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Original article

Effects of light on the quality of honeysuckle during storage based on physico–chemical indicators and chemical composition

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ABSTRACT

Honeysuckle, as a medicinal and edible substance, has been widely used for thousands of years. However, its quality is easy to change during storage by strong light, high temperature, high humidity and other inappropriate conditions. In this study, taking light as an example, the quality changes of honeysuckle during storage were explored by simulated accelerated test. The results showed that after 18 days of storage under light (6000 ± 10 Lx), the color changed from green-white to yellow-white, and the color parameter showed that a^* value increased from 0.522 to 4.061, and L^* value increased from 68.51 to 75.05. The contents of chlorophyll *a*, chlorophyll *b*, and carotenoid decreased by 90%, 78.67, and 75.69%, respectively. Total phenol content decreased from 18.200 to 13.087 GAE $mg{\cdot}g^{-1}$, although total flavonoid and antioxidant activities fluctuated without significant difference. Except quercetin, chlorogenic acid and other nine bio-active compounds contents fluctuated and increased, resulting in higher contents after storage than before storage, which indicated that the honeysuckle still had good utilization value after discoloration. Combined with metabolomics technology, 22 different metabolites related to these changes were screened and identified, such as pheophorbide a, astilbin, and succinic acid. KEGG enrichment analysis showed that the oxidative decomposition of phenolic compounds and pigment components such as chlorophylls and carotenoid caused the chlorosis of honeysuckle. In summary, although the light caused the chlorosis of honeysuckle, it didn't affect the utilization value. Meanwhile, this also systematically explains why the Chinese pharmacopoeia describes the character of honeysuckle as "green-white or yellow-white".

1. Introduction

Honeysuckle, the dried flower bud or opening flower of *Lonicera japonica* Thunb. (Caprifoliaceae), has been widely used for thousands of years as a functional food and traditional Chinese medicine (TCMs). Modern pharmacological studies show that it has a series of biological activities, including antioxidant, anti-bacterial, and anti-inflammatory (Mu et al., 2022; Wang et al., 2021). Chemical composition suggest that it contains rich bio-active compounds, such as flavonoids, phenolic acids, iridoids, organic acids, and volatile oils (Cai et al., 2021; Wu et al., 2023).

At present, a large number of scholars have used advanced technologies such as metabolomics, transcriptomics and genomics to study honeysuckle, mainly focusing on the cultivation, growth and development, origin processing, chemical composition, authenticity identification, pharmacological efficacy, and clinical evaluation of honeysuckle (Wang et al., 2023; Kirina et al., 2021; Cai et al., 2020; Zhang et al., 2020; Xia et al., 2021; Liu et al., 2023). However, its quality is prone to change during storage due to inappropriate conditions such as strong light, high temperature, and humidity. As for the quality change during storage, Xiong et al. used electronic nose technology to detect the quality (Xiong et al., 2014). Wang et al. briefly measured the changes of physico-chemical indicators of honeysuckle caused by different temperature through accelerated shelf test (Wang et al., 2017). Liu et al. established a hyperspectral nondestructive detection model for chlorogenic acid content of honeysuckle combined with storage process (Liu et al., 2019).

For light, it is one of the important factors contributing to the quality change during storage. The accumulation of a toxic alkaloid solanine could greatly reduce tuber value during storage of potato by light (Xiong et al., 2022). The color of wines, as a main quality attribute, was the most factor affected value during storage by light (Arena et al., 2021).

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To our knowledge, there is no report on the effect of light on the quality of honeysuckle.

Therefore, a simulated acceleration experiment was performed aiming to explore the dynamic quality change of honeysuckle caused by light during storage. The physicochemical indicators (including color, chlorophyll, carotenoid, total phenol, total flavonoid, antioxidant activities) and chemical composition (including main bio-active compounds and metabolites) were analyzed with different storage time under the light exposure.

2. Materials and methods

2.1. Chemicals and reagents

Chemical reagents, including folin–ciocalteu phenol, 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid (Trolox), ferric chloride hexahydrate, 2,4,6-tris(2-pyridyl)-strizine (TPTZ), 2,2′-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) diammonium salt (ABTS), 1,1-diphenyl-2-picrylhydrazyl (DPPH), acetic acid, hydrochloric acid, potassium persulfate, aluminum nitrate, sodium carbonate, sodium hydroxide, and sodium nitrite were purchased from Sigma-Aldrich (St. Louis, MO, USA) and Macklin Biochemical Co., Ltd. (Shanghai, China). All reference compounds (neochlorogenic acid, chlorogenic acid, cryptochlorogenic acid, rutin, luteolin, isochlorogenic acid B, isochlorogenic acid A, isochlorogenic acid C, caffeic acid, and quercetin) with mass purities not <98% were purchased from Push Bio-Technology Co., Ltd. (Chengdu, China). HPLC-grade solvents (methanol, acetonitrile, and formic acid) were purchased from Fisher Scientific (Waltham, MA, USA). Analytical-grade methanol was purchased from Fuyu Fine Chemical Co., Ltd. (Tianjin, China).

2.2. Sample preparation

A batch of green-white dry honeysuckle samples was purchased from a Putianhe Chinese Medicine Tablet Co., Ltd. (Hebei, China). Its moisture (6.7%), total ash (4.8%), acid insoluble ashes (1.2%), and bio-active compounds contents (chlorogenic acid, 2.2%; cynaroside, 0.064%) met the requirements of Chinese Pharmacopoeia. After removing a few leaves and impurities, the honeysuckle samples was randomly divided into three groups. Each group was weighted about 100 g, placed them in cartons, and stored at the temperature of 25 ± 0.5 ℃ and relative humidity of 60 ± 1% in a stability incubator (ZSW-HQ250, Shanghai, China). According to our previous study, the quality of honeysuckle does not change under this condition. And then, honeysuckle samples were treated by light exposure with an LED light source illuminated at 6000 ± 10 Lx intensity which was measured by an electronic light sensor.

During storage time, honeysuckle samples were picked up at seven time points at 0, 3, 6, 9, 12, 15, and 18 d. Three replicates of each group were obtained, powdered (particle size: 40 mesh), and stored at –80℃ (DW-86L348, Qingdao, China) in preparation for the following experiments.

2.3. Color measurement

The color parameters of honeysuckle samples with different storage time were determined by a NH-310 colorimeter equipped with CIE color measurement system. Some color parameters, including L^* (lightness), a^* (green to red), and b^* (blue to yellow) were measured, some parameters, including C^* (chroma), h^* (hue), and ΔE (difference), were calculated as described in our previous report (Liu et al., 2023).

2.4. Discoloration and color loss rate

The ratio of the –a/b parameters is usually applied in research focused on the color changes of processed green vegetables. In this study, a^* is a main tone of honeysuckle for its green; L^* can reflect the overall brightness change trend of honeysuckle, so they are used to calculate the discoloration and color loss rate which were calculated according to previous report with some modifications (Yu et al., 2018).

In brief, the discoloration rate of each sample was calculated by plotting a regression line of the natural logarithm of the ratio of $(-L/a)_t$ and $(-L/a)_0$ as a function of storage time in days (d). The formula for calculating the kinetic constant (k) of the discoloration rate was as follows:

$$\ln[(-L/a)_t/(-L/a)_0] = -k \bullet t \quad (1)$$

where $(-L/a)_t$ is the color parameter at any time t, $(-L/a)_0$ is an initial color value at 0 d, and k is the kinetic rate constant (d^{-1}).

The color loss was calculated using the value of (L/a) before and after storage, as follows:

$$\text{color loss}\ (\%) = \frac{(L/a)_t - (L/a)_{t+1}}{(L/a)_t} \times 100\% \quad (2)$$

where $(L/a)_t$ was obtained at the previous time and $(L/a)_{t+1}$ was at the next time.

2.5. Chlorophylls and carotenoids determination

The chlorophyll and carotenoids of honeysuckle samples with different storage time were determined by ultraviolet spectrophotometry. The specific procedure was consisted with our previous literatures (Liu et al., 2023). The chlorophylls and carotenoids were extracted with 95% ethanol and methanol-acetone (80:20, v/v), respectively. The absorbance of chlorophylls was measured at 665 and 649 nm by a TU-1810 UV–visible spectrophotometer, and that of carotenoids at 628 nm. And then, the contents of chlorophyll a, chlorophyll b, total chlorophyll, and carotenoids were calculated.

