Dating: Document

Introduction

In questioned document examinations, the age of a document is often an important criterion to determine the authenticity or highlight a fraud. The following questions are therefore frequently asked to the expert:

- Was a receipt actually produced at the mentioned date?
- Was a contract signed at the same time by the involved stakeholders?
- Was a paragraph added several years after the establishment of a testament?

To answer these questions, it is necessary to determine the date at which a document was written or printed. The time frame may be a few months or several years. As a result, many forensic scientists have investigated the issue of dating documents. Every book about document analysis has a chapter or paragraph about the dating methods [1–8]. The dating can focus on different aspects of the questioned documents. Whereas dating through paper, toner, or handwriting examinations is less considered in the literature, the potential of inks has been more particularly studied and many methods have been proposed so far [9–35]. Several reviews have been published on the topic over the last 30 years [36–45]. Although a number of laboratories apply ink dating in caseworks, the method raises a large amount of controversies and debates among the scientific community [39, 45–62, 112].

The objective of this article is to give a comprehensive overview on the dating of questioned documents, and more particularly their ink. In the first part, three fundamental approaches to dating are presented with examples of dating methods. This article also introduces the problem of method validation and interpretation of dating evidence.

Dating Principles

Three fundamental approaches can be defined and formalized [62–64] (Figure 1). The first, often called the *static approach* [41], focuses on the inherent properties of the ink named *time tags*. Time tags generally yield information on the production date of the document's raw constituents or their introduction on the market. The second, addressed as the *dynamic approach* [42], is based on the aging processes of documents. The third approach aims at reconstructing the chronology among documents or ink entries by ordering them in a sequence. Differentiation between "absolute" and "relative" age of documents is usually made in the literature [15, 16, 18, 46, 50, 52]. Determining the relative age of a document compared to others is equivalent to reconstructing their chronology.

Time Tags

Time tags are transferred to the ink entry or document during the creation of the document and may yield temporal information of varying precision [64]. It may be in the form of a precise transfer time [65] or an interval within which the document was created [67]. In the questioned document field, the information is generally contextual and must be translated in a time estimate by the expert [64].

An estimation of the age of documents can be based on the analysis of stable compounds found in the paper, ink, or toner that are specific to a certain period in time. Fabrication properties change and evolve with time as a function of new industrial developments and progress. For example, ballpoint pens appeared on the market in 1944 and were characterized by oil-based inks up to 1951, when glycols as solvents were introduced in their ink composition [68, 69]. Fluorescent brighteners were introduced in paper composition in the 1950s [67]. Copper phthalocyanine dyes and pigments were introduced in 1954 in blue ink for their good light stability. Gel pens were first found in Japan in 1986 [70]. This approach is generally called *static* in the literature because the measured parameters are invariable in time. It allows the determination of the first possible date of existence for a given composition of ink, paper, or toner found on a questioned document. Incoherencies and anachronisms can be highlighted, as was the case for the paper analysis of Hitler's diaries [67].

The developments mentioned above are quite important and relatively easy to measure, but they occur quite infrequently. Most evolutions in the composition of paper, ink, and toner are less easily

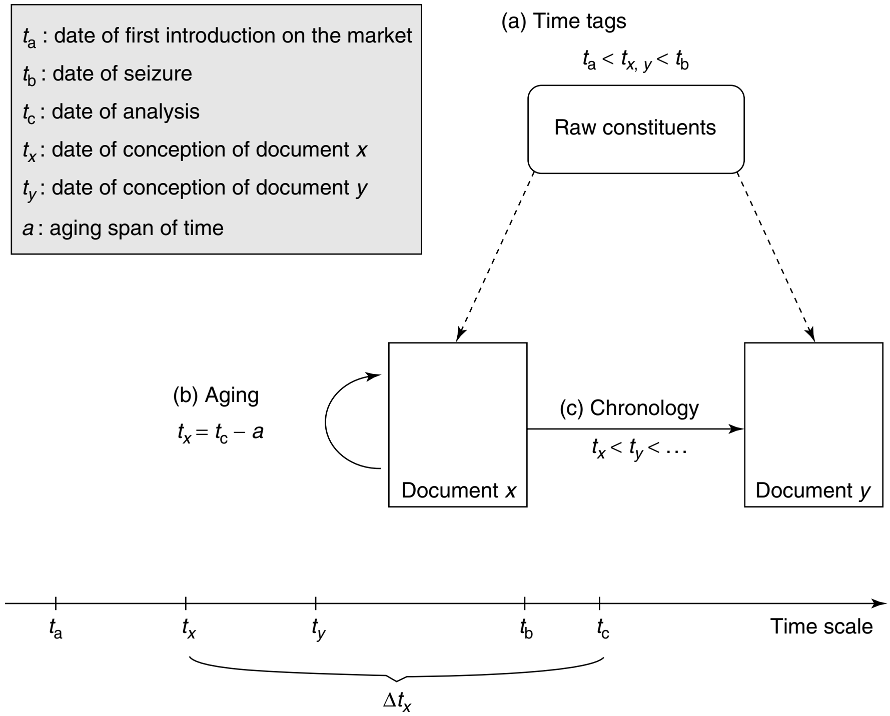

Figure 1 Age determination of questioned documents relying on (a) information yielded by time tags, (b) aging of raw constituents since document conception, and (c) reconstruction of the chronology of document conceptions. The date of first introduction of the market (t_a), the date of seizure (t_b), the date or analysis (t_c), and the aging time span (a) are known and/or yielded by the investigation. These pieces of information then help answer the question about the conception time of the documents (t_x and t_y)

detected and are closely guarded industrial secrets. To highlight the date of introduction on the market of minor characteristics, two conditions must be respected:

- First, the determination of the composition must be based on validated and reproducible analytical methods (*see* **Ink Analysis**) in order to ensure that the selected set of parameters does not alter as a function of time (i.e., their aging is negligible).
- Second, it is essential to have access to a comprehensive database [71–73] containing the selected parameters of all inks, toners, and paper introduced on the market. Ideally, this should cover a broad spatial and temporal range.

