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Preliminary study for aging of latent fingerprints on nonporous substrate

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Abstract: Fingerprints play a crucial role in the identification of potential suspects in criminal cases. However, determining the actual time, i.e., the time at which the fingermark was deposited, is challenging. Herein, we investigated the persistence and aging of fingerprints over time by observing the time evolution of latent fingerprints on a polystyrene box stored in a dark room. Fingerprint samples that were stored for up to two years could be detected with maximum accuracy using a black iron-oxide-based emulsion (black emulsion). To estimate the time of fingerprint deposition, fingerprint aging was studied by analyzing the lipid components of the fingerprints after their development. Cholesterol and squalene were selected as indicators of fingerprint aging, and their ratio was estimated to assess aging. In the case of fingerprint samples stored in a dark room for up to one month after deposition, the cholesterol/squalene ratio was approximately 0.01; it increased gradually to ≥ 0.1 over six months. A substantial reduction in the levels of cholesterol and squalene from the initial levels was also noted. Cholesterol and squalene were not detected after one year of storage. Thus, the extent of aging could be determined by analyzing the aging indicators for up to six months. Two cases that could cause error in the estimation of the fingerprint deposition time, namely, heating of the fingerprint sample before development and storage of the developed fingerprints in a dark room, were also investigated.

Key words: fingerprint, aging, degradation, cholesterol, squalene

1. Introduction

Latent fingerprints are crucial evidence for the identification of potential suspects in criminal cases. There is a fingerprint database of all citizens in South Korea, and fingerprint evidence at a crime scene plays a decisive role in the identification of potential suspect. However, as criminal techniques have become more complex and sophisticated, determining the actual time at which the fingermark was deposited is crucial, especially in situations wherein a suspect has previous legitimate access to the crime scene. Recently, numerous studies have been conducted on the changes in the components of fingerprints due

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to aging.[1-5] Studies wherein fingerprint aging has been evaluated by selecting cholesterol and squalene as indicators of aging, among the various fingerprint components, have shown that the decomposition of cholesterol and squalene depends on the porosity of the substrate on which the fingerprint is deposited. The decomposition of both cholesterol and squalene was found to occur more rapidly on a non-porous substrate than on a porous one.[2,6,7] For instance, squalene was observed to rapidly diminish on glass, whereas the decrease in microfilter substrate followed a much slower rate. Cholesterol diminished at a much slower rate than squalene on glass and did not show any change on microfilters over the time frame studied. When the fingerprints deposited on porous and non-porous substrates were stored in a dark room for 30 days, the squalene/cholesterol ratio decreased over this period.[2] Furthermore, in another study, after fingerprints deposited on glass and polystyrene were stored in a dark room or exposed to sunlight for six months, the fingerprints were developed with TiO_2 powder to analyze their visualization.[8]

This study is based on an actual criminal case wherein highly valuable ceramic articles were stolen from a warehouse located in the basement of a building. The articles were first wrapped in polystyrene boxes and then packed in paper boxes. Based on the fingerprints that were found on the remaining polystyrene boxes at the crime scene, a possible suspect was identified. However, the suspect claimed that he had been employed to transport the articles to the basement of the warehouse, i.e., the crime scene, ten years ago. Thus, by modeling this case, the present study aims to estimate the persistence of fingerprints deposited on polystyrene and the time of fingerprint deposition.

2. Materials and Methods

2.1. Materials

Commercial polystyrene packaging boxes (30 cm × 30 cm × 50 cm) were procured from a market in Seoul for use as substrates for fingerprint deposition. Iron (II, III) oxide (94 %) and emulsion 130K (polyoxyethylene lauryl ether) were procured from Junsei Chemical Company Limited (Tokyo, Japan). Cholesterol (≥ 99 %), squalene (≥ 98 %), and *N,O*-bis(trimethylsilyl)trifluoroacetamide with 1 % trimethylchlorosilane (BSTFA) were procured from Supelco. Potassium hydroxide (85 %, EP) and methyl alcohol (99.8 %, GR) were procured from DUKSAN pure chemicals. In addition, a 6 % KOH/MeOH solution was prepared by dissolving KOH in MeOH.