2.6. Total phenol, total flavonoid, and antioxidant activity determination

Combined with our previous report, total phenol and total flavonoid content was determined using the Folin-Ciocalteu method, $NaNO_2$-Al$(NO_3)_3$-NaOH method, respectively (Saar-Reismaa et al., 2020; Liu et al., 2023). Their absorbance was measured at 765 nm and 510 nm, respectively. Their contents were calculated from standard curves with gallic acid and rutin concentration, respectively. And the results were expressed the results were expressed as mg gallic acid equivalent per g (GAE mg/g) and rutin equivalent per g (RE mg/g).

As for antioxidant activities, it was determined by three methods, including 2,2′-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) diammonium salt (ABTS), 1,1-diphenyl-2-picrylhydrazyl (DPPH), and ferric reducing antioxidant power (FRAP) radical scavenging assays (Szydłowska-Czerniak et al., 2022; Liu et al., 2023). The AA was calculated from a standard curve with Trolox concentration and expressed as μM Trolox equivalent per g (TE μM/g).

2.7. The bio-active compounds quantitative analysis

The ten bio-active compounds, including neochlorogenic acid, chlorogenic acid, cryptochlorogenic acid, rutin, cynaroside, isochlorogenic acid A, isochlorogenic acid B, isochlorogenic acid C, caffeic acid, and quercetin, were analysis by HPLC methods as described in another literature (Liu et al., 2023). Their qualitative and quantitative analysis were combined with the retention time and concentration of the standards to identify and calculate.

2.8. Untargeted metabolomics analysis

The powered sample (50 mg) was extracted using a 400 μL 80% methanol solution with 0.02 mg·mL^{-1} L-2-chlorophenylalanin as an internal standard. The mixture was treated by high throughput tissue

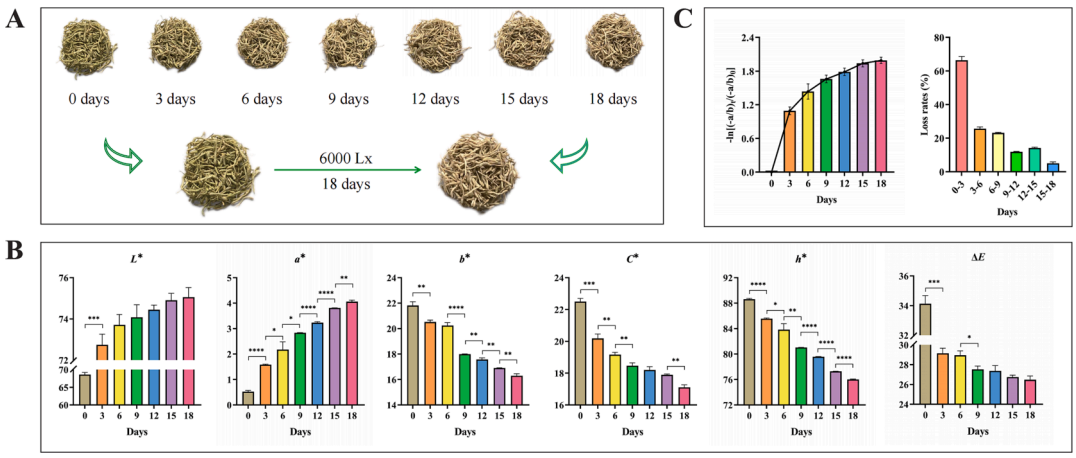

Fig. 1. The appearance (A), color parameters (B), discoloration and color loss rate (C) under light treatment with different storage time. *: $p \leq 0.05$, **: $p \leq 0.01$, ***: $p \leq 0.001$, ****: $p \leq 0.0001$.

crusher (Wonbio-96c, Shanghai, China) at 50 Hz for 6 min at –10 °C, then followed by ultrasonic treatment at 40 kHz for 30 min at 5 °C. The samples were placed at –20 °C for 30 min to precipitate proteins. After being centrifugated at 5,000 rpm for 15 min at 4 °C, the supernatant was made for untargeted metabolomics analysis by UPLC-QE-MS/MS method. In addition, a quality control (QC) sample was prepared by mixing all the tested samples in equal volume.

The instrument platform for this analysis was UPLC-Q Exactive HF-X system of Thermo Fisher Scientific (MA, USA). Chromatographic separation was implemented using a Waters ACQUITY HSS T3 column (100 mm × 2.1 mm, 1.8 μm, Milford, USA) kept at 40 °C. The mobile phases consisted of 0.1% formic acid in water: acetonitrile (95:5, v/v) (A) and 0.1% formic acid in acetonitrile: isopropanol: water (47.5:47.5:5, v/v) (B). The following multi-step gradient elution program was used: 0.0–1.5 min (0–5% B), 1.5–2.0 min (5–10% B), 2.0–4.5 min (10–30% B), 4.5–5.0 min (30–100% B), 5.0–6.3 min (100% B), 6.3–6.4 min (100–0% B), and 6.4–8.0 min (0% B). The injection volume was 2 μL and the flow rate was 0.4 mL·min^{-1}.

The mass spectrometric data was collected using an electrospray ionization (ESI) source operating in both positive and negative ion modes with MS precursor ion scanning from 70 to 1050 Da. The ion source parameters were set as follows: heater temperature, 425 °C; capillary temperature, 325 °C; sheath gas flow rate, 50 psi; aux gas flow rate, 13 psi; ion-spray voltage floating, –3500 V in negative mode and 3500 V in positive mode; and normalized collision energy, 20–40–60 V rolling for MS/MS. Full MS resolution was 60,000 and MS/MS resolution was 7500. Data acquisition was performed with the Data Dependent Acquisition (DDA) mode. In order to monitor the stability of the system and ensure the accuracy of the data acquired, the QC sample was injected at an interval of every six samples.

2.9. Data processing and identification of metabolites

The raw data consisting of *m/z*, retention time, and peak intensity were imported into Progenesis QI software (Waters, Milford, USA) for pre-processing. Internal standard peaks and known false positive peaks (including noise, column bleed, and derivatized reagent) were removed. The peaks that appeared in >80% of the samples in each group were retained. Then, the metabolites were identified by means of the main database of HMDB (https://www.hmdb.ca/), Metlin (https://metlin.scripps.edu/), Majorbio database, and relevant published literature.

The data after the database searches are uploaded to Majorbio cloud platform (https://cloud.majorbio.com) for data analysis. All the data were normalized by total area sums prior to analysis. Meanwhile, the variables with relative standard deviation (RSD) > 30% of QC samples were removed, and log10 logarithm was performed to obtain the final data matrix for subsequent analysis.

2.10. Differential metabolites screening

The data matrix was further analyzed by principal component analysis (PCA) and orthogonal partial least squares-discriminant analysis (OPLS-DA) using SIMCA 14.1 (Umerics, Umeå, Sweden). Pareto scaling was applied to all models for the analyzed data. The quality of the established models was inspected by R^2 and Q^2, whose values > 0.5 indicated good quality of the models and values close to 1.0 indicated an excellent model.

The OPLS-DA model was established with a pairwise modeling method and pairing method was 0 vs 3 d, 3 vs 6 d, 6 vs 9 d, 9 vs 12 d, 12 vs 15 d, and 15 vs 18 d. On the basis of the variable importance in projection (VIP) > 1 for positive ionization mode and VIP > 1.5 for negative ionization mode, together with VIP, *p*-value, and absolute value of fold change, the differential metabolites of honeysuckle samples from different storage time were screened out.

2.11. Statistical analysis

All results were presented as mean ± standard deviation (SD) and analyzed by one-way analysis of variance (ANOVA) using IBM SPSS 26.0 software (SPSS, Chicago, IL, USA). The significant difference was compared by Duncan's multiple range test. The correlation was performed using IBM SPSS 26.0 software. They were visualized by OriginLab Origin 2021 (Origin, San Francisco, CA, USA) and Graphpad Prism 9.0.0 (Graphpad, San Diego, CA, USA).