An alternative method is to introduce chosen markers in the composition that point at the time of introduction on the market. For example, the Bureau of Alcohol, Tobacco, and Firearms (ATF) started a yearly ink tagging program in collaboration with ink manufacturers in the mid-1970s [8, 41, 75]. The objective was to introduce a different tag each year that was easily analyzable. This requires close collaboration with ink manufacturers and a constant monitoring of the introduced tags (which should never repeat) and can generate a nonnegligible increase of the ink costs. Some special papers also enclose their date of production in the form of microimpressions or indirectly through specific watermarks [76]. Additionally, some laser printers mark the printed pages with latent yellow dots in the form of a code which includes the date and hour at which the document was printed [65, 66].

Aging

The second approach is based on measurements of the questioned documents' parameters that change as a function of time. Some older methods such as the migration of chloride and sulfate ions from the ink into the paper [9–12] can no longer be used, as most inks are now free of these ions. Earlier gallotannic inks were also acidic, and caused paper deterioration. Moreover, they contained iron that oxidized, provoking a change of color [1, 2, 13]. The introduction of new composition over the years also meant changes in the aging processes and dating methods.

Paper was studied to a lesser extent than ink [77, 78, **Paper Analysis**], because it starts aging directly after manufacture (or earlier when the tree was felled), and that can be well before the document's conception. Radiocarbon technique was proposed to date recent paper (less than 50 years old) using the strong atmospheric increase in ^{14}C concentration due to the nuclear weapon tests [78].

It is generally expected that ink does not age in the cartridge [79, 80], even though a recent study indicates that ink aging already starts at the tip of the cartridge [80]. Once the ink is applied to paper, it definitely starts to age: dyes fade [13, 15, 16, 19, 22, 81–86], solvents diffuse and evaporate [24–28, 35, 39, 72, 74, 75, 86–89], and resins polymerize [71, 72, 90, 91, 99]. Aging processes of ink follow complex pathways that are considerably influenced by a number of factors other than time, inducing acceleration or quenching of the aging. The influencing factors can be ordered in three main classes [39, 45, 64, 84–88]: (i) initial composition of the ink after transfer (in the cartridge), (ii) physical and chemical properties of the substrate (paper porosity and coatings, etc.), and (iii) storage conditions (temperature, light, air flux, humidity, contaminations etc.). In practice, no information on these factors is generally available. This is why the determination of the absolute age of an ink entry remains truly difficult and in most cases impossible. The more important objective is therefore the determination of a time range rather than a precise date. The considered scale can significantly vary depending on the measured parameters. Thus, while solvents disappear very quickly from the ink, degradation of dyes occurs more slowly over several years. Measured changes are reported as a function of time in order to establish an aging curve (Figure 2).

As fading of dyes is visible to the naked eye, methods based on the aging approach were developed to measure color degradation with time [34, 81, 83, 100]. Unfortunately for forensic scientists, dyes that are unstable in the presence of light do not degrade in the dark, or do so only very slowly [82, 84, 86]. Therefore, dating relying on dye degradation usually is carried out only by comparing ink entries from the same pen on the same paper and stored under the same conditions. Unlike reported in [100], the absence of difference in the degradation states of two ink entries does not necessarily mean that they are contemporaneous. In fact, the inks could also have

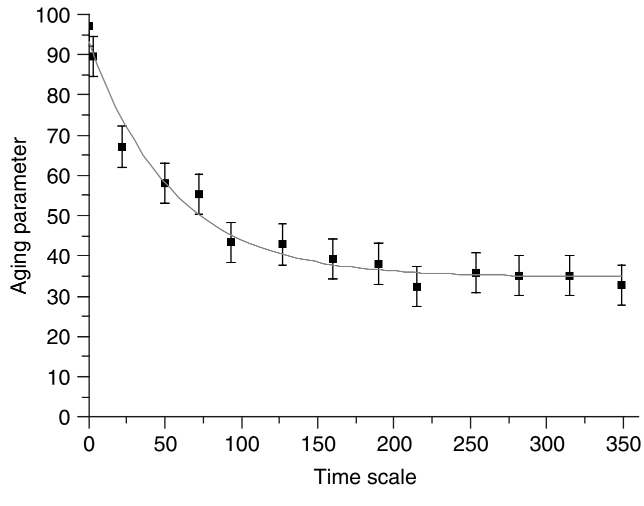

Figure 2 Typical aging curve (exponential decrease of first order). An aging parameter of the ink is plotted as a function of time (for example, in seconds, hours, days, or years) [Reproduced with permission from Polymedia Meichtry SA]

been stored for a significantly different number of years in the absence of light [45].

A second approach based on dye analysis focused on the observation that, when ink dries, it becomes harder to dissolve. This may be due to the polymerization of resins during the fastening of the ink on paper. Resins are high-molecular-weight molecules that are present in small quantities in the ink entries. They are therefore very difficult to analyze [71, 72, 90, 91]. An indirect way to evaluate their hardening thus derived from the measurements of ink extractability over time. Earlier, changes in the extractability of ink have been investigated by measuring the dissolution rates in acids [4]. Later, the sequential extraction of ink dyes was proposed [13, 15–22, 37, 44, 92–96] in order to eliminate the dependence of the extracted mass (i.e., difference in ink thickness resulting in differences in the mass obtained). Thus, two extractions were carried out consecutively: first in a "weak" solvent, and then in a "strong" solvent. Then the extractions were analyzed using thin-layer chromatography (TLC) to quantify dyes in order to calculate an extraction percentage (P). The extracted quantity in the "weak" solvent (M_1) is divided by the total amount extracted in the two solvents ($M_1 + M_2$) in order to obtain the following ratio:

$$P(\%) = \frac{100(M_1)}{(M_1 + M_2)} \quad (1)$$

These methods are based on the following hypothesis: a "weak" solvent extracts only the fraction of the ink that is still fresh, while a "strong" solvent extracts the totality of dyes in the ink entry. Thus, a large P means that the ink is still fresh, while a low P indicates an old ink.

Since then, many authors have shown that the measurements based on dye extraction were not reproducible [47, 48, 51–53, 58]. This is due to the fact that the quantity of dye extracted is dependent not only on the capacity of the solvents to extract them, but also on their initial quantity in the ink and their subsequent degradation. It was observed that each ink required a different extraction solvent, and their aging curves were significantly divergent.

