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Review

Fingerprint composition and aging: A literature review

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ABSTRACT

Fingerprints have a key role in criminal investigations and are the most commonly used form of evidence worldwide. Significant gaps remain however, in the understanding of fingerprint chemistry, including enhancement reaction mechanisms and the effect of environmental variables and time on composition. Determining the age of a fingerprint is also a relatively unexplored area. A successful method, with reliable and quantitative estimates, would have numerous advantages. Previous unreliable methods have predominantly focused on enhancement success based on physical and chemical changes.

This review explores variations in composition due to donor characteristics and environmental variables, and identifies gaps for further research. We also present a qualitative and quantitative summary of the effect of time on composition. Kinetics are presented where known, with summary schematics for reaction mechanisms. Previous studies exploring methods for determining the age of a fingerprint are also discussed, including their advantages and disadvantages. Lastly we propose a potentially more accurate and reliable methodology for determining fingerprint age based on quantitative kinetic changes to the composition of a fingerprint over time.

Contents

Abbreviations: DFO, 1,8-diazafluoren-9-one; GC–MS, gas chromatography–mass spectrometry; LC–MS, liquid chromatography–mass spectrometry; MALDI-MSI, matrix-assisted laser desorption/ionisation mass spectrometry imaging; SIMS, secondary ion mass spectrometry; UV, ultra-violet light/radiation; VMD, vacuum metal deposition

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1. Introduction

Fingerprints are one of the most important forms of physical evidence in criminal investigations [1] and the most commonly used forensic evidence worldwide [2]. Fingerprint examination cases typically match or outnumber all other forensic casework combined [2], with approximately ten times as many cases solved using fingerprint evidence compared to DNA [2].

Previous research exploring fingerprint composition has been within three key areas [3]. Recent advances have focussed on the development of novel enhancement methods and the optimisation of existing reagents [4–11]. Secondly, research has explored intelligence gathering through individualisation from the intrinsic composition. Recent advances in this area are critically examined in this review, including the ability to identify donor age, gender, and race from fingerprint composition [12–26]. Extrinsic composition has also been explored, including drugs, cosmetics, and food contaminants [12,27–32], but is not discussed in this review. Readers are directed elsewhere for further information regarding contaminants. Thirdly research has explored the potential for fingerprint age determination through changes to composition over time [20,33–41]. There are currently no accepted analytical methods for reliably determining fingerprint age. Due to the unreliability of proposed methods, investigators have always distanced themselves from age determination, as speculation is subject to considerable error and is therefore highly dangerous to the reputation of an examiner [42]. The potential to determine a timeframe during which a fingerprint was deposited is however a highly relevant factor in criminal investigations. Convictions can depend on the ability to prove beyond reasonable doubt whether a fingerprint was deposited when a crime was committed or from a previous legitimate visit, as is often claimed by the defence team [1,2]. Several possible methods have been recently proposed, which have focused on physical and chemical changes to fingerprints over time, as well as the effect of these changes on subsequent enhancement with powders or chemical techniques.

This review aims to critically discuss recent findings regarding fingerprint composition and age determination, with a particular focus on the effect of time on composition. Novel developments in the identification of donor characteristics are also discussed, as well as the effect of environmental variables on fingerprint composition. Additionally, this review contains a critical appraisal of fingerprint age determination methods and we propose an optimum methodology based on quantitative changes to the composition of a fingerprint over time. Lastly this review identifies key gaps in scientific knowledge and discusses future requirements and perspectives for fingerprint research. Numerous terms including 'fingerprint', 'fingermark' and 'latent print' [43] have been used in previous research. To minimise confusion this review will use 'fingerprint' throughout, as chemical analysis has only been applied to fingerprints. Additionally, determination of fingerprint age is used to mean identifying when a fingerprint was deposited; opposed to aging a fingerprint or fingerprint aging, which is used to mean leaving a deposited fingerprint to change with time.

2. Fingerprint composition

A fingerprint is composed of sweat secretions transferred onto a substrate, resulting in an impression of the ridge pattern or fingerprint left behind [44]. Sweat composition has been studied extensively from a medical or dermatological view point [45–55], although fingerprint composition is more complex. Fingerprints contain a mixture of substances originating from the epidermis, the secretory glands in the dermis (in a combination of some or all of the three sweat types), intrinsic components including metabolites and traces of medications and drugs; and extrinsic contaminants, such as blood, dirt and grease, make-up, food contaminants, moisturisers, and hair care products [3,12,27–29,31,32,32,56]. The intrinsic and extrinsic constituents can vary significantly between individuals (intervariability), as well as from the same individual from day to day (intravariability) and at different times on the same day [24].

The intrinsic components of a fingerprint are comprised of 95–99% water [57] and organic and inorganic compounds forming a complex emulsion in a three-dimensional matrix [56]. The eccrine component of the fingerprint is composed of approximately 98% water, as well as organic and inorganic compounds [16,58], as shown in Table 1. Studies to date have quantified 20 amino acids in fingerprints [59–66], as shown in Table 2.

Sebaceous sweat is composed of numerous organic compounds, as shown in Table 3. The majority are lipids [56] including fatty acids, glycerides and long chain fatty acid esters; as well as squalene, sterols; such as cholesterol, and numerous lipid esters [16]. A summary of the organic and inorganic components from all three sweat glands is shown in Table 4.

The composition of a fingerprint is subject to numerous factors, which affect the initial composition at both deposition and after aging over time. These are simply demonstrated in the triangle of interaction [68], which demonstrates the relationship between fingerprint composition, the substrate and the environment, as shown in Fig. 1.

These factors affect fingerprint composition in two stages, classed in this review as the '*deposition stage*' and the '*aging stage*', as shown in Fig. 2, and result in a particularly complex and variable matrix [3].

The *deposition stage* is affected by donor characteristics, including age, gender, race and diet; the deposition conditions, including deposition action, contact time, angle and pressure; and the nature of the substrate, including porosity, curvature and surface texture [3]. Due to

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Table 1
Organic and inorganic constituents (average values) in eccrine sweat based on [3,58].

Organic (major)		Organic (trace)	Inorganic (major)		Inorganic (trace)
Amino acids	1.45 mg/L	Creatine	Chloride	3.5 g/L	Magnesium
Proteins	200 mg/L	Creatinine	Sodium	3.3 g/L	Zinc
Glucose	3.5 mg/L	Glycogen	Potassium	0.2 g/L	Copper
Lactate	3.15 mg/L	Uric acid	Iron	3.5 mg/L	Cobalt
Urea	0.75 mg/L	Vitamins	Calcium	2 mg/L	Lead
Pyruvate	0.079 mg/L		Bicarbonate	107 mg/L	Manganese
			Sulphate	10 mg/L	Molybdenum
Organic (lipids)			Phosphate	1.4 mg/L	Tin
Fatty acids	0.055 mg/L		Fluoride	69 μg/L	Mercury
Sterols	0.065 mg/L		Bromide	35 μg/L	
			Iodide	0.85 μg/L	

considerable variations between donors, deposition methods and substrates, the initial composition can vary significantly.

The *aging stage* affects composition immediately after deposition, producing the aged composition encountered at enhancement [3]. During the aging stage fingerprints are affected by the substrate, environmental conditions, such as temperature, humidity and light levels; the enhancement techniques, such as physical, physico-chemical or chemical methods; and the time elapsed since deposition, with longer aging periods resulting in greater degradation of fingerprint components. The final aged composition is therefore a combination of all of the factors from both the deposition and aging stages.

2.1. Variation with donors

Fingerprint composition can vary significantly between donors, due to differences including gender, age and race, as well as medication, psychological state, health, metabolism and diet [3].

Composition can be affected by gender, although there have been differing conclusions as to the significance of the variations observed [16,19,69,70]. Recent findings in small scale experiments indicate fatty acids tend to be present in higher concentrations in male donors, such as saturated C15, C16 and C17 acids [16,19,70], although wide variations in composition were observed due to the sampling protocol [16]. Studies have shown sterols and sterol esters tend to be more concentrated in female donors [58], as well as amino acids, such as alanine, glycine and serine [16,71], although this variation is not statistically significant as only 20 donors were sampled [71]. One study suggested wax esters differ in concentration between male and female donors [72], although this has not since been confirmed and wax esters remain largely unexplored. Larger investigations with increased donors have identified no "statistically significant gender effects" [19]. Additional large scale research is necessary to investigate these variations further.

Donor age can also affect fingerprint composition and considerable research has explored differences between children and adults [15,17,18,25,53,72–75]. Fingerprint composition can alter significantly between birth, puberty and old age [58], as fingerprints from donors prior to puberty are composed of eccrine related compounds, while fingerprints from more developed donors contain sebaceous components, similar to adult donors [23,52]. This difference in volatile components affects the longevity of the fingerprint [3], as a child's fingerprint may completely disappear in as little as 48 h, compared to a week for an adult fingerprint [15]. One study exploring the fingerprint composition of 6 father–son pairs suggested fingerprint longevity is affected by the rate of sebum excretion, which affects the concentration of all sebaceous compounds present in a fingerprint [17]. Studies have indicated this rate is affected by donor age, and causes variations in composition, including fatty acid concentrations, the ratio of wax esters to cholesterol, and the concentrations of cholesterol and carbonyl esters [17,76]. Organic compounds also change with donor age, as a high concentration of lipids have been identified in adult fingerprints, including squalene, wax esters and branched fatty acids [17]. These fingerprints are less volatile compared to fingerprints from child donors (pre-pubescent), which contain cholesterol, cholesterol esters, straight-chain fatty acids and long chain fatty acid esters [17], primarily due to "inactive sebaceous glands" [19]. However these conclusions were obtained from the differences between only 132 fingerprints from 6 children and 6 adults.

Puberty causes numerous changes to fingerprint composition, as the proportion of endogenously synthesised sebaceous lipids, such as squalene, wax esters, and Δ6 fatty acids increases, while the proportion of exogenous lipids, such as cholesterol, Δ9 fatty acids, and the unsaturated omega-6 C18 fatty acid (linoleic acid) decreases [3], as shown in Fig. 3. After puberty, findings indicate there are fewer changes until after middle age [58]. One recent study explored the fingerprint

Table 2
Amino acid composition in latent fingerprints (percentage relative to serine) based on [56,58,67].

Amino acids	Hadorn [64]	Hamilton [65]	Oro [66]	Average	St. dev.
Serine	100	100	100	**100**	0.0
Glycine	54	67	59	**59**	6.1
Ornithine	45	32	45	**41**	6.1
Alanine	35	27	28	**28**	5.4
Aspartic acid	11	22	22	**20**	5.9
Threonine	9	17	18	**16**	4.4
Histidine	13	17	14	**15**	1.8
Valine	10	12	9	**10**	2.1
Leucine	7	10	10	**9**	1.7
Isoleucine	6	8	8	**7**	1.2
Glutamic acid	12	8	5	**7**	3.9
Lysine	5	10	–	**8**	4.1
Phenylalanine	5	7	5	**6**	1.0
Tyrosine	3	6	5	**5**	1.3

Table 3
Organic composition of sebaceous secretions based on [3,58].

Organic (major)	Organic (trace)	
Triglycerides	30–40%	Aldehydes
Fatty acids	15–25%	Ketones
Of which:		Amines
saturated	50%	Amides
monounsaturated	48%	Alkanes
polyunsaturated	2%	Alkenes
Wax esters	20–25%	Alcohols
Squalene	10–12%	Phospholipids
Cholesterol	1–3%	Pyrroles
Cholesterol esters	2–3%	Pyridines
		Piperidines
		Pyrazines
		Furans
		Haloalkanes
		Mercaptans
		Sulphides

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Table 4
Constituents of gland secretions.