2.2. Fingerprint collection

To remove any interfering substances, the polystyrene box was cleaned with 70 % EtOH and then washed with distilled water (DW). It was then dried and cut into 4 cm × 4 cm specimens for use as the substrate for fingerprint deposition. Fingerprints were collected from three healthy Korean adults (two men and one woman) who are in their 50s (IRB No. 906-211116-BR-002-02). The volunteers were instructed to not use any skin care products (such as a lotion) after morning wash; the fingerprints were acquired in the morning. Before fingerprint deposition, the volunteers washed their hands using soap and rubbed their forehead and nose 2-3 times with their thumb or index finger. Thereafter, they placed their fingers on the substrate for fingerprint deposition and pushed the substrate with approximately ± 50 g of weight for approximately 15 s.[4]

2.3. Fingerprint storage

The substrates deposited with fingerprints were placed in a paper box to be stored in a dark room without air conditioning or heating to ensure natural aging during four seasons over a span of approximately two years. The ranges of temperature and relative humidity were 11-26 °C and 21-76 %, respectively.

2.4. Fingerprint development

A black emulsion was prepared by mixing 50 g of iron (II, III) oxide and 5 g of emulsion 130K in 1 L of DW. The mixture was mixed well by shaking to ensure the complete dispersion of iron oxide, following which it was allowed to stand for one day before use.

Then, the fingerprint samples were immersed in black emulsion and shaken sideways 2-3 times for development.[9]

2.5. Visual image analysis and identification

The developed fingerprints were photographed using a DSLR camera (Nikon D810, Japan) with a 105 mm F 2.8 DG MACRO HSM (SIGMA, Japan). Fingerprint images were taken in gray scale after adjusting the white balance (ISO 100, F/11). To examine the minutiae, characteristics, and the matching of minutiae, the inked impression of the volunteers were compared.

2.6. Extraction and derivatization of samples

Fingerprint composition analysis was performed immediately after the development of the latent fingerprints using the black emulsion and subsequent visual image analysis. When compositional analysis could not be performed immediately, it was performed within two days of developing the fingerprint sample.

The polystyrene specimen used for fingerprint deposition was cut using a scalpel to the size of the developed fingerprint with a thickness of approximately 2 mm. Thereafter, the specimen was cut into smaller pieces and placed in a 15 mL test tube containing 3 mL of a 6 % KOH/MeOH solution. After rotational mixing at 50 rpm for 30 min, 3 mL of hexane was added to the tube. After further mixing at 50 rpm for 30 min, the tube was centrifuged at 4000 rpm for 10 min, and the supernatants were transferred to an evaporation tube in an evaporator to remove hexane completely using nitrogen gas at 40 °C. Thereafter, 150 μL of the BSTFA derivatization agent was added to the tube containing the enriched sample (with the lid closed), and the sample was allowed to react at 70 °C for 20 min. For compositional analysis, gas chromatography/mass spectrometry (GC/MS) was performed on 1 μL of the sample. The analysis was performed twice.

2.7. GC/MS analysis conditions

Compositional analysis of the fingerprint extract was conducted using a GC/MS instrument (6890N/5975, Agilent Technologies, Santa Clara, CA) equipped with a nonpolar capillary column DB-5MS (30 m × 0.25 mm id × 0.25 μm thickness). The temperature was maintained at 100 °C for 3 min, which was increased to 150 °C at a rate of 10 °C/min and maintained for 2 min; the temperature was then increased to 160 °C at a rate of 3 °C/min and maintained for 5 min, and finally, increased to 280 °C at a rate of 20 °C/min and maintained for 30 min. The carrier gas, helium, was flown at a constant rate of 1 mL/min, and the inlet temperature was maintained at 250 °C for the injection of 1 μL of the sample at a split ratio of 5:1. The total analysis duration was 54 min. MS was conducted in the electron impact mode at 70 eV, scanning between 50 and 550 amu, following a 5 min solvent delay. The mass spectra obtained from this analysis were identified and evaluated through spectral matching using the Wiley 10th/NIST 2014 mass spectral library (W10N14; John Wiley & Sons, Hoboken, New Jersey, USA).

