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## Original Article

# Effect of shaking process on correlations between catechins and volatiles in oolong tea



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### ABSTRACT

Shaking the tea leaves is the key manipulation to making oolong tea. It contributes to the formation of flavor and fragrance in oolong tea. The dynamic variations of catechins and volatile organic compounds (VOCs) during the shaking process were investigated. The results showed that the contents of epicatechin, epigallocatechin, epicatechin gallate (ECG), and epigallocatechin gallate (EGCG) first decreased after the shaking and then increased to the initial value before the next shaking. Geraniol, linalool and its oxides, and phenylethyl alcohol showed similar variations. The contents of trans- $\beta$ -ocimene, 1H-indole, and 3-hexenyl hexanoate increased after the second or third shaking (the late fermentation stage). However, the contents of aldehydes showed an opposite trend to other VOCs. The abundance of phenylethyl alcohol was positively related to the content of ECG and EGCG during fermentation, whereas the abundance of cis-3-hexenal was negatively related to the content of ECG. The correlations between catechin and VOCs indicated that shaking affected the chemical transformation of the compounds in oolong tea.

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## 1. Introduction

Oolong tea is Taiwan's world renowned semifermented tea (partial oxidation) with an elegant floral aroma and mellow characteristics. The manufacturing process of this semifermented tea includes withering, several rounds of shaking and setting (fermentation or oxidation), firing (fixation), rolling, and drying. Tea fermentation is the most important stage for quality control. An oxidation reaction occurs in this stage; the catechins in tea leaves are oxidized with time [1,2]. In this

process, catechin monomers are polymerized to form theaflavins, thearubigins, or other oxidation products such as theasinensins or oolongthein [1,3,4]. These tea chemical components contribute to the color and taste of oolong tea.

The first step of tea making is withering the fresh tea leaves. As the moisture content of tea flushes decreases, withering reduces the semipermeability of the membranes, thus enabling the catechins stored in the leaf cell vacuoles to flow out of the cytoplasm and come into contact with the oxidase in the cell cytoplasm [5]. From this stage onward, the

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oxidation of catechins starts and tea fermentation begins. In the manufacturing of oolong tea, the withering process includes two stages: solar and indoor withering. In solar withering, other than the loss of moisture, the UV radiation of the sunlight also promotes gene expression of intracellular hydrolytic enzymes,  $\beta$ -primeverosidase and  $\beta$ -glucosidase, and facilitates the hydrolysis of the precursors of volatile organic compounds (VOCs), which are present in the glycosidic form [6,7]. After a short period of solar withering, the tea leaves are moved indoors to continue the withering process. During indoor withering, the tea leaves are shaken three to five times at intervals. Each shaking interval is about 2 hours and is dependent on the temperature, moisture, and the contents of leaves. The tea master decides when to shake the tea leaves, what the intensity should be, and the duration of the shaking by touching and smelling the leaves. The initial shaking is mild, mainly to redistribute the moisture from the stalk to the leaves, and the stomatal conductance of the tea leaf is decreased after plucking and solar withering [7,8]. After a heavy shaking, the catechins are released from the vacuoles to the cytoplasm [5]. The expression of oxidase is also significantly enhanced, indicating significant oxidation [9]. Moreover, the smell of the fermenting tea leaves changes during the entire shaking and setting process. The precursors of the aromatic VOCs undergo oxidation and hydrolysis after the shaking. The hydrolysis of terpene alcohol glycosides generates terpene alcohols as the VOCs, while the oxidation of carotenoids and lipids produces lactones, ketones/enols, and other VOCs [10–12]. Thus, the smell of the fresh tea leaves changes in the manufacturing process. Consequently, the resulting VOCs composition contributes to the unique and characteristic aroma of oolong tea.

The tea master monitors the entire process through his/her senses and decides the timing of each step, which is still an art depending on the traditional master–apprentice model (on-the-job training). This means that good tea can only be made by hand, not by machinery. If we want to produce tea of high quality, we need more information and parameters about tea fermentation. To establish a scientific tea manufacturing process, the changes in the chemical contents of tea leaves during the fermentation process should be elucidated. The fermentation duration of oolong tea can be up to 6–10 hours. We monitored the changes in the chemical contents of tea leaves during the fermentation stage, before and after each shaking step, to understand the changes in catechin monomers and VOCs in the manufacturing process of the oolong tea. We can determine the whole picture of the fermentation process through the change of catechins, the raw materials of tea fermentation, and the VOCs—what tea masters depend on. According to these data, we provide a theory of tea fermentation for procedures to monitor the manufacturing process.