Source\constituents	Inorganic	Organic
Eccrine glands	Chlorides	Amino acids
	Metal ions	Urea
	Ammonia	Lactic acids
	Sulphate	Sugars
	Phosphate	Creatinine
		Choline
		Uric acid
Sebaceous glands	–	Glycerides
		Fatty acids
		Wax esters
		Sterol esters
		Sterols
		Squalene
		Hydrocarbons
		Alcohols
Apocrine glands	Iron	Proteins
		Carbohydrates
		Cholesterol

composition of 63 children between 2 and 11, and determined carboxylic acid salts are relatively stable within fingerprints compared to their esters [73]. It was suggested these acid salts could be used as an enhancement target in children's fingerprints after greater periods of time than currently possible [73].

These differences in composition indicate that it may be possible to distinguish fingerprints from donors of different ages. Some research has explored this possibility through the use of calibration curves to estimate a donor age range [17,18], although larger studies are required to support these conclusions with statistically significant results.

Minimal research has explored differences in composition between donors of different races, despite the potential for advantages in intelligence. A small scale study of 37 donors concluded that the ratio of several fatty acids to their respective methyl esters was significantly different for donors of different races [24]. The Δ6 FAME from the C18 monounsaturated omega-9 fatty acid was found to have a highly variable ratio to the respective unsaturated C18 fatty acid in each race classification [24]. The greatest difference was observed between Caucasian and African American males, although the study acknowledges this finding could be due to the small sample size. Further research with a larger donor set is needed to determine the "true legitimacy of the observed trends" [24].

Diet may also affect composition, as a recent small-scale study identified increased levels of alanine, glycine and serine from vegetarian donors, as well as higher concentrations of saturated C17 and both saturated and unsaturated C18 fatty acids [16].

Previous research has clearly identified the possibility of using fingerprint composition to determine several donor traits, but further large scale research is needed before composition can be used for accurate and reliable individualisation of donor characteristics.

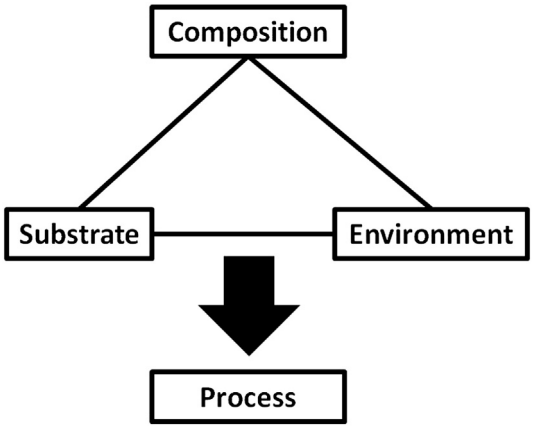

Fig. 1. Triangle of interaction based on [68].

2.2. Other variations

Fingerprint composition is also affected by the deposition conditions, the nature of the substrate and enhancement methods.

The deposition conditions affect the concentration of components within the fingerprint at the *deposition stage* [3,16,20,26,59,77,78]. These conditions include the pressure of the deposition, the duration of the contact, the dimension of the finger in contact with the substrate, the digit used for deposition, and how recently the donor washed their hands [3].

The substrate also affects fingerprint composition predominantly during the *aging stage* [3]. Research has identified high porosity creates adhesion forces between the substrate and the fingerprint, causing greater penetration of components into the substrate [79,80], as shown in Table 5. Findings indicate fingerprints deposited on non-porous surfaces are more susceptible to damage, due to increased exposure to environmental factors [80,81]. Chemical reactions can also occur between the substrate and fingerprint, such as metal corrosion by ionic salts [82]. One study explored 10 metals and determined high concentrations of chloride ions enhanced corrosion of noble metals, such as silver and gold [83]. Research suggests surface texture may also affect composition, as well as the physico-chemical structure, curvature, temperature, electrostatic forces and surface free energy [3], although there have been no studies exploring these factors specifically.

Enhancement techniques also affect fingerprint composition in the *aging stage*. The significance of the effect depends on the fingerprint age [3], although enhancement rarely takes place immediately after deposition. Several studies have explored the effect of indanedione, cyanoacrylate fuming and aluminium powder on composition [35,84–86]. One study focused on changes to squalene, cholesterol and the C14 saturated tetradecanoic acid within fingerprints from 7 donors [35]. Cyanoacrylate fuming and aluminium powder both had no effect on composition, although the powder contaminated the sample [35]. Indanedione enhancement contaminated the fingerprints due to the solvent and through reaction with amino acids. The enhancement reduced the concentrations of squalene, cholesterol and tetradecanoic acid [35]. An awareness of these changes due to enhancement is important, if analysis of composition is to be carried out after enhancement.

3. Effect of time

The composition of a fingerprint is highly variable [3], as significant changes occur after deposition through surface interactions and various decomposition and oxidative mechanisms [14]. Fingerprint composition can be separated into two parts:

1. the *initial composition*, at deposition where compounds within the digit residue are transferred to the substrate in the *deposition stage*
2. the *aged composition*, containing the remaining initial compounds and the degradation products following the *aging stage*.

Comparatively little research has explored changes between the initial and aged compositions and the rate of change with time [16]. Some research has explored the effects of time on reaction mechanisms and kinetics for specific compounds, including amino acids, proteins, fatty acids, squalene, cholesterol and wax esters [3]. This research can be divided into two areas:

1. The development of enhancement techniques [7,31,33,36–39,47,48,132,140]
2. The development of fingerprint age determination methods [7,36,39,51,67,79]

Increased knowledge of fingerprint composition allows for increased comprehension of the chemical mechanisms that occur between

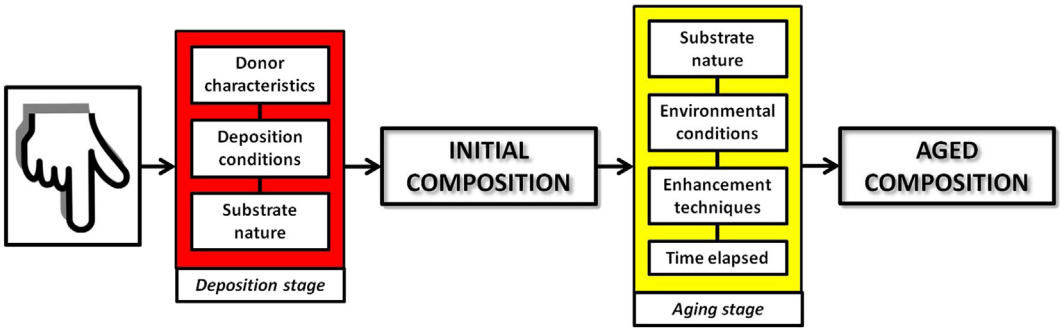

Fig. 2. The variables that affect fingerprint composition prior to and after deposition based on [3].

enhancement reagents and their target compounds, as well as how they interact within the 3D matrix. This allows for improvements to both existing reagents and novel developments with a high level of effectiveness for both fresh and old fingerprints [3]. Existing reagents can vary in their effectiveness on aged fingerprints, such as Nile red which is generally ineffective on fingerprints more than a few days old, due to loss of moisture preventing the partitioning mechanism from taking place [58]. Oil Red O is more effective on fingerprints less than four weeks old, due to diffusion of water-soluble components and degradation of fragile components with time [87]. Physical developer is more effective on aged fingerprints [87].

Fingerprint composition changes through various chemical, biological and physical processes resulting in the *aged composition*. The rate and method of the aging process can vary significantly [88], with initial compounds lost through various processes including degradation, drying, evaporation, metabolism, migration, oxidation and polymerisation [3]. Recent findings exploring these changes are critically discussed in the following sections.

3.1. Loss of moisture & mass

Over time the volatile components of a fingerprint evaporate out of the residue [89,90]. The fingerprint becomes increasingly viscous causing an initial large change in thickness [85]. Findings suggest the ridges become increasingly brittle and topographically irregular [85], through increased susceptibility to physical erosion from friction and air currents that cross the fingerprint surface [89]. As water is lost, the fingerprint becomes less receptive to chemical enhancement, as the remaining mixture of organic and inorganic compounds accumulates within a waxy layer, decreasing the surface area available for contact with enhancement reagents [74].

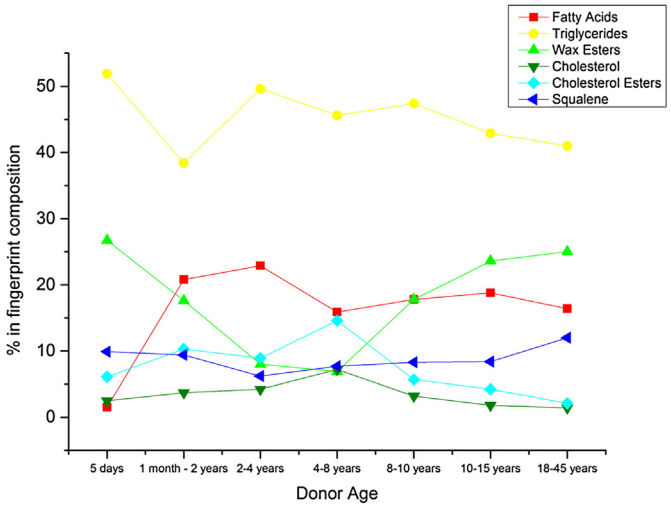

Fig. 3. Effect of donor age on fingerprint composition [58].

The mass of a fingerprint decreases over time, as volatile components are lost. A fingerprint can lose nearly 98% of its original weight within 72 h of deposition [91]. One study identified a loss of mass up to 85% after aging for two weeks [74], possibly due to loss of moisture [3]. An initial increase in detected material has been observed, possibly due to component decomposition [14], although this has not been confirmed in any other studies.

3.2. Organic & inorganic compounds

Both organic and inorganic compounds within a fingerprint are significantly affected by time, although little research has explored this effect [3].

3.2.1. Organic compounds

The majority of research exploring the effect of time on fingerprint composition has focused on lipids, discussed in Section 3.3, although some research has explored other organic compounds.

Research within the last decade determined *amino acids* are relatively stable over time, predominantly due to enhancement success with amino acid targeting reagents on older fingerprints [92]. It was suggested a relationship between the amino acids and the porous nature of the cellulose substrate may have assisted with the preservation [3]. Other research using paper substrates identified a decrease in mass of 0.083 mg/cm^2 to 0.046 mg/cm^2 after 236 days [93]. This indicates that although amino acids may not be completely stable over time, enhancement is still possible, either because amino acid concentration remains high enough for successful reaction or due to the high sensitivity of the enhancement methods. One study exploring the effect of time on *proteins*, used antibodies with albumin to enhance fingerprints on porous substrates. Successful enhancement was possible with both freshly deposited and aged fingerprints (up to 130 days) [94], possibly due to the stability of albumin on paper [3]. Research has also identified a significant decrease in concentration of u*rea* from 0.083 mg/cm^2 to 0.028 mg/cm^2 over a period of 236 days [93].

