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Interpol review of glass and paint evidence 2016-2019

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ABSTRACT

This review paper covers the forensic-relevant literature in paint and glass evidence from 2016 to 2019 as a part of the 19th Interpol International Forensic Science Managers Symposium. The review papers are also available at the Interpol website at: https://www.interpol.int/content/download/14458/file/Interpol%20Review%20Papers%202019.pdf.

1. Introduction

This review chapter covers advances in forensic applications of scientific methods for the examination of paint and glass evidence since the publication of the 18th International Forensic Science Symposium in October of 2016. This chapter covers a review on both of the subjects (paint and glass) using the peer-reviewed literature, published reports, books and book chapters on the subjects as well as highlights of presentations and proceedings from forensic science meetings and symposia published between 2016 and 2019. Forensic examiners should also be aware of the publication of standard practices, guides and test methods (ASTM) as well as the developments within the manufacturing industries including production volumes, production locations, and the current trends in the manufacture of these widely used materials.

2. Overview

The main forensic science journals reviewed for this chapter were the *Journal of Forensic Sciences, Forensic Science International, Science and Justice, the Canadian Journal of Forensic Sciences, the Australian Journal of Forensic Sciences, the Journal of the American Society for Trace Evidence Examiners (ASTEE), the European Paint and Glass (EPG) working group newsletter* and a new Elsevier journal initiated in 2016, *Forensic Chemistry*. In addition, more than fifteen (15) different analytical chemistry or other science journals have published peer-reviewed communications on the advances of forensic paint and glass examinations. The proceedings from several forensic and analytical chemistry conferences are briefly cited here and links to World Wide Web links and resources are also provided.

2.1. Peer-reviewed literature

For this reporting period, manuscripts related to forensic analysis and interpretation of paint evidence were published in a vast variety of peer-reviewed scientific journals. Research and case studies were disseminated in chemistry, physics, analytical, and forensic journals, including: 1) the *Journal of the American Society for Trace Evidence Examiners* (ASTEE), 2) *Talanta*, 3) *Forensic Science International*, 4) *Forensic Chemistry*, 5) the *Journal of Forensic Sciences*, 6) *Applied Spectroscopy*, 7) *Spectroscopy Letters*, 8) *Vibrational Spectroscopy*, 9) *Environmental Forensics*, 10) *Analytical Methods*, 11) *Journal of Raman Spectroscopy*, 12) *Canadian Society of Forensic Science Journal*, 13) *Analytical Chimica Acta*, 14) *Australian Journal of Forensic Science*, 15) *Pigments and Resins Technology*, 16) *Physical Engineering Science*, 17) *Journal of Physical Chemistry*, 18) *Microchemical Journal*, 19) *Journal of Analytical Atomic Spectroscopy* and 20) *Analytical Chemistry*.

2.2. Additional publications

Several books include book chapters devoted to the forensic examination of glass and paint evidence. Of particular interest is the volume published in 2016 and edited by Jay Siegel, *Forensic*

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Chemistry; Fundamentals and Applications, previously reported in the 2016 INTERPOL review. Additional references to standard methods, books and book chapters are provided within each of the paint and glass sections below.

2.3. Conferences/Symposiums

A list of scientific conferences and symposia devoted to the forensic sciences or that included sessions in the forensic examination or interpretation of paint and glass evidence are provided below. The conferences are listed in alphabetical order and include the name, year(s) it was held, and a brief description of the presentations pertaining to paint and glass. A link to the proceedings for the conference are provided below.

- American Academy of Forensic Sciences (2016–2019). Numerous workshops, poster presentations, and oral presentations at AAFS' annual meetings were on the topic of glass analysis. The link to each year's proceedings is as follows: http://www.aafs.org/resources/proceedings/.
- American Chemical Society (2017–2019). LA-ICP-MS, nuclear glass melt for forensic analysis, and use of likelihood ratios in forensics were all presented. The link to the abstracts is as follows: https://www.acs.org/content/acs/en/meetings/national-meeting/about/meetings-archive.html
- American Society of Crime Laboratory Directors Symposium (2019). Poster presentation entitled "The costs of NOT conducting trace evidence analyses in your forensic laboratory", https://www.ascld.org/ascld-annual-symposium/
- Annual IFRI Forensic Science Symposium (2017–2019) (2019 not available) https://ifri.fiu.edu/news-and-events/past-events/index.html
- Forensics @NIST Symposium (2018). Oral presentations on trace evidence and interpretation that were also broadcasted and available at: https://www.nist.gov/news-events/events/2018/11/forensics-nist-2018
- International Forensic Science Symposium (2016). Interpol hosts a Forensics Symposium every 3 years. The 18th International Forensic Science Symposium in 2016 included a session focusing on glass and paint.https://www.interpol.int/en/Ho=w-we-work/Forensics/Forensic-Symposium
- National Institute of Justice, Forensic Technology Center of Excellence: Impression, Pattern and Trace Evidence Symposium (2018). Numerous workshops, posters, podcasts, and oral presentations on forensic analysis and interpretation of glass and paint. The link to the proceedings is as follows: https://forensiccoe.org/workshop/18-iptes/
- RTI International, 2nd Annual Online Symposium: Current Trends in Forensics & Forensic Toxicology. Offered webinars, including oral presentations and posters focused on glass and paint evidence. https://forensicrti.org/2019-online-symposium-current-trends-in-forensic-toxicology/
- SciX (2016–2018). Nuclear forensic glass analysis was presented as well as presentations on Chemometrics within forensics. The link to the proceedings is as follows: https://www.scixconference.org/past-events

3. Paint and coatings examinations

The majority of the publications included in this review focused on architectural, automotive, artistic, and spray paints. New trends in architectural [1,2] and automotive paints were reported [3,4]. For example, multipurpose architectural paints, such as self-priming paint, stain blocking, and hole filling are becoming more prevalent. Also, in addition to new self-cleaning clear coats and matte clear coats, quad-coats have become a trend in some vehicles since 2015. Quad-coats are OEM systems with a four-stage topcoat paint process in which three clear coat layers are applied over a metallic basecoat. For certain finishes, some of the clear coats may be tinted and translucent to add a depth effect in color [5].

The scientific literature addressed the relevance of updated surveys to keep up with market changes. Method validation and assessment of performance rates were described for conventional methods such as microscopy, fluorescence, Scanning Electron Microscopy - Energy Dispersive Spectroscopy (SEM-EDS), UV-Vis Micro-spectrophotometry, Fourier Transform Infrared Spectroscopy (FTIR), and Pyrolysis-Gas Chromatography-Mass Spectrometry (Py-GC-MS). Novel applications were reported using Raman Spectroscopy, Direct Analysis in Real Time - Mass Spectrometry (DART-MS), and Inductively Coupled Plasma (ICP)-based methods. Raman spectroscopy is receiving particular attention in the field of forensic examination of paints, with approximately 30% of the paint literature in the past three years assessing its utility. Therefore, a more widespread adoption at forensic laboratories is likely in the near future.

