

*D. A. Crown,¹ D.Crim.; R. L. Brunelle,² M.S.; and
A. A. Cantu,² Ph.D.*

The Parameters of Ballpen Ink Examinations

Prior to about 1950, the inks encountered on questioned documents were usually fluid inks of the washable dyestuff or iron gallotannate varieties. Analysis of these inks included observation under various wavelengths of light ranging from ultraviolet (UV) to infrared (IR), photography with selected filters, and chemical spot tests to detect metals such as iron, vanadium, and chromium. Since about 1953, the ballpen has taken over the position formerly occupied by the fluid ink pen.

In a paper presented at the July 1951 meeting of the American Society of Questioned Document Examiners, Linton Godown [1] first proposed the use of disk or thin-layer chromatography for the examination of fluid inks. Subsequently, in 1952 Somerford and Souder [2] studied the use of paper chromatography for ink examinations. Shortly thereafter, Brackett and Bradford [3] also reported on the same topic. Two years later, this work was extended to cover ballpen ink writing and the use of electrophoresis by Brown and Kirk [4,5]. In 1966, Tholl [6] introduced a more refined idea of adapting thin-layer chromatography (TLC) to ink examination. In 1961 Crown et al [7] used chemical spot tests to distinguish specific dyestuff constituents in ballpen inks. The works of Godown [8], Dick [9], and Von Bremen [10] on IR luminescence, dichroism, and UV photographic techniques should also be mentioned as further developments of ink analysis.

One of the earlier workers on the dating of ballpen inks was Werner Hofmann of the Zurich Cantonal Police, Zurich, Switzerland, who initiated comparisons of questioned inks with his collection of European ballpen inks standards.³ Prior to this, the dating of inks was mainly limited to determinations of periods of time when gross changes were made in the composition of inks. For example, the change from oil-based inks to glycol-based inks provided a date prior to which certain inks did not exist. The introduction of copper phthalocyanine dyes into inks can also be used to date an ink.

In the mid-1960s the Bureau of Alcohol, Tobacco and Firearms (ATF), Department of Treasury, recognized the need for a more systematic approach to ink analysis and the dating of ink specimens. This work was needed to supplement some of the existing means of dating documents, such as dating watermarks and typewriter typeface designs.

Standard Ink Library

Beginning in 1968, ATF began to systematize procedures for ink analysis and the collection of ink specimens from ink manufacturers. Starting in April 1968, U.S. manu-

Received for publication 13 Feb. 1976; accepted for publication 19 March 1976.

¹ Criminalist, Department of State, Washington, D.C.

² Criminalists, Bureau of Alcohol, Tobacco and Firearms, Washington, D.C.

³ Private communication, 1970.

facturers of ballpen inks have supplied ATF with samples of new ink formulations developed. Additionally, ink formulations developed prior to 1968, which were still on hand, were obtained from the U.S. manufacturers. In addition to the U.S.-manufactured ballpen ink specimens, all the ballpen ink formulations in Hofmann's European collection, manufactured prior to 1972, were also obtained. No permanent sampling procedure for foreign-manufactured inks was established, although samples available in the U.S. are collected periodically.

At this date, the ATF Ballpen Ink Library can be considered complete for all domestically produced inks manufactured after 1968 and predominantly complete for inks produced since 1958. No such claim is made for foreign-produced ink formulations. Presently, the ATF Ink Library includes the following number of different domestic ballpen ink formulations: blue, 239; black, 121; and shades of red, green, yellow, brown, and others, 720. There are 457 samples of foreign-manufactured ballpen inks on hand; however, these include some duplicate ink formulations.

Ink formulations are changed to improve some utilization aspect such as change in available materials and in cost of ingredients. Changes occur in the volatile components or among the nonvolatile components such as dye resins, viscosity adjusters, and fatty acids. The ATF Ballpen Ink Library includes information supplied by manufacturers about the nature of the changes in their ink formulations and the date of these changes. A history of the formulation is maintained as occasionally a change is made in a particular batch without changing the designation of the formula. Such changes are made to meet certain desired specifications and are often detectable. If the difference is detectable with existing technology, the special batches are considered different formulations.

Method of Analysis

In the ATF system, standard inks are initially categorized by color, IR reflectance, and UV fluorescence; secondarily, by solubility and chemical spot tests; and finally by TLC. Adequate separations are achieved using Eastman silica gel plates and a solvent system composed of ethyl acetate:ethanol:distilled water (70:35:30). Both visible dyestuff constituents and UV fluorescent components are separated by using this system.