3.2.2. Inorganic compounds

Minimal research has explored the effect of time on inorganic components. The concentration of *chloride* was observed to decrease slightly from 0.223 mg/cm^2 to 0.217 mg/cm^2 over 236 days [93]. This change could explain why some enhancement techniques, such as silver nitrate which targets chloride ions, are less effective on older fingerprints [3]. This concentration decrease could also be due to diffusion into the substrate over time [95]. Preliminary research suggested changes to this diffusion pattern could be used for exploring fingerprint age [96], although diffusion is significantly affected by environmental variables, such as the storage conditions of the substrate [3], yielding unreliable highly subjective results. As the research findings determined this concentration decrease was not a significant change however, additional

Table 5
Effect of substrate on the absorption of fingerprint residue based on [3,79,80].

Substrate nature	Eccrine compounds	Sebaceous compounds	Examples
Porous	Rapidly absorbed	Slowly absorbed	Paper, cardboard, cotton, wood
Semi-porous	Absorbed slower than porous	Very slowly absorbed (slower than eccrine)	Glossy papers, plastics, laminated wood
Non-porous	Not absorbed — compounds remain on surface until degradation occurs		Plastics, glass, metal, rubber, painted wood

research is required to explore the effect of time further on inorganic compounds within fingerprints.

3.3. Lipids

After deposition, degradation processes result in changes to fingerprint composition over time [14,16]. Unsaturated compounds undergo several shortening and oxidising degradation processes, removing the unsaturated moiety. This removes a target for enhancement reagents, such as fluorescent tags [74]. Previous research exploring these changes has primarily focused on lipid components within fingerprints, as these tend to decrease significantly in concentration over 30 days, such as fatty acids, wax esters, triglycerides, cholesterol and squalene [14,16,20,58,74,97,98]. These various degradation and decomposition processes result in the formation of smaller oxidation products; the details of which are outlined in the following sections.

3.3.1. Fatty acids

Both saturated and unsaturated fatty acids tend to be present in all fingerprints [14]. *Saturated fatty acids* remain relatively stable over time, with one study determining C16 and C18 acids were relatively stable over a 60 day aging period [74]. The concentration of short chain saturated fatty acids was observed to increase over the first 15 days, through the degradation of longer chain fatty acids [14]. The C14 saturated acid increased in concentration over a 20 day aging period, followed by a decrease back to original levels or below after further aging [14], although only 5 male donors were used to obtain these conclusions. Other short chain fatty acids, such as C6, C8 and C9 acids are also present in greater concentrations in aged samples, where they undergo additional reactions to break down further or evaporate [56].

Several different theories have been proposed to explain this increase in fatty acid concentration over the first month [14], as shown in Fig. 4, although no theory has since been confirmed. One theory suggests an increase in two separate phases, with the initial increase over the first 7 to 10 days, followed by a slight decrease, and then phase two occurring between days 15 and 20 [14]. An alternate theory proposes a single concentration increase at different times depending on various factors, including the initial composition of the fingerprint [14]. A third theory proposes competing mechanisms of both production and loss, the equilibrium of which determines the concentration of fatty acids present [14].

It is suggested that the majority of fatty acids present in sweat originate from the hydrolysis of triglycerides in sebum [58]. This observed increase could therefore be due to the decomposition of wax esters and triglycerides, and the subsequent decrease could be due to the volatilisation or chemical degradation of fatty acids. This supports the theory of competing mechanisms at equilibrium within a fingerprint. Research exploring fatty acid production and loss is required to confirm which theory is correct, although enzyme lipolysis has been identified as a possible production mechanism, as triglycerides and methyl esters are broken down into fatty acids [99].

The concentration of *unsaturated fatty acids* has been observed to decrease with time, such as unsaturated C16 and C18 acids. Both acids are significantly reduced in concentration over a 30 day aging period, due to the unsaturated moiety being open to attack through aerobic and anaerobic degradation processes [74], as shown in Fig. 5. Research findings indicate anaerobic conditions cause hydrogenation processes, which transform unsaturated bonds, increasing the concentration of saturated fatty acids and decreasing the proportion of unsaturated fatty acids [100]. Aerobic degradation produces oxidised compounds, such as peroxide linkages, aldehydes and ketones, through a chain reaction process [101].

Tetracosane, a 24-carbon chain alkane, was identified in one study in samples of 'intermediate age' [14], indicating it may be an intermediate in the decomposition of longer chain compounds. No other study has identified this compound and further research is needed to explore possible formation and degradation mechanisms.

Saturated fatty acids remain in fingerprints for longer than unsaturated acids, predominantly due to the lack of a targetable functional group. Lower molecular weight compounds produced from fatty acid degradation are more volatile [58]. One study explored the decomposition of the unsaturated C18 acid over time and used the decomposition products to successfully distinguish three fingerprints of different ages using MALDI-MSI [97].

Further research exploring changes in fatty acid concentrations, as well as the various decomposition products that occur within a fingerprint is therefore clearly warranted, so as to fully understand the processes that occur over time after deposition.

3.3.2. Wax esters

Saturated lipids are less affected by change over time, resulting in higher levels of saturated compounds, such as wax esters [58]. This is predominantly due to the lack of a targetable functional group to aid

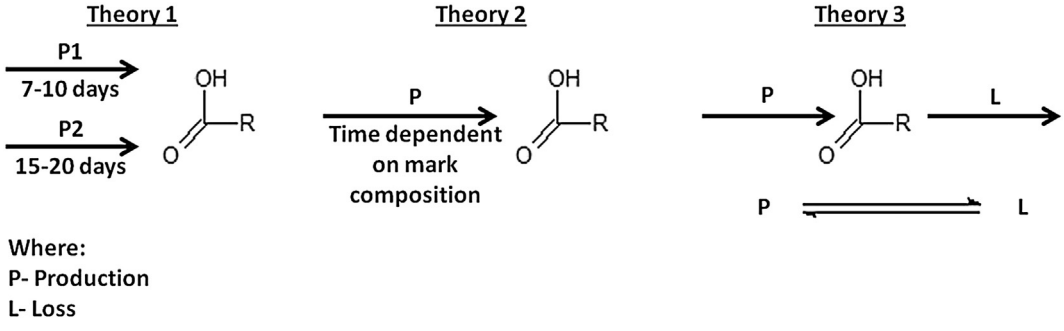

Fig. 4. Three theories for fatty acid concentration variations.

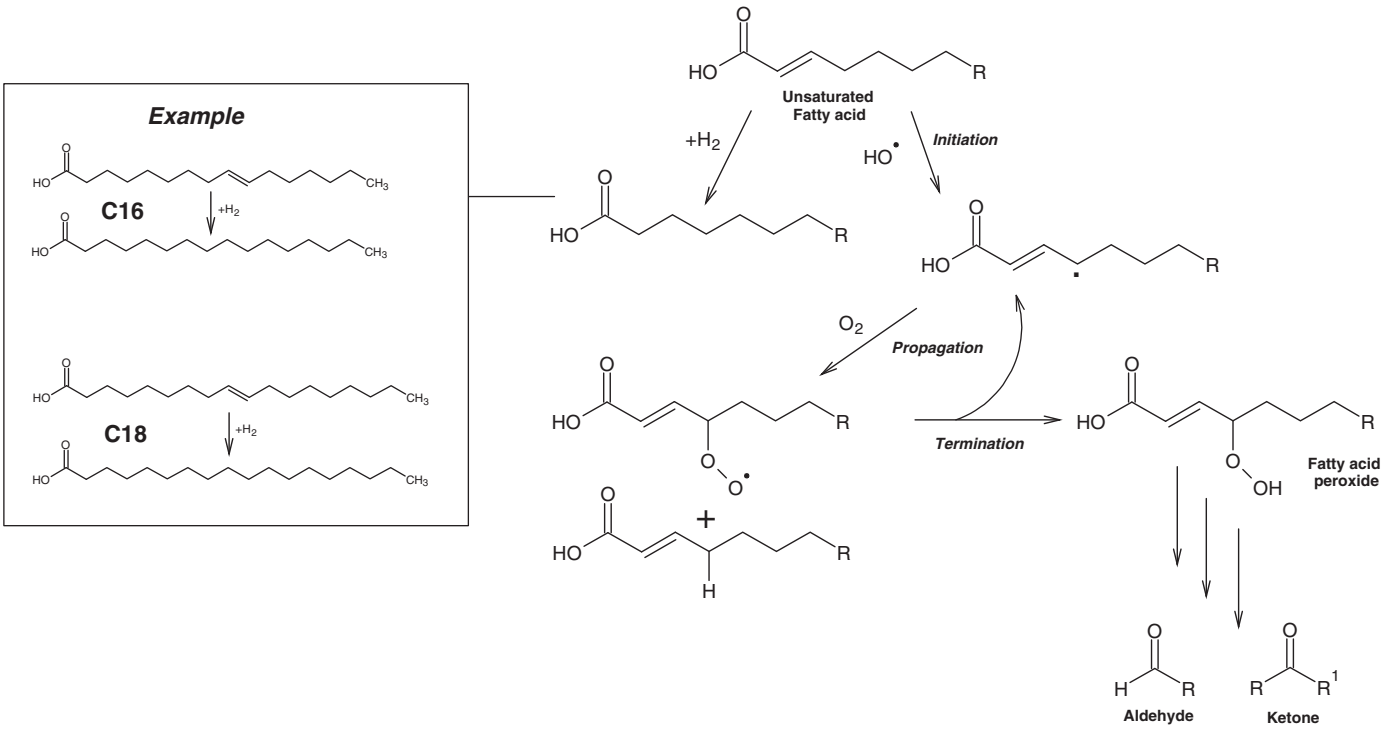

Fig. 5. Degradation of fatty acids through aerobic and anaerobic processes.

decomposition, which may also be why minimal research has explored wax esters in fingerprints. As an increase in the concentration of fatty acids could be due to the breakdown of wax esters [14], further research is clearly required to explore wax esters and their subsequent degradation products in more detail.

3.3.3. Triglycerides

Little research has explored the effect of time on triglycerides, although research in other areas has identified possible degradation mechanisms. Hydrolysis releases fatty acids from the glycerol backbone, resulting in a mixture of saturated and unsaturated fatty acids [100].

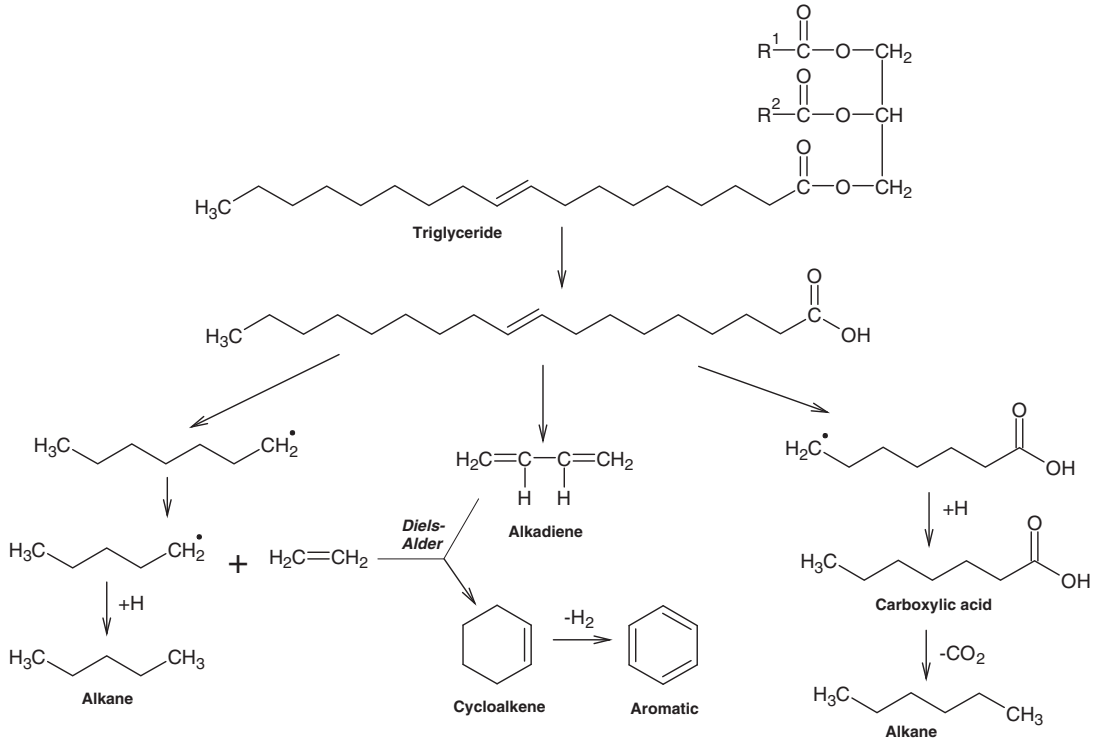

Fig. 6. Thermal decomposition of a generic triglyceride based on [100].

