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ORIGINAL ARTICLE

A metabolomics-based approach for the evaluation of off-tree ripening conditions and different postharvest treatments in mangosteen (*Garcinia mangostana*)

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Abstract

Introduction Metabolomics is an important tool to support postharvest fruit development and ripening studies. Mangosteen (*Garcinia mangostana* L.) is a tropical fruit with high market value but has short shelf-life during postharvest handling. Several postharvest technologies have been applied to maintain mangosteen fruit quality during storage. However, there is no study to evaluate the metabolite changes that occur in different harvesting and ripening condition. Additionally, the effect of postharvest treatment using a metabolomics approach has never been studied in mangosteen.

Objectives The aims of this study were to evaluate the metabolic changes between different harvesting and ripening condition and to evaluate the effect of postharvest treatment in mangosteen.

Methods Mangosteen ripening stage were collected with several different conditions ("natural on-tree", "random on-tree" and "off-tree"). The metabolite changes were investigated for each ripening condition. Additionally, mangosteen fruit was harvested in stage 2 and was treated with several different treatments (storage at low temperature (LT; 12.3 ± 1.4 °C) and stress inducer treatment (methyl jasmonate and salicylic acid) in comparison with control treatment (normal temperature storage) and the metabolite changes were monitored over the course of 10 days after treatment. The metabolome data obtained from gas chromatography coupled with mass spectrometry were analyzed by multivariate analysis, including hierarchical clustering analysis, principal component analysis, and partial to latent squares analysis.

Results "On-tree" ripening condition showed the progression of ripening process in accordance with the accumulation of some aroma precursor metabolites in the flesh part and pectin breakdown in the peel part. Interestingly, similar trend was found in the "off-tree" ripening condition although the progression of ripening process observed through color changes occurred much faster compared to "on-tree" ripening. Additionally, low-temperature treatment is shown as the most effective treatment to prolong mangosteen shelf-life among all postharvest treatments tested in this study compared to control treatment. After postharvest treatment, a total of 71 and 65 metabolites were annotated in peel and flesh part of mangosteen, respectively. Several contributed metabolites (xylose, galactose, galacturonic acid, glucuronate, glycine, and rhamnose) were decreased after treatment in the peel part. However, low-temperature treatment did not show any significant differences compared to a room temperature treatment in the flesh part.

Conclusions Our findings clearly indicate that there is a similar trend of metabolic changes between on-tree and off-tree ripening conditions. Additionally, postharvest treatment directly or indirectly influences many metabolic processes (cell-wall degrading process, sweet-acidic taste quality) during postharvest treatment.

Keywords Mangosteen · *Garcinia mangostana* · Postharvest technology · Metabolic profiling · Gas chromatography–mass spectrometry

1 Introduction

The fruit ripening process is a developmentally regulated process as a complex result of biochemical and physiological changes including firmness, color, taste, aroma, and texture of fruits (Bouzayen et al. 2010; Sonia Osorio et al. 2013;

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Paul et al. 2012; Pirrello 2009). Ripening process of climacteric fruits such as mangosteen, banana, papaya, mango, is characterized by an autocatalytic response of ethylene existence around the fruits (Asif et al. 2014; Fabíola et al. 2009; Lerslerwong et al. 2013; White et al. 2016). Fruit ripening process is affected by various changes in metabolic pathways and sometimes unique chemical changes leading to ripening-associated changes. Detailed study of ripening process is necessary to evaluate optimal harvest maturity and fruit quality (Czarny et al. 2006; García et al. 2015; Guo and Ecker 2004; Wang et al. 2002).

Mangosteen (*Garcinia mangostana* L.) is a tropical fruit with a high market value but with a relatively short shelf-life indicated by rapid changes in its peel color (Paull and Ketsa 2002; Piriyavinit et al. 2011). Mangosteen peel color is a major criterion to judge maturity and for grading of fruit quality (Palapol et al. 2009; Parijadi et al. 2018; Paull and Ketsa 2014; Tongdee and Suwanagul 1989). In the fresh fruit market, mangosteen fruits are harvested when the peel color is light-greenish yellow scattered with a pinkish spot or recognizable as stage 2. Later on, the peel color changed to a mix of red and purple and later, when it is ready to be commercialized, the fruit color will reach deep black purple or recognizable as stage 6. By the time, peel color of deep black purple indicates that the fruit is suitable for consumption (Makhmudah Edris 2010; Ongkunaruk and Piyakarn 2011). Because mangosteen has a relatively short shelf-life, postharvest packaging and handling strategies technology development are necessary to achieve optimal fruit maturity and maintain its quality.

Postharvest technology development is one of the most important processes which greatly affects food quality. Several postharvest technologies (low temperature, edible coating) have been extensively developed to reach the most-favorable condition either for quality improvement or shelf-life improvement in several climacteric fruits such as banana, plum, and peach (Bonghi et al. 2018; García et al. 2014; Raghav et al. 2016; Ren Yinzhe 2013; Valero et al. 2013; De Virginia Vasconcelos Facundo et al. 2012). Existing studies on mangosteen postharvest technologies have been focused on physio-chemical parameter or targeting only a few process related to color changes, texture, and enzymatic assay in the peel part (Dangcham and Ketsa 2007; de Castro et al. 2012; Kondo et al. 2003; Lerslerwong et al. 2013; Manurakchinakorn et al. 2016; Piriyavinit et al. 2011). In recent years, metabolomics approach is widely used as an important tool to support postharvest fruit development and ripening studies including melon, strawberry, apple, grape, sapodilla, litchi, and peach (Ali et al. 2011; Allwood et al. 2014; Biais et al. 2010; Bonghi et al. 2018; Das and De 2015; Hatoum et al. 2014; Kanellis et al. 2016; Moing et al. 2011; Osorio et al. 2011, 2012; Yun et al. 2015).

In order to better elucidate the complex mechanism of metabolic response during postharvest technologies in mangosteen, we performed gas chromatography-mass spectrometry (GC–MS)-based metabolite profiling of mangosteen from different ripening stages in postharvest condition (herein referred to as "off-tree" condition) in comparison with our previous work on mangosteen ripening in natural condition (herein referred to as "on-tree" condition). Furthermore, we also compared the effect of different postharvest treatments to ripening process of mangosteen. The postharvest treatments used were low temperature treatment and stress inducer treatment using methyl jasmonate and salicylic acid.

Multivariate analysis was then performed to correlate specific metabolites with color changes that occur during mangosteen postharvest treatment. This is the first report of application of metabolomics approach for the postharvest study of mangosteen.

2 Materials and methods

2.1 Plant materials

Mangosteen samples used were Raya cultivar, a common commercial cultivar in Indonesia. Mangosteen fruit was freshly harvested in February 2018 in Center of Tropical Horticultural Studies, Bogor Agricultural University, Bogor, Indonesia. Mangosteen plants were grown in an open field (6°38′S × 106°49′E) as a part of an experiment field located in Southeast Asian Regional Center for Tropical Biology (SEAMEO BIOTROP), Bogor, Indonesia and Pasir Kuda experimental field (6°36′S x 106°47′E), Center for Tropical Horticultural Studies, Bogor Agricultural University (Bogor, Indonesia). Cultivation, irrigation, watering, fertilization and pathogen-pest control were performed according to the respective local commercial practices for all samples used in this study.

2.2 Sample treatment

2.2.1 Samples for the study of "off tree" ripening stages

Mangosteen fruit from Bogor, Indonesia corresponding to seven different ripening stages (stages 0–6) were used in this study. The definition of ripening stage follows the description in Tongdee and Suwanagul (1989) and Parijadi et al. (2018). Mangosteen fruits from each ripening stage with three biological replicates were collected from different trees and different cultivation area in random. Sample collection was done in 1 day on February, 19th 2018 in Center of Tropical Horticultural Studies, Bogor Agricultural University,

Bogor, Indonesia. These samples were obtained at the optimum time for harvest, from 9 to 12 am in the morning.