Lately, the interest has shifted to the ink drying processes and solvent analysis [39]. This was first proposed in 1985 by Stewart [24], in which a decrease of the volatile components of ink was measured using gas chromatography/mass spectrometry (GC/MS). Sequential extractions and artificial aging were also proposed for further developments [16, 17]. This resulted in the development of several dating methods that can be classified into three distinct methodologies, all focusing mainly on the solvent phenoxyethanol, because it is the main solvent found in ballpoint pen ink formulation [45, 71, 72, 74, 75]:

- The first group of methods focuses on the diminution of volatile compounds over time [19, 24, 28, 57, 86–89, 97–99]. After a few months, their quantity will become generally very low as solvent evaporation happens promptly after ink application on the paper [57, 87]. This, as well as the limited sample availability, gave rise to reproducibility issues that were partly resolved by the use of a ratio between the solvent content and another stable compound in the ink [19, 21, 26, 57, 97–99]. Recent developments proposed a solvent-to-dye ratio through high-performance liquid chromatography (HPLC) analysis using two detectors [99] or two analyses using GC/MS for solvents and ultraviolet–visible (UV–vis) spectrometry for dyes [97, 98]. These methods have not been reported for application in practical casework yet.
- The second methodology is based on artificial aging [16, 17] and is sometimes referred to the *solvent loss ratio* method. Two ink entries are extracted in a solvent and analyzed using GC/MS. One specimen is analyzed directly (M) and the other is artificially aged at 70 °C during 2 h (M_T) before extraction. A portion of a hypothetical aging curve is then calculated as follows [25, 35]:

  $$R\% = \frac{M - M_T}{M}$$

  However, the reliability of artificial aging has been questioned [45, 46, 50, 86].
- The third methodology is based on sequential extraction and was first proposed by Aginsky [21]. Two extractions were carried out consecutively on the same ink specimen, first in a "weak" solvent (M_1), then in a "strong" solvent (M_2) and analyzed using GC/MS. Similar to dyes, a $P\%$ ratio was then calculated using equation (1). Bügler *et al.* [27] also proposed the sequential extraction of one ink sample using thermodesorption at two temperatures (90 and 200 °C). An alternative approach was studied

without much success using solid phase microextraction (SPME) followed by a liquid extraction in methanol [26]. Later, these approaches were combined to artificial or natural aging (i.e., analysis of several samples artificially or naturally aged) [25, 35, 39, 52]. Aginsky proposed thus the calculation of an additional ratio from equation (1):

$$D\% = P\% - P_T\%$$

Bügler *et al.* proposed the calculation of up to five samples within 3 months to detect a potential trend in the aging curve [[39]].

While some of these methods were applied in casework, several issues remain open and will be discussed below in the sections "Validation" and "Interpretation". The minimum requirements for ink dating analysis should be empirically reached before application in practice [39].

Chronology

This approach aims to determine the relative age of a document in comparison to others (i.e., to order them in chronological sequence). Different methodologies can be applied, such as the study of latent writings by oblique lighting or by an electrostatic detection apparatus (ESDA). If latent writings from a document are found on a second document, then the latter was physically placed under the first one during writing. This information may be useful in sequencing the order of writings.

The determination of the sequence of crossing lines has proved to be very useful in certain cases [29–33, 45, 101–106; **Intersecting Lines: Documents**]. It is, however, not always easy to determine optically which line is above the other. Therefore, chemical [45] and spatial techniques [30–33, 101, 103, 104, 106] were developed and have proved useful in some cases.

Finally, the comparison of ink aging states may help to reconstruct the sequence of apposition of ink entries on documents [15, 16, 18, 46, 50, 52]. This can only be applied for inks on the same type of paper stored under the same conditions (i.e., diaries). The general evolution of the aging curve must be known (Figure 3). If a diminution is expected, it is imperative to ensure that the aging parameters do not increase whatever the conditions.

Validation

The analytical development of dating methods (Table 1) requires a considerable amount of time and resources. It is therefore important not to underestimate the task of ensuring their scientific validity before implementing them in practice. This was confirmed by several court statements, where ink evidence was actually refused:

- at least on one occasion for the time tags approach (*United States of America v. Angelo Bruno et al.* in US District Court, E.D. Pennsylavania, 1971);
- at least on two occasions for sequential analysis of ink dyes (*Regina v. Michael Gurmann* in

Table 1 Summary of document dating possibilities: relevant application time frame and main limitations of dating methods. All methods must be validated before their implementation

Approach	Time variable	Time frame	Main limitations
1. Time tags	Fluorescent tags	1970s – end of project	Limited number of inks with tags
	Laser printer latent code	Not defined	Limited number of printer with this function
	New industrial developments	Precise printing date	Secret of fabrication
2. Aging	Dyes	Months	Unreliable
	Solvents	Weeks	Possible only for a limited number of inks
	Resins	Years	No data
	Paper	Years	Limited to historical document
3. Chronology	Dyes	Months	Same ink and support (e.g., diary)
	Solvents	Weeks	Same ink and support (e.g., diary)
	Line crossings	Years	Aging of the crossings
	Latent writings	Years	Their absence gives no information

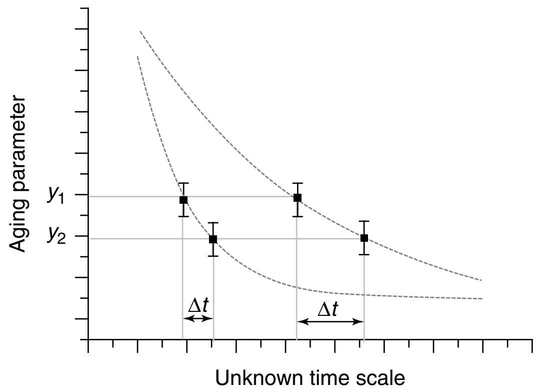

Figure 3 If the aging curve is unknown (dots), the values obtained from two ink entries can be ordered in chronological order at the imperative condition that the general evolution of the curve is known (increase or decrease). Storage conditions, paper type, and ink composition must be the same. Measurement error must be taken into account [Reproduced with permission from Polymedia Meichtry SA]

Ontario, Canada, 1993; Learning Curve Toys, L.P. v. Playwood Toys, Inc. 2000 U.S. Dist. Lexis 5130); and

- at least on one occasion for the solvent analysis (Civil file 5810-08-07 *AmironCTL Financing & Investments v. Wallach*, Central District Court, PetachTikva, Israel (Judge Esther Stemmer), 2007 – verdict given on March 05, 2012).