Thermal decomposition yields a number of compounds, including alkanes, alkenes, alkadienes, aromatics and carboxylic acids [100], as shown in Fig. 6. Decomposition mechanisms are particularly complex, predominantly due to the many structures and potential reactions of varying chain length triglycerides. Research to identify these compounds in fingerprints and their rate of formation would be particularly advantageous for the identification for novel potential targets for enhancement reagents.

3.3.4. Cholesterol

Cholesterol is the most abundant sterol in the body [16]. A small scale study detected cholesterol in all 24 fingerprints investigated and observed a decrease in concentration over time [20], although any decomposition products present were not identified. The focus of the study was on substrate effects and determined the rate of decomposition was dependent on substrate porosity, as concentration decreased at a slower rate on glass compared to polyvinylidene difluoride and did not show any change on porous microfilters over the time frame studied [20].

Few studies have investigated cholesterol decomposition [102], and research exploring changes to cholesterol in fingerprints have only observed a concentration decrease [20]. Research in other areas has determined cholesterol is susceptible to oxidation [103–105] and can form a large number of oxygenated products, including cholesterol oxides and oxysterols through 'autoxidation' [103,104]. The 8 most important [104] are shown in Fig. 7. Cholesterols can also be converted to cholesterol esters, through esterification of the A-ring hydroxyl by the carboxylate group of a fatty acid, as shown in Fig. 8.

One study determined the decomposition of pure cholesterol can be accelerated by triglycerides or fatty acids [104]. The study explored the effect of both saturated and unsaturated C18 fatty acids and determined that the cholesterol oxides formed; 3,5-cholestadiene and cholesta-3,5-dien-7-one, were different from those produced in the presence of triglycerides [104,105]. These oxygenated cholesterol products have not been identified in fingerprints of any age however.

The presence of cholesterol can also influence the decomposition of triglycerides [103], indicating that the stability of lipid components is influenced by numerous interactions among these components and/or their decomposition products. More research is clearly required to explore and identify cholesterol decomposition products within fingerprints, as well as the numerous interactions that occur with other compounds, due to the lack of current research in this area.

3.3.5. Squalene

Squalene is an unsaturated organic triterpene steroid precursor [74] and has been studied in some detail over the last decade [14,16,20,74,98]. Recent findings indicate squalene decomposes rapidly over time and is rarely detected in older fingerprints, such on glass substrates where it is almost undetectable after only one week [98]. The same study also used LC–MS to explore the effect of time and successfully identified a number of oxidation products [98]. The most fully oxidised forms were identified as hexanedioic and pentanedioic acids through oxidation in air [74,98], as shown in Fig. 9. Other intermediate products were identified including epoxides, ketones and a range of hydroperoxides. These included squalene mono-hydroperoxide (SQ-[OOH]) to the

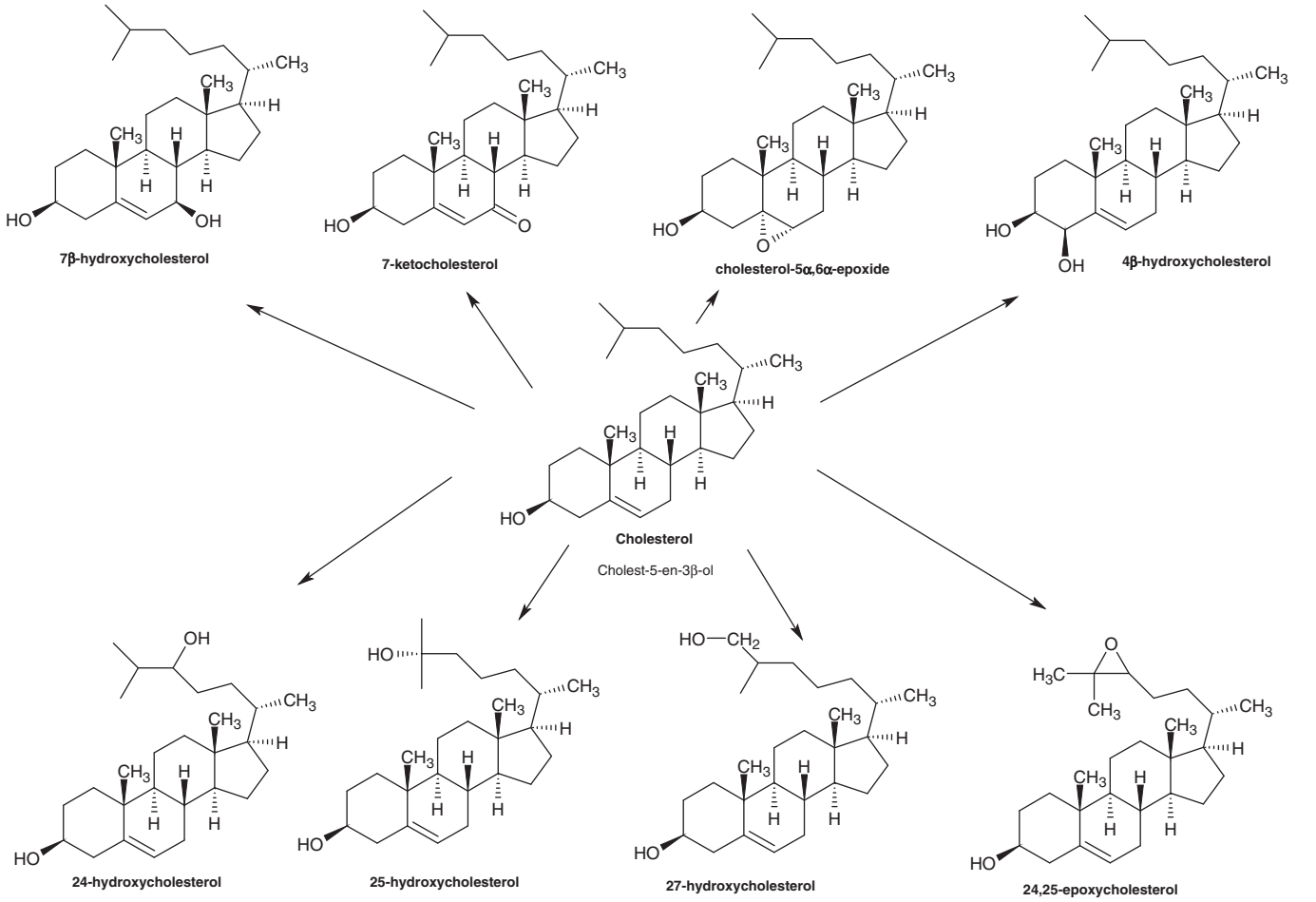

Fig. 7. Decomposition of cholesterol through oxidation to 8 key oxidised forms based on [103].

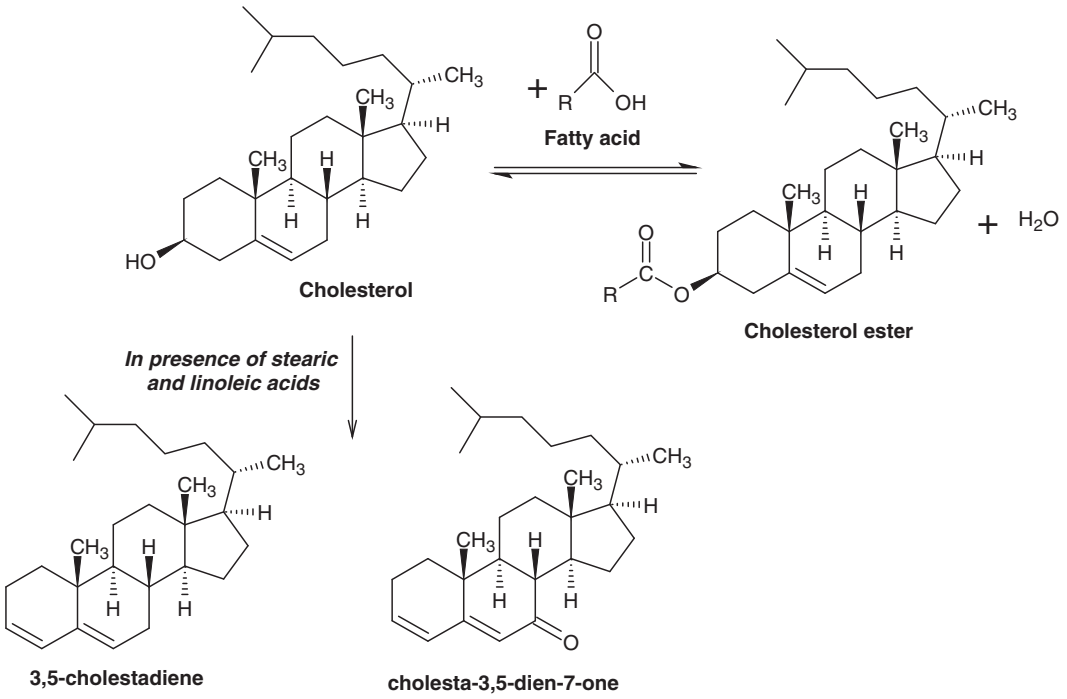

Fig. 8. Esterification of cholesterol to cholesterol ester and decomposition to 3,5-cholestadiene and cholesta-3,5-dien-7-one.

di- (SQ-[OOH]$_2$), tri- (SQ-[OOH]$_3$), tetra- (SQ-[OOH]$_4$), and squalene penta-hydroperoxides (SQ-[OOH]$_5$) [98].

Squalene tetra- and penta-hydroperoxides were still detectable after 20 days [98], indicating they are formed towards the end of the oxidation sequence. One study aged fingerprints for one month under ambient conditions and determined that 10% of each sample was composed of hydroperoxides [3,58]. Squalene epoxide in comparison was detected in freshly deposited marks and increased in concentration over 5 days from deposition [98]. It was undetectable after 7 days however, indicating squalene epoxide undergoes further decomposition [98].