## 2. Methods

### 2.1. Oolong tea manufacturing process

Tea flushes (3 leaves with 1 bud) were plucked from the tea plantations in the Taoyuan County, Taiwan. Two cultivars of

tea (*Camellia sinensis* var. *sinensis*), 'Chin-Hsin-Dah-Pang' and 'Chin-Hsin-Gan-Tzu', were used to make oolong tea in 2011 and 2013. We used the standard oolong tea manufacturing process which was followed by an experienced operator. Samples were collected from fresh tea leaves (F), the leaves after solar withering (SW), and indoor-withering stage which is separated into before each shaking (BSn,  $n^{\text{th}} = 1, 2, 3, \text{ and } 4$ ) and 15 minutes after shaking during the setting period (ASn,  $n^{\text{th}} = 1, 2, 3, \text{ and } 4$ ). The weight of each fresh leaf sample was ~100 g. Three replicates sampled simultaneously were obtained. Twenty grams of samples were dried at 105°C in the oven until a constant weight to measure the water content by the weight loss. Remaining samples were immediately frozen at –20°C and stored until analysis.

### 2.2. High performance liquid chromatography analysis of tea catechins

Freeze-dried tea leaves (20 g) were ground into a powder of < 40 mesh; 0.5 g of the ground tea leaves were extracted with 50-mL boiled deionized water for 20 minutes in a water bath at 90°C. The tea extract was filtered through a 0.45- $\mu\text{m}$  Millipore filter before the analysis. A Jasco High Performance Liquid Chromatography System equipped with PU-2089, AS-2057, UV-2075, and LC-NetII/ADC (Shimadzu Co. Ltd., Kyoto, Japan) was used. A stainless steel Symmetry Waters column (4.6 mm internal diameter  $\times$  250 mm long; 5  $\mu\text{m}$  particle size; WAT 054275, Milford, MA, USA) was used and maintained at a constant temperature of 40  $\pm$  0.5°C. A flow rate of 1.0 mL/min was used during the separation; the injected volume was 8  $\mu\text{L}$ . The mobile phase consists of a combination of solvent A [deionized water with 0.1% (volume/volume) formic acid] and solvent B (acetonitrile). The elution profile for catechin separation was as follows: 0–5 minutes, 100% A; 15 minutes, 90% A, 10% B; 29 minutes, 80% A, 20% B; 35 minutes, 78% A, 22% B; and 40 minutes, 75% A, 23% B. Absorbance at 280 nm was used for the real-time monitoring of peak intensities.

### 2.3. Extraction of volatiles and gas chromatography mass spectrometry analysis

Fresh frozen leaves (5 g) were mixed with liquid nitrogen to homogenize and release the volatiles, and then 0.03 g of the tea fragments were sealed in an airtight vial (30 mL). The samples were heated to 50°C for equilibration. Headspace gas sampling was conducted by solid-phase microextraction (SPME) using 50/30- $\mu\text{m}$  divinylbenzene/carboxen/polydimethylsiloxane (Supelco, St. Louis, MO, USA). A handheld SPME fiber was exposed to a conditioning sample headspace for 15 minutes. The SPME fiber was then injected into the gas chromatograph in the splitless injection mode at 250°C. Gas chromatography mass spectrometry was carried out using a HP 5890 gas chromatograph (Agilent, Santa Clara, CA, USA) equipped with 5972 mass selective detector (Agilent, Santa Clara, CA, USA). A capillary column (DB-5 Hewlett-Packard – Agilent J&W GC Columns) 30 m long and 0.25 mm internal diameter with a 1- $\mu\text{m}$  film thickness was used. The GC-operating condition is as follows: (1) hold at 35°C for 1 minute and then increase to 80°C at a rate of 25°C/min; (2) increase by 3°C/min from 80°C to 120°C, then further increase to 200°C

at 20°C/min; and (3) finally hold at 200°C for 2 minutes with a carrier gas (99.99% helium) at a flow rate of 1.0 mL/min. The injector and detector temperatures were 250°C and 280°C, respectively. The quadrupole mass spectrometer was scanned over the 50–280 amu range at one scan/s with an ionizing voltage of 70 eV. The eluting compounds were identified by their retention time, mass spectra (library matching with NIST 05 and Wiley 2.75 database – Agilent ChemStation Rev. B.04.03GC) and related literature [12,13].

#### 2.4. Chemicals and reagents

Catechin standards were obtained from Sigma/Aldrich: (–)-epigallocatechin (EGC; ≥ 95%), (–)-gallocatechin (GC; ≥ 98%), (–)-catechin (C; ≥ 98%), (–)-epicatechin (EC; ≥ 98%), (–)-epigallocatechin gallate (EGCG; ≥ 95%), (–)-gallocatechin gallate (GCG; ≥ 98%), (–)-epicatechin gallate (ECG; ≥ 98%), and (–)-catechin gallate (CG; ≥ 98%). Caffeine (≥ 98%) and gallic acid (≥ 95%) were obtained from Merck (Darmstadt, Germany), along with liquid chromatography-grade solvents used in the high performance liquid chromatography system, including formic acid and acetonitrile.