Increased attention was also observed on the use of statistical methods for data analysis and interpretation of paint evidence. The primary statistical tools used for paint data included clustering methods (Principal Component Analysis, PCA, and k-Nearest Neighbors, KNN), classification methods (different versions of discriminant analysis (DA) such as Partial Least Square PLS-DA, Linear LDA, and Support Vector Machine SVM-DA), multivariate calibrations (PLS, and Multiple Linear Regression, MLR) and likelihood ratios (LR).

Moreover, there are continuous efforts to improve and assess the performance of the searching algorithms employed in paint databases (i.e., PDQ and EUCAP). Studies have described how automotive paint databases can become handy in forensic investigations to search for potential vehicle make/model or to estimate the rarity of a particular paint system.

Lavine et al. continued a series of studies that use prefilters to predict vehicle-make and to enhance the PDQ library search algorithms when the spectra is collected using ATR-FTIR [6–8]. Also, the ENFSI EWG Paint & Glass Newsletter published a preliminary study to estimate the error rates in the EUCAP database vehicle-make search [9].

This review reports on advances on forensic paint examinations published in the peer-reviewed literature, books, and standard guidelines and methods.

3.1. Standard methods and guidelines

Two ASTM standards were reviewed and published in 2018. One consisted of a guide for using Infrared Spectroscopy (IR) for paint analysis [10] and the other a detailed guide of the sampling, collection, and analytical scheme for the forensic analysis and comparison of paint [11]. These ASTM standard guides were also assessed, balloted and approved through a separate NIST- OSAC (Organization of Scientific Area Committees for Forensic Science) standards approval process. The ASTM standard guides E2937-18 and E1610-18 are now included in the OSAC Registry (https://www.nist.gov/topics/forensic-science/organization-scientific-area-committees-osac/osac-registry/osac-approved).

3.2. Books and chapters

Books with chapters including paint reviews, paint investigations, or instrumental analysis of paint include: 1) Forensic Science: A Multidisciplinary Approach by Katz et al. [12], 2) Forensic Science: A Beginner's Guide. 2nd edition (glass section) by

Jay Siegel [13], 3) Introduction to Forensic Science and Criminalistics by Harris et al. [14], 4) Inorganic Trace Analytics: Trace Element Analysis and Speciation by Vassileva et al. [15], 5) Forensic Chemistry: Fundamentals and Applications by Jay Siegel [16], and 6) the third edition of Forensic Science Handbook by Richard Saferstein and Adam B. Hall [17]. Also, a book focused on forensic examination and interpretation of trace evidence is currently in press [18].

4. Paint measurements

In December 2015, Dolak and Weimer [1] reported the analysis of twenty-six white single layer multipurpose and non-multipurpose architectural paint products. The authors reported the chemical composition of multipurpose paint that is increasing its market demand, and therefore likely to become more prevalent in casework. Intra-brand and inter-brand comparisons were conducted by visual examination, stereomicroscopy, fluorescence microscopy, microchemical and micro-solubility tests, FTIR, and SEM-EDS. Discrimination power of 99.69% was obtained for a total of 325 possible comparison pairs by all the techniques combined. In addition, the authors found the compositions of multipurpose products were different from those of their non-multipurpose counterparts. The major differences found resulted from the amount and type of fillers used in the primers and paints. The presence of elemental zinc was attributed to the anti-mold and mildew architectural products.

Gates [19] reported the study of twenty-eight multi-colored spray paints by FTIR to detect the differences caused by the differential mixing of binder and pigment components. A variety of colors were selected for the study (yellow, gold, beige, brown, green, orange, pink, red, blue, white, gray, black, and clear). The author also described a technique for the sublimation of organic pigments from spray paints for isolation and analysis by FTIR. The results showed variability in the behavior of paint pigments with the amount of mixing before application. This variation was attributed to the absence or presence of carbon black or inorganic extender pigments such as titanium oxide, talc, silicates, and calcium carbonates. The inorganic pigment-loading distribution appeared higher on well-shaken paints than in unshaken paints. To avoid false exclusions, the author recommends the comparison of standards from spray paint in both unshaken and shaken states.

Sloggett [20] published an article on the importance scientific methods and forensic investigations in the analysis of presumably fraudulent pieces of art. The author highlighted the need for evidence policing for situations in which art fraud is suspected; such situations were suggested to be beyond scholarly investigations. The author proposed the use of semi- and non-destructive techniques for a more comprehensive and objective analysis of the materials. Scientific research of the suspected art pieces can provide evidence as to whether the materials and techniques used in the production of the work have been chosen, used or manipulated for deliberative, intentional, or deceptive behavior.

In 2016, Buzzini and Suzuki [21] reported a review of publications showing the use of Raman spectroscopy for the analyses of pigments in paint evidence. The paper consists of a comprehensive review of the forensic applications of Raman spectroscopy for the characterization, differentiation, comparison, and identification of paint evidence. The authors highlighted the capabilities of Raman spectroscopy to detect pigments that are difficult to detect by IR spectroscopy. These pigments' structural features are expected to produce large Raman scattering, which in turn results in intense Raman bands. Raman is expected to unequivocally identify pigments even at very low concentration.

Centeno [22] published a review of publications on the applications of Raman spectroscopy for the analysis of artistic materials (manuscripts, drawings, prints, and paintings) in collections from museums and cultural institutions. The review article aims to show the research progress on Raman spectroscopy applications as well as some challenges and prospects for this type of research. The review is divided by the different components for these types of materials: pigments, ink, and natural organic binding media, adhesives, and varnishes. The need for a comprehensive database for Raman spectra, including naturally and artificially aged materials was suggested. The use of Surface-Enhanced Raman Spectroscopy (SERS) for signal improvement and portable devices for remote analysis would expectedly increase the applications of Raman for the analysis of artistic materials.

Germinario et al. [23] reported the characterization of 45 commercial spray paints used in street art by FTIR, Py-GC-MS and Raman spectroscopy. The analyses were focused on the identification of the synthetic binding media, pigments and additives such as plasticizers and fillers. Some pigments and extenders could be efficiently identified by examination of the FTIR spectra and pyrolysis products. However, for most samples, Raman spectroscopy investigation was required in order to achieve the complete chemical characterization of organic and inorganic pigments, extenders and fillers.

Hibberts et al. [24] published an article on the use of Raman spectroscopy for the analysis of a painting with possible origins in the late fifteenth century. The analysis was conducted prior restoration of the painting. Raman spectroscopic analysis of the pigments placed the painting in the Renaissance period and allowed the identification of several pigments (cinnabar, haematite, red lead, lead white, goethite, verdigris, caput mortuum and azurite) with no evidence of more modern synthetic pigments or of modern restoration. The analysis also allowed to identify the treatment of the canvas substrate with a specific orange-colored resin, as well as the varnish coating of the surface.