Photo-oxidation was also identified as a decomposition method, as it produced very reactive, volatile products including malonaldehyde, formaldehyde, acetone and acetaldehyde [98], which are rapidly lost from fingerprints or undergo further decomposition reactions with other constituents. Acetaldehyde and acetone can be formed through a 6-methyl-5-hepten-2-one intermediate, which can also breakdown to form malonaldehyde [106]. Peroxides are formed through exposure to UV irradiation [107].

Squalene polymerisation through carbon – carbon bond formation has also been observed in one study, which identified the formation of a waxy solid and determined the presence of both higher and lower molecular weight products from the squalene parent compound [14,74].

Further research into the effect of time on squalene concentration, as well as the various decomposition and polymerisation products formed is clearly warranted, so as to fully identify and understand these changes.

3.4. Ratios of compounds

The initial composition of a fingerprint is used to determine the starting point of a potential aging curve [20]. This causes some difficulty when exploring the effect of time on composition, as there can be significant variation between donors. Research focusing on the amounts of specific components relative to each other is potentially more reliable and accurate for age exploration, as it does not require knowledge of the initial concentrations [20], which, in almost all circumstances, is not possible to establish. Successful identification of several compounds and observing their decomposition, allows for comparison of the concentration ratios, which can be used to explore the effect of time more accurately. One study used this method to compare relative concentrations of squalene and cholesterol through the relative peak areas [20]. The relative standard deviation of the ratio of the two (less than 20%) was significantly less compared to the standard deviation of the individual compounds (up to 80%) [20], indicating a more reliable comparison method. This also resulted in more reproducible results over time, as the squalene-cholesterol peak area ratio produced more accurate concentration changes over the first few hours of aging [20]. The use of compound ratios appears to yield more reliable data, which indicates further research is needed to explore this as a method, that has the potential to follow changes to the composition of a fingerprint in a more reproducible way than proposed by previous research [14,108].

Summary schematics of the decomposition processes identified in recent research, as well as degradation products of key compounds present in fingerprints are shown in Figs. 10 and 11. Reaction kinetics or rates of decomposition are not displayed, as these have not been successfully identified for most compounds of interest. Exploration of reaction kinetics requires significant further research, so as to identify reaction products and the rate at which they are formed.

4. Effect of environment

The composition of a fingerprint changes after deposition and is affected by three variables: *donor factors*, discussed in Section 2.1; *transfer conditions*, discussed in Section 2.2; and *environmental factors*, such as air circulation, atmospheric contamination, condensation, dust, friction (handling or other natural movement), humidity, light exposure, precipitation, temperature, ultraviolet and other radiation [14,34]. Fingerprints deposited at crime scenes are on a range of substrates both inside and are therefore subject to environmental variations, which can significantly affect how the composition changes over time.

4.1. Light exposure

Research has identified light exposure can affect composition. One study explored lipid changes over time under a number of different environmental conditions and determined fatty acids were lost less rapidly in dark conditions [26]. The concentration of the monounsaturated

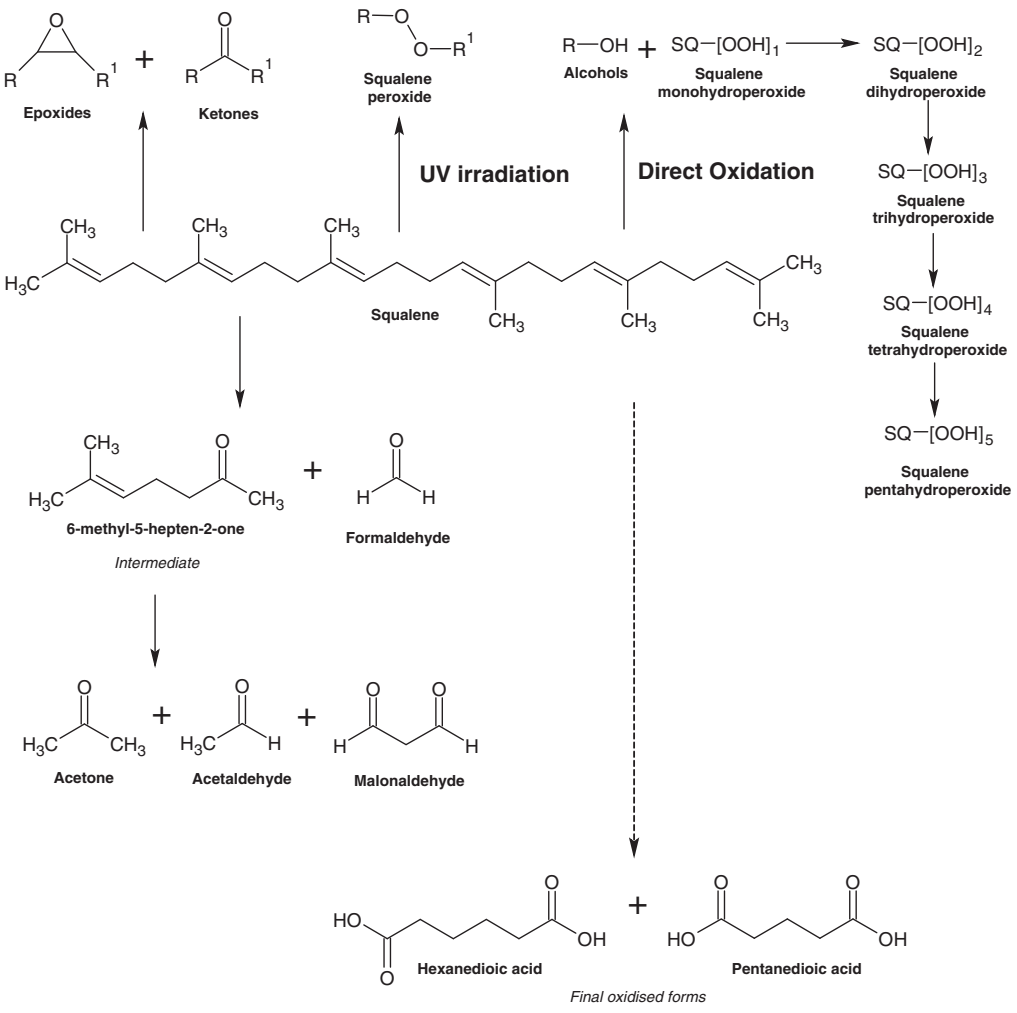

Fig. 9. Squalene decomposition through various processes including oxidation, UV irradiation and direct oxidation to hydroperoxides.

C18 fatty acid increased initially followed by a decrease in dark conditions, but not when aged in light conditions [14], suggesting different decomposition mechanisms occur under different levels of light [14]. Another study explored fingerprints over 1 month [108] and determined fingerprints in dark conditions showed a significant loss of cholesterol and saturated C16 and unsaturated C18 fatty acids [108], indicating decomposition mechanisms that are not light dependent. Results showed squalene concentration also decreased significantly over a 2 week aging period in light conditions [56,108], which is supported through the identification of photo-oxidation mechanisms [98], although squalene concentration decreased when aged in both light and dark conditions [14]. One study determined squalene is not detectable after 9 days in light conditions, compared to dark conditions where it is still detectable after 33 days, although at much lower levels than in freshly deposited fingerprints [14]. Several findings identified squalene decomposition occurs more rapidly in brighter conditions, particularly in the presence of UV radiation, such as in sunlight [14,16,98,107]. This is again supported by recent research determining a photo-oxidation mechanism, which results in the formation of squalene monohydroperoxide and squalene epoxide after only one day [98].

4.2. Temperature & humidity

Fingerprints can also be exposed to temperature and humidity after deposition, with higher temperatures increasing the rate of water loss [109].

Findings indicate high temperature results in increased degradation of amino acids compared to aging at room temperature [61,110]. The study explored pure compounds present in eccrine sweat and determined amino acids undergo thermal degradation as opposed to photo-degradation [110], as the seven amino acids monitored were all degraded after 3 min at 100 °C. These findings have not been replicated using fingerprints. Decomposition products have also been identified, including 3,6-dimethylpiperazine-2,5-dione from alanine and maleimide and 2,5-furandione from aspartic acid [61], as shown in Fig. 12. These decomposition products have been confirmed in several other studies and have been proposed as a possible source of fingerprint fluorescence after exposure to increased temperature [111,112]. One study identified the optimum temperature range to ensure successful enhancement using amino acid reagents as 20–35 °C [113]. The study concluded that if a fingerprint were exposed to heat for prolonged periods of time, enhancement would be much more difficult [113], presumably due to degradation of the amino acid target compounds.

Other compounds are also affected by temperature, such as urea which was identified to decompose more rapidly at higher temperatures [13] and esters which form lower molecular weight product compounds [13,73]. Acid salts are more resistant to higher temperatures and were still detectable, even after heating at 70 °C for 72 h [73]. The effect of temperature on cholesterol in fingerprints has not been explored, indicating further research to determine the effect of temperature on fingerprint composition is clearly warranted.

There has been very little research specifically exploring the effect of humidity on fingerprint composition, although studies suggest that the

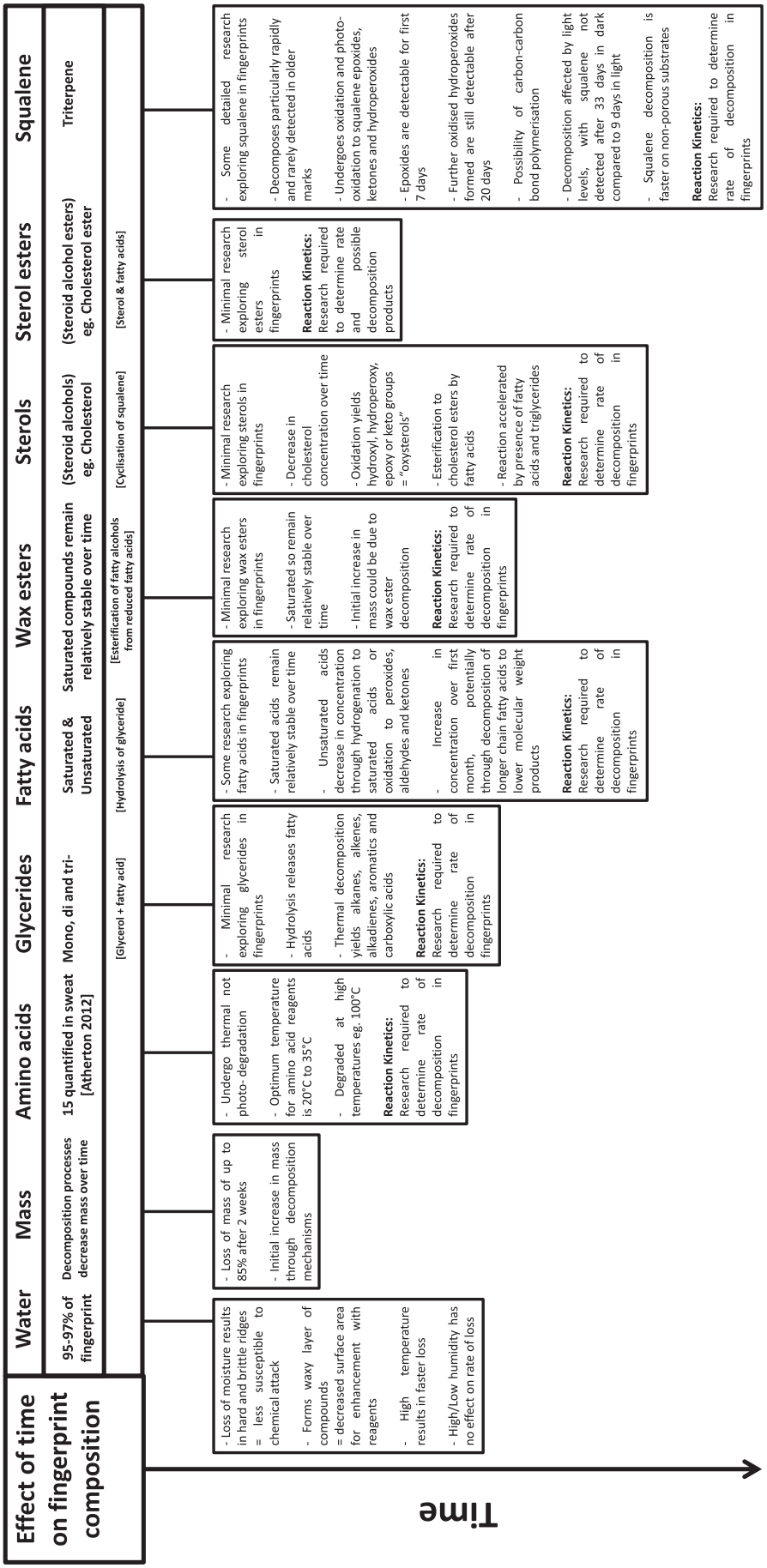

Fig. 10. Summary of effect of time on composition of latent marks.

