyes or pigments, vehicles and resins or polymers (13). Basic dyes, such as Crystal Violet, Methyl Violet, Victoria Blue, Tetramethyl Para Rosaniline, etc., are often found in inks. The basic dyes appear as free base or in different ionic forms. In ballpoint inks, the free base is normally used. These cationic dyes are popular in inks due to their brilliant shades. However, their lightfastness is rather poor, they tend to darken and become dull. In our experiments, a combination of Crystal Violet (CV) and Methyl Violet (MV) has been found in every blue-colored ball pen ink. In some inks, CV and MV are the only dyes detected by HPLC. Figure 1 shows the chemical structure of these two dyes. The only difference between CV and MV is the number of methyl groups bound to the three nitrogen atoms.

On exposure to daylight, both CV and MV decompose. This decomposition seems to imply a successive loss of methyl groups, which are substituted by hydrogen atoms. Crystal Violet thus decomposes into Methyl Violet, which subsequently decomposes into other, structurally similar compounds, by successive loss of methyl groups. On the HPLC system employed in this study, the various decomposition products of CV elute before the CV peak, as the polarity of these molecules increases with the substitution of methyl groups by hydrogen. Figure 2 illustrates the decomposition of CV when exposed to daylight for several weeks. A solution of CV in methanol was applied to a writing paper and stored in daylight and artificial light, indoors. Fresh CV was almost free from other compounds, only small amounts of MV (the peak to the left from CV) were detected. When exposed to light, the amount of CV decreases and a number of decomposition products are detected at 540 nm by HPLC. The prolonged light exposure results in an increase in the relative concentration of the decomposition products. The UV-VIS spectra of the decomposition products obtained by a diode array detector resemble that of CV, but they are shifted with the maxima at shorter wavelength compared to that for CV. The shorter the retention time, the shorter the wavelength maximum, at least for the peaks closest to the CV.

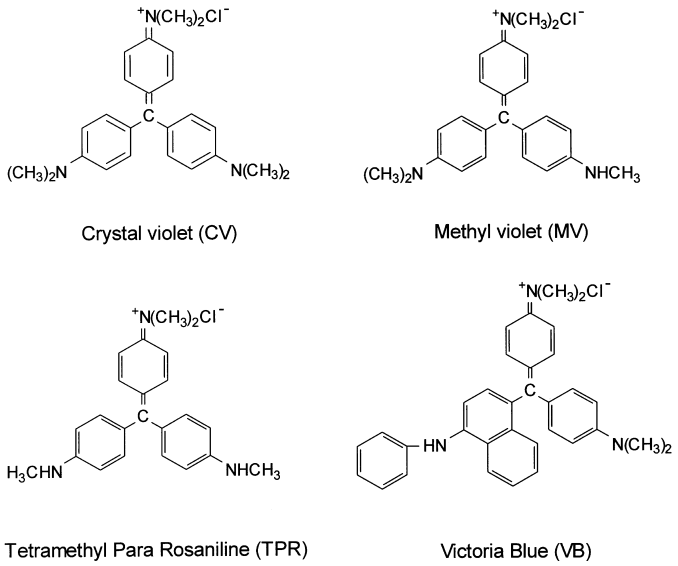

FIG. 1—*Chemical structure of the blue dyes studied in this work.*

Decomposition of Methyl Violet by light is very similar to that observed for CV. However, the sample of MV purchased by our laboratory was found to give three peaks in the HPLC chromatogram. One of the peaks proved to be CV, another one MV and the third one (the peak to the left of the MV peak in Fig. 2) is Tetramethyl Para Rosaniline. Together with CV and MV, this peak is generally present in blue inks and will here be designated as TPR. Exposure of a text written by a ballpoint pen to daylight or artificial light results in formation of various decomposition products of CV, MV, and other dyes in a manner similar to that of pure dyes. Figure 3(*a*) depicts the changes in composition of blue ink upon exposure to daylight inside a laboratory as observed by HPLC analysis and detection at 540 nm. Considerable quantitative changes in composition of the ink were detected already after several hours' storage at indoor daylight (the ink was not exposed to sunshine). For comparison, Fig. 3(*b*), no changes in composition were noted when the same ink was stored in darkness for three weeks.

Evaluation of Quantitative Changes in Ink Composition on Exposure to Light

The three basic dyes—CV, MV, and TPR—show poor lightfastness, as illustrated above. As these dyes are found in practically all the blue colored ball pen inks, the quantitative evaluation of the decomposition due to light exposure should be valuable. Many other dyes, less frequently present in inks, also decompose when stored in light. The changes of concentration of these dyes on exposure to light can be studied and evaluated in a manner similar to that used here for the CV-MV-TPR system.

It can be noted in Figs. 2 and 3 that, when exposed to light, CV decomposes to MV, MV decomposes to TPR, TPR decomposes to other similar substances by the gradual loss of methyl groups. The reaction rates of these decomposition reactions can be written

