

Photoluminescence Spectroscopy for Estimating the Age of the Latent Fingerprints: A New Potential Approach

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Abstract

This study examined the variations in spectral absorption and emission intensity of latent fingerprints over time, employing UV-Vis spectroscopy and photoluminescence spectroscopy. There was a noticeable absorption at 296nm on the samples, and the fluorescence emission intensity showed variations. A total of 1600 samples from various categories were analyzed to validate this hypothesis. The emission intensity observed under photoluminescence spectroscopy varied over time, showing greater intensities in the controlled female and male samples compared to the uncontrolled samples that were exposed to environmental conditions. The emission intensity of the controlled female (CF) sample was the highest at 6341168 CPS, followed by the controlled male (CM) sample at 1270240 CPS. On the other hand, the uncontrolled female (UF) sample exhibited an emission intensity of 5431940 CPS, while the uncontrolled male (UM) sample had an intensity of 100764 CPS. This emission intensity variation proved to be an efficient age profiling marker for the latent fingerprints.

Keywords: Photoluminescence spectrometry; Spectrofluorimetry; UV-Vis spectrometry; Latent fingerprints; Age estimation; Dactylography

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Introduction

Dactylography, often known as fingerprint analysis, is an academic discipline that examines and analyzes the unique patterns formed by the epidermal ridges found on the fingers. The primary objective of this field of study is to use these distinctive patterns for individual identification. The area of fingerprint sciences has evolved from when professionals faced challenges in identifying fingerprints at crime scenes to when individuals who commit crimes outside their nations may also be recognized via an integrated database. As scientific progress continues, the challenges encountered by forensic professionals in crime scene investigations have become more complex. At a given crime scene, forensic specialists encounter several numbers of fingerprint evidence, leading to a time-consuming investigation procedure as they analyze all the fingerprints recovered from the site. The potential ease of estimating the age of a latent fingerprint presents a possible answer to this issue. This capability has the potential to aid fingerprint analysts in eliminating fingerprints that were unrecorded during the criminal activity, thereby aiding in the reduction of the suspect pool.

Photoluminescence spectroscopy, often known as Photoluminescence Spectroscopy, is a cutting-edge method used in forensic science to determine the age of latent fingerprints. This method incorporates various innovative elements that enhance the precision and effectiveness of fingerprint aging analysis. Conventional techniques used to determine the age of fingerprints may be intrusive, posing a risk of modifying or harming the fingerprint. On the other hand, photoluminescence spectroscopy is a non-destructive technique that allows the fingerprint to be preserved for future study. It allows for repeated observations over time without causing any damage to the material, thereby increasing the number of data points and improving the accuracy of age estimation. Chemical changes can be detected using photoluminescence spectroscopy, which is extremely sensitive to the chemical makeup of a material. The device is capable of detecting subtle alterations in the chemical composition of fingerprint residues, including amino acids, lipids, and other organic substances, which occur as the fingerprint matures. This technique yields quantifiable data regarding the chemical alterations, which can be directly linked to the duration that has passed since the fingerprint was left.

Temporal resolution is defined as the ability to accurately measure or distinguish between events or changes that occur over time. Photoluminescence Spectroscopy has the capability to identify alterations occurring within brief time intervals, making it appropriate for approximating the age of recently deposited fingerprints. This is especially valuable in forensic investigations, where establishing the recency of a fingerprint can have a substantial impact on the inquiry. Furthermore, it can be utilized on earlier prints, expanding its scope of application. The approach can be adjusted to examine particular chemical substances that are known to exist in fingerprints. This level of specificity is beneficial for developing a more precise aging model that is based on the deterioration or alteration of these chemicals as time passes.