In the latest case, the judge stated (translation by Prof. Joseph Almog):

> Our matter belongs, apparently, to the scientific domain and (. . .) it seems that the technique (based on solvent analysis) does not meet all the criteria which are essential for the acceptance of a scientific theory. (. . .) Thus, I will only say that at this stage and under the circumstances, I cannot say that "Aginsky's technique" (reported in [25, 35]) is a reliable scientific method for ink dating.

This shows the importance of insuring the scientific validity, not only of the analytical methods but also considering realistic ink specimen (such as may appear in an actual casework) and applying an appropriate interpretation model in a court perspective (see the section below).

Some important aspects of analytical reliability were enounced by Horwitz [111]: reproducibility (between laboratories precision), repeatability (within laboratory precision), systematic error or bias (accuracy), selectivity, and limit of reliability. The robustness of the method is also an important factor.

It is of concern that errors are very rarely mentioned in the literature and are generally not represented in the figures. It is, however, essential to make certain that predicted differences provoked by aging are in fact higher than the measurement errors. Further, the available ink in practical cases is generally not sufficient to repeat analysis several times in order to obtain a mean and a standard deviation [39, 61, 112]. Developments of dating methods are carried out with known ink samples prepared and stored in controlled conditions, and blind testing on realistic samples is therefore imperative. When small quantities are analyzed, such as solvents in ink entries, the detection and quantification limits play an important role in determining the threshold at which the method is not applicable anymore. The most demanding aspect is interlaboratory validation. In fact, to date, most dating methods are used only by one single laboratory. In order to apply dating in casework, the forensic experts should deploy their efforts most particularly in adequate validation of their method [39, 49, 51, 54, 61].

Interpretation

Interpretation of evidence, like in other forensic fields, plays an essential role in the dating of ink and should already be included in method developments. In fact, the question about the age of an ink entry lies perhaps more on the inference of sources rather than on the technological or analytical aspects. Thus, it is important to consider all possible sources (alternative hypotheses) to allow for a balanced interpretation of the evidence [39, 61].

Until now, the interpretation of ink dating results was mainly based on threshold decision. It was observed that ink aging curves level off after some time (see Figure 2) and two options were thus considered:

1. One value is obtained from the questioned ink (e.g., the quantity of phenoxyethanol or a calculated ratio *P*%) [25, 27, 35].
2. Several values are obtained from the aging curves and a second ratio (e.g. *R*%, *D*% [25, 35, 110]) or a trend test was applied (e.g., Neuman test on five to seven values [39]).

In both cases, if the obtained value (e.g., $P\%$, $R\%$, $D\%$, or trend test value) is above a previously defined threshold, the ink is considered fresh (e.g., less than 6 months old). However, if the value is below the threshold, no conclusion is given.

The main limitation of the threshold approach lies in the fact that measurement errors are not considered in the final decision. While the thresholds are defined on a selected population of ink in order to yield no false positive decision (i.e., conclusion that the ink was fresh when it was actually not), it was never (or rarely) tested on realistic samples (for example, on signatures instead of straight lines). One can then expect a significant error rate in the decision-making process. Moreover, the yielded information remains limited to the threshold decision (e.g., the questioned ink is less than 6 months) independently from the casework.

A probabilistic model would actually have the advantages of taking into account the hypotheses of justice as well as the rate of false occurrence. It would not be limited to a time frame (different set of hypotheses can be tested) and may additionally take into account influencing factors. A Bayesian statistical framework was thus proposed to assist in reaching an opinion regarding the date of a document [39, 61]. In this case, when one has to look at the probability of the evidence (E) given the ink entry has been made at a time t_1 (hypothesis of the plaintiff) compared to the probability of this same evidence given the ink entry has been written at a different time t_2 (hypothesis of defense). Then the prior odds of the hypotheses of the plaintiff t_1 and the defense t_2 existing before observation of the evidence E are multiplied by a factor called the *likelihood ratio* (LR) to obtain the posterior odds that account for the new evidence E. The LR is an indication on the strength of the evidence in supporting one of the hypotheses in Bayesian logic [61, 107–109]. It is defined by the probability of E given t_1 is true, divided by the probability of E given t_2 is true:

$$\mathrm{LR} = \frac{P(E|t_1)}{P(E|t_2)}$$

A regression model was proposed to interpret dating evidence [39, 108]. Accepting that the analytical results E obtained for the questioned ink entry (e.g., $R\%$, $P\%$, or $D\%$ values) for a given time t_n are normally distributed, the LR can be calculated as follows:

$$\mathrm{LR} = \frac{P(E|\mu_{t_1}, \sigma^2_{t_1})}{P(E|\mu_{t_2}, \sigma^2_{t_2})}$$

where μ is the mean and σ^2 is the standard deviation of the evidence E. The density probability for a given value of E is generally given by the following function [[39,108,109]]:

$$f(E \left| \mu, \sigma^2 \right.) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left[-\frac{(d-\mu^2)}{2\sigma^2}\right]$$

However, the data necessary to apply this model is not available from the literature yet, and further efforts should be made to allow adequate interpretation of ink dating evidence.

For example, when comparing two ink entries to determine their relative age, one has to evaluate the probability of obtaining the observed results, if the entries have the same age, and the probability of obtaining these same results, if the entries do not have the same age.