3. Results and discussion

3.1. Color characteristics changes

Color attribute is an important criterion for evaluating the quality of some foods such as vegetables, beverages, tea, and wines (Hensel et al., 2023). It was found that buds have higher sensitivity towards light than flowers, while both buds and flowers are sensitive to light (Yan et al., 2018). In this study, the fading of green could be observed obviously under the light treatment for honeysuckle with the extension of storage time (Fig. 1A). There was significant change from 0 to 18 d, which was reflected by the difference of color parameters (L^*, a^*, b^*, C^*, h^*, and ΔE) in Fig. 1B. With an increase in storage time, the values of L^* and a^* increased and of b^*, C^*, h^*, and ΔE declined, suggesting that the honeysuckle became fader under light treatment. Previous study showed that the significant increase of a^* value might correspond to the loss of green color due to phaeophytinization of chlorophyll (Nabechima et al.,

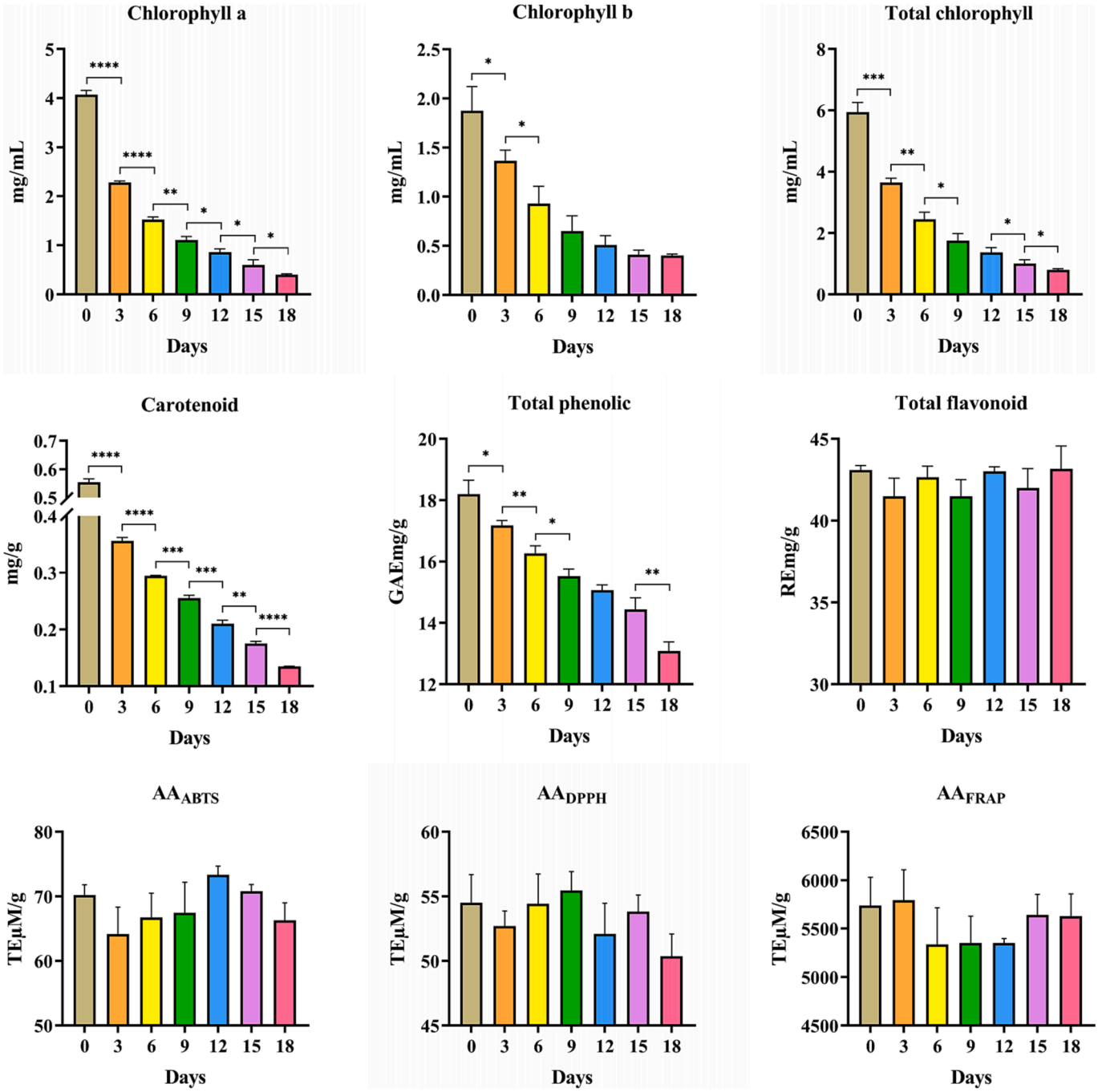

Fig. 2. Physicochemical indicators under light treatment with different storage time. *: $p \leq 0.05$, **: $p \leq 0.01$, ***: $p \leq 0.001$, ****: $p \leq 0.0001$.

2014). Significant difference of color parameters was observed in adjacent comparison pairs of every storage time, especially a^* and h^* values. This was in agreement with the significant color changes of honeysuckle by human eye (Fig. 1A). Our result was consistent with the result that h^* value decreases and L^* value increases of radish after being exposed to light during storage (Xiao et al., 2014).

Additionally, the rates of discoloration and color loss play important roles in color analysis (Yasuda et al., 2019). They had significant difference in different storage time as shown in Fig. 1C. The rates of discoloration and color loss from 0 to 3 d were the largest, of which the later reached 65.13%. Starting from 3 d, the discoloration rate slowed down gradually and color loss rate became smaller. This was also consistent with the results of the most significant changes in color appearance within initial 3 d.

3.2. Chlorophylls and carotenoid degradation

Chlorophylls and carotenoid play key roles in color maintenance of green plants and foods such as tea (*Camelia sinensis*) (Donlao and Ogawa, 2019) and kiwifruit (*Actinidia deliciosa*) (Ampomah Dwamena et al., 2019). Similarly, the content changes in chlorophylls (chlorophyll *a*, chlorophyll *b*, and total chlorophyll) and carotenoid of honeysuckle by light treatment with different storage time were evaluated in Fig. 2. From 0 to 6 d, their average content was greatly declined, chlorophyll *a* from 4.072 to 1.521 mg·mL^{-1}, chlorophyll *b* from 1.877 to 0.931 mg·mL^{-1}, total chlorophyll from 5.950 to 2.452 mg·mL^{-1}, and carotenoid from 0.554 to 0.294 mg·g^{-1}. From 6 to 18 d, their average content was slowly declined, chlorophyll *a* from 1.521 to 0.402 mg·mL^{-1}, chlorophyll *b* from 0.931 to 0.400 mg·mL^{-1}, total chlorophyll from