Lv et al. [25] analyzed 52 automotive colored coating samples by FTIR and Raman spectroscopy. Cu pigments were detected with high frequencies in blue and green samples; Ti was found in all white samples. Bismuth, a substitute for lead in paints, was not detected in the samples under study. Compounds with heavy metals, including TiO_2, phthalocyanine blue, phthalocyanine green, and lead chromate, were frequently detected in the paint samples. Raman complemented FTIR information, particularly in the identification of inorganic pigments and additives, increasing discrimination when both methods are combined.

Lv et al. [26] reported the analysis of paint pigments by confocal Raman spectroscopy in comparison to IR results. Four groups of samples were compared by both Raman and FTIR. Raman spectroscopy provided additional discrimination between samples due to improved pigment characterization. The authors reported that phthalocyanine blue and Vat blue RSN, Pigment Scarlet Powder and Bronze red C, Fe_2O_3 and $PbCrO_4$, Prussian blue and phthalocyanine blue were all successfully identified and discriminated using Raman spectral comparisons.

Maric et al. [27] reported the analysis of the clear coat of 139 Australian and international vehicles by Raman spectroscopy, including 17 manufacturers and 45 different models. PCA resulted in 19 distinct classes that were associated with the vehicles' manufacturer and model, and year when applicable. LDA on the PCA groupings resulted in improved discrimination between the groups, with 96.9% of the calibration set and 97.6% of the validation set correctly classified. The authors reported enhanced discrimination capabilities of Raman spectroscopy compared to IR data for the same clear-coat data set.

Pozzie et al. [28] published a paper of the analysis of closely related molecules by SERS. A binary mixture of red dyes was

analyzed (alizarin, purpurin, carminic and laccaic acids, brazilein) and the spectra were recorded on two different metal substrates (citrate-reduced silver colloids and silver films over nanospheres). Reference materials and red lake oil paint reconstructions were analyzed upon hydrolysis with hydrofluoric acid, to the effect of varying the experimental conditions on dye identification. It was found that, in some cases, the spectral contribution of the second colorant in the mixture goes undetected unless it is present in significant concentrations. The authors confirmed the ability of SERS to detect and identify up to two different colorants in mixtures but concluded that the method was not able to linearly correlate the intensity of the SERS signals with the main die used to color the artifact under study.

Reynolds et al. [3] reported the analysis of 231 automotive paint samples by microscopical examination (stereomicroscopy, compound comparison, and fluorescence microscopy), FTIR, and SEM-EDS. Microscopy resulted in a discrimination potential of 99.97% of the samples. Samples of similar microscopic characteristics were further studied by FTIR and SEM-EDS. Two sample pairs remained undistinguished by all methods; they were manufactured two years apart in the same plant and consisted of the same make and model.

Sandercok et al. [2] reported the analysis of 1028 samples of modern formulations of interior and exterior architectural paint. The samples were characterized by color, FTIR, Py-GC-MS, and SEM-EDS. Visual examination and FTIR combined resulted in a 99.82% discrimination. These methods allowed to identify 700 samples; the 328 remaining samples were divided into 98 groups (956 indistinguishable pairs). The application of Py-GC-MS and SEM-EDS slightly improved the discrimination of a few samples not previously separated by visual and FTIR analyses resulting in 723 uniquely identified samples.

Silva et al. [29] published a paper on the development of a wet block digestion method for architectural paints to determine metals and metalloid using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). Results of the proposed method were compared to the ASTM standard test method D335-85a. Up to 13 elements were studied by the complete solubilization of different bases of paints at low temperature and atmospheric pressure. The limits of quantification ranged from 0.006 to 1.78 mg kg^{-1}. The authors found concentrations of lead that exceeded the threshold established by US legislation (0.009% w w^{-1}).

In 2017, Cesaratto et al. [30] reported the SERS analysis of basic fuchsine, methyl violet, and crystal violet and their degradation products. SERS analysis was not able to discriminate between the two less methylated basic fuchsine homologues, rosaniline and pararosaniline, and between crystal and methyl violet, but it distinguished rosaniline/pararosaniline from new fuchsine, the highest methylated basic fuchsine homologue, and those from crystal/methyl violet. Furthermore, we demonstrate that SERS is a valuable tool to study the photo-induced *N*-demethylation by tracking spectral changes in a series of artificially aged samples.

Cesaratto et al. [31] reported the analysis of acid red naphthol-based azo dyes by Raman spectroscopy, based on the results of the aforementioned publication. Reference dye materials were analyzed by dispersive Raman, FT-Raman, SERS. The reference spectra were used in the study of late 19th century Japanese polychrome woodblock prints. The results obtained by the various Raman techniques were compared. Due to the poor dye-metal interaction, a dispersive Raman approach proved to be more suitable for the effective identification of azo dyes used in art objects. In a case where the fluorescence background was very intense, SERS allowed a firm identification of the colorant.

Chen and Wu [32] reported the use of DART-MS for the study of pigments commonly found in vehicle paints. The authors analyzed twelve common organic pigments including, red, orange, yellow, and purple. Two hit-and-run vehicle accidents cases were investigated by FTIR, as a screening step, and then by DART-MS. Most of the IR information was attributed to the binder and extenders present in paints. DART-MS successfully characterized the organic pigments present in the paints.

De Faria et al. [33] reported the analysis of the pigment Indian yellow and the dye tartrazine by ^{1}H and ^{13}C Nuclear Magnetic Resonance (NMR), SEM-EDS and XRF. The Raman spectrum of Indian yellow and tartrazine had previously been mistaken in literature. This publication makes a distinction between the two and reports the analysis conducted on a genuine Indian yellow sample as an example. The importance of this research lies in the fact that tartrazine is a synthetic dye which was first produced toward the end of the 19th Century, whereas genuine Indian yellow pigment is reported to have been in use since the 15th Century; therefore, the distinction between the two is important for authentication of art works. It was found that the high luminescence showed by Indian yellow does not allow its Raman spectrum to be obtained using excitation in the visible or near infrared at 785 nm, however, in the FT-Raman spectrum (with excitation at 1064 nm) the pigment characteristic bands are clearly observed on an emission background. The genuine sample of Indian yellow was also characterized by SEM-EDX, XRF and ^{1}H and ^{13}C NMR.

Ferreira et al. [34] published an article exploring the potential of Hyperspectral Imaging Visible/Near-Infrared Spectroscopy (HIS-UV/VIS/NIR) combined with PCA as a forensic approach to discriminate automotive paints. A total of 38 samples from twelve different brands and five different colors were analyzed. HIS-UV/VIS/NIR was directly applied to the paint chip's surface. PCA resulted in 100% discrimination of the white, silver, and gray samples. Black paints resulted in 62.5% discrimination because the spectra did not provide enough reflectance suitable for differentiation.

Ferreira et al. [35] reported the use of Raman spectroscopy for the analysis of 36 automotive paint fragments from six different brands and seven different colors. Several parameters such as laser wavelength, exposure time, laser mode, sample substrate, and sample preparation method were evaluated for the use of Raman for paint analysis. PCA was used to assess the grouping of the samples. The results showed that although Raman spectroscopy was found to be accurate in the identification of vehicles, spectral variability must be considered to avoid false database matching and misleading of forensic investigations. The authors suggested the development of separated spectra library for each laser wavelength as well as for each sample substrate.