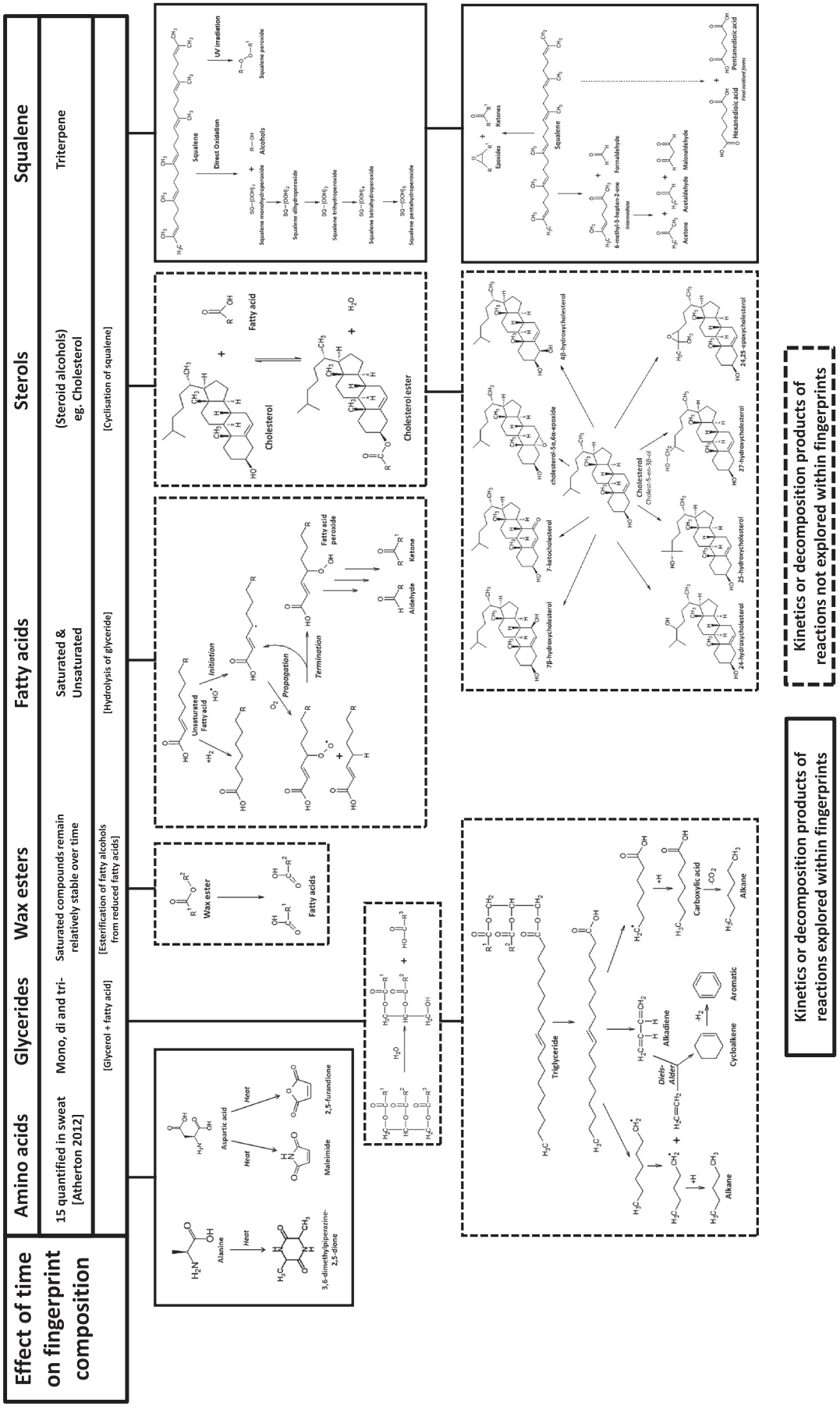

Fig. 11. Summary schematic of possible decomposition processes for compounds present in latent marks.

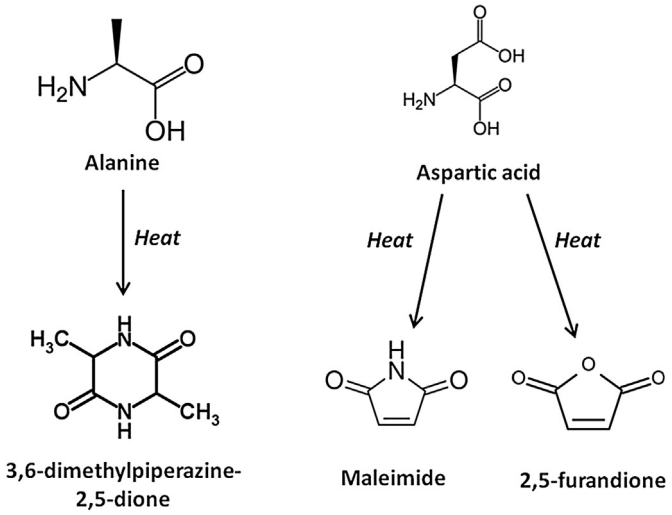

Fig. 12. Decomposition of alanine and aspartic acid under heat.

rate of water loss is not dependent on the relative humidity [85,90]. More research has explored the effect of humidity on enhancement. Research exploring amino acid reagents showed that DFO is less affected by humidity compared to ninhydrin or indanedione, as exposure to high humidity for 1 h did not affect the quality of the enhancement [93,114,115]. This may be due to the general sensitivity of the enhancement methods and the effect of humidity on porous substrates, rather than due to variations in composition or physical characteristics of the fingerprint. Other enhancement techniques are affected by humidity. The quality of fingerprints enhanced with silver nitrate is adversely affected by high humidity levels [93]. Cyanoacrylate fuming quality also varies with humidity, with eccrine constituents most influenced by humidity changes [114], as sodium chloride salt crystals absorb water into the fingerprint ridges at higher humidity [114]. Further research is needed to explore the effect of humidity on fingerprint composition so as to fully understand how composition changes over time [13].

4.3. Vacuum

Fingerprint enhancement can use vacuum or reduced pressure chamber methods, such as VMD. Fingerprint chemistry can also be explored using vacuum based methods, such as SIMS and MALDI [116]. It is important to identify the effect reduced or vacuum pressure can have on fingerprint composition, as changes in composition may affect the efficacy of subsequent enhancement [116].

One study identified a decrease in fingerprint mass when subjected to low pressure vacuum chambers, predominantly due to a reduction in water, as well as several fatty acids, fatty acid esters and squalene [117]. The mass lost was determined to be equivalent to 26% of the mass of the fingerprint, which the study concluded was equal to the equivalent of aging the fingerprint for approximately 5 weeks under ambient conditions [116]. One study identified fingerprints aged over 1 week showed reduced concentrations of the saturated C12 fatty acid, the unsaturated *trans*-C18 fatty acid and squalene under natural conditions [116]. Marks placed within the vacuum chamber, had significantly reduced relative concentrations of the saturated C14 and C15 fatty acids, indicating that vacuum conditions result in reduced concentrations of compounds that are still present under natural conditions after aging for 1 week [116].

This study shows exposure to vacuum pressure can significantly affect fingerprint composition. This is particularly relevant for enhancement techniques, such as iodine fuming, solvent black 3, basic violet 3 immersions and small particle reagents, which target the constituents most readily lost under vacuum conditions [78,116,118]. These changes to composition may result in a decrease in the quality of the enhancement if the fingerprint has been previously subjected to a vacuum system [116].

4.4. Other variables

Research has identified composition is also affected by other factors, such as air circulation, atmospheric contamination, condensation, dust, friction (handling or other natural movement), precipitation and ultraviolet and other radiation [14,34]. Minimal research has explored these variables in any depth, although they are generally accepted as of significance in composition studies [26]. Research into the effect of these various environmental variables on fingerprint composition is clearly required, so as to identify the changes that occur, as well as the respective rates.

5. Age determination

Determining the age of a fingerprint is a relatively unexplored area of fingerprint research. A successful method, with reliable and reproducible age estimates, would have numerous potential advantages for criminal investigations, as well as data privacy. There are currently no accepted analytical methods, although several methods have been previously proposed. These have predominantly focused on physical and chemical changes to fingerprints over time and their effect on subsequent enhancement with powders or chemical techniques. These previous methods are detailed in the following sections, as well as a discussion of the characteristics required for an optimum method, based on quantitative kinetic changes to fingerprint composition over time.

5.1. Previously proposed methods

Several different research groups have explored changes to fingerprints over time [13–15,20,33,34,58,73,74,88,93,95–98,108,119–126],and numerous attempts have been made to determine an accurate method for determining the age of a fingerprint. Some of the methods first proposed are particularly unreliable however, as they focused purely on changes to physical characteristics over time [127]. Research has predominantly focused on four main areas: the success of powdering methods, changes in fluorescence wavelengths and intensity, changes in ridge features, and electrical methods exploring the decay of electrostatic charge.

5.1.1. Powdering & enhancement methods

One proposed method explored the success of ridge detail development using powder, with the age estimation based on how well the powder adheres to the ridges, or the overall clarity of the enhanced ridge detail [34]. One key study explored fingerprints deposited on aluminium drinks cans against a crime scene fingerprint recovered using magnetic powder [128]. Fingerprints contaminated with different ratios of 'relevant contaminants' [34] were deposited on a number of test cans and aged in an attempt to replicate the crime scene conditions [34]. The experimental setup followed the premise that the original enhanced fingerprint obtained was composed of either natural perspiration or food contaminants recently handled by the subject and thereby, failed to recognise other constituents within the fingerprint, as well as the possibility of variations to those compounds over time [129]. The experiment ignored entirely possible differences to fingerprint composition between donors and its subsequent changes over time, the viscosity of the fingerprint, and the quantity of the residue, without which, it would not be possible to determine the evaporative rate of the fingerprint [129]. It was stated in court that the crime scene fingerprint was between 24 and 48 h old [130], although this conclusion was regarded as highly unreliable as the composition of the fingerprint was only 'partially recognised' [129].