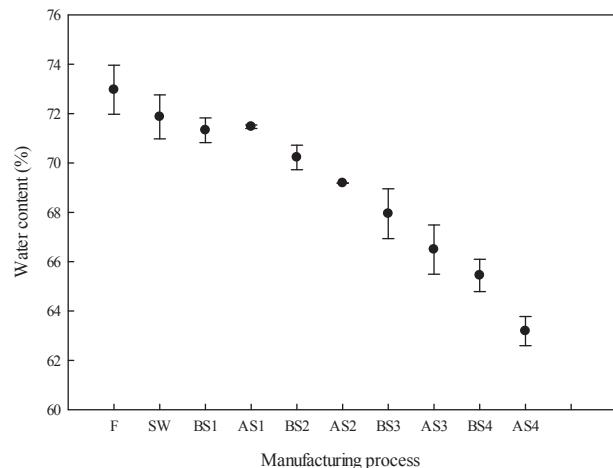
#### 2.5. Statistical analysis

The percentage ratio of variance at each manufacturing stage to the final process of fermentation (AS4) in of EC, EGC, EGCG, and ECG at every stage during the oolong tea manufacturing process was calculated. The variance shows the difference between each stage and the AS4 and basis on the content of the AS4. The relative abundance of VOCs to the AS4 as the basis represents the variation during fermentation. The correlation between the contents of catechins and the abundance of VOCs was analyzed and illustrated using SAS (SAS Institute Inc., Cary, NC, USA) and SigmaPlot 11 (Systat Software, Inc., San Jose, CA, USA) using Pearson correlation coefficient and linear correlation.

### 3. Results and discussion

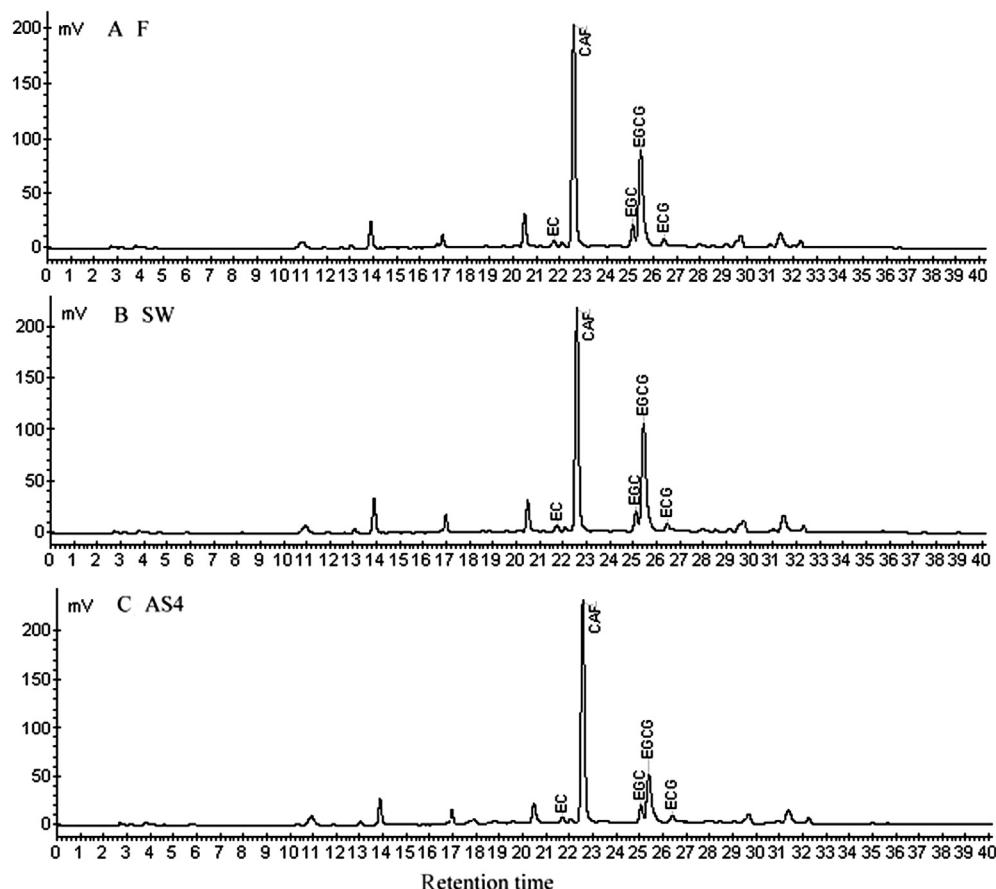
#### 3.1. Dynamic variations in catechin contents during oolong tea fermentation