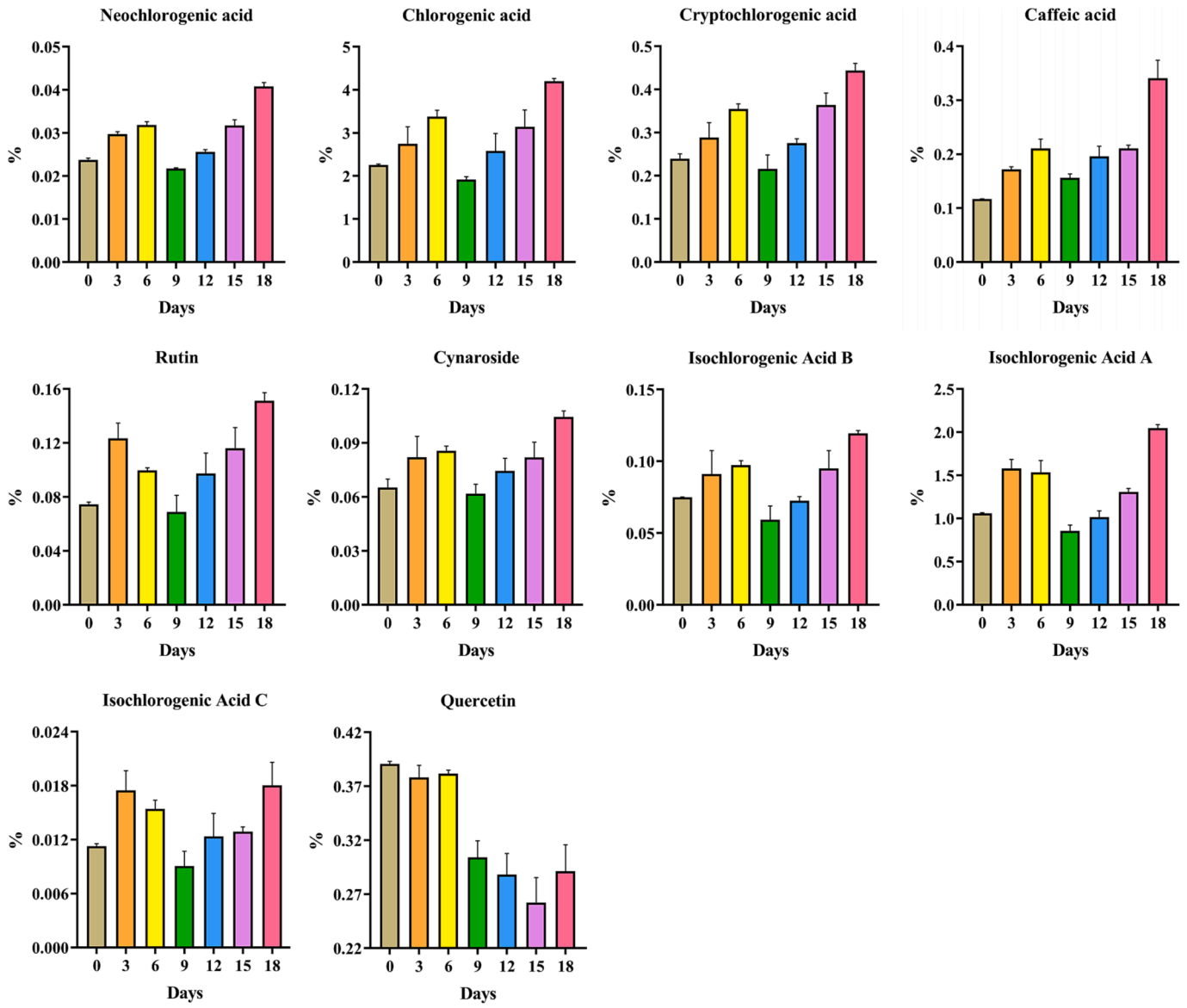

Fig. 3. Ten bio-active compounds during light treatment with different storage time.

2.452 to 0.804 mg·mL^{-1}, and carotenoid from 0.294 to 0.135 mg·g^{-1}. It is indicated that the fading process of honeysuckle under light may be divided into two stages. On the whole, chlorophyll *a* decreased by 90.12%, chlorophyll *b* did by 78.67%, total chlorophyll did by 86.49%, and carotenoids did by 75.69% from 0 to 18 d. These results also indicated that chlorophyll *a* and chlorophyll *b* have different sensitivity to light, and chlorophyll *a* is more susceptible to degradation by light, and the degradation rate of the former is significantly faster than that of the latter under the same light conditions, which is consistent with the change trend of spinach during storage (Huang et al., 2008).

Natural pigments have attracted much attention, but most of these pigments are unstable, especially chlorophyll, which is easily degraded by light, enzyme, heat and other factors during storage. Study found that the loss of carotenoids was accelerated when the samples were stored in an environment with strong light (1875–3000 lx) (Atencio et al., 2022). And photodegradation is one of the main degradation pathways of chlorophyll (Sato et al., 2015; Zhu et al., 2017). In addition, it was found that chlorosis of leaves and other green plants is closely related to chlorophyll degradation (Xiong et al., 2023; Hauenstein et al., 2016) and it was speculated that the fading phenomenon of honeysuckle in this study may be similar.

3.3. Total phenol, total flavonoid, and antioxidant activity changes

The influence of the phenol and flavonoid during the storage of foods has been reviewed recently (Adamenko et al., 2021; Ferreira et al., 2019). However, there are few reports on the effects of light treatment on some indicators of total phenol, flavonoid, and antioxidant activity. Their content and activity of honeysuckle by light treatment with different storage time were shown in Fig. 2. Light caused a significant decline ($p < 0.05$) on total phenol content with different storage time. From 0 to 18 d, the average content of total phenol declined from 18.200 to 13.087 GAE mg·g^{-1}, suggesting that phenol compounds were significantly affected under light treatment. As the main components of honeysuckle, it could be speculated that the oxidative decomposition of phenolic compounds was one of the reasons for its fading color. This is consistent with the result that the total phenol content in jujube decreased after sun-drying treatment (Du et al., 2013). In this study, there was no significant change in total flavonoid, suggesting that the stability of flavonoid compounds of honeysuckle under the light irradiation were good.

The antioxidant activity is a result of the combined action of total phenol and total flavonoid. In this study, the extracts had varying

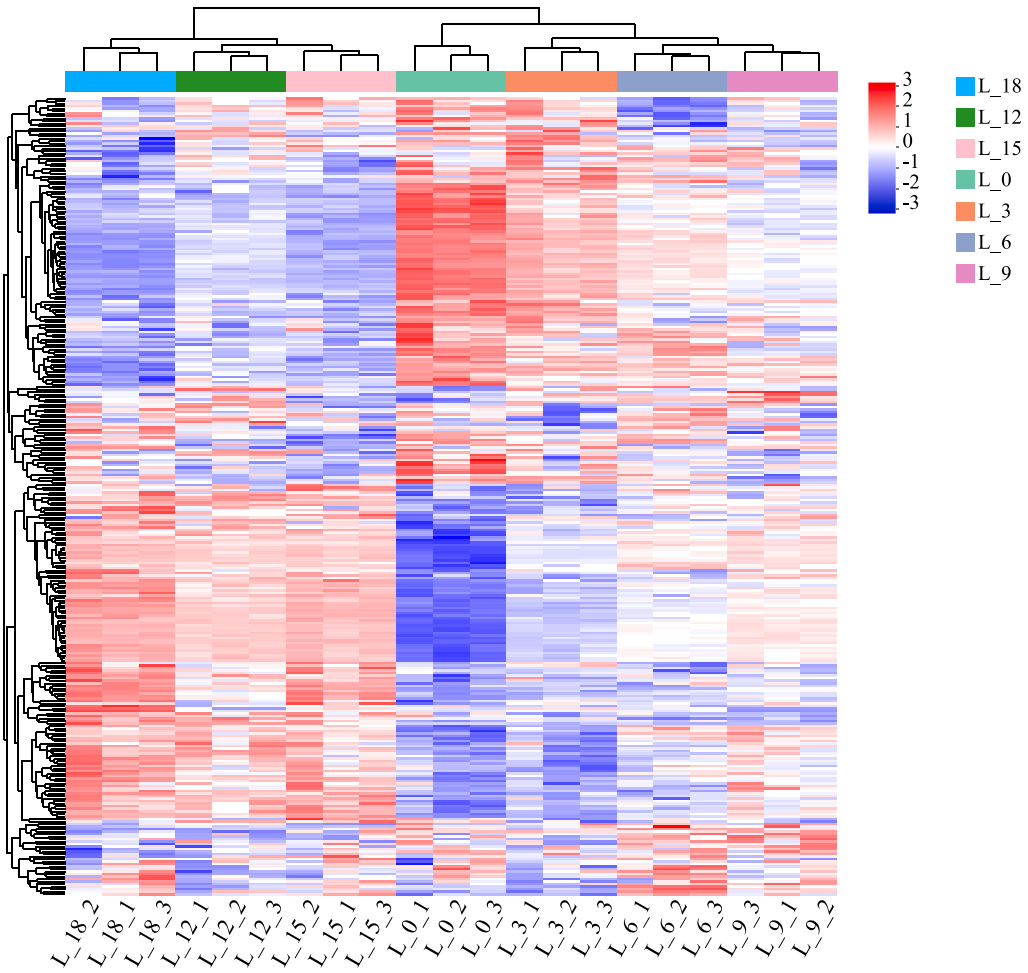

Fig. 4. The hierarchical cluster analysis (HCA) of honeysuckle samples with different storage time.

degrees of antioxidant activity in ABTS, DPPH, and FRAP assays. Although their average values fluctuated with different storage time, there were no statistically significant differences ($p > 0.05$) in means of the results obtained via these methods between 0 and 18 d.