Huang and Beauchemin [36] applied Solid Sampling Electrothermal Vaporization (SS-ETV) coupled to ICP-OES in combination with multivariate statistical tools for the analysis of 32 samples of paints taken from the roof of vehicles. The bulk qualitative multielement analysis of paint fragments by SS-ETV-ICP-OES in combination with LDA allowed to classify samples based on the color, manufacturer, and year of production of an automotive vehicle by monitoring over 15 elements found in the paints. The method is destructive of the sample, requiring 1.5–2 mg, and therefore the authors recommend using it as the last step in the analytical sequence after application of non-destructive methods.

Khandasammy et al. [37] published a review in the recent applications on Raman spectroscopy to forensic science. The review covers the newly published articles on the applications of Raman spectroscopy to several types of evidence, including different types of paints. The manuscript discusses the Raman data analysis by different clustering, classification and multivariate calibration methods.

Maric et al. [38] applied Direct Analysis in Real Time, Time-of-Flight Mass Spectrometry (DART-TOFMS) to clear coats of four vehicles. The samples were also analyzed by Py-GC-MS, as the standard protocol. PCA was utilized for data interpretation. DART-MS provided similar discrimination to Py-GC-MS with the added advantage of reducing the analysis time from 1 h to less than 3 min. In addition, the techniques offered complementary information for samples that were distinguished by one method and not the other. Thermal desorption/pyrolysis DART-MS was also applied, resulting in the discrimination of all the samples based on the distinctive thermal desorption plots.

Zieba-Palus and Kowalski [39] reported a study on the influence of the substrate in spray paint identification. The samples were analyzed by Attenuated Total Reflectance (ATR) FTIR and Raman spectroscopy. Seven spray paint samples placed on metal, glass, fabrics and paper substrates were studied. The results showed that the type of substrate, and thickness of the paint smear, greatly influenced the identification of the paints. The best results were found for highly reflective surfaces: glass and metal. The fabric substrate resulted in interface bands that prevented paint identification.

In 2019, Kruglak et al. [40] conducted a population study to assess the frequency of physical, microscopical, and chemical properties of automotive paint chips. A total of 200 red paint chips were collected from body shops from the Northeastern United States. All samples were analyzed using stereomicroscopy, bright-field, and polarized light microscopy. Further analysis included FTIR, Raman, and ultraviolet–visible (UV–Vis) microspectroscopy. Microscopy alone resulted in 99.995% discrimination of the samples (one indistinguishable pair). Microscopy combined with FTIR and UV–Vis resulted in 100% discrimination. Raman spectroscopy allowed for the identification of 50% of the pigments in the samples.

Palenik et al. [41] conducted a study where they analyzed paint particles not visible by the unaided eye. The authors analyzed particles as small as 40 μm in size by an analytical approach involving a combination of stereomicroscopy, polarized light microscopy, infrared microspectroscopy, Raman microspectroscopy, and SEM-EDS. The results showed evidence of a two-way paint transfer between a blue automobile and a gray painted surface. Three different pigments were identified in the specks of blue paint, and the combination of these pigments was associated with automotive paint. Streaks of gray paint were identified within scratched areas of the known automotive clear coat and elemental analysis demonstrated that these streaks contained pigment-sized particles that are elementally consistent with the components of the known gray paint.

Wang et al. [42] reported the analysis of repainted automotive paint by Optical Coherence Tomography (OCT). The authors developed a custom-built spectral-domain OCT configuration with ~6 μm axial and lateral resolution to obtain three-dimensional (3D) images of an artificially prepared, internally-damaged, repainted automotive paint surface. This technology allowed to recover high-resolution sub-layer images of the repainted automotive paint.

5. Paint interpretation

Hodgins et al. [43] investigated the ability of forensic scientists to use the Paint Data Query (PDQ) database to identify the make and year of a late model motor vehicle from a paint sample. Forensic scientists were provided with a chip of paint from a factory painted motor vehicle manufactured in 2009. The participating scientists (45 respondents) used a combination of stereomicroscopy and FTIR spectroscopy to examine the color and chemistry of the sample, followed by a search of the PDQ database and spectral library using the data collected. Of the 45 respondents, 39 correctly identified the manufacturing plant and model year range from which the paint sample originated, while another 5 respondents were able to search the PDQ database and obtain a hit list that included the correct manufacturing plant and model year, even though each subsequently chose to eliminate it from their result. The errors made by some users demonstrate that they did not consider that the database is representative and not comprehensive.

Wright and Mehltretter [4] published a paper stating the significance of taking into account Original Equipment Manufacturer (OEM) factory repair layer system when making an interpretation statement for the findings of paint analysis. The authors evaluated the frequency of OEM repairs on a data-set of 1057 paint specimens representing vehicles manufactured between 2000 and 2013. Examinations were conducted on different body panels (roof, quarter panel, door, and hood). The results show that the vast majority of samples examined were standard OEM layer systems with no OEM repair (92.2% of the 1057 samples). From the 7.8% of OEM factory repairs, the most common repair system was one additional clearcoat/basecoat application (4 topcoat layers, 6.34% of the samples). Six and eight topcoat layers were less frequent (1.14% and 0.284% of the 1057 samples, respectively. The authors suggested using statements in the forensic reports to draw attention to the rarity of the presence of these additional factory-applied layer systems.

Lambert et al. [44] reported the use a multiblock technique as a chemometric tool for combining spectroscopic data in the forensic analysis of paint. The authors applied the chemometric method to the analysis of domestic red paints. The paints were analyzed by IR and Raman spectroscopy. PCA and Hierarchical Clusters Analysis (HCA) were applied to the spectroscopic data. The authors found that IR spectroscopy showed group patterns related mainly to the binder and extender composition of the paints, whereas Raman spectroscopy data were mainly related to the pigment composition. Common Component and Specific Weight Analysis (CCSWA) was used in order to produce independent PCAs for each block (IR and Raman), and the combined information resulted in a score plot. By applying this method, the authors found an increase number in groups compared to PCA (20 groups vs. 12 IR groups and 7 Raman groups, independently).

Martyna et al. [45] reported a hybrid approach combining chemometrics and likelihood ratio framework for communicating the evidential value of spectra obtained from Raman analyses of automotive paints. The authors used conversion from classical feature representation to distance representation for revealing hidden data peculiarities. Linear discriminant analysis was further applied for minimizing the within-sample variability while maximizing the between-sample variability. Both techniques enabled substantial reduction of data dimensionality. Univariate and multivariate likelihood ratio models were proposed for this data. It was shown that the combination of chemometric tools and the likelihood ratio approach could solve the comparison problem of highly multivariate and correlated data. The results presented the potential of this methodology even for small databases.