This technique may deliver prompt results, often in real-time, which is advantageous in time-critical forensic investigations. This approach has the capacity for portable, in-field applications, enabling forensic experts to conduct initial analysis directly at the crime scene. Upon depositing a latent fingerprint on a surface, the organic constituents within the print experience degradation and chemical alteration. Photoluminescence spectroscopy can be employed to track and see these alterations. For example, the reduction in fluorescence intensity of specific amino acids can provide an indication of the age of the print. The relative concentrations of the amino acids are comparably studied with serine, glycine, ornithine, and alanine in the fingerprint residue and found to be the most abundant¹.

- 1. *Serine* the most abundant amino acid (approximately 100ng), has the absorption property at 200-300nm²
- 2. *Glycine* (The absorption spectra of glycine in an aqueous solution were mainly studied in the near ultraviolet-visible range, specifically between 240 and 600 nm. There was hardly any absorption observed near 270 nm for a 1.0 M solution, but it became significantly stronger as it aged.) (approximately 60)³
- Alanine (Absorption spectra for L-alanine indicate the presence of maximum absorption at wavelengths 361, 353, and 346 nm) (approximately 30ng)⁴

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- 4. *Aspartic acid* (aspartic acids have two absorption peaks at 196nm and 227.0 nm) (approximately 18ng)⁵
- *Tryptophan*: (Absorption 280)- Influences in degradation analysis. (approximately 5ng)⁶

Forensic scientists can estimate the time of fingerprint deposition by establishing a baseline and comprehending the pace of change under different environmental situations.

Background

A study was conducted to estimate the age of fingerprints by utilizing hyperspectral imaging of blood-stained patent-type fingerprints⁷. Hyperspectral imaging combines traditional imaging and spectroscopic techniques to capture both spatial and spectral data from the specimen. The age of the blood-stained fingerprints was determined by analyzing the distinctly visible absorption spectra of the hemoglobin, which falls between the range of 400 and 680nm. The success rate was attained within a 30-day timeframe as a result of the deterioration of blood color in the fingerprints. In another study, mass spectrometry imaging was employed to evaluate the breakdown of triacylglycerols⁸. By analyzing the enhanced images of the fingerprints, an effort was made to determine the age of the fingerprints. This analysis provided insights into the deterioration of fingerprint information with time. However, accurately determining the exact moment of the fingerprints' creation remained challenging. Researchers conducted several supplementary studies to determine the age of the latent fingerprints, but they failed to yield satisfactory conclusions on the matter.

This study examines the natural ability of fingerprint residues to produce a spectrum when stimulated with UV-visible light sources without the need for any developmental or enhancement procedures that could potentially damage the prints. Yielding positive outcomes from analytical techniques such as UV-visible spectroscopy and photoluminescence spectroscopy, it can assist specialists in eliminating fingerprints from crime scene samples that were not recorded during the time of crime conduct.

Methods and Methodology

a. Sample collection:

The samples were collected using the stratified samples collection method with an even interval of 8 days between sample collection for one year, starting December 2021, till December 2022. The undeveloped latent fingerprints were collected from both male and female sexes between the age group of 20 and 25 on the glass slides that were cleaned with ethanol before collection.

b. Sample Conditions:

The samples were collected of both sexes under two conditions: (i) Controlled Samples, where the samples were preserved from environmental contaminations and maintained at the same temperature (37°C) throughout. The samples immediately after collection, were sealed in airtight boxes and preserved. (ii) Uncontrolled Samples: where the samples were exposed to environmental conditions. Here, for instance, a wet laboratory with open windows and no control over the temperature to depict the indoor crime scene scenario.

c. Sample Number:

A total number of 1600 samples were collected under the following categories:

- i. Uncontrolled female **(UF)-** 40 samples were collected from 10 subjects throughout the duration set for sample collection.
- ii. Uncontrolled male **(UM)-** 40 samples were collected from 10 subjects throughout the duration set for sample collection.
- iii. Controlled female **(CF)** 40 samples were collected from 10 subjects throughout the duration set for sample collection.
- iv. Controlled male **(CM)** 40 samples were collected from 10 subjects throughout the duration set for sample collection.

d. Sample preparation:

The samples, both controlled and uncontrolled, and both male and female samples collected on the glass slides were washed using the neutral pH solvent (distilled water). The slides were washed with 3ml

of distilled water using a syringe to apply pressure to remove the sweat composition from the glass slide, which was in the dried condition. The wash was used for the analysis. The wash was first loaded onto the UV-VIS spectrometer to study the absorption wavelength of the existing fingerprint residues. The absorption was found to be strong at 297nm in all the samples. The samples (wash) were then loaded onto the photoluminescence spectrometry, and the emission rate at the wavelength of 297nm was studied.