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One early study used enhancement success to explore the effect of temperature and humidity on fingerprints [127]. Fingerprints were deposited on glass substrates and aged at varying humidity (32%–98%) and temperature (20–30 °C) for seven weeks, and enhanced across the aging time [42]. The study determined humidity had the greatest effect on the quality of enhancement, with high humidity producing the lowest quality of enhanced ridge detail [42]. The temperature range explored was reported to have no effect on enhancement success, indicating that loss of moisture was not a significant factor in the enhancement process [127]. No conclusions identifying fingerprint age were obtained however. A subsequent study explored the effect of environmental variables on fingerprints deposited on a glass window over time [131]. The fingerprints were exposed to heat, cold, humidity and dust and were enhanced with powder and lifted for comparison over three days and after three months [42]. Comparison of the quality of the enhanced fingerprints determined no differences, either visually or microscopically, for any of the ages of fingerprints recovered [131], demonstrating the unreliability of attempting to determine fingerprint age purely on visual differences or the success of enhancement. Environmental variables were also explored in an extensive study using a database of 20,000 fingerprints enhanced using powders from numerous substrates under a selection of environmental conditions [132]. The age of recovered fingerprints was determined through comparison to the database and identifying specific characteristic properties within the ridge detail that were observed to change over time. Although this method identified the importance of exploring the effect of environmental and substrate variables, this method still relies on physical enhancement success and does not take into account fingerprint composition, or possible variations between donors.

A recent study explored fingerprint degradation through exposure to specific monitored laboratory conditions [37]. A number of variables were explored, including temperature, humidity, air currents, light levels and different substrates over 6 months, as well as the difference between eccrine and sebaceous fingerprints [37]. The aged fingerprints were enhanced using a titanium dioxide-based powder and results indicated that fingerprints degraded similarly in both light and dark conditions, with sebaceous marks on glass being the most reliable for identification after a longer aging period [37]. Age estimations for the recovered fingerprints were not proposed however, as enhancement using powders is particularly inaccurate, as many previous studies have determined [42,127].

Some research has explored the effect of time and environmental variables on other enhancement methods. One study explored the effect of water on fingerprints on porous substrates, as a potential method for determining the age of a fingerprint. Fingerprints of different ages were immersed in water prior to chemical enhancement using solvent black 3 [133]. The fingerprint age had little effect on enhancement success however, with only the length of submersion and the resilience of the substrate to moisture being the determining factors [133]. This is not that unexpected, as solvent black 3 targets sebaceous fats within a fingerprint [9], which are less subject to change over time.

The clarity of an enhanced aged fingerprint is the result of the original quality of the mark, rather than the result of changes over time [120,127,134]. It has been clearly shown that using the intensity of a successful enhancement as a guide for age estimation is an unreliable approach, as a fingerprint can “last for weeks at rather extreme conditions and still be easily detectable” [127]. Additionally, this study and several others, report the unreliability of attempting to ascertain age from a microscopic examination, due to the lack of any age dependent processes that affect enhancement, particularly using powders [127,135].

5.1.2. Fluorescence

An alternative method explored the red-shift change in fluorescence wavelength from fingerprints over time. Studies have identified recently deposited fingerprints exhibit green or yellow fluorescence compared to orange fluorescence in older fingerprints [122,124]. Although using the degree of red-shift indicated a potentially quantitative way of determining the age of fingerprints, findings showed variations between donors and over time were too large to allow for accurate age estimations [124]. A recent study was successful in estimating fingerprint age through repeated measurements over several days to obtain multiple measurements for an aging curve [136]. Age estimations were possible up to three weeks after deposition, with a median uncertainty of 1.9 days. The major drawback from this study however, was the inability to estimate ages for every fingerprint. 77% of female and 27% of male fingerprints displayed insufficient fluorescence to attempt the method and of those with sufficient fluorescence, only 55% of those met the criteria for age estimations [136]. Additionally no age estimations were possible for female donors, which the study attributes to the ‘lower excretion of skin components by women’ [136]. One possible advantage of this method however, is that it accounts for substrate effects due to the repeated measurements over several days.

Early research exploring fluorescence to determine fingerprint age was deemed unreliable [42] predominantly because it failed to take into account variations in composition between donors and over time on the fluorescence produced. Repeated measurements over several days to were used to overcome this variation [136], although this method does not take into account the presence of contaminants. Contaminants are frequently present in fingerprints and can dominate the fluorescence produced. Relying on the intensity of the fluorescence or the red-shift of natural constituents alone is unlikely to provide a reliable method for age determination.

One possible approach could be to explore the fluorescence produced by specific compounds and their subsequent degradation through oxidation at known wavelengths, superficially explored by [136]. A ratio of fluorescence from several compounds could be used as a more reliable method for age determination, as this would allow for a single measurement to be taken, as opposed to repeated measurements over several days. Significant further research is therefore required to minimise the potential errors to age estimations due to contaminants, variations in fingerprint composition, as well as the effect of environmental variables affecting the rate of oxidation.

5.1.3. Changes in ridge features

Some research has explored the effect of time on ridge features as a potential age determination method. One very basic method used the rate of healing of a wound on a finger compared to the impression in a deposited fingerprint [137], although this has very obvious limitations for real world applications. A number of studies have explored the degeneration of ridge features and width, with a particular focus on changes to individual pores over time [138,139]. One study identified a decrease in ridge width over 180 days. Indoor conditions showed a decrease from 0.32 mm to 0.26 mm and from 0.30 mm to 0.24 mm for fingerprints aged outdoors [139]. However there were significant overlaps between aged fingerprints, reducing the reliability of the method. Donor blood type was also identified as an influencing factor, as the rate of degradation of both DNA and epithelial cells decreased through blood groups O, A, AB and B [139]. This effect has not been identified in any other study and the exact implications are not fully understood.

Recent research has used technological advances to observe changes to ridge patterns with time on a hard drive platter [38]. Research has used a high-resolution non-invasive chromatic white light optical sensor [39,40] combined with pattern-recognition techniques to determine an aging curve [39,138]. Scanned fingerprints were separated into two categories based on the quality of the ridge detail — younger or older than 4 days [39]. The average error of approximation was determined to be between 13% and 40% [41], indicating further research is required to establish the overall reliability of the method [138], as well as the applicability of the method on real-world substrates.

Recent research has determined a method for age determination using physical characteristics is less viable, as ridge detail is highly

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dependent on the original fingerprint [138]. An age determination method based on physical characteristics requires knowledge of the initial ridge width, pore size and distribution throughout the fingerprint [138], which is, in almost all circumstances, not possible. Therefore, although these approaches can provide age estimations, they are either focused on very limited scenarios or do not produce sufficiently reliable results to be successful as a reliable methodology for fingerprint age determination [40].

5.1.4. Electrical methods

An alternate methodology focuses on the electrical properties of a fingerprint, in terms of changes to electrostatic charge with time [140,141]. One proposed method relies on contact electrification, which studies suggest is independent of both the subject and the transfer method [142]. The decay of the fingerprint charge is thereby only affected by the physical properties of the material and the environmental conditions [142]. The study explored this decay over 14 days and determined image definition remains while the overall level of charge decreases with time [142]. The study found 'very old fingerprints were not visible using charge imaging' [142] and suggested that an estimation of age might be possible for recently deposited fingerprints. One possible option would be to obtain two measurements to produce an aging curve. However, this method is currently limited by very long scan times, a sample area of only 5 μm, and by the substrate; the study explored fingerprints deposited on thin insulators, such as polytetrafluoroethylene (PFTE) and acknowledges issues with thicker samples [142]. Further research to explore this method is clearly required, so as to investigate the charge decay rates on a range of different materials, as well as from different donors.

5.2. Characteristics of the optimum method

Previous research exploring age determination has identified wide variations in a fingerprint's ability to survive under a variety of environmental conditions [42]. A number of unreliable methods have been previously proposed, based on the rate or success of enhancement [42,127], or observations of changes to ridge detail over time. Research has suggested three ways to explore fingerprint age determination [34]: the physical appearance of the developed/undeveloped fingerprint, identifying the environmental factors over a given period of time, or the identification of changes in chemical constituents [34]. The first two methods suffer from the difficulty of reproducing the original conditions [142], meaning the third is considered to be the only viable method [34,42]. One possible approach is to explore how the concentrations of specific components within the fingerprint change over time, through various decomposition processes. The exploration of chemical changes over time is regarded as the most realistic method for developing an accurate and reliable method that will be universally accepted to determine the age of a fingerprint [34].

The initial composition at the point of deposition is a crucial factor, as it is the starting point of an aging curve [20]. Without this knowledge, it is difficult to make definitive conclusions regarding exact changes to composition [14,20]. Additionally, previous research discussed in Section 2 has demonstrated fingerprint composition can vary significantly between donors, as well as due to the effects of environmental variables [14,17,19,24,37]. However this can be overcome through exploration of the relative amounts of constituents and key compounds inherent to fingerprints. Identifying the subsequent decomposition products allows for the potential development of a reliable method [20]. A number of studies have explored the reaction kinetics of decomposition for several compounds within fingerprints [14,16,20,98], the relative amounts of which could be used to produce a method to determine the age of a fingerprint.

5.2.1. Potential compounds for fingerprint age determination

A number of studies have explored variations in lipid concentration over time [14,16], as discussed in Section 3.2. Research has determined unsaturated compounds, decrease more rapidly over time. Fatty acids reduce significantly in concentration over 30 days [74]. Minimal research has explored reaction kinetics, in terms of the rate of decomposition over time, although some decomposition products have been identified. Tetracosane was identified in samples of moderate age [16], as well as volatile lower molecular weight breakdown products [56]. The decomposition of oleic acid has been explored using MALDI-MSI and several epoxides, alcohols and aldehydes were identified [97]. Further research exploring other lipids is required, so as to fully investigate the decomposition and reaction kinetics under a number of different environmental variables.

Eccrine constituents could also be used for age estimations, as one study monitored leucine over 236 days and identified a decrease to approximately 55% of the original concentration [67]. Another study explored the effect of temperature on amino acids and determined higher temperatures result in reduced enhancement [143], possibly due to loss of material. This was supported by a more recent study that identified the optimum temperature range as 20 °C to 35 °C and stated that if a fingerprint were exposed to heat for prolonged periods of time, enhancement would be much more difficult [113]. Recent research determined amino acids undergo thermal degradation, as opposed to photo-degradation [110] and decomposition products for some amino acids have been identified. High temperature pyrolysis-GC–MS at 500 °C was used to identify 3,6-dimethylpiperazine-2,5-dione and maleimide as decomposition products of alanine and 2,5-furandione as the decomposition product of aspartic acid [61], as shown in Fig. 12. The same decomposition products have been identified in recent studies and have been proposed as a possible cause for fingerprint fluorescence after exposure to increased temperature [111,112]. Further research is needed to identify the kinetics of these decomposition reactions, if they are to be used for a reliable quantitative aging methodology.

Cholesterol could be a suitable target compound for fingerprint age determination, as it has been observed to decrease in concentration over time [20], indicating the presence of decomposition mechanisms. Research in other areas, as discussed in Section 3.3.4, has determined cholesterol can form multiple products through oxidation [103]. The identification of these products within a fingerprint with the rate at which they form could allow for an age estimation method.