These samples were obtained to test the robustness of our previously established model for ripening and is herein referred to as "random on tree" condition. These samples were analyzed together with natural ripening condition samples obtained in 2016, in which samples were harvested by monitoring the ripening stages in the course of 2 months and collection was done from three different trees that were tagged since flowering stage (further will be mentioned as "natural on-tree"). Details on sample condition of "natural on-tree" is described in the previous study (Parijadi et al. 2018).

In addition, we obtained additional sample set to imitate postharvest condition ("off-tree" ripening condition). Fruits were selected at green-purple maturity (stage 2) by using color changes parameter. All stage two mangosteen samples were obtained at the optimum time for harvest, from 9 to 12 am in the morning. These stage two mangosteen samples was stored at room temperature for 10 days and daily sampling was conducted to observe the progression of ripening in room temperature. For comparison between on-tree and off-tree ripening conditions, two sample sets collected in 2018 were analyzed together, namely "random on-tree" and "off-tree" ripening conditions. The sample sets used in this study are described in Fig. 1. All mangosteen samples were fast quenched in liquid nitrogen and stored in − 20 °C prior to extraction and homogenization process.

2.2.2 Postharvest treatment samples

Fruits were selected at green–purple maturity (stage 2) by using color changes parameter and were treated with several postharvest treatments (Fig. 1). Stage two was selected as an initial stage for postharvest treatment samples to imitate the fresh fruit market condition. Four sets of experimental treatments were adopted during this experiment: low temperature (LT; 12.3 ± 1.4 °C); 0.5 mM methyl jasmonate (MeJa), and 5.0 mM of salicylic acid (SA) treatment. Room temperature (RT; 27 ± 3.4 °C) storage was used as a control treatment.

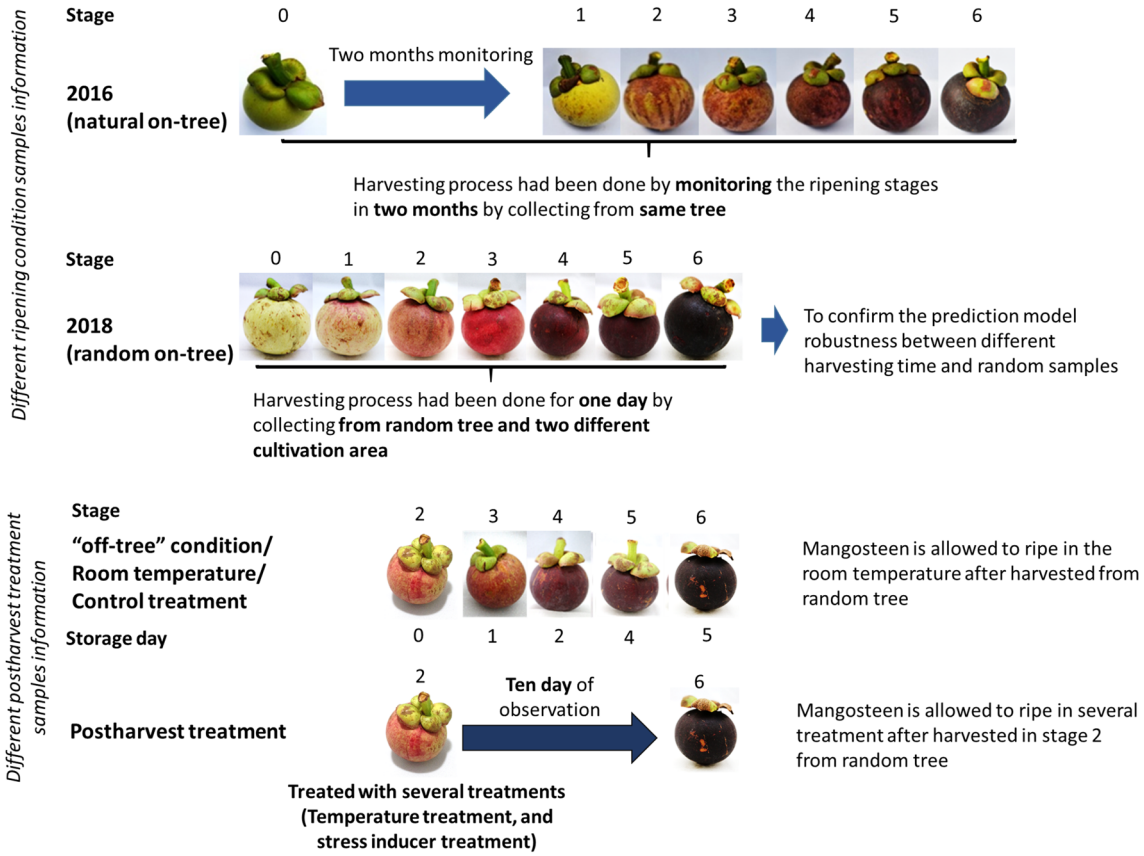


Fig. 1 Samples information of this study. Seven ripening stages were collected from the different ripening condition. "Natural on-tree" condition samples were collected from 2016 as described in the previous study (Parijadi et al. 2018). "Random on-tree" samples were randomly collected from different tree and different cultivation area in 2018. The number shown in the figures represent the ripening stage. Both ripening condition samples were analyzed together using GC–MS. The "off-tree" condition was performed by using stage two as an initial stage which allow to ripen in the room temperature until reach stage six of ripening. "Off-tree" samples will further used as a control in postharvest treatment. Stage two was randomly collected and treated with several postharvest treatments

These postharvest treatments were previously reported as an effective treatment to prolong mangosteen shelf life (Mustafa et al. 2018; Piriyavinit et al. 2011). For temperature treatment, the fruits were stored at RT and LT for 10 days and fruits were randomly collected for each treatment every day. For stress inducer treatment, treated and untreated fruit (control) were allowed to dry at room temperature after treatment and was stored at room temperature for 10 days. Similarly, with temperature treatment, sample collection was performed daily for each treatment. All mangosteen samples were fast quenched in the liquid nitrogen and stored in − 20 °C prior to extraction and homogenization process.

2.3 Color changes observation

Color changes in fruit peels were measured using Konica Minolta CM-2500d before homogenization and extraction of mangosteen as described in the previous study (Parijadi et al. 2018). Measurement of color changes was performed using the previously established CIELAB method with illuminant D65, observer angle 10^{o} (Tosetti et al. 2014; Voss 1992). The measured data was proceed using Spectramagic NX software (Konica Minolta, Tokyo, Japan). CIELAB method (dLab color space value) was used as the color solid representative value to represent mangosteen color changes in the ripening process. L* indicates the lightness of the samples that were measured. a* and b* values are chromaticity diagram that describes red-green color for a* values and yellow-blue color for b* values.

2.4 Samples extraction and derivatization

Two different parts of mangosteen, namely flesh, and peel were separated and homogenized. Homogenized mangosteen samples were extracted by following the procedure of liquid–liquid extraction using methanol:chloroform: water (5:2:2) as described in the previous study (Parijadi et al. 2018). In this protocol, mangosteen samples, blank samples, and quality control (QC) samples are extracted together and lyophilized on a single day. Ribitol as an internal standard was added into each sample during the extraction. QC samples were prepared by collecting small aliquots of each biological samples obtained in this study. The use of QC samples is to represent sample type under analysis. The number of QC samples were decided as described in the previous study (Dunn et al. 2011). Lyophilized samples were derivatized by following the procedure as described in the previous study (Parijadi et al. 2018). Briefly, oximation and silylation were performed using methoxyamine hydrochloride (20 mg/ml in pyridine) and *N*-methyl-*N*-trimethylsilyltrifluoroacetamide (MSTFA), respectively.