3.4. Bio-active compounds dynamic analysis

In theory, the change in the color of honeysuckle should be the result of a comprehensive change in the type and/or content of its chemical compositions. In this study, an HPLC method was established to determine simultaneously the content of 10 bio-active compounds in the storage process, aiming to judge whether the honeysuckle under the light treatment could still be used as medicine and food.

Ten bio-active compounds were identified by comparing retention times and absorbance spectra with their reference compounds. With the extension of storage time, their content as shown in Fig. 3. Except quercetin, the changes of 9 compounds were divided into 2 stages. In detail, it was showed that their content first increased and then decreased within the initial 9 d, and increased continuously from 9 to 18 d. This trend has been similarly reported in other study. For example, in the storage process of Dongzao, most of flavonoids gradually increased in the first 5 days of storage, and began to decline after 5 days (Niu et al., 2023). It is speculated that it is related to phenylpropane metabolism and flavonoid biosynthetic pathway.

It was interesting that their content on 18 d is significantly higher ($p < 0.05$) than that on 0 d. For chlorogenic acid and cynaroside prescribed by Chinese Pharmacopoeia, the content increased from 2.26% and 0.06% to 4.20% and 0.10%, respectively. Studies have shown that chlorogenic acid, as a natural potent antioxidant, is not degraded under light irradiation (Rivelli et al., 2010). Moreover, caffeic acid has stronger antioxidant activity than chlorogenic acid (Sato et al., 2011). With the increase of light intensity, the content of rutin increased, which indicated the importance of light to the accumulation of rutin (Kuo et al., 2020). These findings can explain the content changes of corresponding compounds in this study to a certain extent. As for quercetin, it showed a gradual decrease within the initial 6 d and sharp decline from 6 to 18 d. It was showed that quercetin was decomposed under light treatment in a report (Golonka et al., 2020), which was consistent with our result. On the one hand, these results explain why faded honeysuckle is still used as medicine and food. On the other hand, these results prompt us to explore other chemical components in depth.

3.5. Metabolites profiling analysis

The UPLC-QE-MS/MS method was established in this study to acquire large-scale untargeted metabolites of honeysuckle in both positive and negative modes. Fig. S1 showed the total ion chromatograms (TIC) of honeysuckle samples under light treatment. Taking the positive ion mode as an example, the overall profile of the TIC with different storage times was similar. On the one hand, this showed the extraction and detection of the metabolites were repeatable and reliable and the instrument was stable. On the other hand, the metabolites did not change significantly during the whole storage process.

For the samples in the storage process, most of the influences caused by time and environmental conditions are studied, and the TIC chromatograms of the same samples at different time point are basically

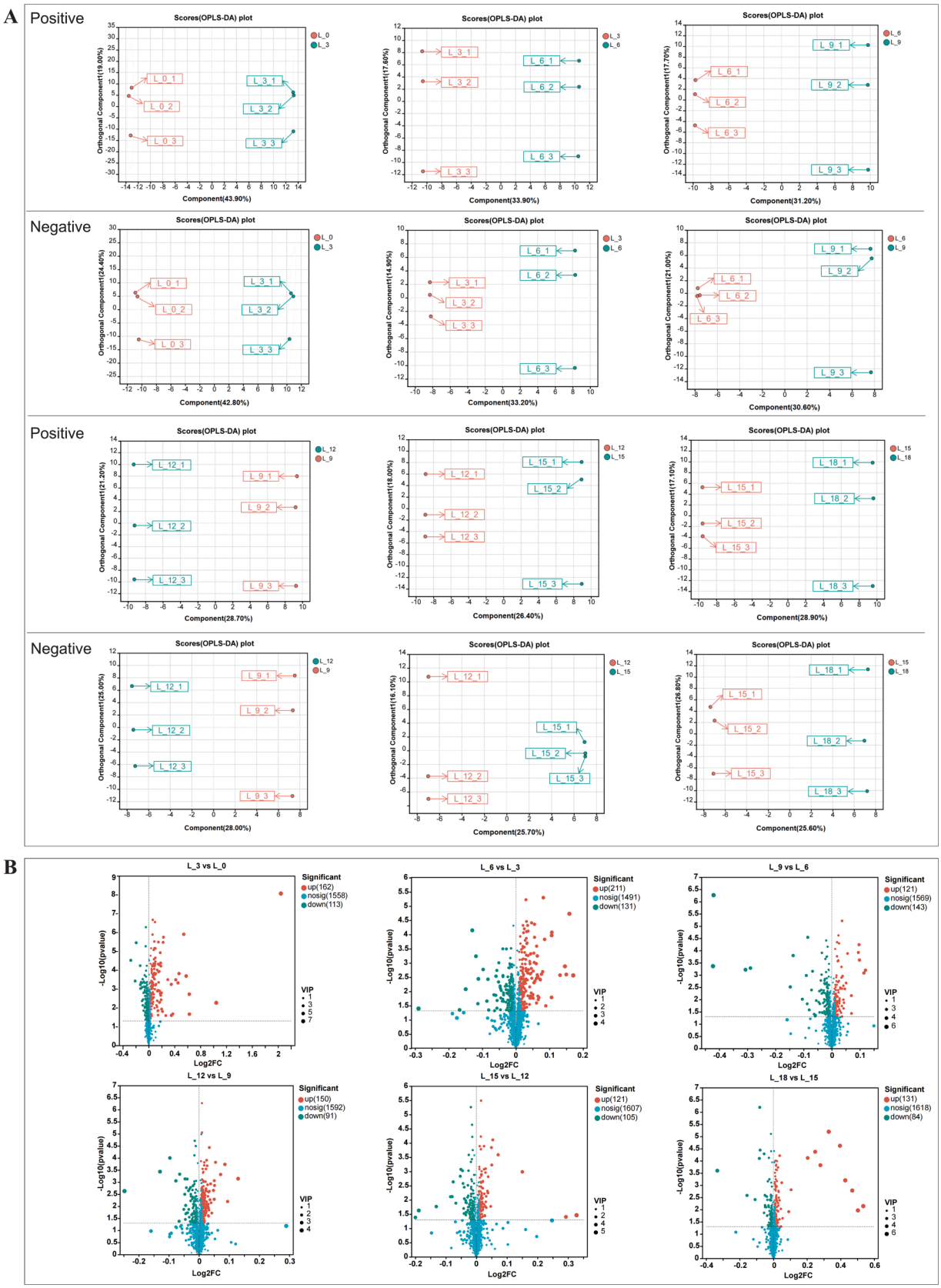

Fig. 5. The score plots of OPLS-DA (A) and volcano plots (B) of differential metabolites with different storage time.