De Roy et al. [9] published a study that estimated the error rates in the EUCAP database vehicle-make search. Fifty automotive paint samples were subjected to a blind car-make identification using a modified multilayer search routine on the BioRad software (KnowItAll, 2015 version). The samples selected for the study were not part of the EUCAP database. Two search strategies were conducted, resulting in 10% false-positive identifications when the correlation algorithm was applied and 16% false positives when the 1st derivative Euclidian match algorithm was used. The combination of these algorithms reduced the false positive error rate to 8%. The results of the study highlight the need to assess the capabilities,

limitations, and reliability of the searching and comparison algorithms.

In 2017, Lavine et al. [6] reported the development of a search engine for the IR spectral libraries of the PDQ database. The authors applied a pattern recognition approach using pre-filters and a cross-correlation library search algorithm. The cross-correlation library searching algorithm in conjunction with the search pre-filters outperformed OMNIC.

Kwofie et al. [7] used transmission infrared imaging microscopy for the forensic examination of automotive paints. Concatenated IR data from all paint layers in a single analysis was collected by scanning across the cross-sectioned layers of the paint sample using an FTIR imaging microscope. A multivariate curve resolution method was applied to obtain the IR spectrum of each automotive paint layer. Comparing the reconstructed IR spectrum of each layer against the IR spectral library of the PDQ database allowed the identification of the correct model of the vehicle from these reconstructed spectra. The use of this IR imaging method allows direct analysis of paint chips without the need to separate the paint layers, saving time, and simplifying the sample preparation.

Lavine et al. [8] applied pattern recognition techniques to the IR spectra of the PDQ database to differentiate between non-identical but similar IR spectra of automotive paints. Prefilters were developed to identify the vehicle make from the IR spectrum of a paint sample recovered at the crime scene. To develop these search prefilters, IR spectra from the PDQ database were preprocessed using the discrete wavelet transform to enhance significant features in the IR data. Wavelet coefficients characteristic of vehicle make were identified using a genetic algorithm for pattern recognition and feature selection. By using prefilters, the search results were reduced to a smaller number of hits.

Michalska et al. [46] published a paper in which likelihood ratio (LR) approach is applied to Raman data of blue automotive paint samples. Different analytical parameters were tested to determine their significance to the likelihood ratio determination. For the construction of the LR models, two variables were tested: areas under selected pigments bands and coefficients derived from discrete wavelet transform procedure (DWT). It was found that objective magnification played an important role in the performance of the LR models. The effects of laser power and time of radiation were also explored. Time of irradiation upon established laser power did not affect solving the comparison problem with the use of the LR test. In the same manner, upon the established time of irradiation 5% or 10%, laser power could be used interchangeably without affecting conclusions.

5.1. Paint weathering and degradation

Jost et al. [47] published a study on the degradation of spray paint samples, illustrated by Optical, FTIR and Raman measurements. Unlike automotive paint, which are designed for improved outdoor exposure and protection, spray paints are affected by solar radiation, temperature and humidity. Six different spray paint samples were exposed to outdoor UV-radiation for a total period of three months and both FTIR and Raman measurements were taken systematically during this time. Results were later compared to an artificial degradation using a climate chamber. The IR analyses suggested that spray paints are rapidly affected by degradation and the differences began to appear after a few days already. These are rapidly increasing until two months, where the degradation becomes more stable and follows a linear trend. Raman results suggested that the pigments, on the other hand, are much more stable and did not show any sign of degradation over the time of this study. As a conclusion, spectral variations due to oxidization products are likely to appear in FTIR spectra, while Raman spectra were found to be more stable. Care should still be taken when comparing two samples to assess a common origin, and degradation issues should be kept in mind to explain any significant difference that may appear between two paint samples.

Van der Pal et al. [48] published a paper of the effects of environmental degradation on the characterization of automotive clear coats by IR spectroscopy. Three samples collected from different vehicles were tested. The samples exposed to the outside environment revealed no changes in model predictions over a 175-day period; however, incorrect predictions were observed following 435 days of exposure. Inspection of the corresponding infrared spectra revealed that these changes were likely due to the hydrolysis and photodegradation of polymer chains present in the clear coat, which were not observed in samples stored inside over one year. Analysis of previously weathered samples using synchrotron infrared microscopy found these changes occurred on the surface of the clear coat. This indicates that weathering may affect the surface characterization of clear coats overtime, but the targeting of deeper portions of the clear coat layer may still be useful. The authors recommend obtaining the spectra from the middle of a paint cross-section to reduce the influence of weathering and migration of the color coat into the clear coat.

In 2018, de Oliveira et al. [49] reported the use of Raman spectroscopy for the analysis of weathering effect on automotive paints. Vehicles were exposed to the outdoor environment for over seven years. Paint samples were extracted from two vehicle panels of different degradation levels for chemical comparison. In situ IR and Raman spectra were taken from the surface of the paint chips. Raman images of cross-sections were also acquired to show an effect of alternation in stratigraphy and the composition of paint layers on the routinely used in situ analysis. The authors reported significant differences between less and more degraded samples in terms of spectroscopic spectra and remodeling of the layers.

6. Glass examinations

Books and book chapters that were published on glass within the last three years include *Forensic Analytical Methods,* published in 2019 by Thiago Paixão et al. [50] It contains a chapter on laser-induced breakdown spectroscopy (LIBS) and how it applies to glass samples. Bernard Robertson et al. published *Interpreting Evidence: Evaluating Forensic Science in the Courtroom 2*nd *edition* in 2019 [51]. This book has multiple chapters of interest such as interpreting scientific evidence, explaining the strength of evidence, and assigning likelihood ratios. Overall Robertson et al., explains general principles of the scientific method, how to analyze the data, how to explain the data to a courtroom, and provides actual cases as examples to show how the case could have been affected by the evidence if presented in a different way. In 2019, Craig Adam published Forensic Evidence in Court: Evaluation and Scientific Opinion [52]. Adam has two chapters relevant to this review paper: Case Studies in Expert Opinion and Trace Evidence, Databases and Evaluation. The first chapter covers real court cases and how expert opinion makes a difference. The second chapter is explaining trace evidence (how it is made and analyzed) and what tools are used to interpret the data.

Black and Daeid published 30-Second Forensic Science in 2019 which contains a chapter on a brief overview of glass evidence [53]. In 2019, Elkins published Introduction to Forensic Chemistry that contains a chapter on trace evidence, including glass. It talks about analytical techniques and practices of the forensic science field [54]. Harris and Lee introduced *Introduction to Forensic Science and Criminalistics, Second Edition* in 2019 [55]. This book contains a chapter on material evidence, which contains glass. The chapter covers techniques in collecting and analysis. Katz and Halamek

wrote a book in 2016 titled Forensic Science [56]. It gives background on forensic science and covers many topics including glass. It covers scanning electron microscopy-energy dispersive X-ray (SEM-EDX), X-ray fluorescence spectrometry (XRF), inductively coupled plasma-optical emission spectrophotometry (ICP-OES), and inductively coupled plasma-mass spectrometry (ICP-MS).