The controlled condition samples are free from contamination, and hence, they also serve as a control sample to measure the influence of contamination on the uncontrolled samples. Considering that both the controlled and uncontrolled samples have the highest absorption at around 297nm, the contamination found on the uncontrolled samples has little to no influence on the age estimation of the latent fingerprints.

e. Instrumentation

- i. UV-VIS-NIR Spectrophotometer (Make: Varian; Model: 5000): This approach is widely used in the domains of chemistry and biochemistry. It is utilized to study the absorption of ultraviolet and visible light by chemical substances. This apparatus comprises a light source, typically a deuterium lamp for the UV range and a tungsten lamp for the visible range. It also includes a monochromator for precise wavelength selection, a sample holder, and a detector, which can be either a photodiode array or a photomultiplier tube. Electronic transitions occur when a molecule absorbs ultraviolet (UV) or visible light; these transitions involve the transfer of electrons from lower energy orbitals to higher energy orbitals. The absorbed energy corresponds to the energy disparity between the ground state and the excited state of the molecule. The UV spectrum comprises the wavelengths 200-400 nm, whereas the visible region spans 400-800 nm.
- ii. *Spectrofluorometer (Make: Jobin Yvon; Model: Fluorolog FL3-11):* The present study experimented with spectrofluorimetry to assess the age of latent fingerprints that were in an undeveloped state. Spectrofluorimetry is a scientific equipment that employs a light source to transmit light through a given sample, enabling

the detectors to analyze the material's reaction to the excitation caused by the light source.

The research was conducted on two sets of undeveloped latent fingerprint samples: conditional and unconditional samples (kept at a controlled temperature and exposed to the environment, respectively). It demonstrated the efficacy of sophisticated scientific equipment methodologies in criminal investigations while also addressing a common challenge faced by fingerprint analysts.

Results and Discussions

The samples, categorized as UF (uncontrolled female samples), UM (uncontrolled male samples), CF (controlled female samples), and CM (controlled male samples), were collected throughout the course of one year, from December 2021 to December 2022. The acquisition process established a conventional methodology and used clean glass slides. The samples were examined to determine if they exhibited any intrinsic potential to emit luminescence, which can be used to distinguish the fingerprints based on the time range. In order to observe luminescence, the samples must possess the ability to absorb and emit light, as specified by the principle. To attain this objective, the absorbance of the 160 samples was examined using UV-Vis spectroscopy. All the samples exhibited absorption at wavelengths of 297nm and 356nm, with 297nm showing a greater level of absorption. In addition, to analyze the emission capability of the samples, photoluminescence spectroscopy was used to assess all the samples. Table no.1 displays the mean maximum and mean minimum intensity values acquired from the photoluminescence spectroscopy analysis.

	Maximum Intensity Data (Unit in CPS)				Minimum Intensity Data (Unit in CPS)			
Week No.	UF	UM	CF	СМ	UF	UM	CF	СМ
1	118720	100764	164800	85310	22592.5	13026	44968	10033
2	128920	112302	174056	97728	26676	12712	47770	12442
3	125040	123418	155144	129954	19542	28864	30384	28108
4	91400	124022	99498	109162	14648	29360	15680	204902
5	93738	181242	254796	98302	8743	40563	60082	21162
6	95772	171220	199021	109122	9110	28891	66700	44911
7	120012	172001	113471	110292	11938	9883	56610	39221