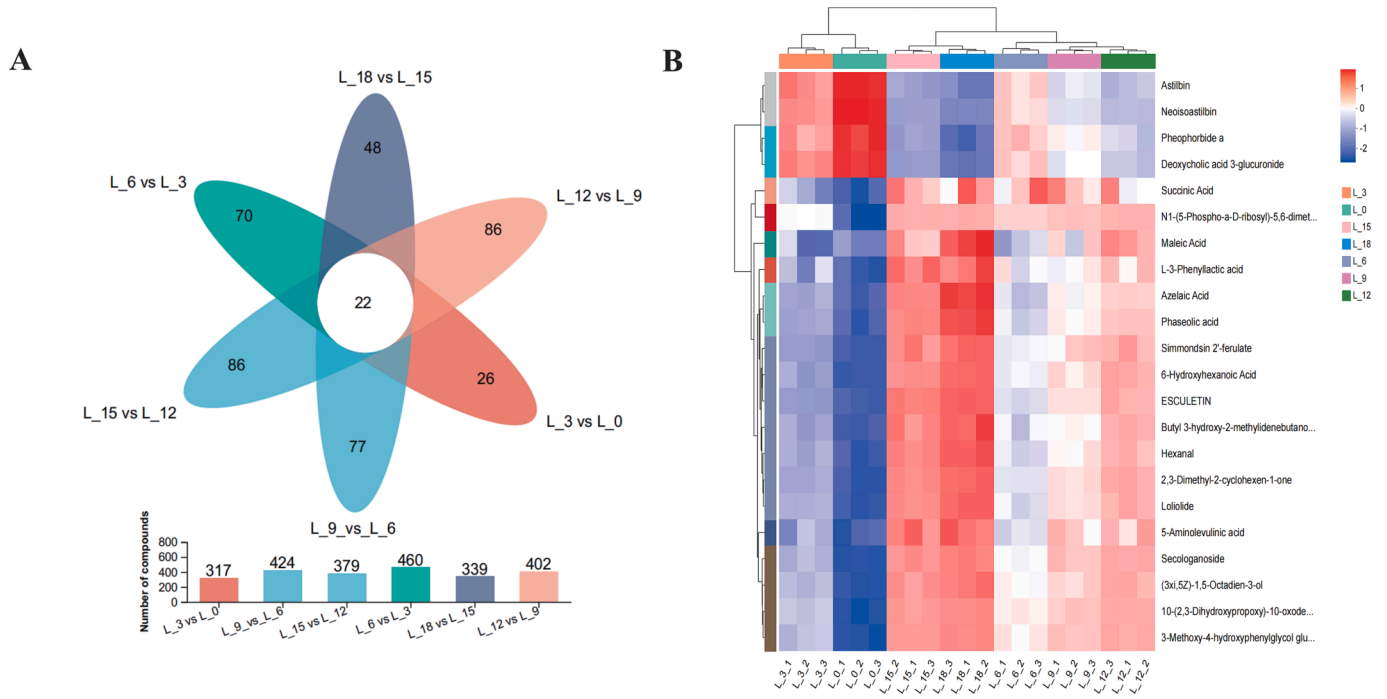

Fig. 6. The venn plot (A) and heat-map (B) of differential metabolites under light treatment with different storage time.

similar (Li et al., 2021; Xiao et al., 2021). These data need to be analyzed and compared by further multivariate statistical analysis. In fact, almost all metabolomics reports are carried out in this way (Chen et al., 2024; Hou et al., 2023). For obtain a more visible comparison of the chromatograms from different storage times, further data mining steps are essential based on the obtained metabolic fingerprints.

3.6. Metabolomic data analysis

Chemometric analysis is always a critical step for screening differential metabolites. Two separate data matrices containing 959 features for ESI^+ mode and 874 features for ESI^- mode were obtained, whose species and numbers were more abundant than those found in earlier reports of honeysuckle (Wang et al., 2023). To observe overall clustering effect and classification trend of the data matrices, an unsupervised PCA was firstly performed and displayed in Fig. S2A-B. The duplicate QC samples were grouped together and located on the center of score plots. These results once again demonstrated the robustness of the developed UPLC-QE-MS/MS analytical procedure and guaranteed the repeatability and reliability of the metabolomic data. Meanwhile, these results showed that the honeysuckle samples had great difference at different storage time.

To reduce system noise and demonstrate the discrimination of above data, supervised PLS-DA model was built for honeysuckle classification. As shown in Fig. S2C-D, honeysuckle samples from seven points of storage time were well-differentiated in both positive and negative modes, which was consistent with the classification result of PCA model. These results indicated that the honeysuckle under light treatment in the fading process could be distinguished based on its untargeted metabolomic fingerprints data.

In addition, an accumulation pattern of metabolites among honeysuckle samples can be visualized through a heat-map hierarchical cluster analysis and the result was showed in Fig. 4. The heat-map showed that three replicates of each group with different storage time were cluster together, indicating the good homogeneity between three replicates. The samples of 3, 6, and 9 d were grouped into one category, and the samples of 12, 15, and 18 d were grouped into another category. The results indicated that the fading process of honeysuckle in storage time might be divided into two stages, which was consistent with those results of the content of chlorophylls and bio-active compounds. In addition, this heat-map showed that, some metabolites of honeysuckle were up- or down- regulated when stored under light, suggesting that honeysuckle might undergo significantly different metabolic processes.

This method combining PCA, PLS-DA and HCA to judge the consistency and difference within and between sample groups has become very popular, such as the color change of black tea from Huangjinya during processing (Zhou et al., 2022), the calcification of chestnuts during storage (Xiao et al., 2021), and the automatic oxidation of hazelnut oil during storage (Gao et al., 2022).

3.7. Differential metabolites screening

For screening differential metabolites from the seven honeysuckle groups, an OPLS-DA model was performed with pairwise contrasts among the samples and the score plots were shown in Fig. 4A. In this model, R^2X and R^2Y represented the interpretation rates of the *X* and *Y* matrices, respectively, and Q^2 did the prediction ability. All pairwise comparison results showed that the R^2Y and Q^2 scores were higher than 0.9, indicating that the model was suitable. This model was verified through 200 times permutation test and its result confirmed that the model was meaningful (Fig. S3). In Fig. 5A, all pairwise groups were performed good separations.

Subsequently, the difference in the expression level of metabolites between two groups could be visualized through volcano plots (Fig. 5B). The fold change > 1, VIP > 1, and *p*-value < 0.05 were combined to screen out the differential expressed metabolites. Then, 317, 424, 379, 460, 339, and 402 characteristic variables were selected for 0 vs 3 d, 3 vs 6 d, 6 vs 9 d, 9 vs 12 d, 12 vs 15 d, and 15 vs 18 d, respectively. Twenty-two characteristic variables were found to be present in all six groups as shown in Fig. 6A. They can used as the differential metabolites for the discrimination of different storage time samples.

The heat-map about the differential metabolites in honeysuckle samples was shown in Fig. 6B, the content change was shown in Fig. 7, and the detail information was shown in Table S1. There were 4 metabolites with down-regulated levels including astilbin, neoisoastilbin, pheophorbide a, and deoxycholic acid 3-glucuronide and 18 metabolites