3. Results and Discussion

To model a real criminal case, the fingerprints deposited on the surface of a polystyrene box and stored in a dark room were developed using a black emulsion to determine the duration over which they could be visualized. The components of the fingerprint were then analyzed using the developed samples to estimate the time of fingerprint deposition.

3.1. Identification of latent fingerprints stored for different durations

The fingerprints deposited on the surface of the polystyrene box were stored in a dark room for up to two years; the latent fingerprints were developed after different time intervals using the black emulsion technique. As indicated in *Fig.* 1, we could clearly visualize the latent fingerprints on the polystyrene box after one, three, and six months, and even after two years of storage, by developing them using the black iron (II, IIII) oxide-based emulsion. In a previous study,[8] the greasy latent fingerprints on polystyrene could not be identified after 170 days of dark room

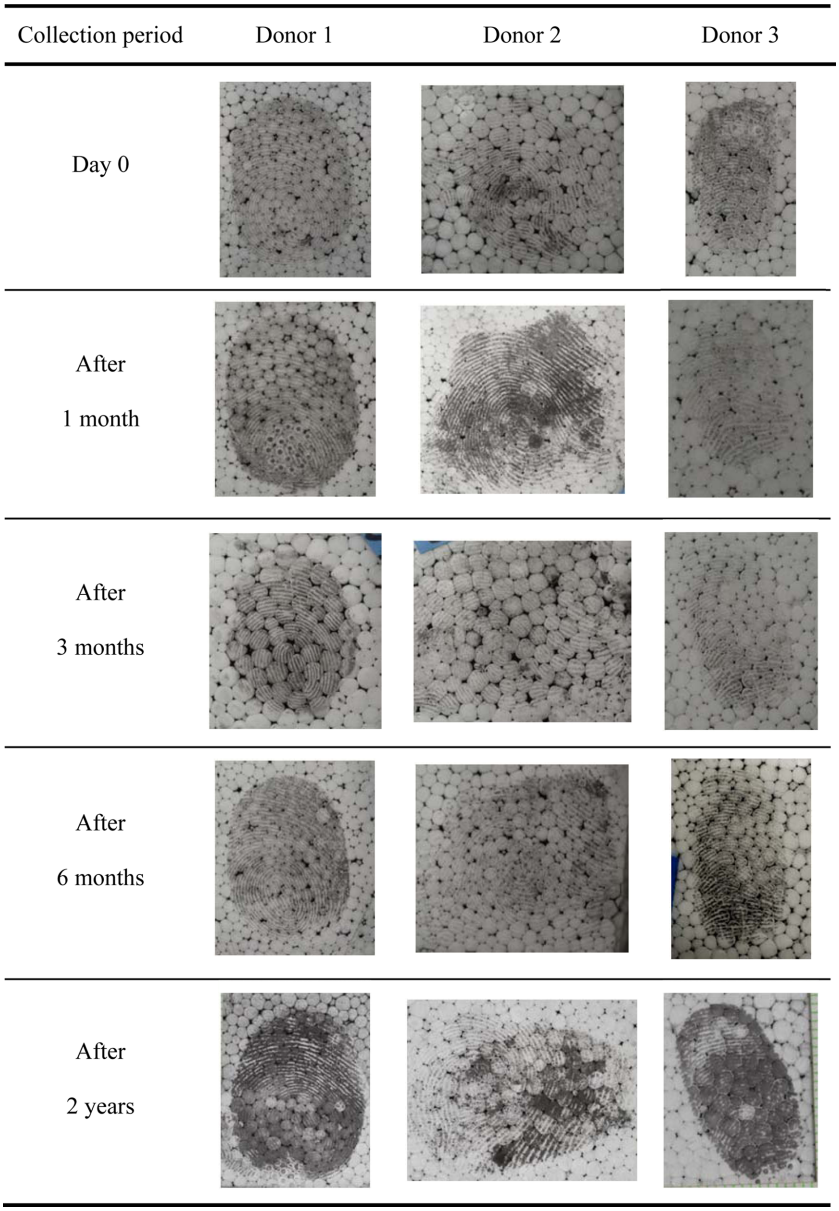

Fig. 1. Visualization of the developed latent fingerprint samples after different aging durations

storage when titanium dioxide powder was applied using a squirrel hairbrush for development to prevent over-powdering. The minutiae and other characteristics could not be distinguished, although faint ridges were observed. In contrast, in this study, fingerprint ridges were also visualized in the fingerprint samples stored for two years. This can be attributed to the result of fingerprint aging processes such as drying and fixation. Visualization is also affected by the development method.

To identify the developed fingerprints, the minutiae and other features were analyzed by comparison with the original fingerprint. As shown in *Fig.* 2, the fingerprints of the first volunteer provided 19 and 17