Oolong tea was made of 'Chin-Hsin-Dah-Pan' in 2011 and 2013 and 'Chin-Hsin-Gan-Tzu' in 2013. Shaking was performed four times in every oolong tea manufacturing process, except 'Chin-Hsin-Dah-Pan' which was used in 2013. The water content during the whole manufacturing decreased from 72.97% to 63.19% at the end of the fermentation process (Figure 1). The water content decreased slowly until BS2, and after BS2 the constant decline at the sequential processing. The redistribution from the stalk to the leaf and the evaporation caused continuous water loss during the tea manufacturing process [8]. The withering made catechin oxidation possible. Gallic acid, caffeine, and eight catechins, including GC, C, EC, EGCG, EGC, GCG, ECG, and CG, were analyzed. Among all the chemical compounds, the contents of EC, ECG, EGC, and EGCG show more obvious variations (Figure 2). These four catechins were detected as major



**Figure 1** – The variation of water content (%) during the oolong tea manufacturing process using 'Chin-Hsin-Gan-Tzu' in 2013 and 'Chin-Hsin-Dah-Pan' in 2011. Bar means  $\pm$  standard error. ASn = after  $n^{\text{th}}$  shaking for 15 minutes (setting period); BSn = before  $n^{\text{th}}$  shaking; F = fresh tea leaves = SW: solar withering.

components in oolong tea and the oxidation substrates of the fermented tea leaves mainly comprise of these four catechins [14,15]. During the fermentation of oolong tea, sampling was conducted before the shaking and after the setting of the leaves for 15 minutes. The results of different tea cultivars and leaves harvested in different years showed that the contents of EC, ECG, EGC, and EGCG were reduced after each shaking (Figures 3 and 4). The tea master ends the tea fermentation process depending on the flavor of the leaves after the last shaking. Because the shaking times of 'Chin-Hsin-Gan-Tzu' in 2013 and 'Chin-Hsin-Dah-Pan' in 2011 were both four times, we take the each catechin content in AS4 of each process as the base line to validate the variation ratio between each manufacturing process. The variation ratio of content in EGC with smaller standard error shows consistent alternation in the former fermentation process (SW, AS1, and AS2), but variation in the ECG with relative small standard error shows in the later process (BS3, AS3; Table 1). The different patterns of these two decreasing EGC and ECG contents suggest that each catechin might result from the different oxidative activities. Theasinensins, which are composed of EGC and EGCG, are a group of valued oxidative compounds during oolong tea manufacturing under the light fermentation [1]. However, the theaflavins, which are the oxidative dimers from the combination of EC, ECG, EGCG, and EGC, were usually found in the oolong tea after heavy fermentation or black tea [14]. The contents of EC (Figure 3A), ECG (Figure 3B), EGC (Figure 4A), and EGCG (Figure 4B) varied in fluctuation under our tea making monitoring. The contents of four catechins, EC, ECG, EGC, and EGCG, were decreased after each shaking (Figures 3 and 4). In previous research, the increase of polyphenol oxidase activity was detected after each shaking [8]. The oxidation of catechins is supposed to be activated with the high polyphenol oxidase activity after the shaking process. Before

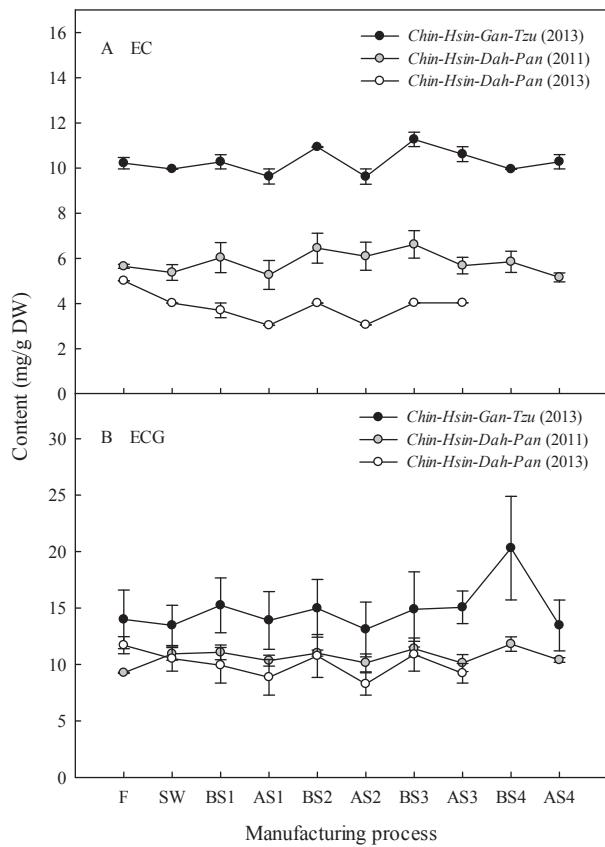


**Figure 2 – High performance liquid chromatography profiles (0–40 minutes) of oolong tea manufacturing process at: (A) fresh leaves (F); (B) after solar withering (SW); and (C) after the fourth shaking process at 280 nm. Caffeine (CAF) and the four major catechins, (-)-epicatechin (EC), (-)-epigallocatechin (EGC), (-)-epigallocatechin gallate (EGCG), and (-)-epicatechin gallate (ECG) are indicated.**