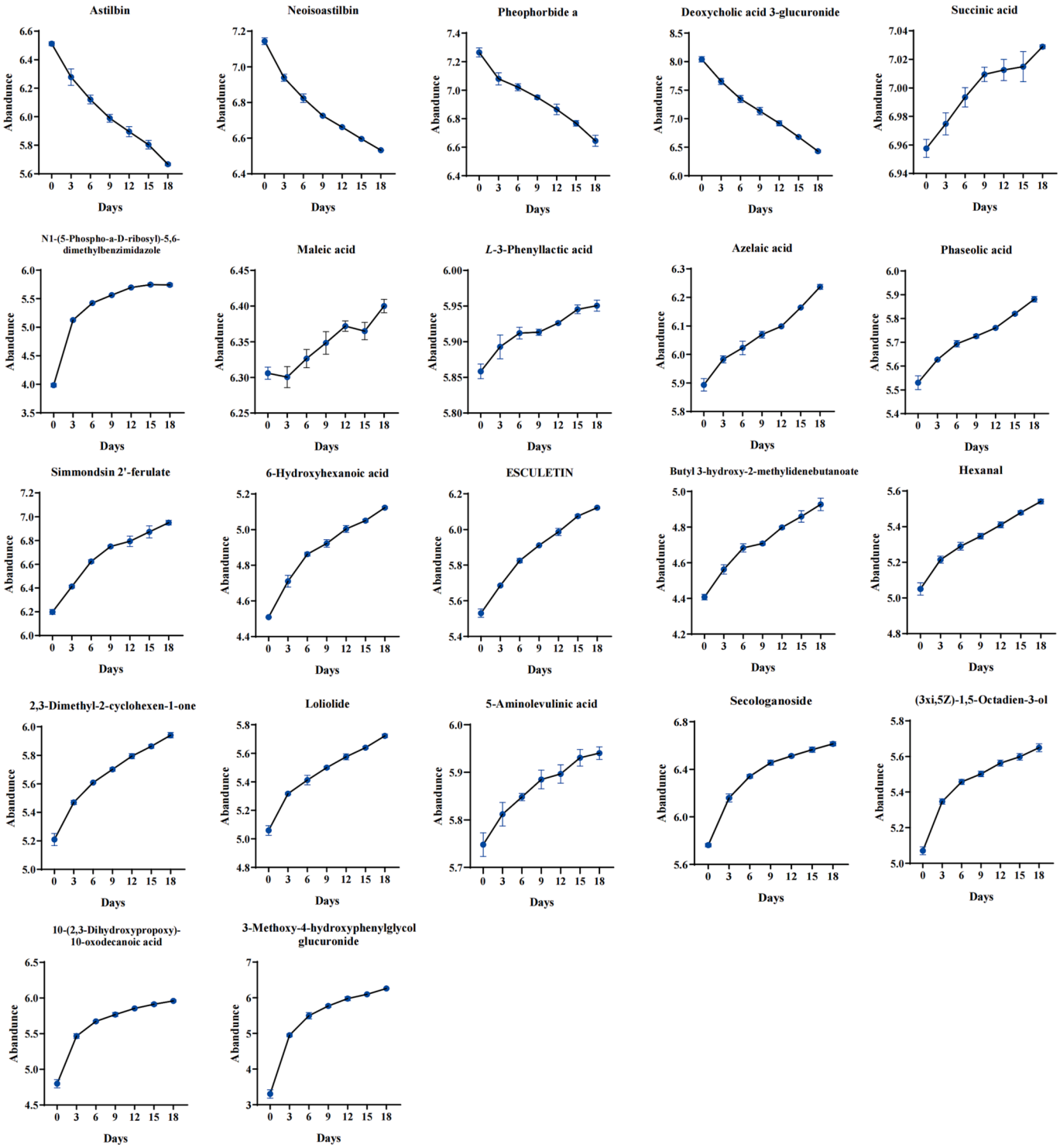

Fig. 7. The content change of differential metabolites under light treatment with different storage time.

with up-regulated levels such as secologanoside, maleic acid, and succinic acid. For down-regulated differential metabolites, astilbin and neoisoastilbin are dihydroflavonol glycosides that widely exist in plants and functional food, with various biological activities such as selective immunosuppression, antioxidant, anti-inflammatory, and hypolipidemic (Ding et al., 2023). Astiltin is unstable in light and should be stored away from light (Sui and Wang, 2015).

Chlorophyll is cleared by two enzymes to produce pheophorbide a, as an intermediate product, which continues to be rapidly broken down (Das et al., 2018). Combined with the mechanism of chlorophyll degradation in previous reports, pheophorbide a oxygenase could degrade pheophorbide a into unstable red chlorophyll catabolite and eventually form colorless primary fluorescent chlorophyll catabolite. This is a key step of degradation metabolism in the chlorophyll degradation pathway (Aubry et al., 2020; Jockusch and Kräutler, 2020). This explains the content decrease of chlorophyll and pheophorbide a in the fading process under light treatment of honeysuckle.

For up-regulated differential metabolites, loliolide, a metabolite from carotenoid, is a signaling chemical in allelopathic interaction (Li et al., 2020). In this study, the increase of loliolide is the result of

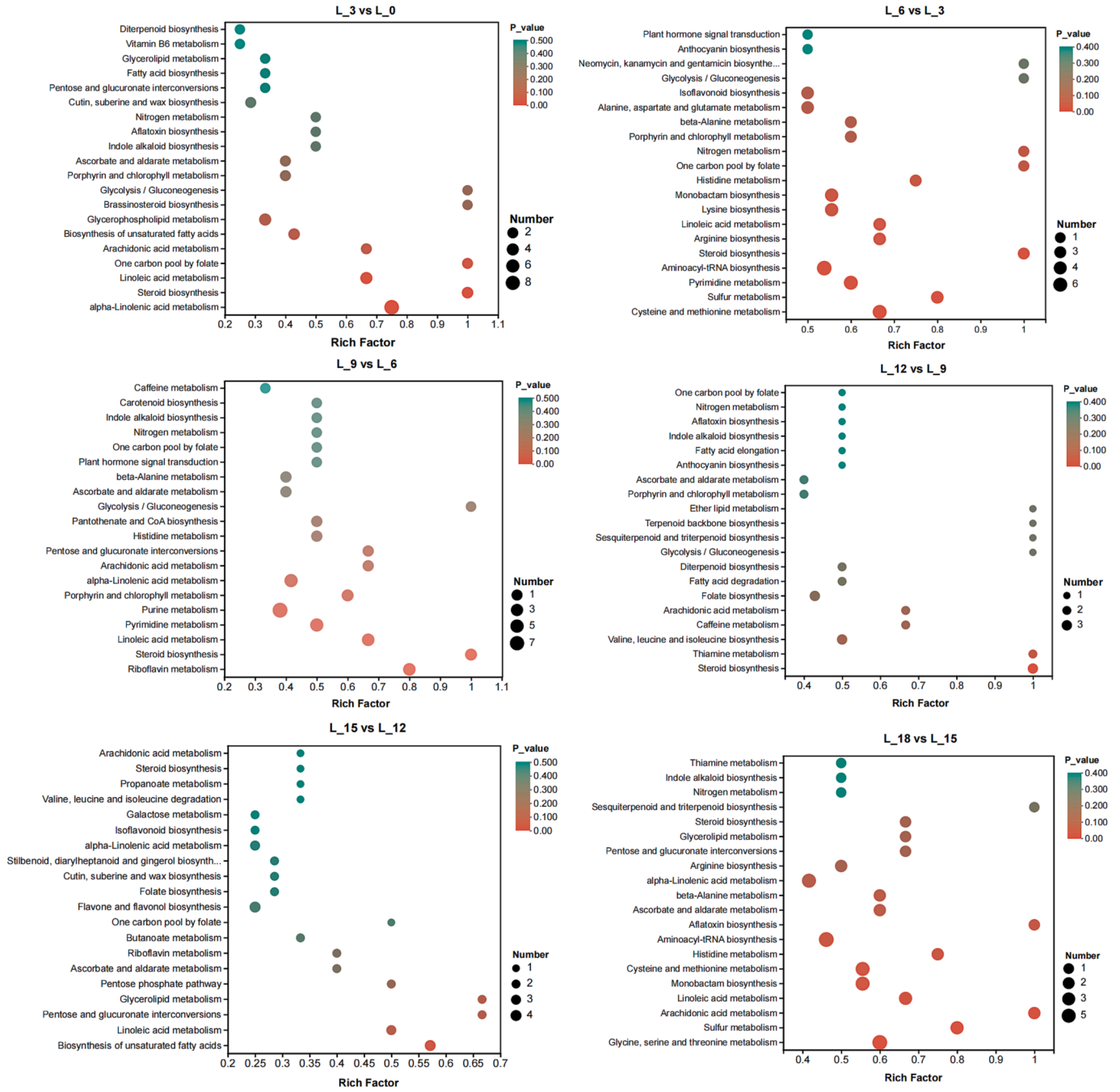

Fig. 8. KEGG enrichment pathways analysis (Top 20) of differential metabolites under light treatment with different storage time.

carotenoid metabolism under light treatment, which corresponds with the decrease of carotenoid content in section 3.2.

3.8. *KEGG annotation and enrichment analysis*

The metabolic pathways of these differential metabolites in each pairwise comparison were explored by Kyoto Encyclopedia of Gene and Genomes (KEGG) database. As shown in Fig. 8, the first 20 metabolic pathways were annotated and enriched. Porphyrin and chlorophyll metabolic pathways were enriched in 0 vs 3, 3 vs 6, 6 vs 9, and 9 vs 12. Chlorophyll metabolism was considered to be the main influence leading to the fading within 0 to 9 d. This result was consistent with the obvious color change of honeysuckle under light treatment. It was not been found in both 12 vs 15 and 15 vs 18 in main pathways. This pathway was not considered to be a dominant role although the content of pheophorbide a and chlorophyll continued to decline from 12 to 18 d. This result was consistent with the non-obvious color change during this period.

There were some pathways were overlapped such as porphyrin and chlorophyll metabolism and phenylpropanoid biosynthesis in all metabolic pathways. Porphyrin and chlorophyll metabolism serves as a basic step for chlorophyll synthesis and degradation, which was closely related to the tolerance of plant to light (Yamatani et al., 2022). It was found that the physiological phenomenon of plant and food was related to the degradation of chlorophyll, such as varying degrees of chlorosis (Xiong et al., 2023). These results indicated the chlorophyll degradation could cause honeysuckle fading based on metabolic pathways under light treatment during storage once again.