Fig. 2. Matching of minutiae determined by comparison. Left: developed sample of the latent fingerprint after 6 months of storage; right: inked impression of the volunteer.

matches for minutiae after six months and two years of storage, respectively. The fingerprints of the second volunteer provided 24 and 25 matches for minutiae after six months and two years of storage, respectively. Our findings confirmed that suspect identification is possible even when the samples were developed after storing for more than two years. The fingerprints of the third volunteer, however, exhibited four or fewer matches for minutiae after one month or greater periods of storage (six months or two years). Thus, identification was not possible. This is attributed to donor-specific sample variation rather than being an effect of fingerprint aging over time. The results thus suggested that latent fingerprints on polystyrene could remain intact for up to two years of storage in a dark room and that they could be visualized using an effective method for development. Thus, for the development of latent fingerprints, the samples should be immersed in a liquid to ensure the accurate visualization and identification of fingerprints without the deterioration of the ridges by the brush used to apply the powder developer.

3.2. Estimation of the fingerprint deposition time

After developing latent fingerprints and the subsequent identification, the samples were used for compositional analysis. To investigate fingerprint aging, cholesterol and squalene were selected as the indicators of aging, as reported previously.[6,7]

To determine the interfering effects of the develop-

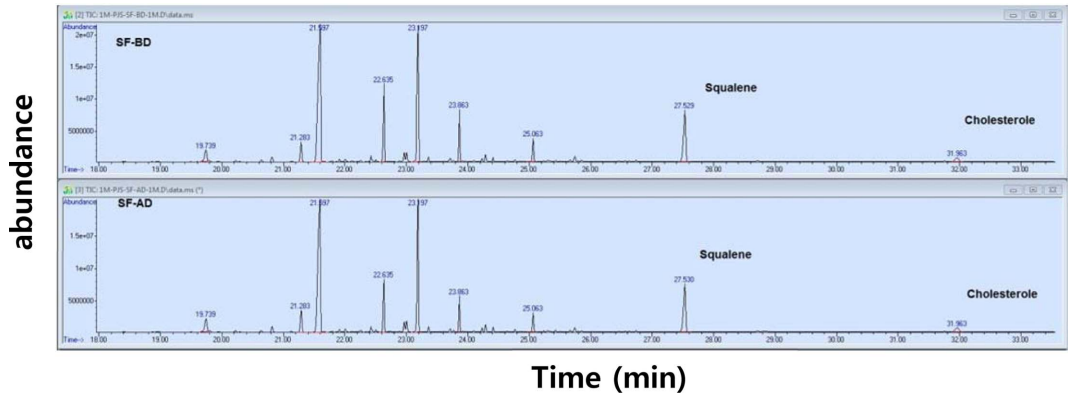

Fig. 3. TICs before (top) and after (bottom) development.