each shaking, the contents of these catechins did not change significantly (Figures 3 and 4). The variation indicates that during the setting period, which was ~1.5–2 hours, the levels of catechins returned to a relatively high level. The content under the dry weight basis seemed to fluctuate within a range. It means that the shaking process may release the free catechins for the next oxidation reaction. However, the water content decreased with time so the concentrate of catechins could be condensed to a high level. The condensation might cause a trigger to further reactions for more intermediates or oxidative products. The change of EGCG during oxidation is most clearly among these four catechins. The dynamic oxidized variations of different cultivars were not affected by the initiation concentration of EGCG (Figure 4B). The mode of oxidative variations during the shaking process was independent to the original concentration in the fresh tea leaves. The cyclic oxidation in EGCG was illustrated by a cyclic voltammetry model to the formation of theaflavins and thearubigins [16]. The similar periodic variations of EGCG and other catechins we investigated was the first monitoring data to show the changes during oxidation in living tea leaves. The opposite

up-and-down variation was also found in polyphenol oxidase activity [8]. The fluctuating variations in the catechin oxidation during the oolong tea process and polyphenol oxidase activity displayed that the shaking manipulation should be a critical process of tea oxidation, and it was the key to the tea quality control.

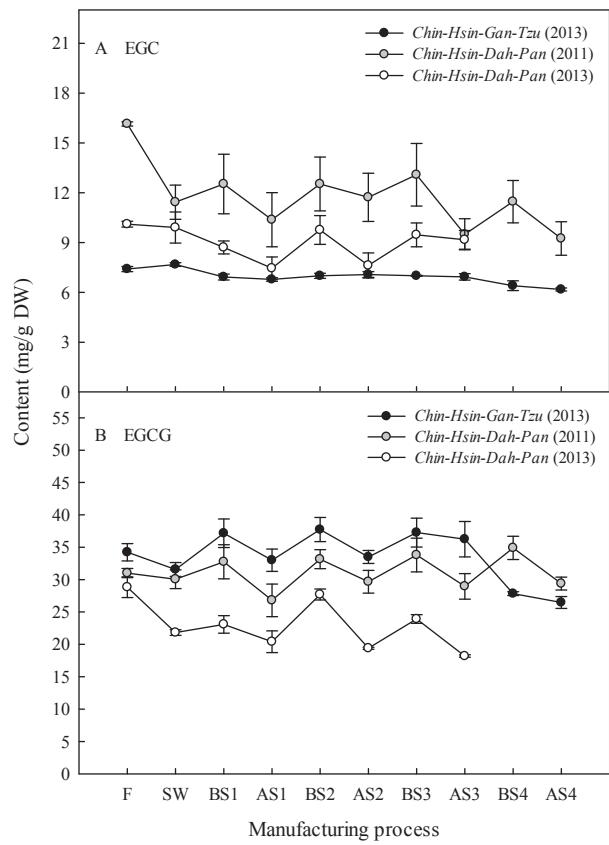
The correlation coefficients between all catechins are shown in Table 2. The correlation coefficient means the level of synchronous between either of the two compounds. Among all these catechins, we focus on EGCG, EGC, and EC, because of the correlation coefficients of EGCG and EGC, EC and EGC, and EGCG and EC during the fermentation were 0.91, 0.77, and 0.73, respectively (Table 2). EC with EGC were recognized as the source of the oxidation of theaflavins, and EGCG with EGC were oxidized to other intermediates, theasinensins, or oolongtheinins [1,17]. A small amount of other oxidation products, catechin dimers, were also obtained from the reaction of GC with EGC [14]. The formation of different oxidation products can be attributed to the structural isomerization of the catechin monomers. Therefore, changes in the contents of the catechin monomers can be attributed to the sum of the oxidized catechins (polymer) and freed catechins (monomer).