	Maximum Intensity Data (Unit in CPS)				Minimum Intensity Data (Unit in CPS)			
Week No.	UF	UM	CF	СМ	UF	UM	CF	СМ
8	133001	168819	129901	99019	11128	11739	33919	38713
9	129947	156110	122101	110921	18891	19918	33718	32991
10	139911	114990	130010	138812	8991	21100	34771	45110
11	1503740	654612	281082	513490	130030	153980	35547	49273
12	2071208	359310	345567	590042	178947	47880	39557	51310
13	331378	412820	261228	529518	33427	32700	26643	41343
14	412140	322984	316924	1270240	44410	37310	36023	70200
15	354838	199021	306788	1164880	35023	46067	26030	177920
16	631594	439000	266030	426850	46795	45225	42237	49813
17	366674	575210	1321166	437401	46386	40670	39143	59696
18	780313	414384	2911370	341496	80200	37197	87737	35753
19	521216	405646	2387886	335314	32700	28313	37767	45310
20	424752	411628	6341168	300452	57603	62140	45860	46330
21	5431940	156202	4284500	252870	10190	3608	10446	8320
22	289834	222128	3144750	298910	4050	4678	2600	9346
23	2993070	250292	495380	303870	6250	5814	4100	9754
24	179234	175694	1410930	302067	4628	4308	4726	10338
25	162438	660190	197243	278050	4348	5620	3236	10086
26	175890	174196	165432	156408	4092	3838	3674	4238
27	191334	153756	152092	160344	4232	3752	3446	4094
28	151698	159148	167938	150647	3442	3944	3792	3442
29	168324	157632	173764	160629	3462	4166	4834	4693
30	172648	1700111	172478	149356	3122	3616	4074	3250
31	1141842	183966	836510	159694	4508	3876	5270	3772
32	196020	1102380	174872	150304	4900	3998	4238	3894
33	108920	1115552	151330	177036	3980	4698	4096	6192
34	275002	1115752	164930	186386	23810	3636	2392	5354
35	1779500	1164392	177840	157350	5810	5158	2658	4450
36	121650	1311962	135898	125297	3486	7338	2024	9910
37	205520	184502	143838	157782	5736	4370	2586	3692
38	173424	191338	143584	153660	3246	3544	2868	3888
39	181674	197976	158112	153092	2910	5182	3310	3688
40	2011791	197388	189913	155794	4550	4558	2239	3564

Table 1: *Maximum Intensity*- The mean values of maximum emission observed in the samples collected at each time duration at 297nm, and *Minimum Intensity*- The mean values of minimum range at which the samples started to emit luminescence. (i.e. in the 1st week, 10 samples were collected under the UF category, the 10 samples were analyzed and their mean values were extracted for the analysis; Week No.1 denotes the mean values of the maximum emission by the samples collected in the 1st week of the sample collection procedure)

Table 1 demonstrates a comprehensive understanding of the emission variations among the categories UF, UM, CF, and CM. Initially, the samples demonstrated a higher degree of variability during the warmer months and a reduced degree of variability during the cooler months. This can be ascribed to the notably larger quantity of perspiration generated in the warmer and the contrasting trend observed in the colder months.

Upon comparing the maximum variation within the samples, the emission rates over time were found to be higher in the controlled female and controlled male samples compared to the uncontrolled female and uncontrolled male samples that were exposed to environmental conditions. In this study, the controlled samples were utilized as the reference samples, while the uncontrolled samples served as the comparative samples. The uncontrolled samples exhibited emission variation comparable to the controlled samples when analyzed using photoluminescence spectroscopy, with the main distinction being the intensity of the emission. The controlled female samples exhibit the highest emission with an intensity of around 6341168 CPS (counts per second), while the uncontrolled female samples show a maximum emission intensity of roughly 5431940 CPS.