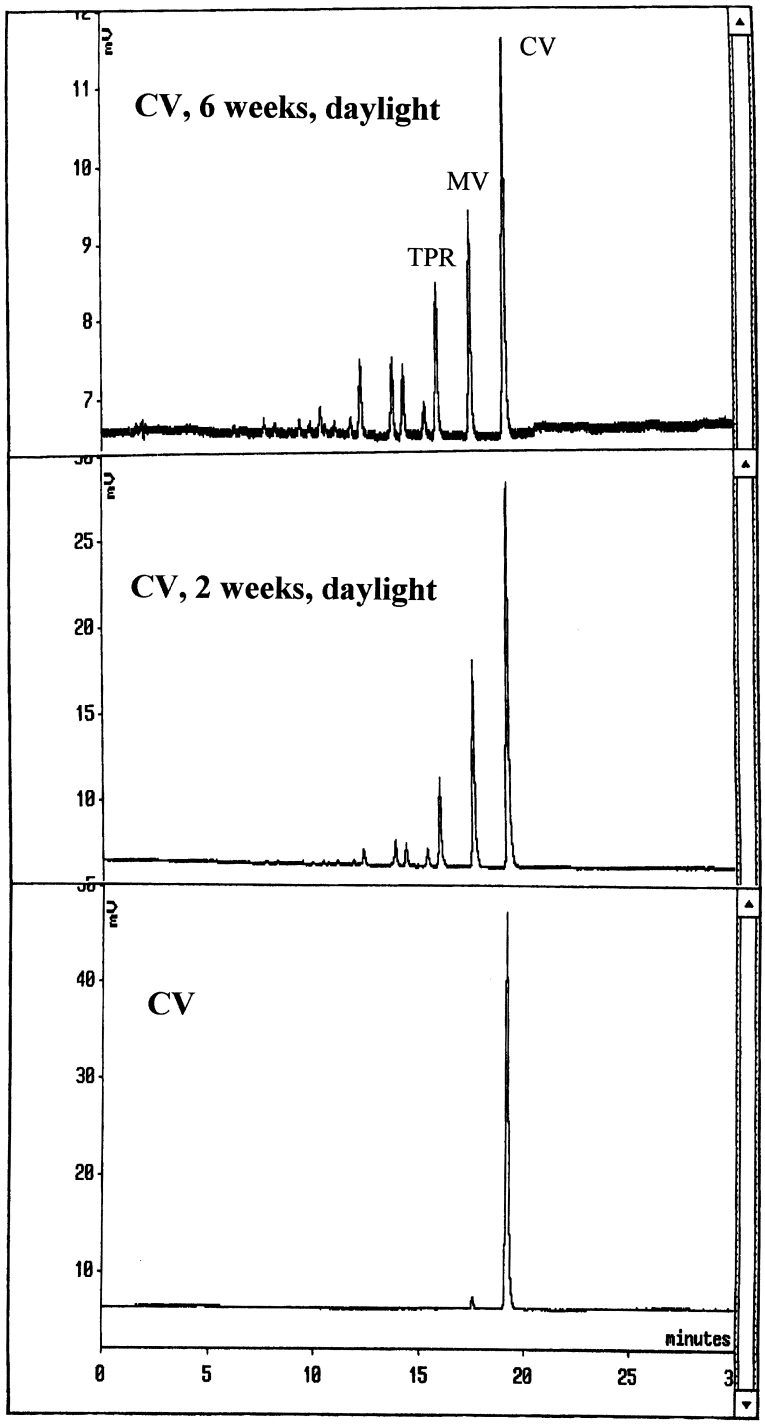

FIG. 2—*Changes in the composition of Crystal Violet (CV) when exposed to light. Methyl Violet (MV) is the first peak on the left side from CV. Next peak, on the left side from MV, is Tetramethyl Para Rosaniline (TPR). Note the decrease in the peak height of CV exposed to daylight. HPLC chromatograms were obtained at the detection wavelength of 540 nm by using a variable wavelength detector connected to a Varian Liquid Chromatograph.*