Ink samples are removed from written ink lines using either a blunted hypodermic needle or scalpel (Fig. 1). Anywhere from 1 to 3 μg of ink (0.5 to 1.5 cm of ink line or 8 to 10 punched-out samples of ink) are dissolved in pyridine and then spotted on the TLC plate. If a new standard ink formulation cannot be differentiated from other similar standard ink formulations using this plate/solvent system, then the TLC procedure is repeated with Merck silica gel glass plates using the above solvent system. This usually suffices for distinguishing ink formulations; however, if necessary, additional TLC can be performed using Merck plates and a solvent system with different polarity, such as *n*-butanol:ethanol:distilled water (50:10:15). The Merck plates are very sensitive and provide excellent resolution and differentiation among ink formulations (Figs. 2-4).

Because of the sensitivity of the chromatographic plates to ambient conditions, it is best to analyze questioned and standard inks side by side on the same TLC plate. To obtain further characteristics one can determine the relative ratio of detectable components on the plates using densitometric techniques, use fluorometric techniques to compare fluorescent components, or apply liquid chromatography to analyze resins or fatty acids. The ATF procedures are covered in greater detail in Refs 11-13.

Semantics of Ballpen Examination

At this time, with the advent of standardized techniques for TLC separation of ballpen ink dyestuff constituents, we can standardize some of the statements that can be