2.5 Gas chromatography–mass spectrometry

Intracellular metabolites of all samples were measured by Shimadzu Ultra QP-2010 GC–MS with an InertCap 5 MS/NP (30 m, 0.25 mm i.d., 0.25 µm film thickness, GL Sciences, Japan). Tuning and calibration of the mass spectrometer were done prior to analysis (Supplementary Table S1). The derivatized samples were injected to GC–MS. GC and MS conditions were performed by following the procedure as described in the previous study (An et al. 2014; Parijadi et al. 2018). The netCDF files along with associated metadata have been deposited into the Metabolomics Repository of EBI MetaboLights (Note: Link soon to be attached together after it is verified by Metabolights team).

2.6 GC–MS data extraction, alignment, and peak identification

The raw chromatographic data were converted into ANDI files (Analytical Data Interchange Protocol, *.cdf) using the GC–MS solution software package (Shimadzu, Kyoto, Japan) for GC–MS analysis. Peak detection, baseline correction, and peak alignment of retention times were performed on the ANDI files using MetAlign (Wageningen; can be downloaded freely at http://www.wageningenur.nl/). Detailed metAlign parameter information is described in supplementary Table S2. All of the converted data was aligned together with QC samples as a reference. The peak intensity of each metabolite was normalized based on the ribitol internal standard.

The detected peaks were tentatively identified by comparing the retention indices and unique mass spectra with our in-house library database, AIoutput2 (version 1.30) annotation software (Tsugawa et al. 2011). Detailed AIOutput parameter information is described in supplementary Table S3. Retention indices of all detected metabolites were calculated based on standard alkane mixture (C10–C40). Additionally, the retention time of each tentatively identified metabolites was compared with the National Institute of Standards and Technology (NIST) library. Tentative identification of metabolites refers to Metabolites Standard Initiatives (MSI) level 2 identification procedures (Fiehn et al. 2007).

2.7 Multivariate data analysis

Multivariate analysis was done using SIMCA-P + version 13 (Umetricts, Umea, Sweden) for principal component analysis (PCA) and partial least square (PLS) regression by following the procedure as described in the previous study (An et al. 2014). Hierarchical cluster analysis (HCA) was performed to further clarify metabolite distribution during mangosteen ripening condition and postharvest treatment

using MeV: Multi-Experiment Viewer (download freely at http://www.tm4.org/mev.html). The derived data from PCA was statistically analyzed using negative Pearson's correlation with gene and sample leaf optimization and average linkage clustering without any cut-off value applied to see the correlation between ripening stages in HCA.

3 Results and discussions

3.1 Metabolic changes during mangosteen ripening process in different ripening condition

The seven mangosteen fruit ripening stages were randomly collected in 1 day from different trees and from two different plantations. The mangosteen ripening stages were classified based on our previous study (Parijadi et al. 2018). These samples ("random on-tree") were compared with natural ripening condition ("natural on-tree") (Fig. 1) to evaluate whether the same ripening trend occur in different harvesting year, different collection method and different plantations.

Colorimetric analysis revealed increasing color solid representative (dLAB value) as peel color shifted in both ripening stages from green to purple–black in both ripening conditions (Supporting Information, Table S4). We also noted there is no significant difference in dLAB value between "random on-tree" ripening condition and "natural on-tree" ripening condition, corroborating previous studies (Palapol et al. 2009; Parijadi et al. 2018).

Metabolic changes between "random on-tree" and "natural on-tree" ripening condition were furtherly compared with GC–MS analysis. Three fruits from all ripening stages in "random on-tree" and "natural on-tree" ripening condition were used and the analysis was performed separately for peel and flesh part of mangosteen. We tentatively identified 70 metabolites from flesh and 60 metabolites from peel part in both ripening conditions using the National Institute of Standards and Technology (NIST) and our laboratory in-house library (Supporting Information, Table S5, and S6).

Two PCA models of the GC–MS-derived dataset were generated, the first based upon the fruit peel samples and second based upon the fruit flesh samples, the first two PCs within each model provided a total explained variance (TEV) of 49.8%, and 49.47%, respectively (Fig. 2a). The results showed the trend of PC1 describes the metabolite changes trends during mangosteen between "random on-tree" and "natural on-tree" ripening process from raw to ripen stages. PCA results also clustered based on metabolites distribution as explained in the previous study (Parijadi et al. 2018).

Hierarchical cluster analysis (HCA) was performed to further clarify metabolite distribution between "random on-tree" and "natural on-tree" ripening process. HCA resulted in clustering of metabolite intensity showing identical or highly similar patterns of intensities throughout all GC–MS datasets of different ripening condition (Supplementary Information, Fig. S1). In accordance with PCA results, HCA results showed a clear clustering based on its ripening stages in both ripening conditions. These results also successfully reproduced the previous study which describes heat-map results in "natural on-tree" ripening process (Parijadi et al. 2018). Therefore, we used the "random on-tree" samples as they are collected in the same harvesting time in 2018 with "off-tree" samples for the next comparison between "on-tree" and "off-tree" ripening stages. In "off-tree" samples, stage 2 mangosteen was allowed to ripen in room temperature over the course of 10 days observation. Based on color changes parameter, we assigned the ripening stage of mangosteen from stage 2 to stage 6. The progression of ripening was much faster in "off tree" samples compared to the ripening that occurred naturally "on tree". When harvested in stage 2, the peel color was light-greenish yellow scattered with a pinkish spot. At day one after harvest, the fruits ripened to stage 3, then reached stage 4 at day two, stage 5 at day four and reached fully ripen stage 6 at day five. The peel color then remained constant until day 10 (Fig. 1).

Metabolic changes between "on tree" and "off tree" ripening condition were furtherly compared with GC–MS analysis. Three fruits from different ripening stages in "on-tree" and "off-tree" ripening condition were used and the analysis was performed separately for peel and flesh part of mangosteen. We tentatively identified 57 metabolites from flesh and 72 metabolites from peel part in both ripening conditions using the NIST and our laboratory in-house library (Supporting Information, Table S7). Two PCA models were generated, the first based upon the peel samples and second based upon the flesh samples, the first two PCs within each model provided a total explained variance (TEV) of 41.5%, and 56.3%, respectively (Fig. 2b). The results showed the trend of PC1 describes the metabolite changes trends during mangosteen between "on-tree" and "off-tree" ripening process from raw to ripen stages. The ripening progression in "on-tree" ripening condition shown in Fig. 2b is consistent with the previous PCA model (Fig. 2a). In the comparison between "on-tree" and "off-tree", the trend is different in samples of stage 4, 5 and 6 of peel part. Meanwhile, the trend of ripening progression in flesh part is similar between the two ripening conditions (Fig. 2b). For detailed investigation on metabolite changes that are associated with ripening in these two conditions, PLS regression was performed.