ment reagent on the analysis of cholesterol and squalene, two fingerprint samples from the same volunteer on polystyrene were analyzed before and after development using the black emulsion. As shown in *Fig.* 3, squalene and cholesterol were detected at retention times of 27 and 31 min, respectively, for both types of samples (before and after development). Furthermore, the total ion chromatograms (TICs) of the two samples were identical, and the cholesterol/squalene (Ch/Sq) ratios before and after development were the same at 0.01. This result clearly suggests that the iron (II, III) oxide and emulsion 130K used in the black emulsion technique do not interfere with the compositional analysis of the fingerprint, i.e., they only physically interacted with the fingerprint without chemically reacting with the components.

Furthermore, as expected, the fingerprint components varied according to aging. The intensities of the peaks of squalene and cholesterol decreased with increasing storage time owing to their degradation (*Fig.* 4). The peak area ratio(Ch/Sq) was approximately 0.01 for up to one month of storage. After six months, the ratio increased to ≥ 0.1 because the degradation rate of squalene exceeded that of cholesterol (*Fig.* 5). The levels of cholesterol and squalene also exhibited a trend of substantial reduction with time. After one year of storage, squalene was not detected in one sample, whereas it was detected in two samples,

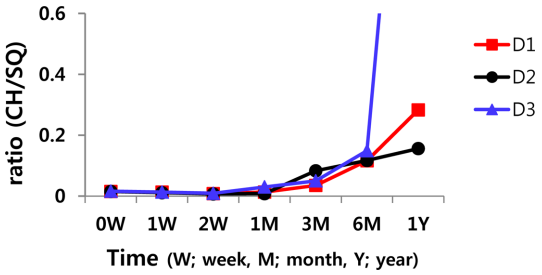

Fig. 5. Ratio of cholesterol and squalene with aging. D1, D2, D3: volunteers (each point represents the average of two replicates).

although at a considerably lower quantity compared to the initial level. When the storage time exceeded one year, neither squalene nor cholesterol was detected in any of the samples.

3.3. Aging by heating

Fingerprint experts sometimes develop fingerprints by applying heat (artificial aging) to enhance development. To monitor the artificial aging phenomena in various fingerprint samples, the samples were heated at 70 °C in an oven for approximately 11 h. *Fig.* 6 shows that the fingerprints sample with heat applied appeared a little more clearly than the sample without heat. This might be attributed to the effects of drying and fixation of the fingerprint components.