The Organization of Scientific Area Committees for Forensic Science (OSAC) approved two standard methods for glass analysis to the OSAC Registry within the last 3 years: the "Standard Test Method for Forensic Comparison of Glass Using Micro X-ray Fluorescence (μ-XRF) Spectrometry" (E2926-17) and the "Standard Test Method for Determination of Trace Elements in Soda-Lime Glass Samples Using Laser Ablation Inductively Coupled Plasma Mass Spectrometry for Forensic Comparisons" (E2927-16e1). These ASTM methods were revised during the reporting period (2016–2019) and approved by OSAC to place on the registry (https://www.nist.gov/topics/forensic-science/organization-scientific-area-committees-osac/osac-registry/osac-approved).

6.1. Industry

The National Glass Association (NGA) [57] is an industry-supported information center for manuals, publications, classes, and many more resources. In 2017, the NGA released a glass information bulletin that lists the current editions of "industry consensus and federal flat glass standards", as well as physical and mechanical properties of soda lime float glass. The Glass Magazine [58] is a journal where any article the NGA publishes may be found. The NGA and Glass Magazine have founded a world map containing information on all float glass plants and glass manufacturers across the world call the World of Glass Map [59]. The World of Glass Map reports 1million tons of flat glass manufactured per week with over 90% of it being using in construction and automotive industries. This global production results from 204 float glass plants and 176 glass fabricators in operation as of August 2019.

The 2019 annual report by Devlin and Dick [60] states that over the last 25 years the float glass lines have expanded from 150 to over 500, from 1992 to 2018. A majority of the production used to be from the "European Union (EU), United States of America (USA), and Japan". Today, glass production is mainly from China, although the region's production is expected to decline due to saturation. It is estimated that the glass manufacturing industry will reach $232.4 billion worldwide in 2020 with India being the most promising market [59,60].

Asia had the most plant openings and/or expansions this year. Currently, China has 55 float glass plants open [59]. Poland, with 4 plants operational today, staid stagnant. It did however start constructing a new plant in Częstochowa. Glass companies such as Şişecam Group, Fuyao Glass, and Xinyi glass have expanded globally. Şişecam Group in Turkey acquired a plant in Italy that increased their capacity by 220,000 tons per year.

North American glass production has declined from 44 lines in 2005 to 34 in 2015 but began to rebound and had 38 lines in operation at the beginning of 2019. The USA lost 3 lines in 2017, decreasing the domestic capacity by 8.5% due to fires, yet has started to stabilize in the last 2 years as lines that were being repaired became operational once again, with only 24 plants being currently active according the World of Glass Map [59]. It is estimated that the float glass industry will increase 20–45% through 2022 in developed countries [55]. The reports on USA production are contradictory, depending on the source as forecast to increase the fastest of the developed countries [61] and as the market showing signs of slowing [62].

7. Glass measurements

Seyfang et al. [63] determined the composition of glass fractionators that replace antimony sulfide in bullets primers of 0.22 rimfire bullets by scanning electron microscope-energy dispersive X-ray spectrometry (SEM-EDS), time of flight-secondary ion mass spectrometry (ToF-SIMS), and Sensitive high-resolution ion microprobe (SHRIMP). The elemental and isotopic compositons changed throughout the population. ToF-SIMS had a discriminating power of 94.1%, SEM-EDS had 79.4%, and SHRIMP (when composition with the other two techniques) had 95.6% discrimination between brands. The authors did measure refractive index to demonstrate each cartridge only has one population.

In a later study, Seyfang et al. [64] also assessed other sources of particles to see if glass-containing GSR (gGSR) is not commonly found naturally. The authors studied fireworks, matches, and nail gun cartridges to see the prevalence of gGSR. The analysis was run using a backscattered-SEM-EDS (BS-SEM-EDS) and was found that nail gun created particles indistinguishable from gGSR, while the matches and fireworks created no particles similar to gGSR.

Seyfang et al. [65] also published in Forensic Science International about the different methods use to discriminate gGSR. The authors studied methods to analyze low caliber rimfire ammunitions due to the lack of antimony and tin, as this will change the likelihood ratios. Rimfire ammunitions do however contain a frictionator consisting of ground glass. Seyfang et al. analyzed unfired gGSR with SEM-EDS, Focused Ion Beam (FIB), and ToF-SIMS. The authors reported that FIB followed by ToF-SIMS or ToF-SIMS using ion sputtering offers a higher discrimination.

Harshey et al. [66] studied the pattern of fractured window panes (of varying thickness) by 4.5 mm lead pellet fired through a 4.5 mm caliber Air Rifle. The authors found the hole diameter to range from 4.77 to 7.5 mm. The Chi-Square test showed consistency in the fractures, supplemented by graphical representation, which can lead to distinguishing weapons by fracture pattern.

Tiwari et al. [67] varied thicknesses of glass to study the consistency of multiple fracture patterns when shot with an air rifle loaded with round nose pellets. Goodness of fit was used to analyze the data and found consistency within the fractures.

Srivastava et al. [68] studied fracture patterns made in glass by 4.5 mm round and flat nose lead pellets from an air gun. The metal framed glass was kept at a fixed distance. To analyze the data, graphical representation was used and was found to have significant trends.

Baca et al. [69] reported that 60 glass panes, 60 glass bottles, and 60 plastic tail lights all had different patterns when compared to each other. It is noted that more studies need to be repeated to achieve statistical significance to this theory.

Panadda et al. [70] used the Stoke's law to replace the sink-float method of analyzing glass density since it uses toxic solutions. The authors examined lab glassware, glass bottles, car glass, architectural glass, and kitchenware glass. To ensure the technique worked, Panadda et al. compared their values to ASTM C693-93. The preliminary findings were that it "is possible but with some limitations".

Cook et al. [71] developed a synthetic nuclear glass melt to try to mimic an authentic sample. The synthetic sample was irradiated in a high-flux isotope reactor in Oak Ridge National Lab. The sample was counted twice, and analyses were performed so improvements could be made on subsequent batches.

Reading et al. [72] developed a novel technique to create "homogeneous, flux-free glass beads of geochemical reference materials, uranium ores, and uranium ore concentrates". The process uses 9 parts of high purity synthetic enstatite and 1 part of sample. They are fused on an iridium strip resistance heater under argon.

The resulting bead was then analyzed using LA-ICP-MS.

Bonamici et al. [73] used samples from the Trinity nuclear test ("trinite") to create a dataset consisting of the major elemental composition to determine the mechanism of which glassy fallout is created. The CaMgFe component is largest in these samples and shows volatility-controlled condensation from plasma.

Nizinkski et al. [74] produced synthetic debris that was tested against trinite using electron microscopy and x-ray diffraction. It was shown to be similar to trinite and surrogate glass melt but was different for individual cities. The authors believe that this debris could serve to advance and validate existing nuclear forensic analytical methods. (20).

Nogami et al. [75] developed a new method to analyze forensic soil by focusing on the trace elemental composition of volcanic glass with in the sample. The analysis was conducted with LA-ICP-MS and resulted in 2 samples (one from a forest in japan and one from a car) from varying places were found to have the same origin. The authors demonstrated that volcanic glass is useful for soil identification in Japan.