3.2 Prediction model of mangosteen ripening process in two different harvesting condition

PLS analysis was used to regress PCA variables (metabolite intensity) on color change values, allowing us to verify whether color changes can well predict metabolite-based

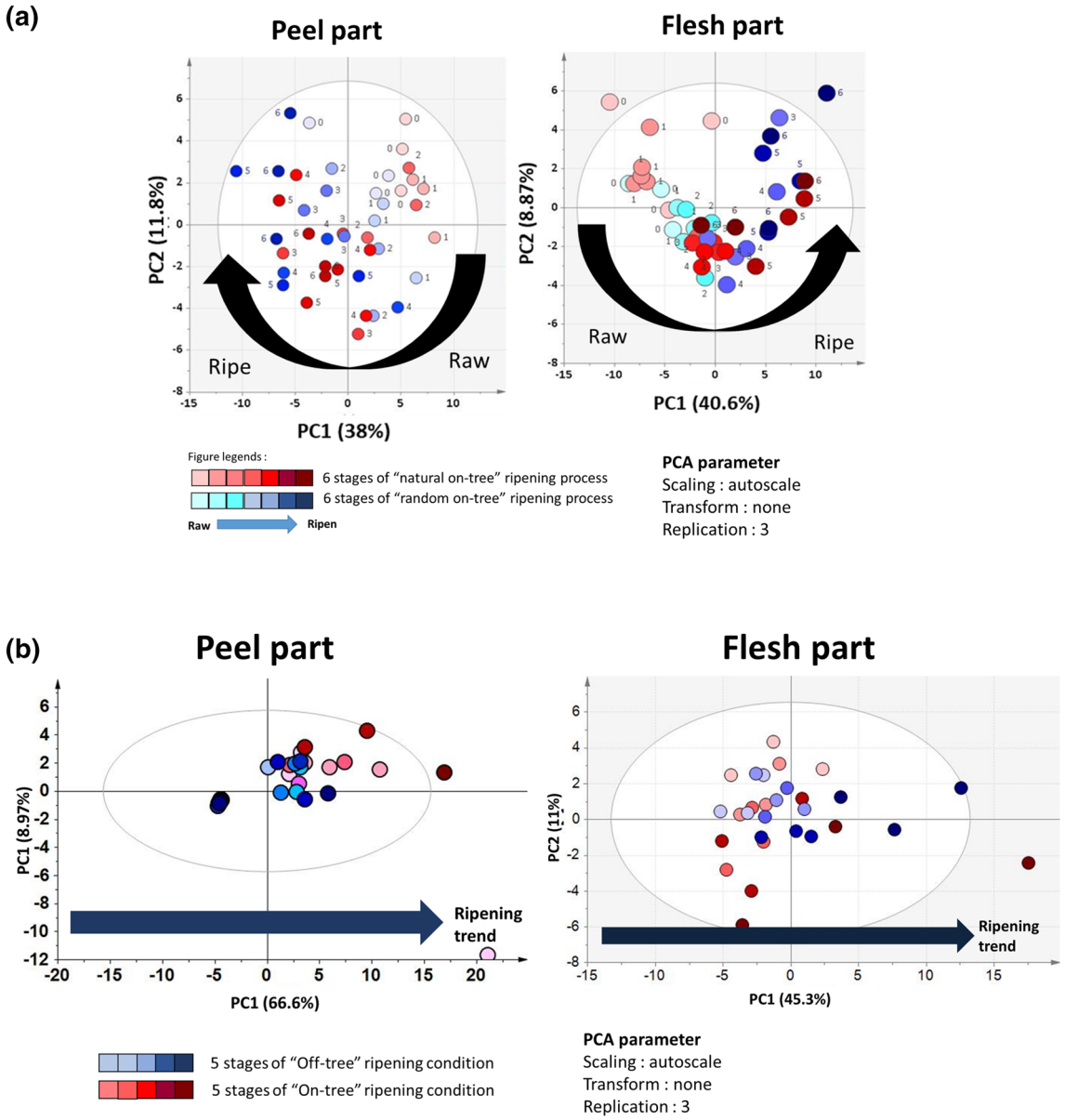


Fig. 2 Metabolites abundances during mangosteen ripening process in peel part and flesh part using PCA. **a** PCA results showed the trend of PC1 describes the metabolite changes trends during mangosteen between "natural on-tree" and "random on-tree" ripening process. **b** PCA results showed the trend of PC1 describes the metabolite changes trends during mangosteen between "on-tree" and "off-tree" condition. Scaling method, UV scale; transformation, none; $n = 3$

ripening stages. PLS analysis was conducted using data from color changes as the response variable (y-variable) and metabolite peak intensity values as the predictor variables (x-variables). Representative samples of three major ripening stages (stage 0, 2, 4, and 6), referring to the previous study (Parijadi et al. 2018), were selected as a training set, and the remaining stages were used as a data set to validate the prediction model. PLS analysis was performed in both harvesting condition to confirm their contributing metabolite differences. The constructed models have linear coefficient (R^2) and prediction ability (Q^2) greater than 0.7, both thresholds for a valid model with a good fit (Supporting Information, Table S8). The results of the PLS model indicated that different metabolites were giving large contributions to color change depending on the mangosteen part. Contributing metabolites were selected based on the five highest variable importance in projection (VIP) scores.

Primary metabolites were selected from both harvesting condition in the peel part as ripening associated metabolites (Supporting Information, Table S8). Galactose and galacturonic acid were repeatedly reported in both harvesting conditions in peel part. These metabolites have been reported

in the previous paper (Parijadi et al. 2018). Galacturonic acid and galactose were reported to have a contribution to cell wall degradation which gives an effect to fruit softening during the ripening process. This cell-wall breakdown causes a drastic decrease in firmness when the fruit is ripe (Piriyavinit et al. 2011). For example, an aspect of ripening involves an ethylene-triggered breakdown of pectin, hemicellulose in cell walls to form simple metabolites such as galactose, xylose and galacturonic acid (Hayama et al. 2006; Merchante et al. 2013). Here, we found that galacturonic acid and galactose abundance increased considerably during mangosteen ripening, implying the same process occurs in this fruit between two different harvesting conditions. Interestingly, shikimic acid was shown to be one of the important metabolites in the peel part of "on-tree" condition. Shikimic acid was repeatedly reported as a precursor metabolite for pigment and lignin production in transcriptomics studies of peach and pear fruit development (Liu et al. 2017; Zhang et al. 2016). Similar study in mangosteen reported that lignification was induced during "on-tree" fruit maturation upon hypoxic condition treatment using capillary water (Noichinda et al. 2017). Lignification in mangosteen fruit affected peel hardness which increased fruit firmness and such phenomenon occurred under low oxygen conditions or upon physical damage (Dangcham and Ketsa 2007; Siriwan Dangcham et al. 2008; Ketsa and Atantee 1998).

Additionally, primary metabolites were selected in the flesh part as ripening associated metabolites in both "on-tree" conditions (Supporting Information, Table S9). It is also mentioned that listed amino acids act as precursors for fruit aroma metabolites. Previous report mentioned several amino acids have a contribution to several volatile metabolites biosynthesis in melon (Allwood et al. 2014), These findings have a good agreement with the information of several amino acids might have a role as a precursor metabolite for plant volatile metabolites that had been reported in the previous studies (Ä Herna Ä Ndez-orte et al. 2002; Allwood et al. 2014; MacLeod and de Troconis 1982; Tieman et al. 2006). A number of flavor volatile metabolites were also found in abundance of mangosteen using headspace-solid phase microextraction (HS-SPME) which are related to specific aroma metabolites (Laohakunjit et al. 2007). In addition to amino acids, metabolites such as quinic acid, lyxose, and arabionose that were previously reported as sweet-acid related metabolites in coffee and direct precursors of volatile metabolites in several fruits were also selected as ripening associated metabolites (El Hadi et al. 2013; Jumhawan et al. 2013; Trinh et al. 2010; Zou et al. 2017).

Furthermore, phenylalanine consistently appeared as the fifth highest VIP score obtained from the PLS model in both harvesting condition in the flesh part. The metabolite has been previously reported to regulate reactive oxygen species (ROS) activity whilst also regulating pH, cell homeostasis, and structural integrity of membranes and cell walls (Schmidt et al. 2016). The roles of ROS had also been reported in fruit development and ripening. ROS has association with the increase in polygalacturonase activity, peroxidase activity and the concentration of peroxide during fruit softening process of papaya, and banana using proteomics approach (Du et al. 2016; Huan et al. 2016; Pandey et al. 2013; Yang et al. 2008).