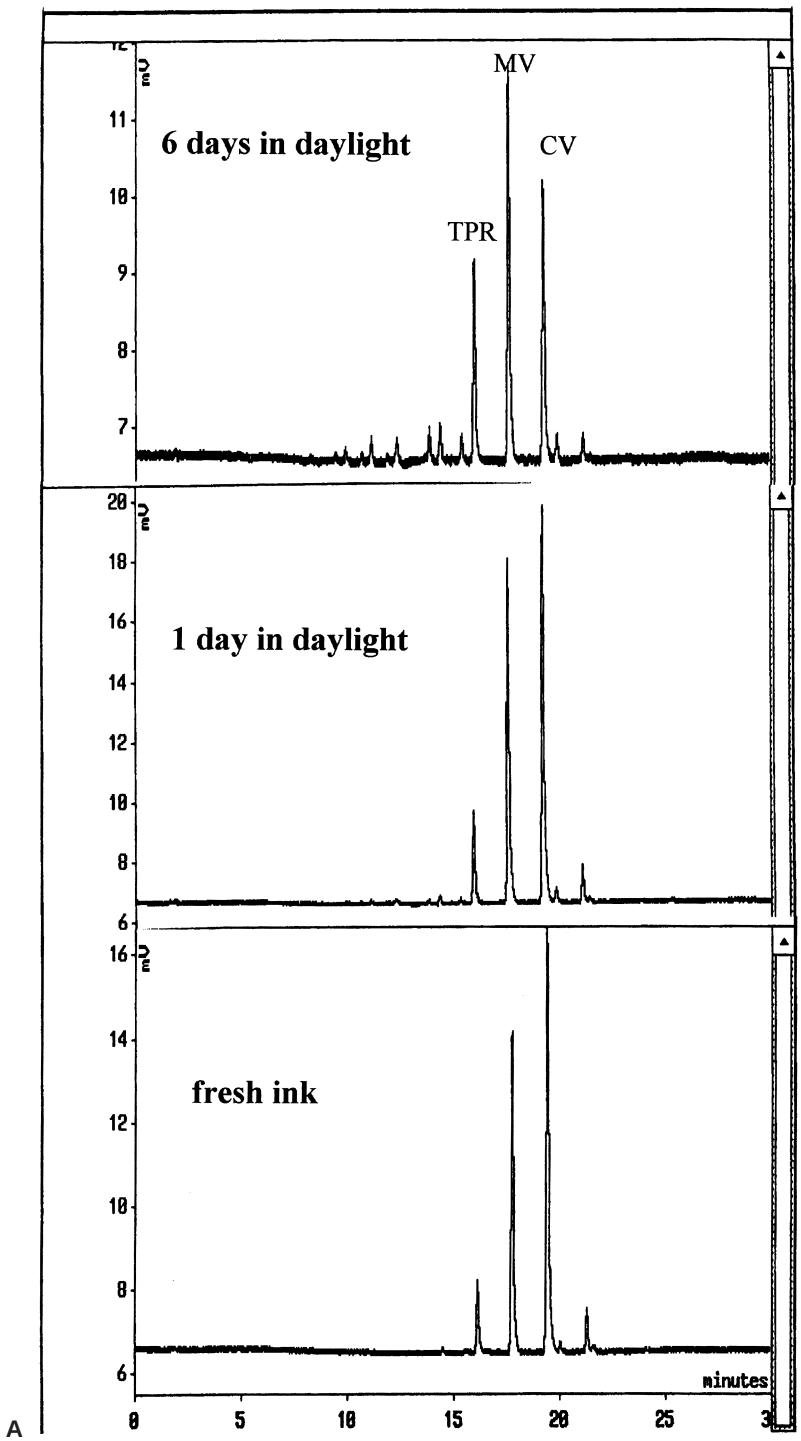

FIG. 3—*Effect of daylight on the composition of a blue ball pen ink. Detectable quantitative changes in composition were observed already after several hours of storage in daylight inside a laboratory. A Varian Liquid Chromatograph with a variable wavelength detector operating at 540 nm was used. The tendency of the changes in the CV-MV-TPR system is always the same—the concentration of CV decreases whereas that of TPR increases on exposure to light.*

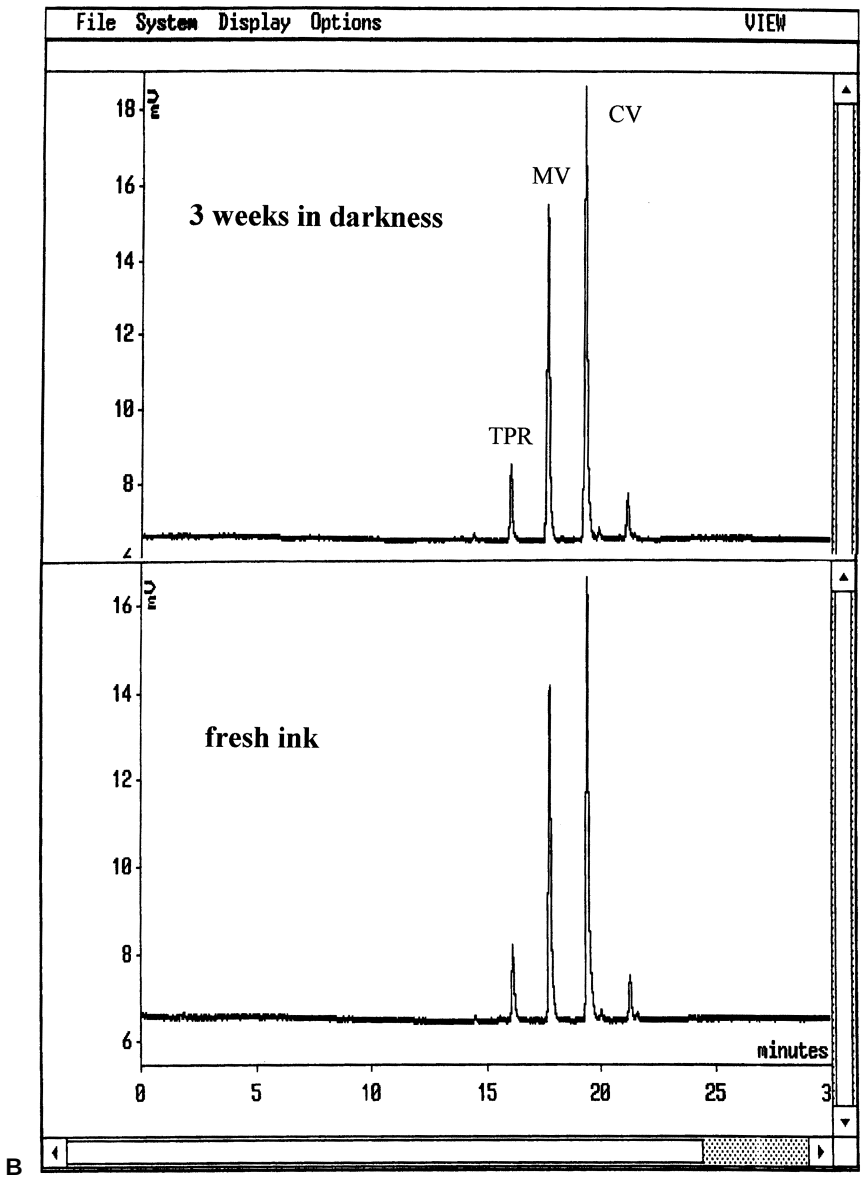

FIG. 3—*(Continued.)*

mathematically and the various rate constants calculated, but the exact mechanism and kinetics of each of the reactions is not known. Also, the intensity of light and the extent of the light exposure during the storage are normally unknown. Instead of kinetic calculations, we have used ternary diagrams in this study. In these diagrams, changes in composition of CV, MV, and TPR are easily illustrated. The dyes CV, MV, and TPR are plotted to each other in such a manner that the sum of the peak areas for these three dyes detected at 540 nm is 100%, regardless of the presence and concentration of other compounds. This way of plotting only concerns how the relative concentration of these three dyes change compared to each other. This does not reflect that the amount of all three dyes could decrease with time.