In 2017, Montoriol et al. [76] analyzed bone lesions using a SEM-EDS and found window and mirror glass particles. The authors experimented on human rib fragments that they cut with fragments of window and mirror glass to simulate an injury involving glass. They did however find that boiling and defleshing the bones created a loss of particles.

Michalska et al. [77] conducted an analysis on sample preparation for a SEM-EDX since embedding is "impractical for small glass fragments". When using likelihood ratios it is found that laying a smooth, flat glass sample on a SEM tab is viable. The authors compared results using likelihood ratio models and found no significant differences in accuracy, precision, reproducibility, and false answer rates when comparing embedded vs nonembedded glass standards. (24).

Almirall and Trejos [78] published on LA-ICP-MS and how its application pertains to forensic science for trace elemental analysis. The technique is applicable to numerous samples such as ink, paper, soil, adhesive tapes, and glass. LA-ICP-MS can perform both qualitative and quantitative measurements of elemental and isotopic components. It also can be applied to food authentication, and gold and diamond provenance.

In 2016, Lee et al. [79] used LA-ICP-MS and linear discriminant analysis (LDA) to discriminate 35 side window samples. The samples came from 5 car manufacturers and 2 different glassmakers. The authors also analyzed 120 side mirrors from the same suppliers. Light rare earth elements were found to be statistically different from each glass maker, making LA-ICP-MS a viable technique for forensic science. The side mirrors could not be discriminated.

Heydon et al. [80] conducted an experiment on float glass with LA-ICP-MS to test for heterogeneity. The authors believe that the heterogeneities are caused by flaws within manufacturing. These flaws may cause a Type I error when combined with a 4 standard deviation criterion. Heydon et al. recommend distributing the ablation spots evenly throughout the thickness of the glass to detect the heterogeneities.

Corzo [81] defended and published her dissertation in 2018 using likelihood ratios on glass samples analyzed by LA-ICP-MS. She, with the help of Hoffman [83], created a database of 420 windshield samples. Corzo then analyzed these for elemental concentrations to later interpret likelihood ratios. The author developed an R code to allow of easier interpretation and used both their database and a database from the BKA to train the model being used. The result was a database that could determine likelihood ratios with less than 0.1% random match between vehicles.

Hoffman et al. [82] conducted an inter-laboratory exercise with 10 different laboratories. The labs analyzed forensic glass by using the standardized method ASTM 2927-16e1. This was done to evaluate the rate of misleading evidence. To calculation the likelihood ratio 3 different databases were used. There different exercises were performed. The first had 34/36 labs associate the known with the questioned correctly, while the other two exercises had all the labs submit correct association. The random match probability was calculated to be ~0.1%.

Hoffman [83] also published her dissertation on the analysis of glass samples with LA-ICP-MS to create a database. She collected samples from cars from IIHS and created a database of 420 samples. Hoffman then found the elemental composition and determined the likelihood ratio by comparing to this database as well as the BKA data base in Germany.

In 2019 Latkoczy et al. [84] used an interlaboratory study to compare different LA-ICP-MS systems. He used NIST SRM 610 and 612. He cross-examined laser ablation systems with different ICP-MS systems to determine which was best. He found less than 10% deviation.

Lehmann and Arruda [85] compiled a review of different analytical techniques to see which methods require the least amount of sampling and sample preparation. These techniques include x-ray spectrometry, LA mass spectrometry, laser-induced breakdown spectrometry (LIBS), ICPMS, optical emission spectrometry (OES), and Mössbauer spectrometry. Raman spectroscopy and ambient ionization mass spectrometry are also mentioned.

Walke and Rajan [86] also published a review of forensic science methods. Their reasoning for publishing the review is that methods, tools, and instruments have all advanced.

Fakiha [87] reviewed "scanning electron microscopy, DNA fingerprinting, alternative light photography, facial reconstruction, and LA-ICP-MS" to determine how best to apply these techniques. It was determined that forensic investigations have improved immensely from these techniques. The authors recommend that scholars aid each other for a better application of techniques and knowledge, and to apply forensic genetics to more than genetic material.

Kammrath et al. [88] reviewed glass evidence as a whole; not only the past, but also offered suggestions for the future of glass evidence. Glass evidence should be classified, discriminated, and/or individualized if possible. The most commonly used techniques to measure elemental analysis are XRF, ICP-OES, ICP-MS, and SEM-EDX. Analysts also look at physical and optical properties, according to this review.

A study was conducted by Auxier et al. [89] to expedite nuclear melt glass analysis by coupling a gas chromatograph (GC) to a time-of-flight ICPMS. This was done to shorten the dissolution time, expedite chemical separation, and improve analysis of nuclear melt glass. The GC and ICP-TOF-MS together decreased the separation and analysis time. They also provided a more detailed elemental and isotopic analysis.

In 2018, Bode et al. [90] discovered that Neutron Activation Analysis (NAA) can analyze large samples without need of pretreatments. This allows less error or contamination to occur. The authors cover the basic concept of NAA as well as elaborate on applications for the technique.

Acharay and Pujari [91] complied a review on NAA, Prompt Gamma-ray NAA (PGNAA) and Particle Induced Gamma-ray Emission (PIGE) to demonstrate use within forensic science. The samples these techniques can analyze (i.e. food, cloth, glass, and soil) need high precision and accuracy for elemental concentrations. These techniques are demonstrated to have application in forensic science.

Funatskui et al. [92] identified glass manufacturers in Japan by analyzing automobile windows with refractive index (RI), X-ray

Absorption Fine Structure (XAFS), and XRF. They determined the concentrations of compounds such as CeO2 and Al2O3 to discriminate manufacturers. This study identified the manufacturers of all 75 samples.

Laser Induced Breakdown Spectroscopy (LIBS) was used in a 2016 study conducted by Devangad et al. [93] They used this technique to determine the concentrations in phosphate glass. The authors also reported "very good linear regression coefficient (R2) values. The leave-one-out method was applied to predict its analytical ability. The correlation of uncertainty between LIBS and certified ratios were reported to be low values, confirming that LIBS has a large potentiation for quantitative analysis.

Khalil and Morsy [94] used double pulse (DP) -LIBS and electron paramagnetic resonance (EPR) to analyzed borate glass for copper composition. The 266 nm and 1064 nm pulses were used to predict the electron's temperature and density. Since a double pulse laser is being used the intensities are higher than a single pulse laser. The authors proposed different proto cols that allow DP-LIBS to detect trace copper.

In 2016, Jantzi et al. [95] reviewed sample preparation and treatment of LIBS. LIBS has had sample preparation developed for it to improve the analytical performance. All forms of samples are discussed to allow better application of the technique.

Weis [96] reviewed LA-ICP-MS on glass samples at the Bundeskriminalamt laboratory. He compared case work and how their analyses aided in investigations. The author used LA-ICP-MS and likelihood ratios to determine the weight of the evidence.

Gupta et al. [97] conducted an experiment for intra-day and inter-day variation when analyzing with LIBS. The authors used standard reference glass and explains the conclusions drawn from the data.