Furthermore, the Ch/Sq ratio of the heated sample

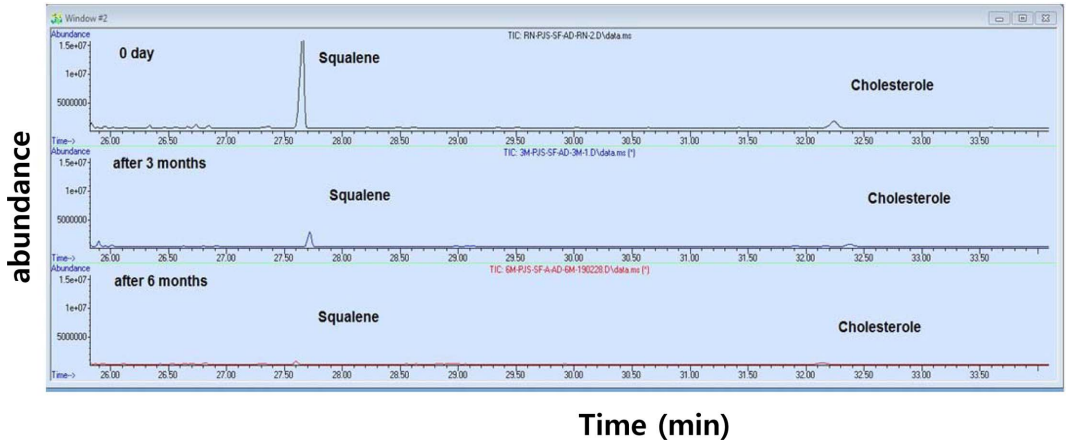

Fig. 4. TICs of the fingerprint samples according to the storage time (dark room storage). Top: immediate; middle: 3 months after development; bottom: 6 months after development.

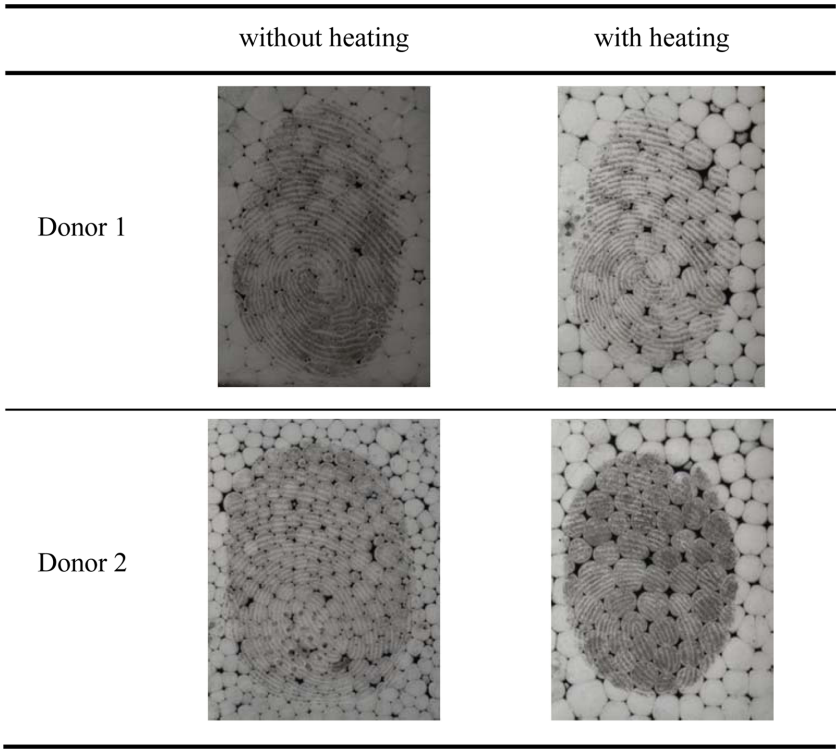

Fig. 6. Visualization of the developed latent fingerprints from two volunteers before and after aging by heating.

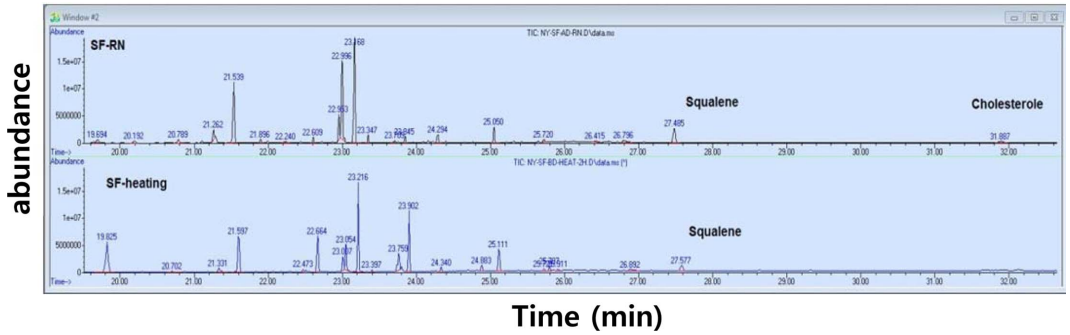

Fig. 7. Comparison of the TIC before (top) and after (bottom) heating.

was in the range 0.02-0.08, which is greater than the initial ratio, thereby indicating the progression of aging. As shown in *Fig.* 7, heating reduced the intensities of the squalene and cholesterol peaks; this indicates the heat-induced degradation of the fingerprint components. Hence, fingerprint samples developed with heating may cause an error in the estimation of the deposition time.