Figure 4 shows a ternary diagram with results obtained from several different Ballograf inks. Note that the triangulation points represent compositions other than for pure substances, the diagrams are expanded in this way, for making changes in composition of the inks more pronounced. The results of the two series of experiments, described above, are depicted in Fig. 4. For the samples stored in daylight (inside our laboratory), the samples were taken at about ten day intervals. The experiments were carried out in wintertime, when daylight in Sweden is much shorter and weaker compared to summertime. For the samples exposed to light from a fluorescent tube at a short distance, the samples were generally taken at 2 h intervals. Arrows mark the initial compositions of the inks examined. It can be seen in this figure, that the relative concentrations of CV, MV, and TPR change quite significantly with exposure to daylight or light from fluorescent tubes. These changes follow curves, starting from the left, going upwards and to the right. Small changes can be approximately estimated by straight lines with the slope increasing for inks containing less CV and MV (compositions towards the right corner of the triangle). The general feature of these curves can easily be seen in the figure and the curves may be obtained for other ink compositions. When exposed to light, CV will decompose mainly to MV, which in turn decomposes mainly to TPR. For inks with a relatively high content of CV

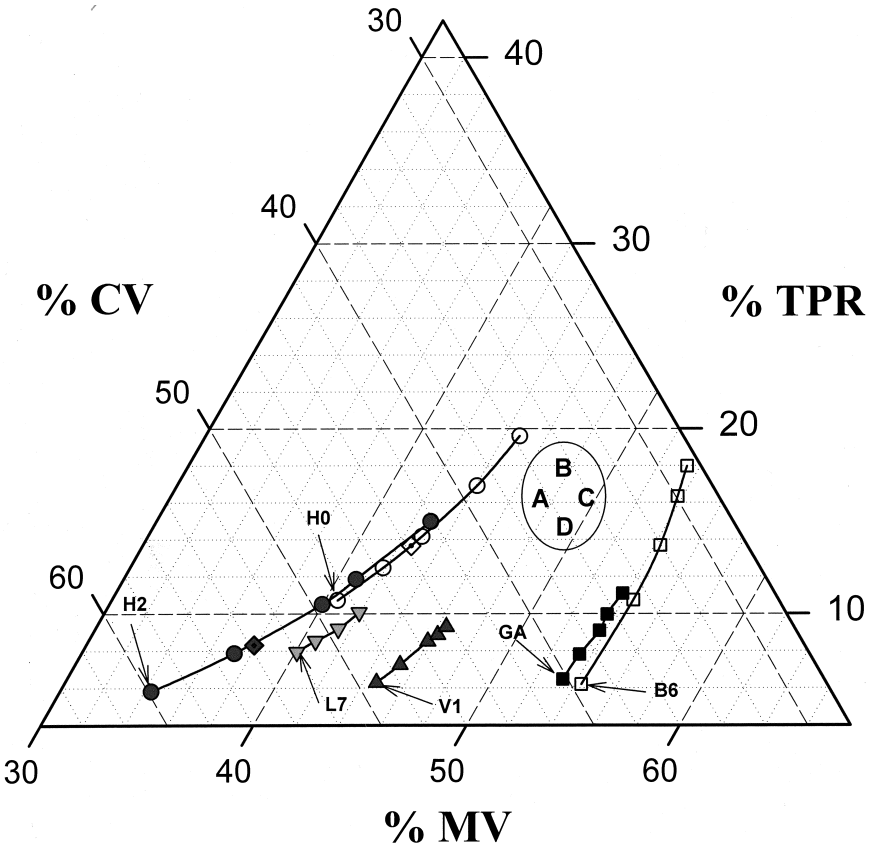

FIG. 4—*Ternary diagram constructed for the system CV-MV-TPR. Ball pen inks manufactured by Ballograf have been used in these experiments. Arrows mark the initial composition of fresh inks, unexposed to light. The inks H2 (•), H0 (○), and B6 (□) were exposed to daylight inside a laboratory. Inks (as asterisks on paper) were simply stored inside the laboratory and samples for analyses were taken at about ten day intervals. The experiments were carried out during November and December, when the daylight is weak and combined with artificial ceiling lighting. The inks V1 (▲) and GA (■) were exposed to light from a fluorescent tube from about 5 to 10 cm distance. The samples for analyses were taken at about two hour intervals. The symbols ◊ and ◆ represent the composition of the inks H0 and H2 exposed to the light from a fluorescent tube for four hours. Changes of the ink L7 (▼) are due to normal aging, sampled at about one year intervals for a period of three years. The letters A, B, C, and D represent the hypothetical composition of four inks. Which of the inks can be differentiated from the others is discussed in the text.*

compared to MV (e.g., the inks H2, L7), MV will be initially enriched in this system. With prolonged exposure the concentration of MV will reach a maximum and even start to decrease. For inks with lower content of CV compared to MV (e.g., inks B6, V1), the amount of MV formed by decomposition of CV will not compensate for losses by its decomposition to TPR. The apparent concentration of MV is nearly constant and decreases slightly with further light exposure. The enrichment of TPR and depletion of CV is a common feature.

Some of the inks in Fig. 4 were studied in both series of experiments—exposure to daylight and exposure to light from a fluorescent tube. The changes in the ternary composition CV-MV-TPR were found to follow exactly the same curves. The effect of direct sunshine on the composition of inks was not studied in this work, but it is known, that prolonged exposure to sunshine changes the appearance of blue ink substantially. It could be difficult to relate the composition of ink exposed to direct sunshine for a longer time to the original composition. Our ternary diagrams were constructed for the purpose of comparison of documents not intentionally exposed to sunlight or other strong light source.

Two of the inks presented in Fig. 4, namely ink H0 and H2, represent an interesting pair. The inks differ clearly in their composition when not exposed to light. When the ink H2 is exposed to light for sufficiently long time, its composition will reach that of the ink H0, unexposed to light (about 49% CV, 41% MV, and 10% TPR). Figure 5 shows HPLC chromatograms obtained for this particular situation. With prolonged exposure to light, the ink H2 will follow approximately the same curve as that observed for the H0 when it starts to be exposed to light. Both inks contain another unidentified strong peak detected at 540 nm, but of an indistinguishable amount. It may be difficult to discriminate between these two inks by HPLC in casework, unless special attention is applied to minor peaks.