**Figure 3 – Dynamic variations of: (A) (–)-epicatechin (EC); and (B) (–)-epicatechin gallate (ECG) during oolong tea fermentation. Data are the average of three batches of manufacturing process by using ‘Chin-Hsin-Gan-Tzu’ in 2013, ‘Chin-Hsin-Dah-Pan’ in 2011 and 2013, respectively. Bar means  $\pm$  standard error. ASn = after  $n^{\text{th}}$  shaking for 15 minutes (setting period); BSn = before  $n^{\text{th}}$  shaking; DW = dry weight; F = fresh tea leaves; SW = solar withering.**

### 3.2. Changes in VOCs during oolong tea fermentation

In the manufacturing process of Chin-Hsin-Dah-Pan, the dynamic changes in the abundance of VOCs were monitored in the manufacturing process. Although each VOC changed independently before and after the shaking, three types of changing patterns were observed. The first pattern was observed as a high-to-low fluctuation after each shaking process in some major monoterpenoids, such as geraniol, linalool and its oxides, and phenylethyl alcohol that were responsible for the floral note during fermentation. Those VOCs were higher before each shaking and decreased after the setting period (Figure 5A). Although floral VOCs did not always increase in the next shaking stage, the relative abundance of VOCs obtained after the final round of shaking (AS4) was more than that in the fresh leaves. This shows that the decision of each shaking time by tea masters was dependent on the smell of tea. The shaking should be taken when it reached a balance and the balance was broken by



**Figure 4 – Dynamic variations of: (A) (–)-epigallocatechin (EGC); and (B) (–)-epigallocatechin gallate (EGCG) during oolong tea fermentation. Data are the average of three batches of manufacturing process using ‘Chin-Hsin-Gan-Tzu’ in 2013 and ‘Chin-Hsin-Dah-Pan’ in 2011 and 2013, respectively. Bar means  $\pm$  standard error. ASn = after  $n^{\text{th}}$  shaking for 15 minutes (setting period); BSn = before  $n^{\text{th}}$  shaking; DW = dry weight; F = fresh tea leaves; SW = solar withering.**

each shaking. However, shaking also increased the intense of floral flavor. The second pattern was shown by trans- $\beta$ -ocimene, 1H-indole, and 3-hexenyl hexanoate (Figure 5B). The relative levels of these VOCs clearly increased after the second or third shaking; however, the shaking did not significantly cause the fluctuation (Figure 5B). Aldehydes with a grassy odor showed the third changing pattern during the fermentation of oolong tea. The relative abundance of mainly cis-3-hexenal and trans-2-hexenal varied during the first and second shaking in the early fermentation stage, while that of 2,4-hexadienal varied after the third shaking process (Figure 5C). The grassy odor increased after the shaking. This change is opposite to that shown by the other VOCs. By these data, we find out the flavor compounds in the floral flavor and the grassy odor that were monitored by tea masters. The shaking not only increased the floral flavor but also decreased the grassy odor. Each shaking broke the balance between the target flavor and aversion odor and then brought a new better balance after shaking.

**Table 1 – The variance of (–)-epicatechin (EC), (–)-epicatechin gallate (ECG), (–)-epigallocatechin (EGC), and (–)-epigallocatechin gallate (EGCG) between each manufacturing stage and the final process (AS4) during oolong tea manufacturing.**

%	EC	ECG	EGC	EGCG
F	4.45 <sup>a</sup> ± 5.06	–3.48 ± 7.44	47.24 ± 27.31	17.40 ± 11.90
SW	0.53 ± 3.67	2.59 ± 2.59	23.99 ± 0.39	10.78 ± 8.50
BS1	8.47 ± 8.49	9.86 ± 3.34	23.78 ± 11.65	25.91 ± 14.46
AS1	–2.12 ± 4.19	1.36 ± 1.94	10.99 ± 1.18	7.93 ± 16.74
BS2	15.71 ± 9.36	8.55 ± 2.67	24.38 ± 11.07	27.67 ± 14.85
AS2	5.90 ± 12.27	–2.55 ± 0.09	20.70 ± 6.08	13.77 ± 12.80
BS3	18.95 ± 9.33	10.09 ± 0.47	27.40 ± 14.04	27.89 ± 12.89
AS3	6.73 ± 3.44	4.64 ± 7.24	7.48 ± 4.77	17.76 ± 19.21
BS4	5.10 ± 8.29	32.20 ± 18.63	13.81 ± 10.15	11.95 ± 6.79
AS4	0.00	0.00	0.00	0.00

ASn = after  $n^{\text{th}}$  shaking for 15 minutes (setting period); BSn = before  $n^{\text{th}}$  shaking; F = fresh tea leaves; SW = solar withering.

<sup>a</sup> Percentage ratio of variance was calculated by the difference between each stage and the AS4 stage (final processing of fermentation) and basis on the content of the AS4. The data showed as means ± standard error.