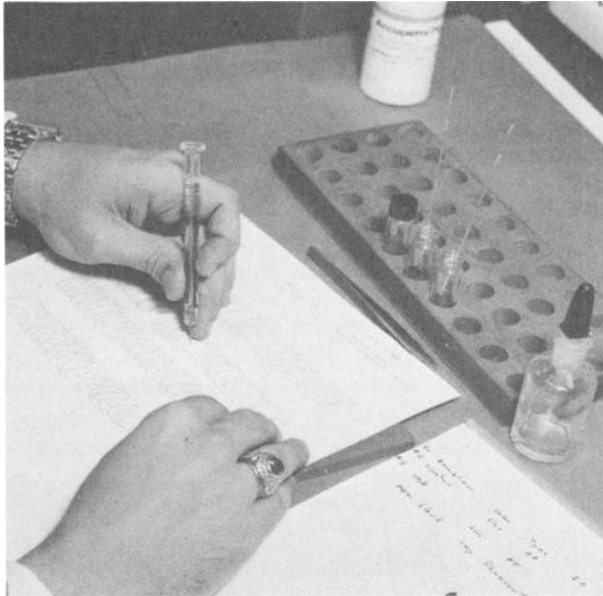


FIG. 1—*Removing ink from an ink line with a blunted hypodermic needle.*

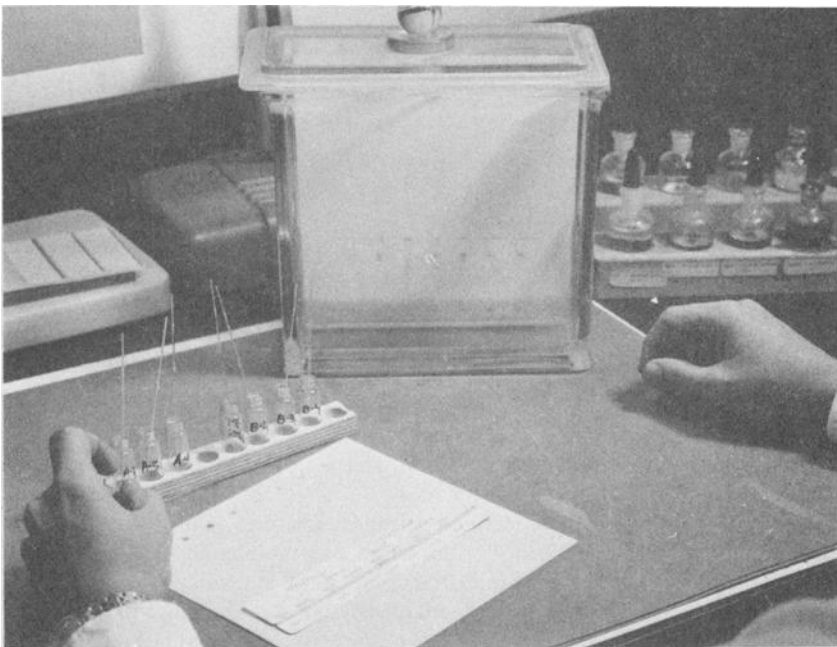


FIG. 2—*Running a chromatogram.*

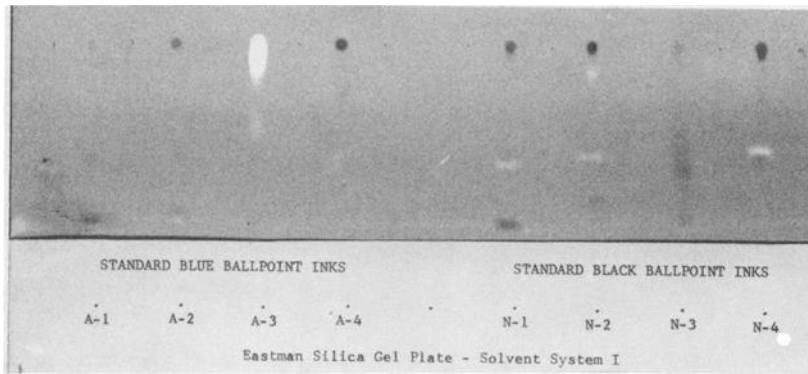


FIG. 3—*Eastman silica gel chromatogram photographed under ultraviolet light.*

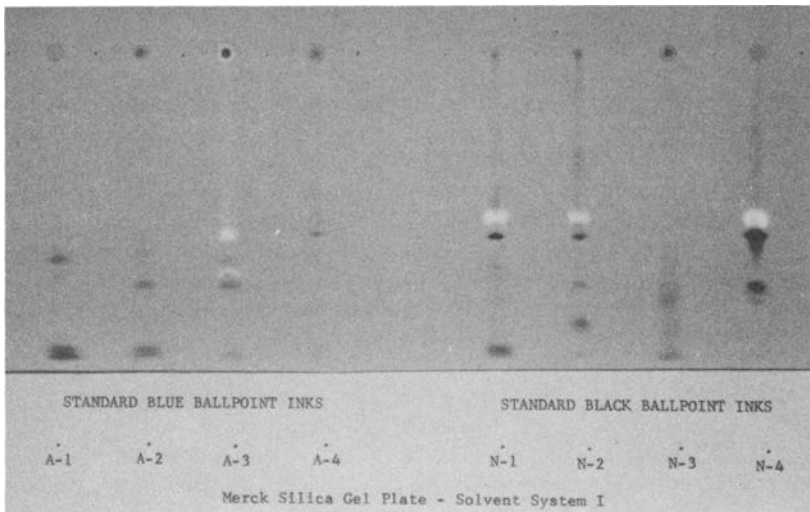


FIG. 4—*Merck silica gel chromatogram photographed under ultraviolet light.*

made about the significance of TLC ink comparisons. It would be well to assure that statements are in line with current forensic science and generally accepted forensic semantics.

Ballpen inks, like other manufactured chemical mixtures, cannot ordinarily be individualized. Many pens are filled with ink from one production, and it is not usually possible to differentiate the ink from different batches if the batches are carefully made according to a specific formula with standardized sources of supply, that is, if there is consistent quality control. Individualization, in this case, means connecting an evidential object with a unique source. The types of evidence that can be individualized include (a) aspects of a unique person, such as friction ridge surfaces, handwriting, and voices, (b) striated markings resulting from a harder object scraping across a softer surface; (c) random breaks or tears of a single object (jig-saw matching); and (d) manufactured items

which through use or accident become individual. Ballpen ink, as Crown [14] said of paint,

must be considered "class" evidence until basis has been established to individualize it and differentiate it from all others of the same class. Establishing a narrow class rather than a broad class does not provide basis for definite identification or individualization. Agreement in all observable aspects, no matter how finely measured, does not provide basis for definite individualization.

In considering inks as a type of class evidence, very narrow classes (formulations) of distinct evidential value can be established by using the ballpen ink dyestuffs and the UV fluorescing additives as the differentiating criteria. A finite number of ballpen ink formulas exist, as opposed to other types of evidence which involve an infinite number of unique items such as writers and handwriting. The TLC technique is considered a valid technique in court for separating ink constituents. The technique and principles are as scientific as the techniques and principles involved in the examination of soils or paints. In the case of soil, which is also class evidence, not every soil has been, nor could be, tested.

The exact finite number of different ballpen ink formulations is yet to be determined. The files of the ATF Laboratory are complete for U.S.-manufactured ballpen inks manufactured after 1968. The ATF files for ballpen inks manufactured prior to 1968 are incomplete. Additionally, the ATF files are incomplete for foreign-made inks. It is not accurately known how many different brands of ballpens or how many different ballpen inks have entered the U.S. since 1945, the year they were commercially introduced.