Figure 6 illustrates the differences between the inks shown in Fig. 5 in the amount of minor peaks formed by decomposition of CV, MV, and TPR. The peaks were numbered from 1 to 5. These peaks are identical with some of the decomposition peaks in Figs. 2 and 3. The amount of decomposition is higher for the ink H2 by comparison with the ink H0. To illustrate the importance of measuring minor decomposition peaks in ink comparison, a diagram presented in Fig. 7 was constructed. For simplicity, the amount of decomposition products in inks was expressed as the sum of peak areas of the peaks 1 to 5 detected at 540 nm. The amount of TPR was then plotted against this amount. Figure 7 depicts the results achieved for the inks from the ternary diagram (Fig. 4). In the range of the light exposure employed in this study, the amount of TPR increased apparently linearly with the amount of decomposition products. The straight lines in Fig. 7 are parallel with sim-

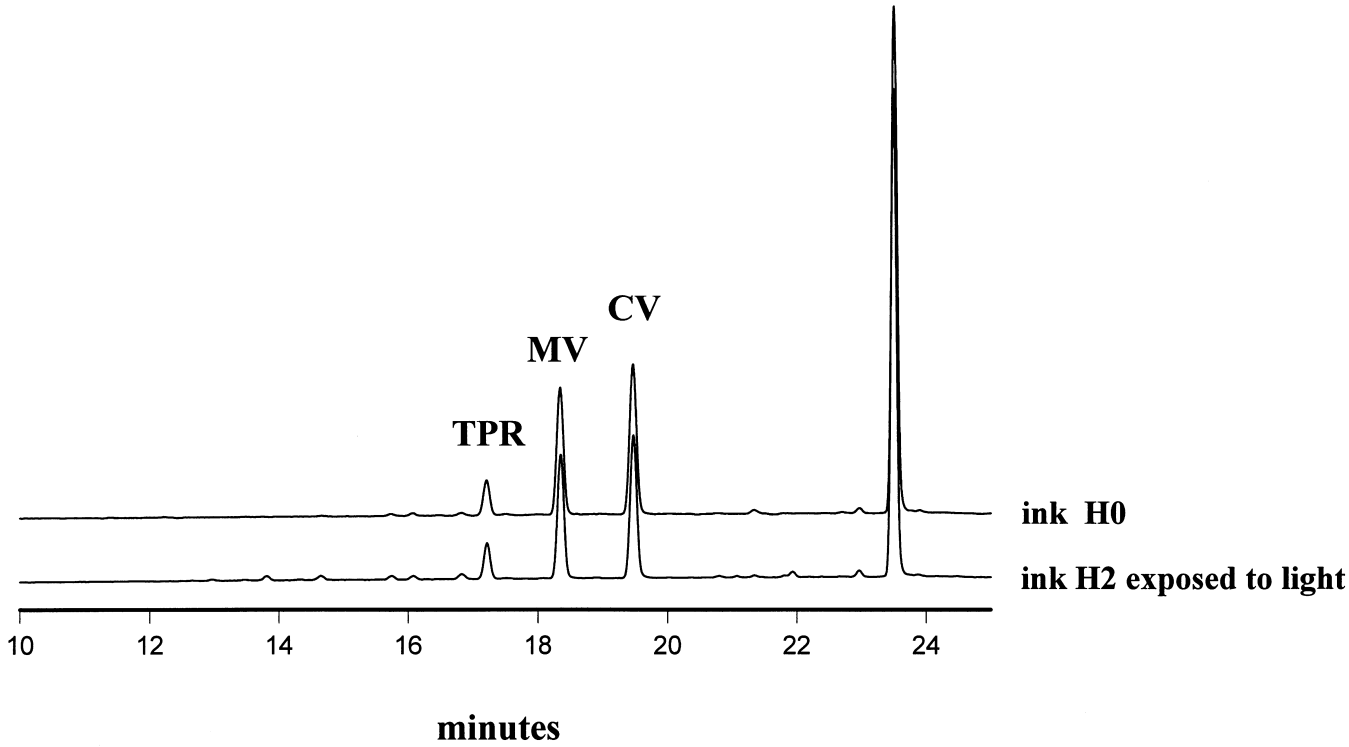

FIG. 5—*HPLC chromatograms obtained for the ink H0 (not exposed to light) and the ink H2 exposed to daylight for about four weeks. The composition of these inks, initially quite different, seems to be hardly distinguishable. Diode array detector, detection at 540 ± 60 nm.*