**Table 2 – Correlation coefficient (R) of variations between catechins during oolong tea fermentation.<sup>a</sup>**

	EC	ECG	EGC	EGCG	C	CG	GC
EC	—						
ECG	0.61** <sup>b</sup>	—					
EGC	0.77**	0.40**	—				
EGCG	0.73**	0.91**	0.62**	—			
C	0.46**	0.38**	0.34*	0.41**	—		
CG	0.28*	0.66**	0.07 ns	0.58**	0.39**	—	
GC	0.65**	0.70**	0.32*	0.49**	0.29*	0.11 ns	—
GCG	0.71**	0.65**	0.55**	0.76**	0.28*	0.47**	0.40**

C = (–)-catechin; CG = (–)-catechin gallate; EC = (–)-epicatechin; ECG = (–)-epicatechin gallate; EGC = (–)-epigallocatechin; EGCG = (–)-epigallocatechin gallate; GC = (–)-gallocatechin; GCG = (–)-gallocatechin gallate; ns = no significance.

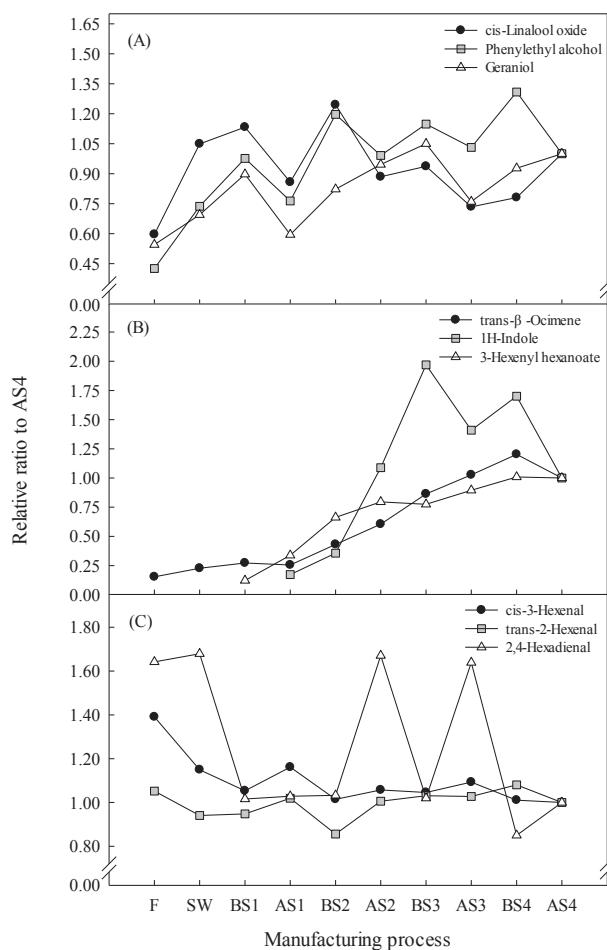
<sup>a</sup> Data are based on the manufacturing oolong tea using 'Chin-Hsin-Dah-Pan' in 2011,  $n = 66$ .

<sup>b</sup> \* $p < 0.05$ ; \*\* $p < 0.01$ ; ns = no significance.

### 3.3. Correlations between catechins and VOCs

The main purpose of the first and second shaking processes occurred during the early stage of fermentation is to remove the moisture from the fresh tea leaves [8]. At this time, the content of EC showed a more clear variation than the other catechin isomers (Figure 3A). The oxidation of EC with polyphenol oxidase preferentially oxidizes EC to EC-quinone. EC-quinone reacted with EGC to afford theaflavin [18]. For VOCs, the abundance of cis-3-hexenal, trans-2-hexenal, and cis-linalool oxides with a grassy or greenish odor also significantly changed during the initial fermentation stage (Figure 5A; 5C). The small molecules were the aldehydes, and the content of linalool correlated to the bioconversion of aldehyde and alcohol [19]. The chief purpose of the third and fourth shaking process was to provide a pure aroma to the oolong tea. At this time, the difference in the ECG and EGC contents before and after the shaking was more obvious than during the early stage of fermentation (Figure 3B; 4A), while the relative abundance of trans- $\beta$ -ocimene, 1H-indole, and 3-hexenyl hexanoate also significantly increased at the late stage of fermentation (Figure 5B). These chemical compounds that varied in the late fermentation stage were the essential oxidation substrates and components of oolong tea [20].