The controlled male samples exhibit the highest emission with an intensity of approximately 1270240 CPS (counts per second), while the uncontrolled male samples show a maximum emission intensity of roughly 100764 CPS. The controlled female samples have the lowest intensity of 2024CPS, followed by the uncontrolled female samples with 2910CPS, the controlled male samples with 3250CPS, and the uncontrolled male samples with 3544CPS. The controlled samples exhibited a narrower range of lowest intensity compared to the uncontrolled ones. The graphs below depict the maximum and minimum differences in intensity between the UF, UM, CF, and CM samples. (as shown in graphs 1 and 2).



MAXIMUM EMISSION INTENSITY VARIATIONS BETWEEN UF, UM, CF, AND CM SAMPLES

Graph 1: Maximum emission intensity variation between the samples UF, UM, CF, and CM with respect to the time period of 1 year (From December 2021-December 2022)





Graph 2: Minimum emission intensity variation between the samples UF, UM, CF, and CM with respect to the time period of 1 year (From December 2021 to December 2022).



Graphs 3,4,5 and 6 are the reference spectrograms retrieved under the PL Spectrometer of the samples (under categories UF, UM, CF, and CM) collected on the 1st week of the sample collection.

The following graph, 7, shows the maximum intensity of the emission spectrum by the fingerprint residue samples during the warmer duration of the year. Graphs 8 and 9 show the maximum intensity of the emission spectrum by the fingerprint samples during colder durations of the year, the starting of the sample collection (December 2021), and the ending of the sample collection period (December 2022).



Graph 7: Maximum emission intensity variations between UF, UM, CF, and CM samples (during the warmer times of the year)



Graphs 8 and 9: Maximum emission intensity variations between UF, UM, CF, and CM samples (during the colder times of the year)

To assess the statistical significance of the study, a correlation analysis was conducted on the samples. The results indicated that the minimum intensity data of the fingerprints exhibited a consistent positive correlation with time. However, the maximum intensity data displayed both positive and negative correlation values to some extent. (Table 2)

Maximum	UF	UM	CF	СМ
UF	1			
UM	-0.048373863	1		
CF	0.353388071	-0.086922501	1	
СМ	0.113157464	-0.030978731	0.08742983	1
Minimum	UF	UM	CF	СМ
UF	1			
UM	0.744020011	1		
CF	0.472502566	0.498654186	1	
СМ	0.284612063	0.425182348	0.31048849	1

Table 2: Correlation table for the maximum and minimum intensity data range of the samples UF, UM, CF, and CM.

Regression Statistics for Maximum Intensity Data:						
0.47182						
0.22261						
0.13377						
10.88048						
40.00000						
	s for Maximur 0.47182 0.22261 0.13377 10.88048 40.00000					

ANOVA	df	SS	MS	F	Significance
Regression	4	1186.5338	296.6335	2.505673	0.05
Residual	35	4143.4662	118.3847		
Total	39	5330			
	Coefficients	Standard Error	t Stat	P-value	_
Intercept	16.13	3.32	4.86	0.00	_
UF	0.00	0.00	0.93	0.06	
UM	0.00	0.00	2.89	0.01	-
CF	0.00	0.00	-0.05	0.039	_
СМ	0.00	0.00	-0.97	0.04	_

Table 3: Regression Model analysis performed on Maximum emission intensity data.

Regression Statis						
Multiple R 0.796						
R Square	0.634					
Adjusted R sq	0.592					
Standard Error	7.468					
Observations	40.000	_				
		_				
ANOVA	df	SS	MS	F	Significance	
Regression	4.000	3377.953	844.488	15.142	0.000	
Residual	35.000	1952.047	55.773			
Total	39.000	5330.000				
	Coefficients	Standard Error	t Stat	P-value	_	
Intercept	30.697	1.787	17.182	0.000		
UF	0.000	0.000	0.780	0.040		
UM	0.000	0.000	-0.453	0.054		
CF	0.000	0.000	-5.605	0.000		
СМ	0.000	0.000	-2.445	0.020	_	