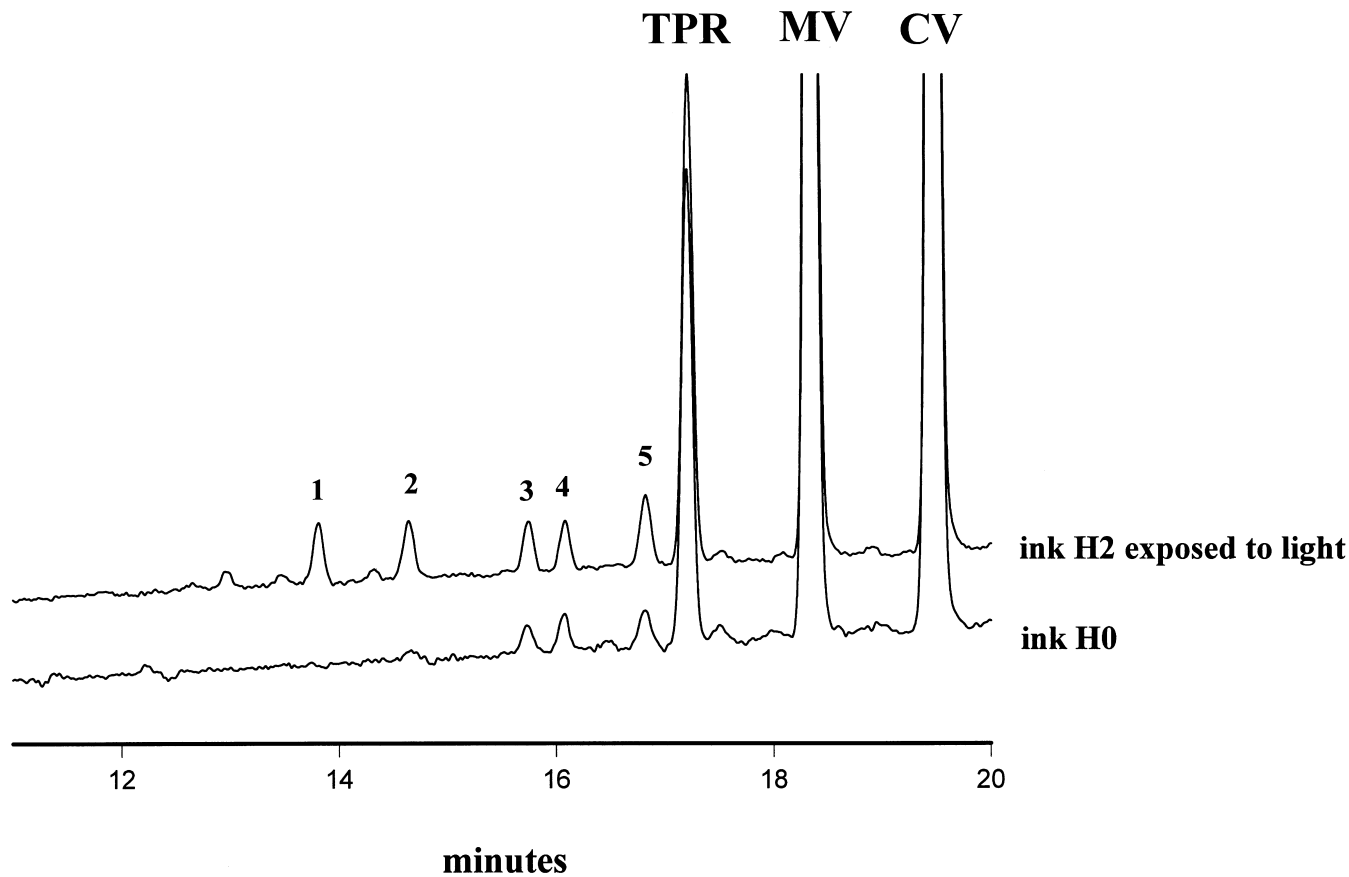

FIG. 6—*An expanded part of the chromatograms shown in Fig. 5. The peaks designated by numbers 1 to 5 represent various decomposition products formed from CV, MV, and TPR. On exposure to light the amount of these products increases. The peaks can be used to differentiate between the H2 and H0 inks.*