If the dye compositions of the two ink entries were determined and found to be nondifferentiable, at least three explanations should be taken into account. In fact, the analysis of two ink entries having the same age and the same initial composition, paper, and storage conditions would logically lead to nondifferentiable results. However, two inks having different ages but the same initial composition, on the same paper and stored in the dark, would also lead to nondifferentiable results, because dye fading occurs very slowly in the absence of light [82, 85]. Finally, two inks having different compositions and/or stored in different conditions did sometimes also yield nondifferentiable results [45]. The probability of each of these alternative actually has to be estimated in order to reach a correct conclusion.

Conclusion

From its beginning, the field of questioned documents has been concerned with dating. Proposed methods usually lean upon complex processes, and controversy among the scientific community is still high. Every document dating method whose objective is to be applied in forensic caseworks must fulfill validation requirements. Moreover, source inference must also be taken into account in the interpretation of the dating evidence. To date, most methods still fail to be adequately validated, and should be applied with extreme caution. The limitations of the methods used must be adequately disclosed and documented.

Acknowledgments

The author would like to thank all those who helped her achieve this work, and especially Prof. Bernhard Spengler of the University of Giessen, Dr. Dieter Kirsch of the Bundeskriminalamt in Wiesbaden, and Prof. Pierre Margot of the University of Lausanne. She also wishes to sincerely thank her PhD students Agnès Koenig and Matteo Gallidabino who indirectly contributed to this work through their research on ink dating and interpretation. Finally, she wishes to thank the Swiss National Foundation for supporting her dating researches in forensic science (Fund No. PP00P1_123358).

References

[1] Osborn, A.S. (1910). *Questioned Documents*, The Lawyers' Co-operative Publishing Company, Rochester, NY.

[2] Brewster, F. (1932). *Contested Documents and Forgeries*, The Book Company, Calcutta.

[3] Grant, J. (1937). *Books and Documents – Dating, Permanence and Preservation*, Grafton & Company, London.

[4] Locard, E. (1959). *Les faux en écriture et leur expertise*, Bibliothèque Scientifique, Paris.

[5] Harrison, W.R. (1966). *Suspect Documents – Their Scientific Examination*, Sweet & Maxwell, London.

[6] Brunelle, R.L. & Reed, R.W. (1984). *Forensic Examination of Ink and Paper*, Charles C. Thomas Publisher, Springfield.

[7] Ellen, D. (1997). *The Scientific Examination of Documents – Methods and Techniques*, *Forensic Science Series*, Taylor & Francis, Salisbury.

[8] Levinson, J. (2001). *Questioned Documents – A Lawyer's Handbook*, Academic Press, London, CA.

[9] Metzger, D., Rall, D. & Heess, W. (1933). Ein neues Verfahren, Identität und Alter von Tintenschriften festzustellen, *Archiv für Kriminologie* **92**, 105–113.

[10] Türkel, S. (1933). Eine Schnellmethode zur ersten Orientierung welche von zwei Tintenspuren auf einem Papiere jüngeren oder älteren Datums ist, *Archiv für Kriminologie* **92**, 224–226.

[11] Heess, W. (1935). Ein neues Verfahren, Identität und Alter von Tintenschriften festzustellen-2. Mitteilung, *Archiv für Kriminologie* **96**, 13–17.

[12] Heess, W. (1937). Ein neues Verfahren, Identität und Alter von Tintenschriften festzustellen-3. Mitteilung, *Archiv für Kriminologie* **101**, 7–37.

[13] Sen, N.K. & Ghosh, P.C. (1971). Dating iron-base ink writings on documents, *Journal of Forensic Sciences* **16**(4), 511–520.

[14] Frei-Sulzer, M. (1971). Messungen zeitlicher Veränderungen der Schreibrillentiefe, *Kriminalistik – Ein Hilfsmittel bei der Alterfeststellung von Urkunden* **7**, 348–349.

[15] Brunelle, R.L., Breedlove, C.H. & Midkiff, C.R. (1987). Determining the relative age of ballpoint inks using a single-solvent extraction technique, *Journal of Forensic Sciences* **32**(6), 1511–1521.

[16] Cantu, A.A. & Prough, R.S. (1987). On the relative aging of ink – the solvent extraction technique, *Journal of Forensic Sciences* **32**(5), 1151–1174.

[17] Cantu, A.A. (1988). Comments on the accelerated aging of ink, *Journal of Forensic Sciences* **33**(3), 744–750.

[18] Brunelle, R.L. & Lee, H. (1989). Determining the relative age of ballpoint ink using a single-solvent extraction, mass-independent approach, *Journal of Forensic Sciences* **34**(5), 1166–1182.

[19] Aginsky, V.N. (1993). Some new ideas for dating ballpoint inks – a feasibility study, *Journal of Forensic Sciences* **38**, 1134–1150.

[20] Aginsky, V.N. (1994). A new version of the internal standard method in quantitative thin layer chromatography, *Journal of Planar Chromatography* **7**, 309–314.

[21] Aginsky, V.N. (1994). Determination of the age of ballpoint pen ink by gas and densitometric thin-layer chromatography, *Journal of Chromatography A* **678**, 119–125.

[22] Aginsky, V.N. (1995). A microspectrophotometric method for dating ballpoint inks – a feasibility study, *Journal of Forensic Sciences* **40**(3), 475–478.

[23] Brunelle, R.L. & Speckin, E.J. (1998). Technical report with case studies on the accelerated aging of ballpoint inks, *International Journal of Forensic Document Examiners* **4**(3), 240–254.

[24] Stewart, L.F. (1985). Ballpoint ink age determination by volatile component comparison – a preliminary study, *Journal of Forensic Sciences* **30**(2), 405–411.

[25] Aginsky, V.N. (1996). Dating and characterizing writing, stamp pad and jet printer inks by gas chromatography/mass spectrometry, *International Journal of Forensic Document Examiners* **2**(2), 103–116.

[26] Andrasko, J. (2003). A simple method for distinguishing between fresh and old ballpoint pen ink entries (Abstract), *Forensic Science International* **136**(1), 80–81.

[27] Bügler, J.H., Buchner, H. & Dallmayer, A. (2008). Age determination of ballpoint pen ink by thermal desorption and gas chromatography-mass spectrometry, *Journal of Forensic Sciences* **53**(4), 982–988.