Furthermore, changes in the contents of EGCG, geraniol, and phenylethyl alcohol maintained a constant pattern during the entire fermentation process, showing a trend of higher contents before the shaking and lower contents after the shaking process (Figure 4B; 5A). Many studies have revealed the presence of catechins and VOCs during the manufacturing process; however, the relationship between them has not been established [7,20–22]. In the present work, the ECG content had a negative linear relationship with the abundance of cis-3-hexenal (Figure 6A) during the fermentation process of semifermented tea. By contrast, the ECG (Figure 6B) and EGCG (Figure 6C) contents showed a positive linear relationship with the abundance of phenylethyl alcohol; the R values were 0.89 and 0.87, respectively. Such correlations showed that the VOCs and catechin contents had a similar varying pattern. The changes in the catechin contents before and after the shaking process decreased with the grassy odor and increased with the floral aroma. This result indicates that the shaking not only affected the changes in the oxidation of catechins but also the formation of the smell. These two changes indeed had a connection. Although the direct evidence might be insufficient to the whole picture about how they connected to each other, we show scientific evidence about why the smell change could be the principle for a tea master monitoring to

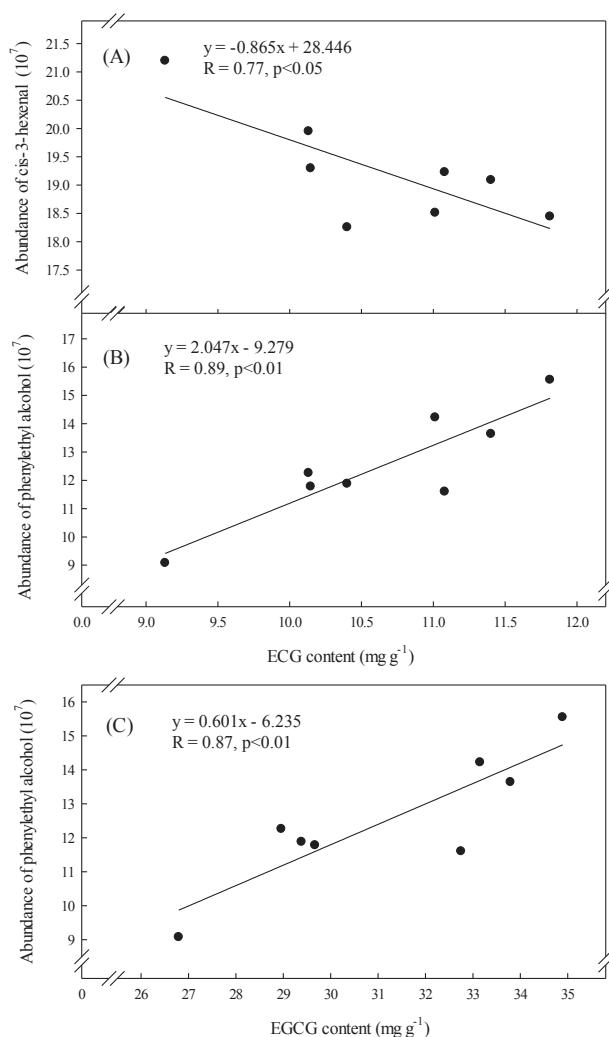


**Figure 5 – Variations of volatile organic compounds during oolong tea fermentation using ‘Chin-Hsin-Dah-Pan’ in 2011. (A) cis-Linalool oxide, phenylethyl alcohol, and geraniol; (B) trans- $\beta$ -ocimene, 1H-indole, and 3-hexenyl hexanoate; (C) cis-3-Hexenal, trans-2-hexenal, and 2,4-hexadienal. Data was shown as the relative abundance to the final stage (AS4) of the process. ASn = after  $n^{\text{th}}$  shaking for 15 minutes (setting period); BSn = before  $n^{\text{th}}$  shaking; F = fresh tea leaves; SW = solar withering.**

decide the shaking timing and to keep a consistent quality of oolong tea. The shaking timing is unique for decreasing what we are undesired and increasing what we prefer in good oolong tea quality.

#### 4. Conclusion

This study reveal that in the oolong tea manufacturing process, the concentration of the main oxidation materials (EGCG, ECG, EGC, and EC) of catechins showed fluctuations because of the shaking and setting processes. These fluctuations may be crucial for removing undesired flavors and for obtaining the characteristic aroma of oolong tea.



**Figure 6 – Relationship between catechins and volatile organic compounds including (A) the (–)-epicatechin gallate (ECG) contents and the abundance of cis-3-hexenal; (B) the ECG contents and the abundance of phenylethyl alcohol; and (C) the (–)-epigallocatechin gallate (EGCG) contents and the abundance of phenylethyl alcohol, respectively. Data were the average of three batches manufacturing of ‘Chin-Hsin-Dah-Pan’ in 2011. DW = dry weight.**

Our results also show a correlation between the variation patterns shown by the catechins and VOCs. Therefore, the correlations between EGCG, ECG, EGC, EC and phenylethyl alcohol or cis-3-hexenal indicate that the shaking affected the chemical transformation of the compounds in tea and helped in imparting the characteristic features to oolong tea. The tea master can properly adjust the time for the shaking and setting treatments on the basis of the changes in the aroma. Thus, our research shows the dynamic changing patterns of catechins and VOCs and provides a scientific and credible basis for tea processing. This information may assist to improve tea production technology.

## Conflicts of interest

The authors have nothing to disclose.

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