Squalene has been explored in some detail within fingerprints and a number of decomposition products have been identified [14,16,20,74,108], as discussed in Section 3.3.5. The concentration of squalene decreases rapidly over time and is rarely detected in older fingerprints [14,98], making it a potentially useful target compound for age determination. The presence or absence of squalene epoxide could also be used for age estimation, as the concentration increases up to 5 days after deposition, but is undetectable after 7 days [98]. A range of hydroperoxides are also produced over time, including the main oxidation product squalene mono-hydroperoxide to squalene penta-hydroperoxide, through the di-, tri- and tetra-hydroperoxide versions [98], as shown in Fig. 9. Findings showed squalene tetra- and penta-hydroperoxides were still detectable after 20 days [98]. This is particularly useful for age estimations, as the concentrations of squalene, squalene epoxide and squalene hydroperoxides could all be compared. A recently deposited fingerprint would have greater concentrations of squalene, squalene epoxide and the mono-hydroperoxide form compared to an older fingerprint, which would have lower concentrations of squalene and the epoxide, and higher concentrations of several hydroperoxides. More research is required to explore squalene reaction kinetics in greater detail, as well as to determine the effects of substrate interactions and environmental variables, so as to establish a reliable methodology for determining the age of a fingerprint.

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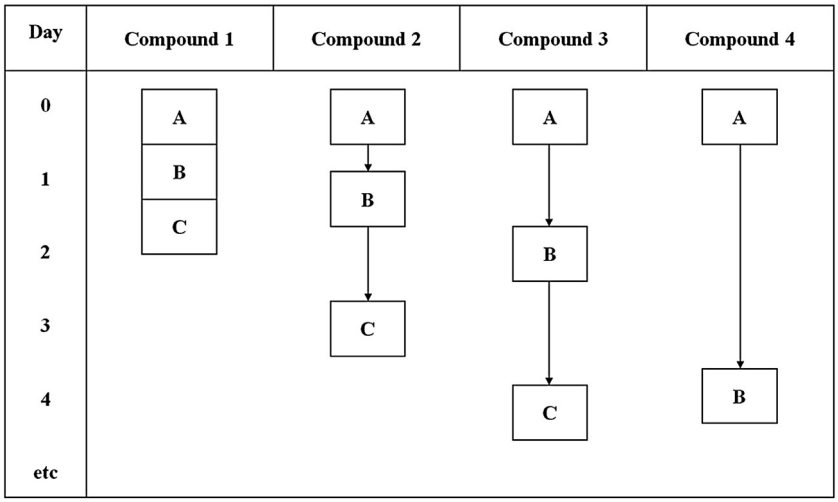

Fig. 13. Graphical representation of proposed aging method with 4 compounds with different rates of decomposition.

5.2.2. Points to consider

We propose the optimum method for age determination is likely to involve monitoring changes to the composition of a fingerprint over time, through the exploration of key constituents within the recovered fingerprint. Detailed knowledge of the precise reaction rates of various decomposition mechanisms, would allow for the concentrations of starting parent compounds to be compared to those of the decomposed products. Knowledge of several rates from decomposition mechanisms would increase the reliability of the method, as the parent to product ratio could be determined for several different decomposition mechanisms within the fingerprint eg. squalene, cholesterol, fatty acids. The age estimation would be based on the concentrations of several different chemical species detectable within the fingerprint, as shown graphically in Fig. 13, all with different decomposition rates and affected differently by environmental variables. This would yield a significantly more accurate method than a focus on the degradation of a single chemical species. The decomposition mechanisms explored would need to be predominantly driven by exposure to oxygen, with minimal variations in decomposition rates through exposure to UV radiation or fluctuations in temperature; alternate mechanisms would be too inconsistent to establish a reliable methodology.

The effect of environmental variables and substrate characteristics on the rate of decomposition for all the compounds of interest would need to be thoroughly explored. A statistical model could then be developed to determine fingerprint age based on known environmental factors, such as an approximate temperature range, or duration of light exposure. Using this model, an age or age range could be provided, based on the concentrations of specific constituents within the fingerprint. The model would also allow for the use of statistical probability to quantitatively estimate fingerprint age to a certain confidence level, as the estimate would be based on known and reproducible decomposition mechanisms of specific key compounds.

Current analytical methods often require preparatory steps, such as derivatisation for mass spectrometry and are destructive. For the proposed optimum method to be implemented into current practices, it would be necessary to identify a non-contact quantitative analytical method, which could non-destructively determine the concentrations of specific compounds within a fingerprint. The ridge detail of the recovered fingerprint would then be preserved for comparison against suspect fingerprints.

One limiting factor with this method is the use of enhancement reagents which target specific constituents within the fingerprint, thereby altering the composition. This composition change would need to be determined, although the best approach would be to identify compounds that are unaffected by the various enhancement processes currently in use. This would allow the proposed methodology to be reliably applied after all required enhancement procedures.

6. Conclusion

Fingerprint research has been carried out for many years, but there remain significant gaps in scientific knowledge. Future advances should therefore remain a priority. The exploration of the effect of time on composition is one such gap. Further research allows for the exploration of new and novel enhancement reagents targeting previously unknown compounds within a fingerprint, as well as potential improvements to existing methods and formulations through a greater understanding of reaction mechanisms. Donor characteristics could also be determined, which would greatly benefit intelligence gathering. Current findings can determine donor characteristics, such as the gender of the individual [19], race characteristics [24] and donor age [17]. This suggests the eventual possibility of profiling an offender, purely from the chemical content of a recovered fingerprint.

Intelligence gathering would also benefit from the knowledge of when the fingerprint was deposited. There is no currently accepted method for determining the age of a fingerprint. Previous research, as summarised in Table 6, has so far failed to identify a method that is sufficiently reliable and accurate across the wide range of variables commonly encountered. One possible approach is to explore the effect of time on fingerprint composition. This review proposes identifying specific components and their rates of reaction for decomposition mechanisms. Identification of the degree of decomposition present in a recovered fingerprint would allow for age estimation. Decomposition mechanisms that are driven by oxygen exposure would be advantageous, so as to reduce the variability in age estimation due to the effect of other environmental variables.

Naturally considerable research is still required, but we believe this method has merit as a way to quantitatively determine the age of a fingerprint across a range of substrates, as the flexibility of the methodology could accommodate such a complex set of variables.

7. Future perspectives

As discussed, further research exploring fingerprint composition is necessary to advance both understanding and the role of fingerprints in criminal investigations, as well as to potentially develop a reliable age determination methodology. The following sections discuss 7 key areas identified as in need of further research and discuss potential future developments.

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Table 6
Summary of previously explored aging methods with proposed best approach methodology.

Aging methods	Previous methods				Optimum method
	Powdering & enhancement methods	Fluorescence	Changes in ridge features	Electrical methods	Analysis of composition
Description	Degree of success of development of ridge detail using powder	Exploration of repeated measurements and red-shift change over time	Observation of changes through degradation to ridge features using optical sensors	Observation of electrostatic charge and decay remaining on surface after deposition	Exploration of chemical changes to fingerprints over time through comparison of parent compounds and degradation products
Advantages	Simple observations of degree of success of enhancement	Potentially quantitative, non-destructive method	Simple observation of ridges to estimate age, non-destructive	Independent of subject and transfer method	Qualitative and quantitative, reproducible, potentially non-destructive if method identified
Disadvantages	Unreliable — no account for variations in composition between donors, presence of contaminants, or changes to composition over time	Unreliable — no account for variations in composition between donors, changes over time or fluorescent contaminants	Unreliable — large average errors with preliminary results, no account for variations in composition between donors or over time	Minimal research carried out and limitations of technique not yet known	Relatively unexplored — large amount of research required to determine effect of substrates. Potentially destructive method required for quantitative analysis
Further research	Unlikely due to unreliability of method and numerous studies identifying major unreliability in the method	Exploration of fluorescence ratios of key compounds within fingerprints	Exploration of changes on range of substrates with large scale tests required	Exploration of decay on range of substrates with large scale tests required	Exploration of decay on range of substrates with large scale tests required, as well as the effect of enhancement reagents on fingerprint chemistry

7.1. Key areas to explore

For the identification of donor characteristics from composition and for the exploration of compositional change over time to be a viable aging method, there are several key areas that need to be fully explored:

1) Identification of decomposition products

To fully understand the chemistry of fingerprints, the reactions and mechanisms that occur over time need to be determined, as well as all the possible reaction intermediates and final end products.

2) Determination of the reaction kinetics

The kinetics of the chemical breakdown reactions for the identified compounds and their intermediates need to be determined. From this, several compounds can be selected which have sufficiently different decomposition rates, so as to identify the fingerprint composition at specific time intervals after deposition.

3) Determination of compound ratios

Specific compounds and their decomposition products can be used to produce reagent-product ratios. The effect of time on these ratios can then be explored, which will allow for more accurate age estimations, compared to concentration changes of single compounds alone.

4) Determination of the effect of environmental variables and substrates

Fingerprint chemistry is affected by a multitude of variables, which affects the decomposition kinetics. Exactly what these effects are needs to be determined, so as to identify how composition is affected and to further develop an accurate model for age estimations.

5) Determination of the effect of chemical enhancement

To explore donor characteristics and age estimations in a real-world application requires the exploration of fingerprint composition after chemical enhancement. The effect of individual and numerous sequentially performed enhancement processes needs to be explored, so as to identify changes to fingerprint chemistry. This allows for improvements to be made to age estimations, as the model can be additionally developed to include the effect of enhancement.

6) Large scale tests to explore significance of findings

Lastly large scale blind tests and operational trials are required, so as to fully explore the relationship between composition and donor traits, and to test the model for determining fingerprint age. In both cases it is important to determine the significance of an identified trend, as well as identify the accuracy, precision and reliability of the proposed method for determining the age of a fingerprint.

7.2. Potential future developments

One possible benefit to increased knowledge of fingerprint composition and the changes that occur with time is the development of novel enhancement reagents, which could be specifically tailored for determining fingerprint age. This could occur in two ways. Firstly, an enhancement reagent could only target fingerprints of a specific age, through selective chemical reaction with compounds that are only present for a known duration and then rapidly decompose. Selective enhancement using squalene or squalene epoxide as the target compound could be explored, as both are detectable in freshly deposited fingerprints but decrease in concentration over time, with squalene epoxide undetectable in latent marks after 7 days. This would allow an enhancement reagent to selectively develop fingerprints relevant to when a crime was committed. One example of this could be a shop burglary, which occurred at a known time and where potential surfaces regularly receive a large volume of deposits. Successfully distinguishing recent from older background fingerprints would be of significant benefit, as it would reduce the volume of evidence requiring processing and result in faster intelligence for suspect identification.

Secondly, an enhancement reagent, or multi-step enhancement process, with the ability to selectively enhance numerous compounds could be used. As specific composition changes with age, this could be used to develop a reagent that reacts with several different components present in the fingerprint at different times to form different coloured complexes. This would allow for immediate assessment of the fingerprint age, purely based on the colour of the enhanced fingerprint. Squalene hydroperoxides could be potentially used, as they are potential receptors for chemi-luminescent development techniques and are present

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in different concentrations over time, as discussed in Section 3.3.5. Potential reactions with contaminants would naturally need to be investigated to reduce the possibility of incorrect age determination, although a highly selective chemical method would reduce the possibility of false colour enhancement occurring.

Currently both of these methods remain unexplored, but the identification of potential benefits to enhancement through increased fingerprint knowledge clearly highlights the requirement for further research. Until a methodology for age determination has been fully explored and is routinely used in investigations however, attempts to determine the age of a fingerprint should be explored with care, so as to prevent damage to the credibility of an examiner or the use of fingerprints as evidence in criminal investigations generally.

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