[28] Brazeau, L. & Gaudreau, M. (2007). Ballpoint pen inks: the quantitative analysis of ink solvents on paper by solid-phase microextraction, *Journal of Forensic Sciences* **52**(1), 209–215.

[29] Poulin, G. (1996). Establishing the sequence of strokes: the state of the art, *International Journal of the Forensic Document Examiners* **2**, 16–32.

[30] Kasas, S., Khanmy-Vital, A. & Dietler, G. (2001). Examination of line crossings by atomic force microscopy, *Forensic Science International* **119**, 290–298.

[31] Berx, V. & De Kinder,J. (2002). A 3D view on the 'crossing lines' problem in document investigation, in *Investigative Image Processing II, Crossing lines problem in document investigation, in Investitive Image Processing II, Proceedings of SPIE*, Z.J. Geradts & L.I. Rudin, eds, Orlando, FL (July 19 2009) Vol. 4709 pp. 102–110. DOI:10.1117/12.4747.

[32] Berx, V. & De Kinder, J. (2003). Line crossings and laser profilometry (Abstract), *Forensic Science International* **136**(1), 81.

[33] Schirripa Spagnolo, G. (2006). Potentiality of 3D laser profilometry to determine the sequence of homogenous crossing lines on questioned documents, *Forensic Science International* **164**(2–3), 102–109.

[34] Grim, D.M., Siegel, J. & Allison, J. (2002). Evaluation of laser desorption mass spectrometric and uv accelerated aging dyes on paper as tools for the evaluation of a questioned document, *Journal of Forensic Sciences* **47**(6), 1265–1273.

[35] Gaudreau, M. & Brazeau, L. (2002). Ink dating using a solvent loss ratio method, *60th Annual Conference of the American Society of Questioned Document Examiners*, San Diego, CA.

[36] Brunelle, R.L. & Cantu, A.A. (1987). A critical evaluation of current ink dating techniques, *Journal of Forensic Sciences* **32**(6), 1522–1536.

[37] Stewart, L.F. & Guertin, S.L. (1991). Current status of ink age determination, *International Criminal Police Review* **429**, 9–12.

[38] Brunelle, R.L. (1992). Ink dating – the state of the art, *Journal of Forensic Sciences* **37**(1), 113–114.

[39] Weyermann, C., Almog, J., Buegler, J. & Cantu, A.A. (2011). Minimum requirements for application of ink dating methods based on solvent analysis in casework, *Forensic Science International* **210**(1–3), 52–62.

[40] Stewart, L.F., Becker, J.L. & Fitzgerald, M. (1995). Multi-agency response to the advancement of ink age determination, Brief Communication. 47th AAFS Meeting, Seattle.

[41] Cantu, A.A. (1995). A sketch of analytical methods for document dating. Part I. The static approach: determining age independent analytical profiles, *International Journal of Forensic Documents Examiners* **1**(1), 40–51.

[42] Cantu, A.A. (1996). A sketch of analytical methods for document dating. Part II. The dynamic approach: determining age dependent analytical profiles, *International Journal of Forensic Documents Examiners* **2**(3), 192–208.

[43] Ezcurra, M., Góngora, J.M.G., Maguregui, I. & Alonso, R. (2010). Analytical methods for dating modern writing instrument inks on paper, *Forensic Science International* **197**(1–3), 1–20.

[44] Brunelle, R.L. & Crawford, K.R. (2003). *Advances in the Forensic Analysis and Dating of Writing Ink*, Charles C. Thomas Publisher, Springfield.

[45] Weyermann, C. (2005) Mass spectrometric investigation of the aging processes of ballpoint ink for the examination of questioned documents, PhD Thesis, Justus-Liebig University, Giessen, at http://geb.uni-giessen.de/geb/volltexte/2006/3044/, (last access May 2012).

[46] Lyter, A.H. (1994). Relative aging of ball pen ink: natural aging vs. artificial aging, *American Academy of Forensic Sciences Meeting*, San Antonio.

[47] Starrs, J.E. (1994). Forensic science in law enforcement – ink analysis, *The Scientific Sleuthing Newsletter* **18**(4), 11.

[48] Hicks Champod, T., Khanmy, A. & Margot, P. (1995). Ink aging: perspectives on standardization, in *Proceedings of the 13th Meeting of the International Association of Forensic Sciences, Düsseldorf, 1993, Advances in Forensic Sciences 3, Forensic Criminalistics 1*, B. Jacob, W. Bonte, W. Huckenbeck & P. Pieper, eds, Verlag Dr. Köster, Berlin, pp. 304–309.

[49] Margot, P., Hicks Champod, T. & Khanmy, A. (1995). Ink dating up-dated and up-ended – correspondence of Prof.Starrs, *International Journal of Forensic Document Examiners* **1**(1), 2–5.

[50] Stewart, L.F. & Fortunato, S.L. (1996). Distinguishing between relative ink age determinations and the accelerated aging techniques, *International Journal of Forensic Document Examiners* **2**(1), 10–15.

[51] Aginsky, V.N. (1996). Accelerated aging – its use in methods for dating ink, *International Journal of Forensic Document Examiners* **2**(3), 179–181.

[52] Aginsky, V.N. (1998). Measuring ink extractability as a function of age – why the relative aging approach is unreliable and why it is more correct to measure ink volatile components than dyes, *International Journal of Forensic Document Examiners* **4**(3), 214–230.

[53] Andermann, T. & Neri, R. (1998). Solvent extraction techniques – possibilities for dating ball point pen inks, *Journal of Forensic Sciences* **4**(3), 231–239.

[54] Starrs, J.E. (2000). Review: the many use of the forensic sciences – document examination, *The Scientific Sleuthing Newsletter* **24**(1), 12–17.

[55] Fortini, A. (2000). *Tester la reproductibilité des mesures de l'une des méthodes de datation des encres d'Aginsky (GC/MS)*, *Séminaire de 4ème Année*, Institut de Police Scientifique et de Criminologie, Université de Lausanne, Lausanne, Switzerland.

[56] Weyermann, C., Kirsch, D., Costa-Vera, C. & Spengler, B. (2003). Dating of ink entries by MALDI/LDI-MS and GC/MS analysis: reality or utopia? (Abstract), *Forensic Science International* **136**(1), 71–72.