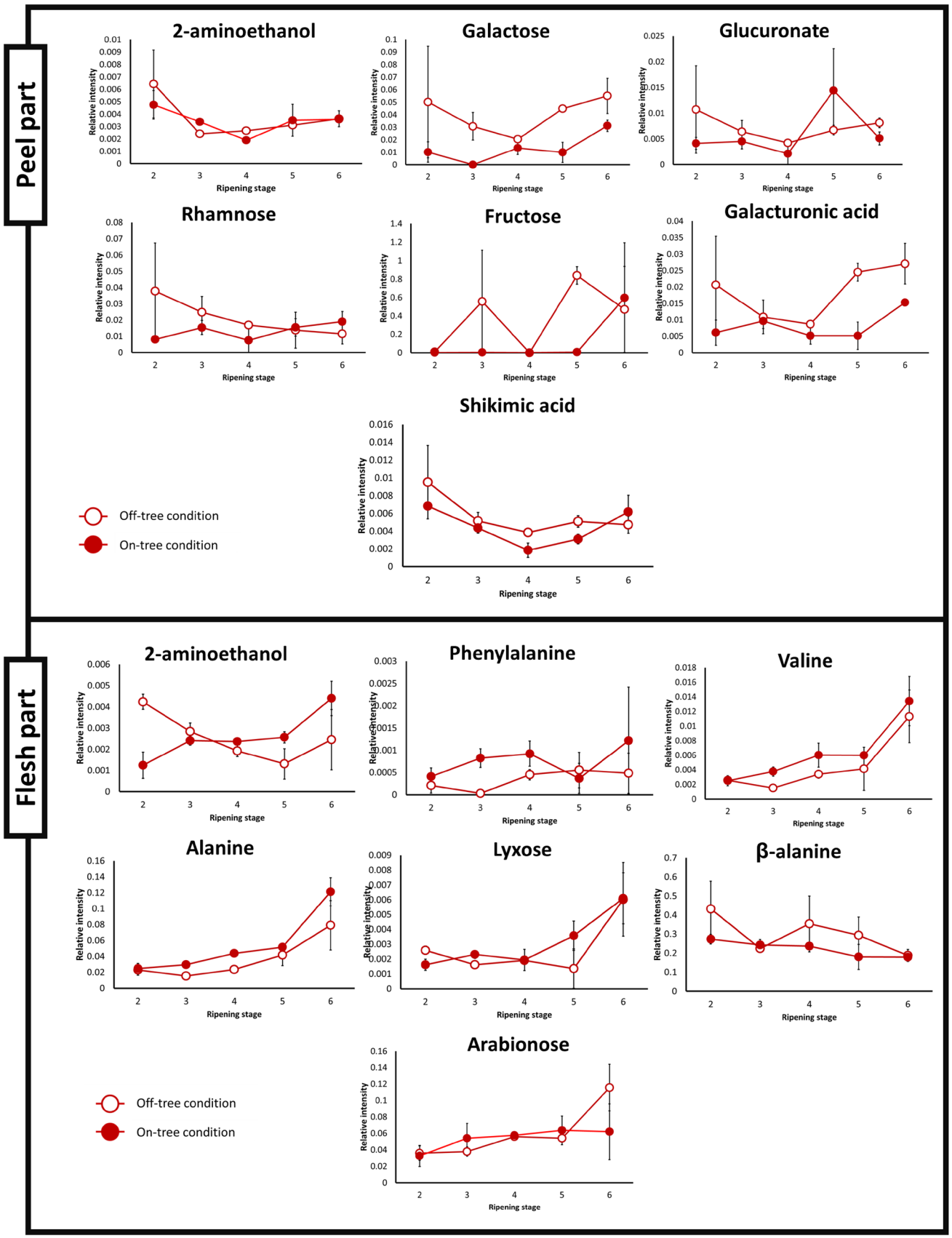
The increase of amino acids during ripening indicated that ROS activity was present during mangosteen ripening. Moreover, phenylalanine was reported as one of low oxygen stress response metabolite in the legume after waterlogging treatment (António et al. 2016). The accumulation of several amino acids can also be caused by the hypoxia condition occurring in the flesh of mangosteen since the oxygen gradient in flesh might be lower than the peel part as previously reported in pear and apple (Ho et al. 2006, 2010).

The additional PLS analysis was performed to confirm their robustness between different ripening condition. Samples of "natural on-tree" harvesting condition (stage 0–6) were selected as a training set, and the "random on-tree" samples were used as a data set to validate the prediction model. This model is herein referred to as "on tree" combined model. The constructed model has a linear coefficient (R^2) and prediction ability (Q^2) greater than 0.6 in both parts (peel and flesh part) (Supporting Information, Table S10, Fig. S2).

In order to verify our results, we added the test set into the model wherein they fit perfectly in the predicted regression line (Supplementary Information, Table S9). Moreover, the root means square error (RMSEE) was calculated to determine how well the observed color changes during ripening stages matched with the actual color changes during ripening stages of mangosteen. Result showed that the root mean square error of estimation (RMSEE = 6.90 and 5.89 in peel and flesh part, respectively) was considered as not significantly different from the root mean square error of prediction (RMSEP = 13.08 and 9.72 in peel and flesh part, respectively), thus indicating that the regression model was valid.

In mangosteen peel, similar ripening associated metabolites (galactose and galacturonic acid) with "natural on-tree" and "random on-tree" as contributing metabolites in combined prediction model (Supporting Information, Table S10). In mangosteen flesh, we identified similar metabolites as "natural on-tree" and "random on-tree" as an abundant metabolite in "on-tree" combined prediction model.

The relative intensity of major metabolites between "natural on-tree" and "random on-tree" ripening condition was compared using a line chart. Top metabolites (based on VIP score from both ripening condition) individual mangosteen parts were analyzed separately (Supporting Information, Table S8, S9). The line chart shows that the relative


Peel part
2-aminoethanol
Galactose
Glucuronate
Rhamnose
Fructose
Galacturonic acid
Shikimic acid
Off-tree condition
On-tree condition
Relative intensity
Ripening stage
Flesh part
2-aminoethanol
Phenylalanine
Valine
Alanine
Lyxose
β-alanine
Arabionose
Off-tree condition
On-tree condition
Relative intensity
Ripening stage

◂**Fig. 3** Line graph of metabolite relative intensity changes analysis using GC–MS between two ripening condition of mangosteen ripening process in **a** peel part; **b** flesh part. The relative intensity of metabolites that are important between "on-tree ripening" condition and "off-tree" ripening condition were explained using a line chart. The peak intensity of each metabolite was normalized based on ribitol internal standard. Y axis represents metabolites relative intensity and X axis represents ripening stage. Black circle represents "on-tree" ripening condition and white circle represents "off-tree" ripening condition

intensities of top metabolites were correlated with ripening stages in both ripening conditions, in accordance with ripening progress (Supplementary Information, Fig. S3).

Major metabolites relative intensities are increased in accordance with ripening stages in all conditions. Contributed soluble sugars (galactose, fructose, glucoronate, rhamnose, lyxose, and arabionose) were accumulated at the end of the ripening process between both harvesting conditions. Additionally, contributed amino acids (phenylalanine, threonine, β-alanine, alanine, isoleucine, glycine, and valine) follow the same trends during the ripening process of both harvesting conditions. Meanwhile, 2-aminoethanol relative intensity is relatively constant between the stages in all mangosteen parts.

In conclusion, we successfully developed a robust prediction model of mangosteen ripening process based on metabolome data in two different harvesting conditions with data taken in different harvesting year, sample collection and plantations.