8. Glass interpretation

Morrison and Poh [98] tested 3 techniques that shrank the value of the likelihood ratio (LR) closer to 1. The techniques were uninformative priors, empirical lower and upper bounds, and regularized logistic regression. The authors compared with Linear Discriminant Analysis (LDA). They tested these techniques on face imagine, glass fragments, and voice recordings.

Aitken [99] reviewed Software for the Analysis and Implementation of Likelihood Ratios' (SAILR) software package used for analysis and implementation of likelihood ratios in forensic science. He reviewed the history, purpose, and background of the program.

McNevin [100] investigated prosecution hypothesis' (HP) and defense hypothesis' (HD) effect on LR. He states that since forensic science has begun providing posterior ratios, but the prior ratio is neglected that the posterior ratio is in fact unknown. McNevin presents criterion for determining limitations of LR and that a frequentist interpretation estimates only the denominator of the LR.

Franco-Pedroso et al. [101] explores a widely used multivariate approach to forensic analysis, kernel distribution function (KDF) and how it compares to Gaussian mixture model (GMM). The authors determined that GMM is a better fit for LR due to the between-source variation and is a provides a better calibrated LR.

Meuwly et al. [102] suggested a method to validated forensic analyses using LRs. They cover questions from a workshop presented before the publication of this paper as well as validation standards, strategy, methods, and a protocol in reporting. The authors use these topics as the source level of evidence.

Van Es et al. [103] evaluated the analysis of LA-ICP-MS on glass evidence with different approaches, such as the *t*-test or LRs. The authors present that an LR system is robust, empirical upper and lower bound method is ideal for density models, and empirical cross-entropy is viable. The rates of misleading evidence were reported to be less than 0.5%.

Vergeer et al. [104] investigated LR extrapolation errors as they occur outside of the data set range. This in turn limits the L values. The authors proposed to find these extrapolation errors by combining normalized Bayes error-rate and introducing the LRs to increased strength to purposely mislead the system.

Biedermann et al. [105] measures LR by its two components, probability of the proposition and probability density of the evidence. If both are true, then the LR becomes a single value.

Gittelson et al. [106] considered an argument by another group, Lund and Iyer (L&I). L&I argued that an LR presented in court should have further consideration outside of the expert witness' presentation. The authors also state that L&I say that LR should not be practiced, which the authors believe that "no one advocates" for. Gittelson et al. also conclude that LR provides an informative summary of the weight of evidence and that courts should be informed of how LRs provide that information.

Corzo et al. [107] experimented with using databases from LA-ICP-MS' to calculate LRs when presented with glass evidence. The authors state that a match criterion followed by a verbal scale is the typical approach to analyzing glass evidence and how that approach has many flaws. Corzo et al. used a multivariate kernel model to calculate the LR of 2 different glass databases. They found the rates of misleading evidence was <1.5% for same source evidence and <1.0% for different source evidence.

Hoffman et al. [82] used 3 databases to calculate LR for an inter-laboratory study in 2018. The random match probability of glass evidence was 0.1%.

Bovens et al. [108] explains that while chemometrics within forensic science has provided an enormous tool, it also is demanding in an everyday work scenario. The authors provide an overview to data handling and chemometric methods to improve evaluation, as well as workflow. They also will design a software tool to help forensic scientists.

Kumar and Sharma [109] review chemometrics in forensic science. They compare approaches, discuss history, and ponder applications within various disciplines. The authors propose new techniques and methods to help forensic analysts to get more confident statistical results.

Armstrong [110] defended his dissertation on the development of a Kernel-based model. This model allows for high-dimensional data and determining sources for multiple samples. The author experimented with SEM-EDX data on dust and microspectrophotometry on colored fibers.

Morrison et al. [111] argue against the use of a two-stage procedure for evaluation of forensic evidence. The first stage being match or non-match process. The second stage would be an assessment of sensitivity and false acceptance rates. The authors do explain that evidence that are "continuously-valued and have within-source variability" are not to use this two-step process and gives more appropriate procedures.

Biedermann et al. [112] discusses cut-off values for forensic analysts and explain when, and why, values are not appropriate. The authors challenged the use of cut-offs for ease and simplicity. There is discussion of logical cut-offs when using a standard measure and says when cut-offs are incompatible.

Ramos et al. [113] reports on the cross-entropy function that is used to classify performance and optimization. This publication analyzes prior knowledge and LR on the cross-entropy function. The authors also discuss discrimination and calibration within the function. They also give theoretical interpretations of cross-entropy. Lastly, the present an Empirical Cross-Entropy (ECE) plot.

Marquis et al. [114] reviews discussions held when developing and implementing a verbal scale. First, the authors published

arguments for verbal qualifies and mentions that help with communication on all sides of LRs. Secondly, the authors discuss the arguments in favor of the verbal scale proposed. Third, disadvantages of the verbal scale are mentioned. The authors recommend not using the verbal scale alone in a written statement. Lastly, if all parties can understand LRs then verbal qualifies may be abandoned.

Corzo [115] defended her work on evaluation of glass in 2018. She determined likelihood ratios for a glass database of 420 samples and created the R code to do so.

Hoffman [116] defended her graduate experiment on glass databases. She used vehicle windows to interpret likelihood ratios of evidence to present in court. The database was 420 samples and used the code produced by Corzo [115].

Park [117] used LA-ICP-MS data collected using the ASTM E2927-16e1 method to analyze a number of glass fragments including glass samples originating from the same "ribbon" collected over two weeks of manufacture and conclude that "Random Forests" analysis performs better than the comparison criteria recommended by the ASTM method. A close review of this paper [118] unveils serious errors in its experimental design and of poor quality of the underlying data collected. The authors selected a sample set that included glass produced within consecutive days to evaluate a "false positive rate" and incorrectly stated that these samples should be considered "different" by elemental composition when analyzed by LA-ICP-MS. The dataset choice is problematic as the false positive rate is greatly overestimated and misleading. Another flaw in the experimental design is the lack of independence between the training/validation set and the test set. For instance, several pairs of samples were collected on the same day, one of which was used in the training/validation set and the second was used in the test set. Therefore, the test sets and the training/validation data sets are in large parts essentially the same. Finally, close analysis of the elemental data shows poor quality lithium data, again leading to incorrect conclusions.

Disclaimer

This is a republication in journal form of a conference proceeding that was produced for the 19th Interpol Forensic Science Managers Symposium in 2019 and was originally published online at the Interpol website: https://www.interpol.int/content/download/14458/file/Interpol%20Review%20Papers%202019.pdf. The publication process was coordinated for the Symposium by the Interpol Organizing Committee and the proceeding was not individually commissioned or externally reviewed by the journal. The article provides a summation of published literature from the previous 3 years (2016-2019) in the field of glass and paint evidence and does not contain any experimental data. Any opinions expressed are solely those of the authors and do not necessarily represent those of their agencies, institutions, governments, Interpol, or the journal.

Declaration of Competing Interests

Jose Almirall is the Co Editor-in-Chief of Forensic Chemistry. The authors have no other competing interests to declare.

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