[57] Lociciro, S., Mazzella, W., Dujourdy, L., Lock, E. & Margot, P. (2004). Dynamic of the aging of ballpoint pen ink, *Science & Justice* **44**(3), 165–171.

[58] Jahns, K. (2004). Altersbestimmung von Schreibmittln durch chemische Analysverfahren, in *Mannheimer Hefte Für Schriftvergleichung*, P.E. Baier, ed, Universität Mannheim, Schmidt/Römhild, p. 29, Jahrgang.

[59] White, P. (2004). The age of an ink: an age old problem, Oral Presentation at the 17th International Symposium on Forensic Science, ANZFSS, Wellington.

[60] Schirripa Spagnolo, G., Zavattaro, D., De Santis, M. & Gennaro Ferrillo, L. (2007). Max Frei theory revisitation: does really strokes depth change along time? *Forensic Science International* **167**, 163–166.

[61] Weyermann, C., Schiffer, B. & Margot, P. (2007). A logical approach to ballpoint ink dating, *Science & Justice* available online. DOI: 10.1016/j.scijus.2007.10.009.

[62] Weyermann, C. (2008). Revue des méthodes de datation de l'encre: est-il possible de déterminer l'*â*ge d'un document en analysant l'encre? *Revue Internationale de Criminologie et de Police Technique et Scientifique* **3**(LXI), 359–376.

[63] Weyermann, C., Gueissaz, L., Lefeuvre, A., Curchod, J., Milliet, Q. & Voisard, R. (2009). Reconstruire, *Revue internationale de criminologie et de police technique et scientifique* **LXII**(2), 37–48.

[64] Weyermann, C. & Ribaux, O. (2012). Situating forensic traces in time, *Science & Justice* **52**(2), 68–75.

[65] EzcurraGondra, M. & Gravalos, G.R. (2012). Sistema de protección contra la falsificación en fotocopiadoras e impresoraslásercolor, *Análisisforense de documentos – Sistemas de impresión y sus tintas*, 1st Edition, Ediciones La Rocca, Buenos Aires, Vol. 2, Chapter 5.

[66] Khamsi, R. (2005) *Laser Printer Dot Code Revealed: Nature News*, at http://www.nature.com/news/2005/051019/full/news051017-9.html (last accessed July 2012).

[67] Grant, J. (1985). The diaries of Adolf Hitler, *Journal of the Forensic Science Society* **25**, 189.

[68] Hilton, O. (1957). Characteristics of the ball point pen and its influence on handwriting identification, *Journal of Criminal Law Criminology and Police Science* **47**, 606–613.

[69] Hung, P.S. & Leung, S.C. (1995). Some observations on the morphology of a ballpoint pen stroke, *International Journal of Forensic Document Examiners* **1**, 18–31.

[70] Mazzella, W.D. & Buzzini, P. (2005). Raman spectroscopy of blue gel pen inks, *Forensic Science International* **152**(2–3), 241–247.

[71] Bügler, J., Buchner, H. & Dallmayer, A. (2005). Differenzierung von Kugelschreiberpasten durch Thermodesorption und Gaschromatographie – Massenspektrometrie, *Archiv für Kriminologie* **214**, 141–148.

[72] Bügler, J., Buchner, H. & Dallmayer, A. (2005). Characterization of ballpoint pen inks by thermal desorption and gas chromatography-mass spectrometry, *Journal of Forensic Sciences* **50**(5), 1–6.

[73] Neumann, C., Ramotowski, R. & Genessay, T. (2011). Forensic examination of ink by high-performance thin layer chromatography–the United States Secret Service Digital Ink Library, *Journal of Chromatography A* **1218**(19), 2793–2811.

[74] Cantú, A.A. (2012). A study of the evaporation of a solvent from a solution – application to writing ink aging, *Forensic Science International* **219**(1–3), 119–128.

[75] LaPorte, G.M., Wilson, J.D., Cantu, A.A., Mancke, S.A. & Fortunato, S.L. (2004). The identification of 2-phenoxyethanol in ballpoint inks using gas chromatography/mass spectrometry – relevance to ink dating, *Journal of Forensic Sciences* **49**, 155–159.

[76] Caywood, D.A. (1995). Watermarks and the questioned document examiner – an historical retrospective on watermarks and document examination, *International Journal of Forensic Document Examiners* **4**(1), 399–304.

[77] Stewart, L.F. (1982). Artificial aging of documents, *Journal of Forensic Sciences* **27**(2), 450–453.

[78] Zavattaro, D., Quarta, G., D'Elia, M. & Calcagnile, L. (2007). Recent documents dating: an approach using radiocarbon techniques, *Forensic Science International* **167**(2–3), 160–162.

[79] Grim, D.M., Siegel, J. & Allison, J. (2002). Does ink age inside of a pen cartridge, *Journal of Forensic Sciences* **47**, 1294–1273.

[80] Andrasko, J. & Kunicki, M. (2005). Inhomogeneity and aging of ballpoint pen inks inside of pen cartridges, *Journal of Forensic Science* **50**(3), 542–547.

[81] Andrasko, J. (2001). HPLC analysis of ballpoint pen inks stored at different light conditions, *Journal of Forensic Sciences* **46**(1), 21–30.

[82] Andrasko, J. (2001). Changes in composition of ballpoint pen inks on aging in darkness, *Journal of Forensic Sciences* **47**(2), 324–327.

[83] Grim, D.M., Siegel, J. & Allison, J. (2001). Evaluation of desorption/ionization mass spectrometric methods in the forensic applications of the analysis of inks on paper, *Journal of Forensic Sciences* **46**(6), 1411–1420.

[84] Weyermann, C., Kirsch, D., Costa Vera, C. & Spengler, B. (2006). Forensic investigation of ageing and degradation of ballpoint dyes on paper by LDI- and MALDI-MS analysis, *Journal of the American Society for Mass Spectrometry* **17**, 297–306.

[85] Weyermann, C., Kirsch, D., Costa Vera, C. & Spengler, B. (2009). Evaluation of the photodegradation of crystal violet upon light exposure by mass spectrometric and spectroscopic methods, *Journal of Forensic Sciences* **54**(2), 339–345.