3.3 Prediction model of different ripening condition in mangosteen

Additional PLS analysis was performed to confirm the robustness of "on tree" ripening prediction model using "off-tree" samples as test set Samples of "random on-tree" ripening condition (stage 2–6) were selected as a training set, and the "off-tree" samples (stage 2–6) were used as a test set to validate the prediction model. This model is herein referred to as "on tree" and "off-tree" combined model (Supporting Information, Table S9). The constructed model has a linear coefficient (R^2) and prediction ability (Q^2) greater than 0.6 in peel part. However, validation using RMSEE and RMSEP value of peel part prediction model was not successfully performed since the result showed a significant difference (RMSEE = 1.12, RMSEP = 227.272) between two different ripening conditions. Furthermore, the constructed model of flesh part has the prediction ability (Q^2) lower than 0.5. This indicated that different metabolites were not giving large contributions to color change depending on the mangosteen part. In two ripening conditions, significant difference may be observed if comparison was performed using all metabolites. Such observation is understandable due to very different physiological conditions between "on-tree" and "off-tree" conditions.

Despite low robustness between the two ripening conditions in PLS model, we examined the trends of ripening associated metabolites in "on-tree" and "off-tree" ripening condition as shown in Fig. 3. Interestingly, all of general ripening associated metabolites are increased in accordance with ripening stages in all ripening conditions. Degradation of pectin to galactose and galacturonic acid in peel part during mangosteen ripening stages did not show any difference between two different ripening conditions. Additionally, amino acids that were listed in the previous section also accumulated in flesh part. The accumulation of amino acids indicates similar hypoxic condition occurs in the flesh part of two different ripening condition.

The results suggest that rapid progression of ripening that occurred in "off-tree" samples did not result in any significant difference with regards to the changes in ripening associated metabolites. This is the first report that mention about the "on-tree" and "off-tree" ripening condition of mangosteen. These results can also be used as a basis to indicate the difference between ripening condition in mangosteen and can act as useful feedback for postharvest technology study.

3.4 Comparison of different postharvest treatments to prolong mangosteen shelf-life based on its physical appearance

Physical appearance (color changes) of all treated-mangosteen was evaluated during the treatment period for 10 days. Mangosteen color changes were selected based on the determination of ripening index as explained in the previous studies (Palapol et al. 2009; Parijadi et al. 2018). Peel color is also reported as an important marketing attribute of mangosteen and influences both consumer acceptance and sales. Different mangosteen samples sets were used for postharvest treatment. Stage two of mangosteen ripening was used as an initial stage before it is treated with several postharvest treatments, namely low temperature and stress inducer treatment using either salicylic acid or methyl jasmonate. All treated samples were allowed to ripe and evaluated for 10 days storage.

The peel color during treatment is shown in Fig. 4 and Supporting Information Fig. S2. When harvested in stage 2 (herein defined as day 0 after harvest), the color was light-greenish yellow scattered with a pinkish spot. On day 2 after harvest, control sample (storage in room temperature (RT) at 27 ± 3.4 °C) showed rapid change of peel color to red and purple. By day 5, it reached blackish purple color and the color remained constant until day 10. Among all postharvest treatment, low temperature (LT) showed the most significant effect to delay ripening based on the peel color, whereas stress inducer (SI) treatment did not show

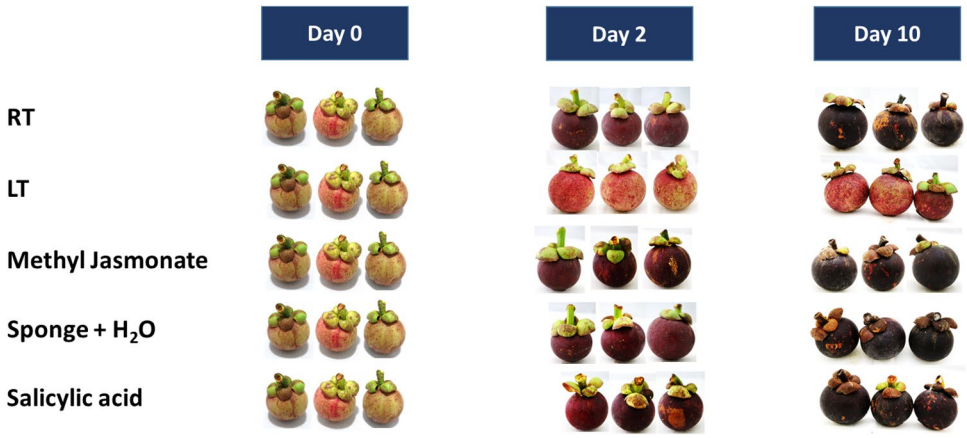


Fig. 4 A photograph of physical appearance comparison between room temperature and several temperature treatment after 10 days of observation. Fruits were selected at green-purple maturity (stage two) by using color changes parameter as an initial stage and were treated with several postharvest treatments. The fruits were then stored inside the containers at room temperature and low temperature (12.3 ± 1.4 °C) for 10 days

any significant effect compared to control. Similar physical parameter changes after LT treatment have been reported in the previous study with similar treatment (de Castro et al. 2012; Piriyavinit et al. 2011) and is considered as a suitable treatment for prolonging mangosteen shelf-life based on peel color with good reproducibility.

LT treatment effects in the peel part were furtherly evaluated by using a metabolomics approach to confirm its metabolite changes, especially for the fifth highest VIP score metabolites. We did not further analyzed the metabolite changes data of peel part after SI treatment because it did not show any effect to prolong mangosteen shelf life based on color changes. However, we analyzed the metabolite data on flesh part for all postharvest treatments to confirm the detailed effect in fruit metabolite changes.

3.5 Effect of postharvest treatment in metabolite changes of mangosteen

A total of 71 metabolites were annotated in peel part of mangosteen samples treated with LT versus control (Supporting Information, Table S10). After GC–MS analysis, the data set of LT treatments and control were subjected to multivariate statistical analysis. PCA model was generated, with each explaining an accumulated TEV for the first two PCs of 42.7% (Fig. 5a), attributable to the metabolite variation in the peel of the fruits stored under the two different temperature regimes. PC1 describes the difference of metabolite changes trends between LT treatment and control from raw to ripen stages in the peel part (Fig. 5a). As can be seen in the PCA score plot of peel part, LT treatment showed a markedly different metabolite changes with that of control. HCA analysis results (Fig. 6a) in the peel part allows the classification of two treatment into three major groups that explained raw to ripen stage after storage treatment. HCA resulted in clustering of low-temperature treatment together with day 1 and 3 in room temperature. Low-temperature treatment seems to delay the ripening process based on HCA result of peels part.

The relative intensity of metabolites with highest VIP score obtained from all PLS models during LT and control were compared using a line chart in a time course manner for 10 days. Top metabolites (Supporting Information, Table S8, S9) and flavor-related metabolites from individual mangosteen parts were analyzed separately. Relative intensities of important metabolites in LT treatments and control were shown in Fig. 7. These metabolites all had lower relative intensity in LT treatment compared to control treatment in both parts of mangosteen.

In mangosteen peel part, we identified several metabolites related with citric acid cycle (oxalacetic acid, isocitric acid + citric acid, and succinic acid) were decreased together with the storage time (Fig. 7). The metabolites have a strong correlation with the respiration process. It is suggested that LT treatment decreased the respiration process in mangosteen, implying the same process occurs in other fruit such as banana, tomato, sapodilla, litchi, and peach (Bonghi et al. 2018; Bustamante et al. 2012; Das and De 2015; De Virginia Vasconcelos Facundo et al. 2012; Yun et al. 2015; Zou et al. 2017). Previous research has indicated the reduction of ACO activity during 15 °C storage of mangosteen which related to anoxia condition during storage treatment (Dangcham and Ketsa 2007; Dangcham et al. 2008; Piriyavinit et al. 2011). In conclusion, this study suggests that LT treatment is the most effective treatment to prolong mangosteen shelf-life based on color appearances and metabolites changes in the peel part.