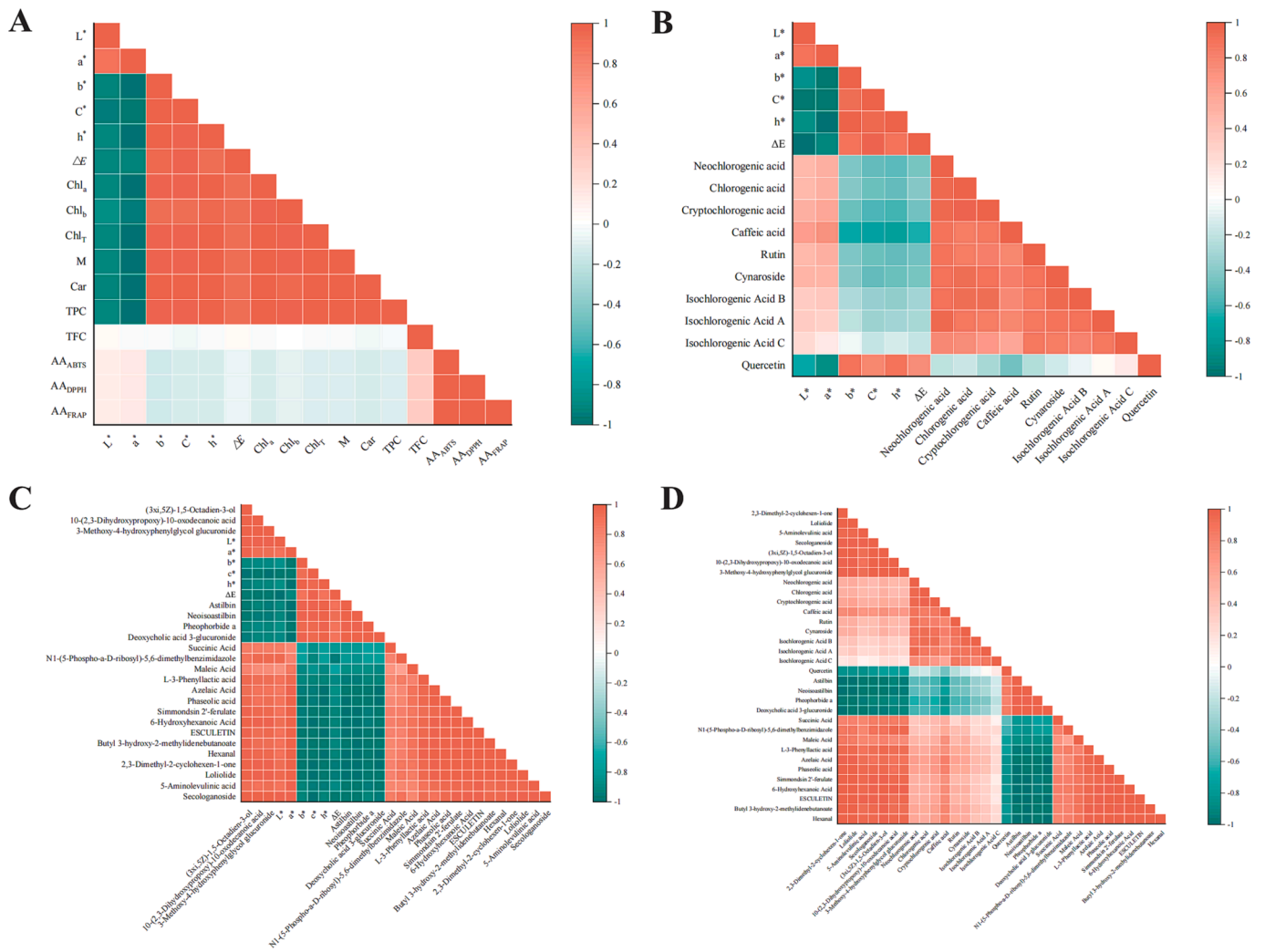

Fig. 9. Correlation of honeysuckle in the fading process. (A) Color parameters and physiological indicators, (B) color parameters and bio-active compounds, (C) color parameters and differential metabolites, (D) bio-active compounds and differential metabolites.

The phenylpropanoid biosynthesis pathway, a major secondary metabolism in plant, was activated under abiotic stress conditions (light, drought, heavy metal, salinity, and/or temperature) to result in accumulation of numerous antioxidants such as flavonoid and phenol (Glagoleva et al., 2022; Sharma et al., 2019). In this study, phenylpropanoid biosynthesis was enriched under light treatment with different storage time. Interestingly, the content of many differential metabolites from phenylpropanoid biosynthesis was found to increase, such as scopoletin, syringin, coumaric acid, and *L*-tyrosine. Similar results have been reported during food storage process (Xiao et al., 2021; Xu et al., 2022). Moreover, light has been shown to stimulate the activity of phenylalanine aminolyase, a key enzyme in the phenylpropanoid metabolic pathway that promotes chlorogenic acid and its derivatives synthesis (Lamb et al., 1977). Similarly, light could enhance the accumulation of phenylpropanoids and flavonoids (Cuong et al., 2018). In addition, phenylalanine, tyrosine, tryptophan biosynthesis and phenylalanine metabolism pathways were enriched in the whole process of our study. And more, LED light source irradiation could improve the transcription level of mRNA and promote the accumulation of phenolic compounds such as rutin, chlorogenic acid, and kaempferol (Thwe et al., 2014). These findings provided strong evidence for the increase of chlorogenic acid and other nine bio-active compounds in our study. Meanwhile, this fully explained why the content of total flavonoid and antioxidant activity does not decrease significantly during the whole process under the light.

3.9. Correlation analysis

In general, color attributes have significant correlations with chemical composition in foods and TCMs. The chemical composition of camel colostrum had high correlation with the color parameters a^* and b^* (El-Hatmi et al., 2023). The antioxidant activity of purple chrysanthemum tea was significantly stronger than that of yellow chrysanthemum tea (Han et al., 2019). ABTS scavenging capacity of coffee pulp was positively correlated ($p < 0.05$) with ΔE but negatively correlated with L^*, a^*, and b^* (Lu et al., 2023).

To assess the relationship between the color and chemical components in the fading process, correlation analysis was carried out and shown in Fig. 9. For physiological indicators, chlorophylls, carotenoid, and TPC were negatively correlated with L^* and a^* but positively correlated with b^*, C^*, h^*, and ΔE. Both TFC and AAs had little correlation with these color parameters. For bio-active compounds, only quercetin had significant negative correlation ($p < 0.01$) with L^* and a^* while weak positive correlation with b^*, C^*, h^*, and ΔE. Twenty-two differential metabolites were positively correlated with L^* and a^* except astilbin, neoisoastilbin, pheophorbide a, and deoxycholic acid 3-glucuronide. Besides, there was a weak correlation between differential metabolites and 10 bio-active compounds.

On the whole, most compounds of honeysuckle were considered to the light-sensitive substances because their content had a high correlation with color parameters. Meanwhile, both color and differential

metabolites were weakly correlated with the bio-active compounds. These bio-active compounds were not significantly affected by light treatment although their color had significant change.

4. Conclusion

In this study, the effects of light on the storage process of honeysuckle were explored in combination with physico-chemical indicators and chemical components. Through simulated accelerated tests, the significant color changes (green-white to yellow-white) were observed in only 18 days under light irradiation, accompanied by rapid degradation of pigment components including chlorophyll and carotenoids. Even so, combined with changes in antioxidant activity, total flavonoids and bio-active compounds, its value had not decreased, and it could still be used for medicinal and edible purposes. The present study provides comprehensive information on the quality changes of honeysuckle under light during storage, fully explains the reasons for color change but quality remains stable, and also provides data support for the characteristics described in the Chinese Pharmacopoeia.

CRediT authorship contribution statement

Zhenying Liu: . **Yunxia Cheng:** Formal analysis, Investigation, Software, Validation. **Zhimao Chao:** Validation, Resources, Conceptualization, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.arabjc.2023.105595.

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