The complete collection of ballpen inks manufactured in the U.S. since 1968 is complete insofar as all known manufacturers have been contacted and have cooperated. We believe there are no known manufacturers that have not cooperated, and all known ink formulations after 1968 have been reported to the ATF laboratory.

A key issue in the TLC technique for ballpen ink examination is the evaluation of matches and dissimilarities with the ATF files. If there is no match with the ATF ballpen ink files, it is possible that (a) the questioned ink was manufactured prior to 1968 in the U.S. or overseas, (b) the questioned ink was manufactured overseas sometime after 1968, or (c) that there has been some significant physical deterioration of the questioned ink sample.

If there is a match between chromatograms from a questioned ink and an ink on file in the ATF laboratory, it is possible that the questioned ink is of the same class as a dated ink formulation on file in the ATF laboratory. The possibility also exists that there is another ink formulation made prior to 1968 in the U.S. or overseas at any time that produces chromatograms matching an ink manufactured in the U.S. after 1968. The degree of probability of there being another ink of the same formulation as one of the inks on file at the ATF laboratory must be estimated by the ink chemist or questioned document examiner performing the examination.

An exclusionary statement that a document could not have been prepared on the date claimed because the ink used matches a known ink formulation manufactured after the date of the document cannot be made unless there is evidence that the pen allegedly used was filled with a U.S.-made ballpen ink formulated after 1968. Any indication that the pen used could have been filled with a pre-1968 formulation or a foreign-made ink formulation would provide basis for claiming a match with a formulation not on file in the ATF laboratory.

As the ATF files on foreign and pre-1968 pens increases, the likelihood of an ink not on file at ATF being encountered in comparisons decreases, and we will be moving from reasonable possibilities to theoretical possibilities. While it is not possible to make a definite statement that a document was back-dated, based only on matching ink form-

ulations, such data may be combined with other evidence by the judge or jury in reaching a reasoned determination of fact.

References

- [1] Godown, L., "Differentiation and Identification of Writing Inks by Chromatographic Analysis," presented at the Annual Meeting of the American Society of Questioned Document Examiners, Rochester, N. Y., 1951.
- [2] Somerford, A. W. and Souder, J. L., *Journal of Criminal Law, Criminology, and Police Science*, Vol. 43, No. 1, 1952, pp. 124-127.
- [3] Brackett, J. W. and Bradford, L. W., *Journal of Criminal Law, Criminology, and Police Science*, Vol. 43, No. 4, 1952, pp. 530-539.
- [4] Brown, C. and Kirk, P. L., *Journal of Criminal Law, Criminology, and Police Science*, Vol. 45, No. 3, 1954, pp. 334-339.
- [5] Brown, C. and Kirk, P. L., *Journal of Criminal Law, Criminology, and Police Science*, Vol. 45, No. 4, 1954, pp. 473-480.
- [6] Tholl, J., *Police*, Vol. 2, No. 55, 1966.
- [7] Crown, D. A., Conway, J. U., and Kirk, P. L., *Journal of Criminal Law, Criminology, and Police Science*, Vol. 52, No. 3, 1961, pp. 338-343.
- [8] Godown, L., *Journal of Criminal Law, Criminology, and Police Science*, Vol. 55, No. 2, 1964, pp. 280-286.
- [9] Dick, R. M., *Journal of Forensic Sciences*, Vol. 15, No. 3, July 1970, pp. 357-363.
- [10] Von Bremen, U., *Journal of Forensic Sciences*, Vol. 10, No. 3, 1965, pp. 368-375.
- [11] Brunelle, R. L. and Pro, M. J., *Journal of the Association of Official Analytical Chemists*, Vol. 55, 1972, pp. 823-826.
- [12] Kelly, J. D. and Cantu, A. A., *Journal of the Association of Official Analytical Chemists*, Vol. 58, 1975, pp. 122-125.
- [13] Brunelle, R. L. and Cantu, A. A., *Forensic Science*, American Chemical Society Symposium Series, Vol. 15, 1975, pp. 134-141.
- [14] Crown, D. A., *The Forensic Examination of Paints and Pigments*, Charles C Thomas, Springfield, Ill., 1968, p. 6.

R. L. Brunelle
 Bureau of Alcohol, Tobacco and Firearms
 P.O. Box 7615
 Washington, D.C. 20044