In order to evaluate the effect of postharvest treatment in the flesh part, metabolomics approach was performed in LT and SI treatment separately. Two PCA models were generated, with each explaining an accumulated TEV for the

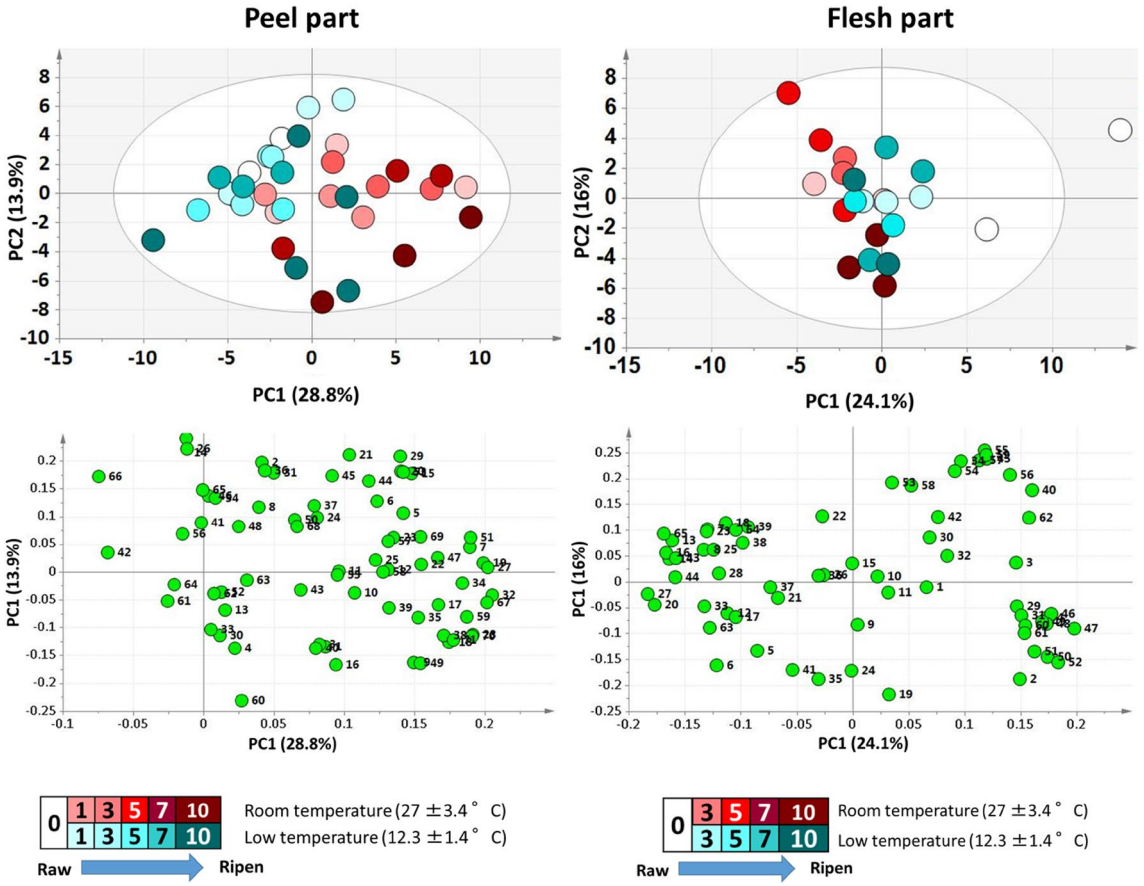


Fig. 5 Metabolites abundances during mangosteen LT treatment in **a** peel part; **b** flesh part. PCA score plot results showed the trend of PC1 describes the metabolite changes trends during mangosteen between room temperature and low temperature storage condition. PCA loading plot shows the contributions of each metabolite on the changes during treatment in score plot according to the distance to the origin. Day 1 sample of flesh was removed from analysis because it is an outlier. PCA parameter is described as follows, scaling method, UV scale; transformation, none; $n = 3$. The detailed information about metabolites name is described in the Supplementary Table S10 for peel part and Table S11 for flesh part

first two PCs of 40.1% (Fig. 5b) and 42.6% (Supplementary Information, Fig. S5), attributable to the metabolite variation in the flesh of the fruits stored under the two different temperature and stress inducer regimes, respectively. Similarly with PCA result, HCA result also confirmed all postharvest treatment was not effective to halt ripening-associated metabolite increase in the flesh part (Fig. 6, and Supplementary Information, S4). The flesh part is the consuming part in mangosteen and might affect the organoleptic properties such as fruit firmness and taste–aroma. Therefore, more detailed investigation on the metabolites associated with organoleptic properties were also performed.

In mangosteen flesh part, glucoronate and several amino acids that are reported to be aroma metabolites precursors (β-alanine, isoleucine, valine, alanine) seem to be inhibited in LT treatment (Fig. 7). The inhibition of amino acid biosynthesis was presumably caused by hypoxia condition that was more profound in flesh part after LT treatment. Previous research has indicated the decrease of oxygen concentration in flesh part compared to peel part as reported in apple and pear (Ho et al. 2006, 2010). Meanwhile, most of metabolites relative intensity related to sweet-acidic taste quality (glucose, sucrose, fructose, citric acid, malic acid, quinic acid, and lactic acid) did not show any significant differences between LT treatment and control during mangosteen storage.

Moreover, the relative intensity of important metabolites between SI and control were compared using a line chart in a time course manner between day 0, 2, and 10. Most of the important metabolites relative intensity were not affected by SI treatment (Supporting Information, Fig. S6). This finding further strengthen that stress inducer treatments did not show any effect to prolong mangosteen shelf-life based on metabolite abundance. Based on our results, the