Table 4: Regression Model analysis performed on Minimum emission intensity data

Further, to strongly display the significance of the study, regression model analysis (Tables 3 and 4) and ANOVA were performed on the study, which showed significant results. The significance, P-value, for the maximum emission among the different categories with respect to the time is found to be 0.050, and the P-value for the minimum emission among the different categories is found to be 0.000, both of which have P<0.05, demonstrating that the use of UV-Vis spectrometry and Photoluminescence Spectroscopy for the age

Category:	Overall	UF	UM	CF	CF
Mean	20.50	23713.29	20930.97	22943.85	30512.64
Standard Error	1.85	5639.23	4297.51	3543.54	6703.40
Median	20.50	9050.50	8610.50	13063.00	10212.00
Mode	-	-	-	-	-
Standard Deviation	11.69	35665.62	27179.86	22411.30	42396.03
Kurtosis	-1.20	10.13	14.27	0.14	9.80
Skewness	0.00	3.00	3.24	0.88	2.94
Range	39.00	176036.70	150436.00	85712.70	201652.00
Minimum	1.00	2910.00	3544.00	2024.00	3250.00
Maximum	40.00	178946.70	153980.00	87736.70	204902.00
Sum	820.00	948531.40	837238.90	917753.80	1220505.60
Count	40.00	40.00	40.00	40.00	40.00
Confidence Level (95.0%)	3.74	11406.42	8692.54	7167.48	13558.91

estimation of the fingerprint samples are possible with a standard error of the maximum emission intensity being 1.848. (Table 5)

Table 5: Descriptive statistical data of the maximum emission of the samples with respect to time

Moreover, to strongly support the hypothesis, the regression model was performed between 2 categories, and the Variance-inducing Factor (VIF) was studied. Table 6 shows the VIF values were derived from the regression model's R square value using the following formula:

VIF value = 1 / (1-R sq value)

Variance Inducing Factor from the regression models:

	Maximum Ir	ntensity Data	Minimum Intensity Data		
Category	R Sq Value	VIF Value	R Sq Value	VIF Value	
UF:UM	0.00234	1.00234552	0.55356578	2.23997164	
UF:CF	0.1248831	1.14270451	0.22325867	1.28742989	
UF:CM	0.0128046	1.0129707	0.08100403	1.08814405	
UM:CF	0.0075555	1.00761304	0.248656	1.33094827	
UM:CM	0.0009597	1.0009606	0.18078003	1.22067337	
UF:CF	0.007644	1.00770286	0.0964031	1.10668818	

Table 6: VIF values from the Regression model analysis

The variance-inducing factor above 0 indicates that there is a variance between the categories, and regardless of the sexes, the age

estimation of the fingerprints with the help of UV-Vis spectrometry and Photoluminescence Spectroscopy is possible.

Conclusion

Latent fingerprints predominantly comprise remnants deposited by sweat, oils, and other substances on the skin. These residues might include chemicals that possess luminescent characteristics. One of the major reasons why the fingerprint residue was able to emit fluorescence is because of the presence of amino acids. It is estimated that the average amino acid content of a fingerprint is 250ng, with an absorption range between 200 and 360nm. The fingerprint residue shows an absorption spectrum under the UV-Vis spectrometer near 296nm; they show both the maximum emission rate and also minimum emission rate. The minimum intensity refers to the lowest intensity at which the amino acids can emit fluorescence.

Currently, there is a lack of studies on this effective technique for estimating the age of latent fingerprints using photoluminescence spectroscopy. The use of Photoluminescence spectroscopy in calculating the age of latent fingerprints is novel and notable for its non-destructive nature, high sensitivity, capacity to offer speedy and quantitative results, and potential for both short-term and long-term aging studies. These characteristics render it a potent instrument in forensic science, augmenting the precision and efficacy of crime scene investigations. This research establishes a theoretical and practical basis for the current exploration and favorable outcome of this technique. Furthermore, it is necessary to address additional issues, such as fluctuations in environmental circumstances and sex-related factors, in further investigations.

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