[86] Weyermann, C. & Spengler, B. (2008). The potential of artificial aging for modelling of natural aging processes of ballpoint ink, *Forensic Science International* **180**, 21–31.

[87] Weyermann, C., Kirsch, D., Costa Vera, C. & Spengler, B. (2007). A study of the drying of ballpoint pen ink on paper by GC/MS, *Forensic Science International* **168**(2–3), 119–127.
[88] EzcurraGongora, M., Góngora, J.M.G., Maguregui, M. & Alonso, R.M. (2010). Evaluation of loss of phenoxyethanol from a ball point pen ink with time by GC-MS depending on the location of the signature on the document, *Journal of American Society of Questioned Document Examiners* **13**, 9–14.
[89] EzcurraGongora, M., Góngora, J.M.G., Maguregui, M. & Alonso, R.M. (2009). Analysis of ball point pens BIC cristal medium inks using gas-retention time locking chromatography, thin layer chromatography and liquid chromatography tandem mass spectrometry, *Journal of American Society of Questioned Document Examiners* **12**(2), 57–68.
[90] Kirsch, D., Weyermann, C., Koehler, F. & Spengler, B. (2005). Mass spectrometric investigation of the aging of ballpoint inks, *53rd ASMS Conference*, American Society for Mass Spectrometry, San Antonio, TX.
[91] Kirsch, D., Koehler, F. & Spengler, B. (2006). Mass spectrometric investigation of resins in ballpoint inks, *4th European Association of Forensic Science Conference*, Helsinki, Finland.
[92] Mitchell, A.C. (1920). Estimation of the age of ink in writing, *The Analyst* **XLV**(435), 246–258 (reprinted in the Journal of Forensic Document Examiners 1991 (1991) (1995) 1956–1962).
[93] Kikuchi, Y. (1959). Estimation of the age of the writing in blue black ink (I), *Japanese Police Science Laboratory Report* **12**(3), 379–386.
[94] Kikuchi, Y. (1960). Studies on the age of iron–gallotannate ink writing (II) the chromatic study of ink stain, *Journal of Criminology* **26**(2), 39–59.
[95] Kikuchi, Y. (1963). Estimation of age of blue black ink writing (III), *Japanese Police Science Report* **16**(1), 83–86.
[96] Brunelle, R.L. (1995). A sequential multiple approach to determining the relative age of writing inks, *International Journal of Forensic Document Examiners* **1**(2), 94–98.
[97] Wang, Y., Yao, L., Zhao, P., Wang, J. & Wang, Y. (2005). Determination of the writing age of blue ballpoint ink by gas chromatography, *Chinese Journal of Chromatography* **23**, 202–204.
[98] Wang, Y., Yao, L., Zhao, P., Wang, J. & Wang, Y. (2006). Determining the relative age of blue ballpoint ink by gas chromatography, *Frontiers of Chemistry in China* **1**, 223–226.
[99] Kirsch, D., Guillou, V., Anheier, B., Seiler, P. & Köhler, F. (2010). Different analytical methods of dating ball point ink entries, *ENFSI, 6th European Document Examiners Working Group (EDEWG) Conference*, Dubrovnik, Croatia.
[100] Hofer, R. (2004). Dating of ballpoint pen ink – case report, *Journal of Forensic Sciences* **49**(6), 1–5.
[101] Gal, T., Sandor, J. & Karoly, A. (2007). Determining the sequence of crossed lines by FT-IR-ATR-microscopy, *Global Forensic Science Today* **1**, 8–11.
[102] Aginsky, V.N. (2002). Determining the sequence of non-intersecting media on documents: ball point pen ink and laser toner entries, *Journal of the American Society of Questioned Document Examiners* **5**(1), 1–3.
[103] Redjah, H., Mazzella, W. & Margot, P. (2010). Détermination de la séquence chronologique entre les impressions électrophotographiques (imprimantes laser et photocopieurs) et les traits de stylos à bille, sans croisement. *Revue internationale de criminologie et de police technique et scientifique* **LXIII**(3), 379–376.
[104] Magdalena EzcurraGondra (2008). Is atomic force microscope a suitable tool for studying crossing lines? *Journal of the American Society of Questioned Document Examiners* **11**(2), 41–49.
[105] Montani, I., Mazzella, W., Guichard, M. & Marquis, R. (2012). Examination of heterogeneous crossing sequences between toner and rollerball pen-strokes by digital microscopy and 3-D laser profilometry, *Journal of Forensic Science*, **57**(4), 997–1002.
[106] Bojko, K., Roux, C. & Reedy, B.J. (2008). An examination of the sequence of intersecting lines using attenuated total reflectance – fourier transform infrared spectral imaging. *Journal of Forensic Sciences* **53**(6), 1458–1467.
[107] Evett, I.W. (1987). Bayesian inference and forensic science: problems and perspectives, *The Statistician* **36**, 99–105.
[108] Gallidabino, M., Weyermann, C., Romolo, F. & Taroni, F., Estimating the time since discharge of spent cartridges: a logical approach for interpreting the evidence, *Science & Justice* available on http://dx.doi.org/10.1016/j.scijus.2011.12.004 17 January 2012.
[109] Biederman, A., Taroni, F. (2006). A probabilistic approach to the joint evaluation of firearm evidence and gunshot residues, *Forensic Science International* **163**, 18–33.
[110] Aginsky, V.N. (2002) Current methods for dating ink on documents, *60th Annual Conference of the American Society of Questioned Document Examiners*, San Diego, CA, at http://www.rileywelch.com/ink.htlm (accessed 2005).
[111] Horwitz, W. (1982). Evaluation of analytical methods for regulation of foods and drugs, *Analytical Chemistry* **52**, 67A–76A.
[112] Weyermann, C., Mazzella, W., Margot, P., Commentary on: Berger-Karin, C., Hendricks, U. & Geyer-Lippmann, J. (2009). Comparison of natural and artificial aging of ballpoint inks, *Journal of Forensic Sciences* **53**(4), 967.

CÉLINE WEYERMANN