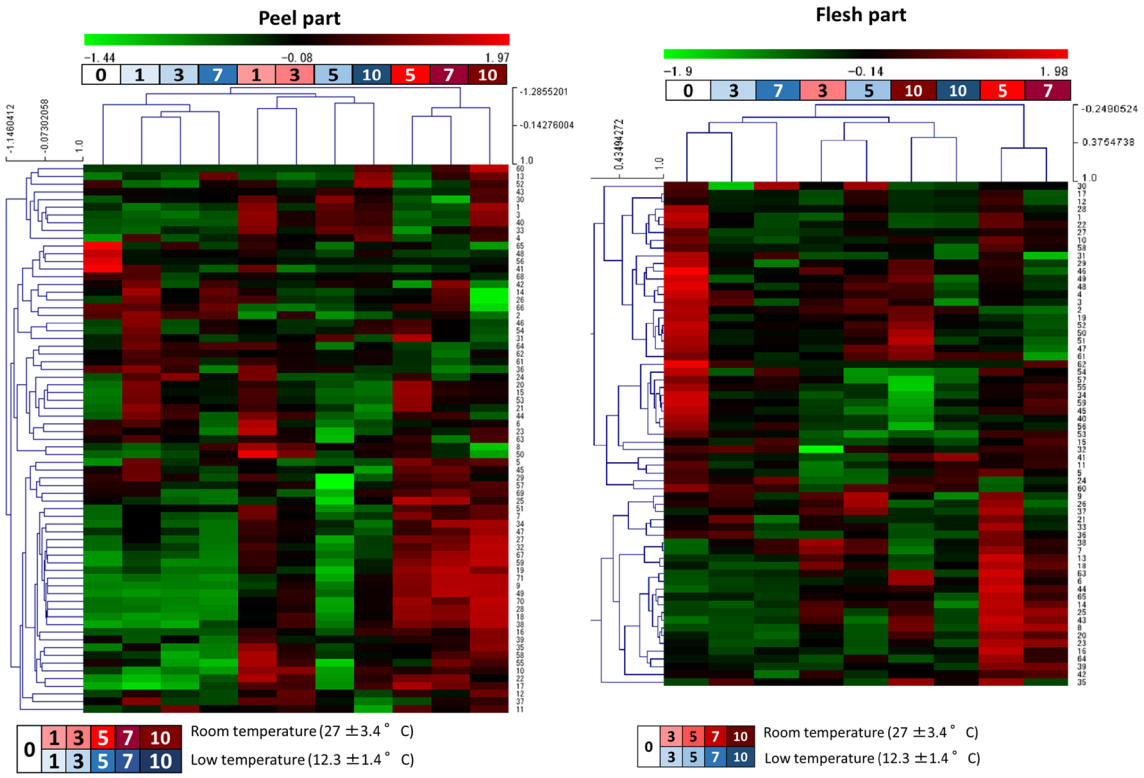


Fig. 6 Distribution of metabolites analyzed using GC–MS during mangosteen LT storage treatment in **a** peel part; **b** flesh part. Stored-mangosteen was observed for 10 days. Metabolite distribution during storage was employed and analyzed using HCA. Day 1 sample of flesh was removed from analysis because it is an outlier. HCA parameter *correlation: Pearson correlation, average linked cluster analysis. Red color represents room temperature treatment. Blue color represents low temperature treatment. The number shown in the figures represent the storage day of each treatment. Both treatments were analyzed together using GC–MS. The detailed information about metabolites name is described in the Supplementary Table S10 for peel part and Table S11 for flesh part

LT treatment showed the most effective method to prolong mangosteen shelf-life and further development of postharvest technology in mangosteen should be introduced in the future.

Although previous reports mentioned that a combination of several postharvest treatments (1-MCP, methyl jasmonate, and salicylic acid, edible coating) were effective to prolong mangosteen shelf life (de Castro et al. 2012; Mustafa et al. 2018; Piriyavinit et al. 2011), our results showed that when tested individually, only LT treatment showed a profound effect. Further studies to assess the effect of postharvest treatment to sensory attributes such as flavor, taste, and aroma should be investigated to confirm the consumer acceptance. In addition, application of other omics approaches may validate and strengthen the results obtained in this study.

Finally, the study of mangosteen postharvest technology using a metabolomics approach provides further information about the chemical mechanism of several physio-chemical results such as pericarp hardening during mangosteen storage treatment. Metabolomics approach offers a great advantage to discover metabolites and metabolic associations correlated with fruit quality traits such as color, texture, and flavor. The present study marked the first investigation using GC–MS based metabolomics approach to evaluate the effect of different harvesting and ripening conditions as well as postharvest technology in mangosteen. In addition, this is the first report of the use of PLS analysis to correlate metabolite intensity and color changes as a physical parameter for maturity indices of mangosteen in different ripening process conditions. The results from this study can provide a better understanding of mangosteen postharvest strategies and quality control by controlling changes in metabolites involved in ripening.

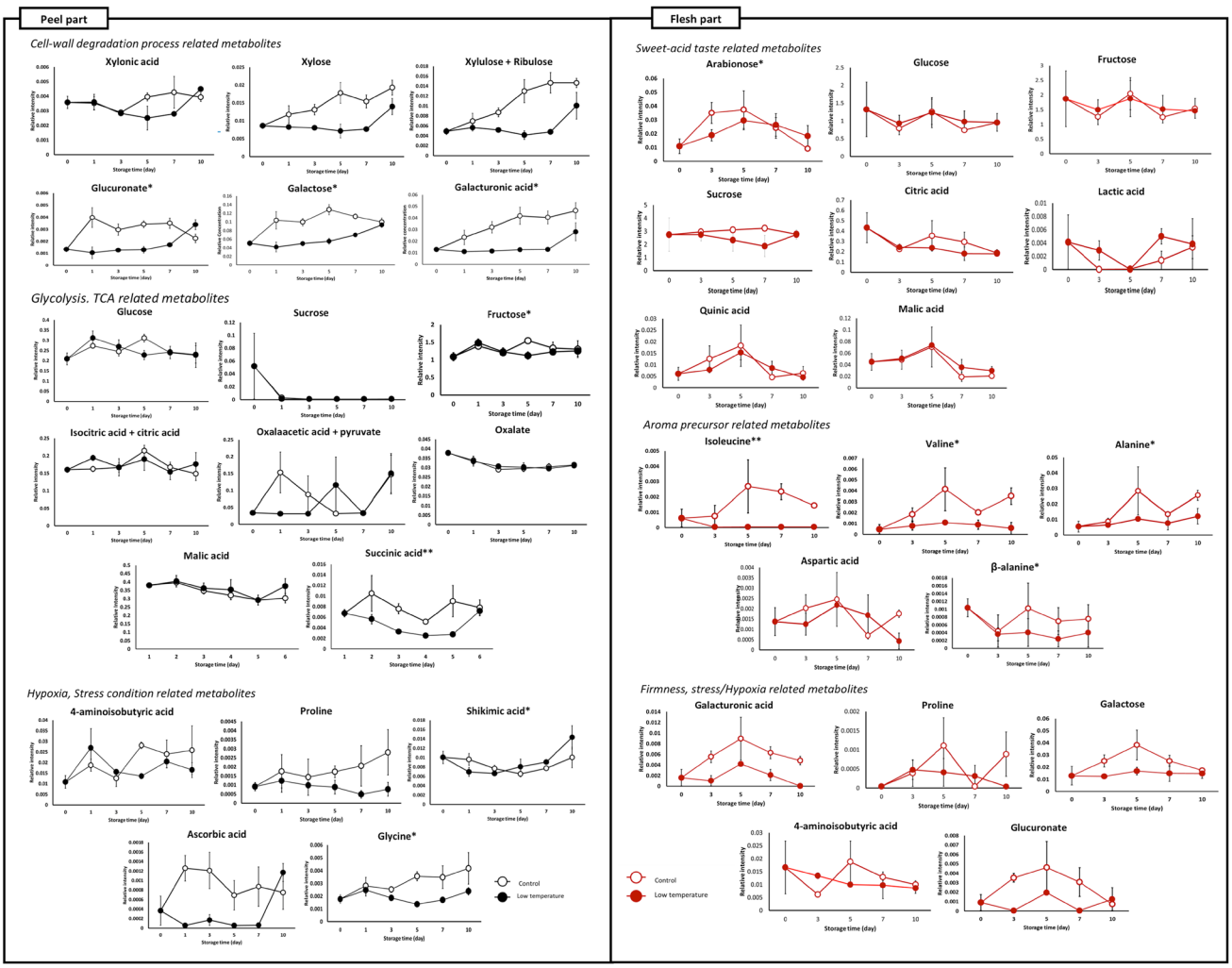


Fig. 7 Line graph of metabolite changes analysis using GC–MS during mangosteen LT treatment in **a** peel part; **b** flesh part. The relative intensity of metabolites that are important during mangosteen postharvest treatment at low temperature and room temperature were explained using a line chart. The metabolites were dominated by saccharides, organic acids, and amino acids. The peak intensity of each metabolite was normalized based on ribitol internal standard. Y axis represents metabolites relative intensity and X axis represents day after storage. White circle represents room temperature treatment and black circle represents low temperature treatment. Day 1 sample of flesh was removed from analysis because it is an outlier. The metabolites were marked with asterisk are listed as ripening-associated metabolites

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Authors contribution AARP designed the study, performed the metabolome analysis and data analysis, and drafted the manuscript. SPP participated in the design of the study, coordination, and writing of the manuscript. SR advised the selection of postharvest treatment and participated in the design of the study. FMD aided in data interpretation and participated in the design of the study. EF conceived of the study and participated in its design and coordination. All authors read and approved the final manuscript.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Research involving human and animal participants This article does not contain any studies with human participants or animal performed by any of the authors.

Informed consent Informed consent was obtained from all individual participants included in the study.

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