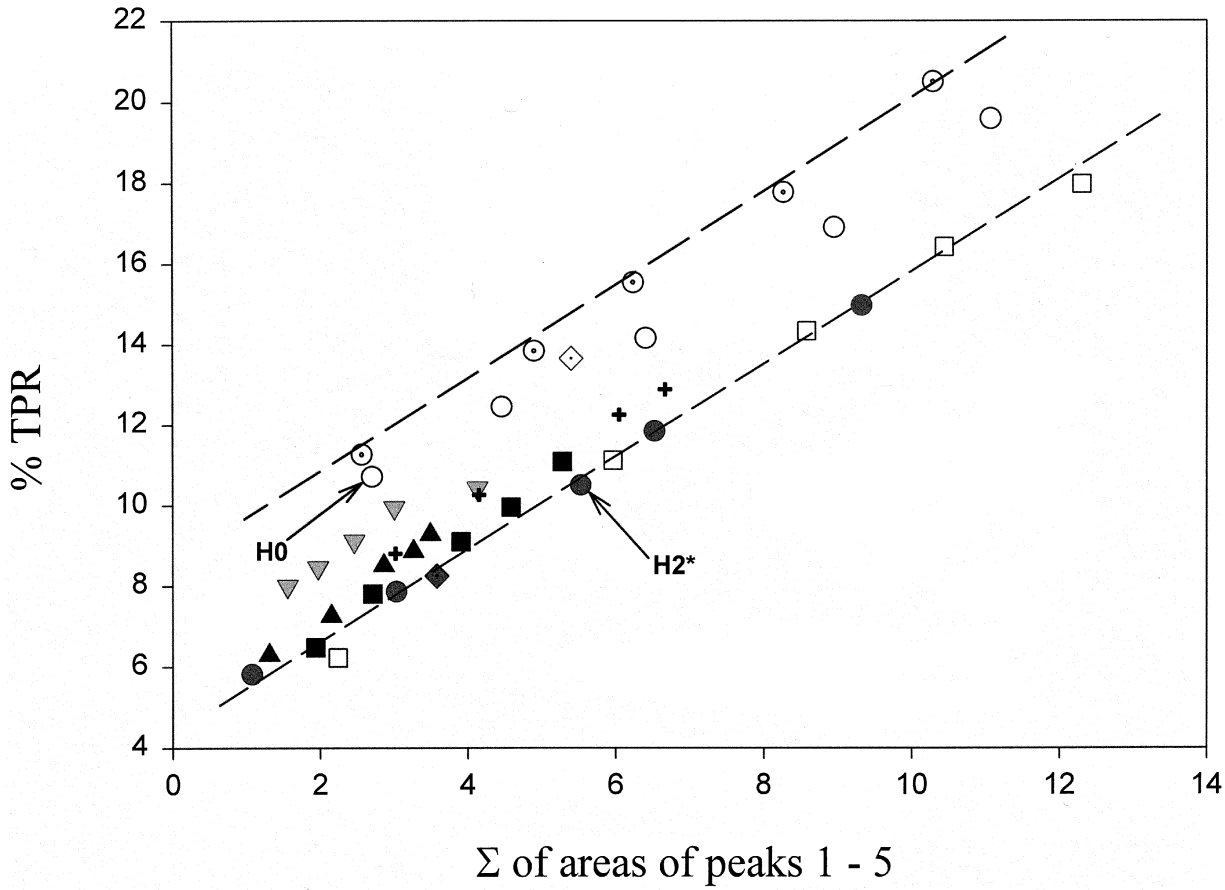

FIG. 7—*A plot of TPR as a function of the sum of peak areas of the decomposition peaks 1–5 (for designation, see Fig. 6). This diagram is a complement to the ternary diagram in Fig. 4. The inks are marked by the same symbols as in Fig. 4. —H0 (○), H2 (•), B6 (□), L7 (▼), V1 (▲), GA (■). In addition, the inks M1 (⊙) and E5 (+) were measured. Arrows mark the composition of the fresh H0 ink and of the H2 ink exposed to daylight, as shown in the chromatograms in Figs. 4 and 5. The symbols ◊ and ◆ represent the composition of the inks H0 respective H2 on exposure to the light from a fluorescent tube for 4 h. The concentration of TPR seems to increase approximately linearly with the sum of peak areas 1 to 5. The dashed lines are aimed to lead the eye in the diagram.*

ilar slopes. The lines do not pass through the origin of coordinates. The reason for these features is that inks not exposed to light contain various amounts of both the decomposition products and TPR, presumably already from production. Two clearly separated points in Fig. 7 represent the composition of the inks H2 and H0 shown in Figs. 5 and 6 and indistinguishable in the ternary diagram. H2* represents point in Fig. 4 where ink H2 (after exposure to light) reaches composition of ink H0 not exposed to light (49% CV, 41% MV, and 10% TPR). The points do not belong to the same straight line, which indicates that inks have different formulations.

Figure 7 gives an additional dimension to the ternary diagram (Fig. 4). By the use of this fourth dimension (amount of decomposition products in inks), many inks exposed to different amounts of light can be distinguished.

Not only exposure to light causes changes in the CV-MV-TPR system, but also normal aging of inks will result in decomposition of CV, MV, and formation of peaks, such as those designated 1 to 5 in this study. This process is, however, much slower for inks not exposed to light. Figure 4 shows changes in composition of an ink under a period of several years. To our knowledge the ink was not unnecessarily exposed to light. The changes in composition of this ink seem to have the same tendency as for inks exposed to light. This normally aged ink is also presented in Fig. 7.

Use of the Proposed Diagrams in Comparison of Inks

In forensic investigation of ballpoint pen inks, the storage of the inks to be examined is generally not known. An analysis of ink entries on different documents may reveal quantitative differences in chemical composition as well as differences in optical properties. This may result in the inks being classified as different. In spite of that, the inks may be of the same kind, but stored under different light conditions. That can easily be verified by plotting a ternary diagram for the system CV-MV-TPR as well as the diagram presented in Fig. 7. If the positions of two inks to be compared are such that light exposure may convert the composition of one of the inks into the other one, it cannot be excluded that the inks are the same. The ink exposed to light to higher extent will be found on the right side and diagonally upward, compared to the other ink. In Fig. 7, the inks will be represented by points on the same line, with the more exposed ink having higher *x*- and *y*-values. This procedure is, of course, necessary only if the qualitative composition of the inks is the same. For illustration, compositions of four hypothetical inks, marked as A, B, C and D, are included in Fig. 4. With guidance from the curves describing the changes in the composition of the various inks in this figure, some conclusions may be done directly. When exposed to light, the ink A might be transferred to the position B, but never to the position of C or D. While B can char-

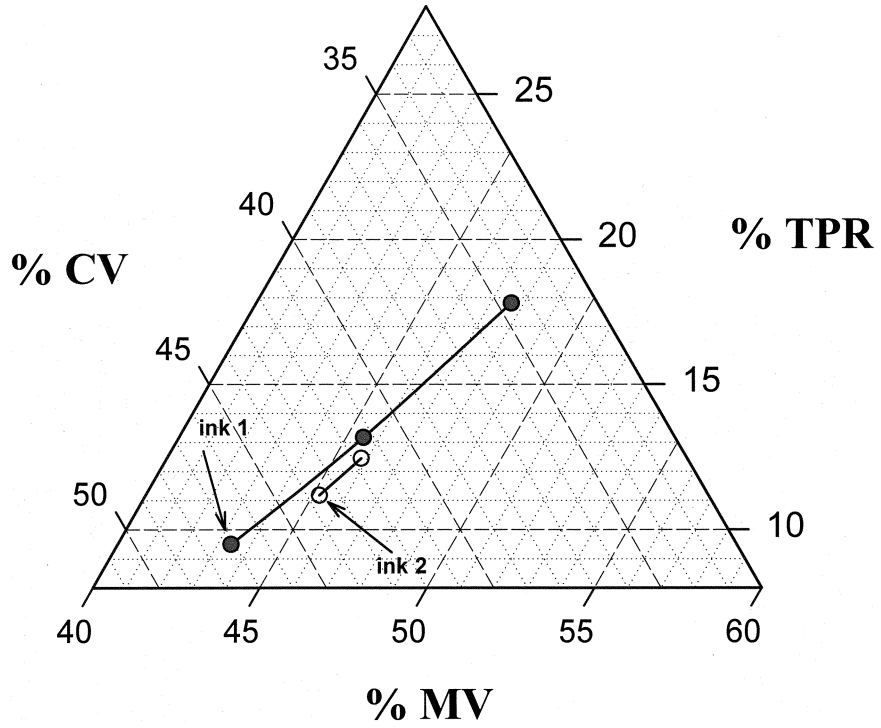

FIG. 8—*Ternary diagram constructed for the system CV-MV-TPR. Arrows mark the initial compositions of two inks encountered in casework. The inks differed slightly on an optical examination. The diagram shows the changes in the composition of ink 1 on exposure to artificial light for 3 and 6 h, respectively. Ink 2 was exposed to light for 1.5 h. The aim of the various light exposures was just to control if the curves describing the changes of the concentration in the CV-MV-TPR system intersect each other or not. They seem not to do it. Thus, although similar to each other, the inks were considered different.*