3.4. Aging of the developed samples

After the fingerprints were developed, the developed samples were stored again in the dark room, and the fingerprint components were analyzed after 18 months. The results revealed that neither squalene nor cholesterol could be detected due to the degradation (*Fig.* 8), thus confirming the aging of fingerprints over time, even when the samples were stored after development.

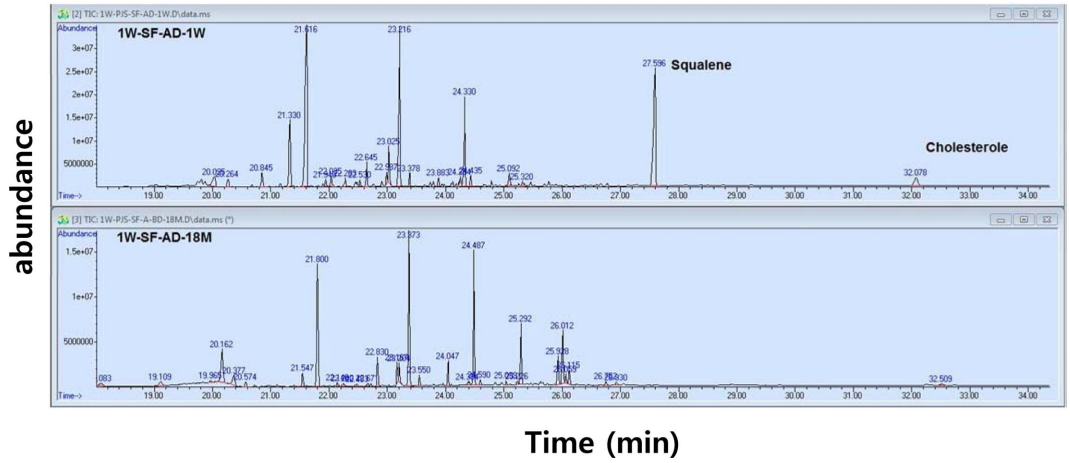

Fig. 8. Comparison of the TIC immediately after development (top) and after 18 months (bottom) following development.

4. Conclusions

In this study, we investigated the aging of fingerprints over time by modeling a previous criminal case. The latent fingerprints on a polystyrene box were examined after storage in a dark room. Fingerprint identification could be performed by developing a fingerprint sample after two years of storage using a black emulsion. The components of the fingerprints were analyzed from the developed fingerprint samples because the reagents used in the black emulsion did not interfere with the compositional analysis of the fingerprints. To estimate the time of fingerprint deposition after aging in a dark room, cholesterol and squalene were selected as indicators and their peak area ratio (Ch/Sq) was estimated. For the fingerprint samples stored for up to one month in a dark room, the Ch/Sq ratio was approximately 0.01; it increased significantly to ≥ 0.1 after six months. The levels of cholesterol and squalene also exhibited a significant decreasing trend compared to the initial levels, and neither cholesterol nor squalene was detected after storage for approximately one year. Therefore it is difficult to estimate the exact timing of fingerprint deposition, but it is thought that a rough estimate of fingerprint aging is possible depending on the criminal cases. In both the cases of heat-based fingerprint deposition and long-term storage of the developed fingerprint, the aging of the fingerprint prevented the estimation of the time of fingerprint deposition by compositional analysis.

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