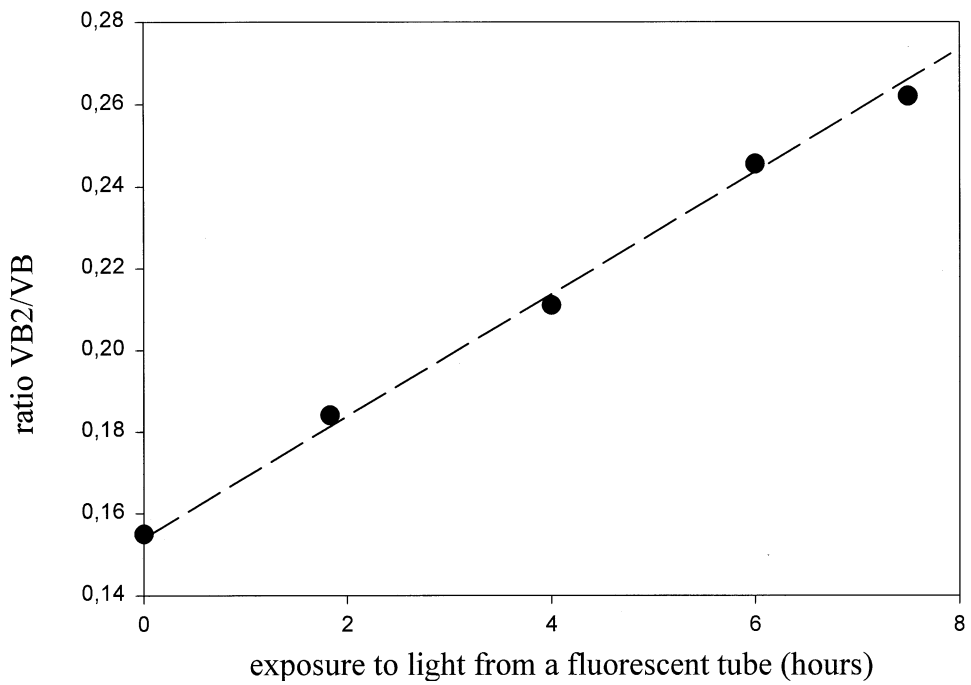

FIG. 9—*Decomposition of Victoria Blue (VB) in a blue ball pen ink exposed to light from a fluorescent tube. The decomposition peak has been designated as VB2. The experimental arrangements are the same as for the inks shown in the CV-MV-TPR diagrams.*

acterize the ink A exposed to a certain amount of light, the compositions C and D represent inks different from A. In addition to that, B represents ink different from D. The ink D can characterize the ink C exposed to light.

It may sometimes be difficult to judge the exact direction of changes in the composition of inks after exposure to light. The slopes of the curves describing these changes in the ternary diagram (Fig. 4) vary with the position in the triangle. In such a case, it is advisable to perform additional analyses of the inks after exposure to light under controlled conditions. The procedure is illustrated by an example from our casework.

Ink entries on two separate documents were suspected of being written by the same pen. An optical examination revealed slight differences between the two inks. HPLC analysis of the inks showed they were qualitatively the same, but they differed in their quantitative composition. The relative concentration of CV, MV, and TPR was different and no other dyes were detected. Figure 8 shows the ternary diagram for the system CV-MV-TPR with the composition of the two inks. The differences in composition are such that the exposure of ink 1 to light may cause an agreement with ink 2. Therefore, additional samples were taken from the suspect documents. The samples were exposed to light from a fluorescent tube for three and six hours. Afterward the samples were analyzed by HPLC. The result of these analyses is included in Fig. 8. It seems that, exposed to light, the composition of ink can be made almost indistinguishable. By plotting the amounts of TPR against the total amount of the decomposition peaks (peaks 1 to 5), the two inks appear to belong to the same line and may represent the same ink. The two curves in Fig. 8 seem, however, to be slightly shifted from each other. Our experiences with analysis of inks by HPLC are that the repeatability is very good. As all the HPLC analyses were carried out one after the other, we considered the inks being different. If the analyses were performed on different occasions, we would probably not find these inks different. The case also illustrates that the tendency of compositional changes in the system CV-MV-TPR is the same for other ink manufacturers.

The CV-MV-TPR system employed in this study is not the only system, which can be used for an evaluation of compositional changes of ball pen inks exposed to light. Many other changes occur in inks when they are exposed to light. Victoria Blue (VB) is an example of another dye with poor lightfastness. It should be mentioned here, however, that the lightfastness of VB is much higher than that of MV and CV. Victoria Blue decomposes in a manner similar to that of CV, but only one decomposition product (designated here as VB2) is observed when moderately exposed to light. Figure 9 shows this decomposition graphically. All such factors must also be considered in forensic examination of inks. The CV-MV-TPR system is, however, applicable to every sample of blue colored ball pen ink and should be considered first.

Conclusions

It is not always straightforward to state that two ink entries on different documents are different. If the differences between the ink entries are not of qualitative character, storage of the documents containing the same ink at different light conditions might have caused the observed differences. Both optical properties (color shade, IR luminescence) and chemical composition will change and the changes occur already after rather short exposure to daylight (a few hours at daylight). The presence of various amounts of decomposition products indicates that the documents may have a different history of storage. Ternary diagrams constructed for the CV-MV-TPR system together with plots of TPR against the total amount of decomposition products may reveal if the inks can be differentiated or not. Many times, it may be advisable to expose the investigated inks to light and follow the changes in the composition in the diagram to decide if the inks can be discriminated or not.

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Additional information and reprint requests:
Jan Andrasko, Ph.D.
SKL, National Laboratory of Forensic Science
S-581 94 Linköping, Sweden