ORIGINAL PAPER

Effects of heating on the total phenolic content, antioxidant activities and main functional components of simulated Chinese herb candy during boiling process

Qianqian Yin1,2 · Haibo Mu3 · Maomao Zeng1 · Daming Gao1 · Fang Qin1 · Jie Chen1,2 · Zhiyong He1,2

Received: 7 July 2018 / Accepted: 24 October 2018 / Published online: 29 October 2018 © Springer Science+Business Media, LLC, part of Springer Nature 2018

Abstract

The effects of heating (100–140 °C/10–180 min) on functional component contents, total phenolic content (TPC) and antioxidant activities of six simulated Chinese herb candies containing honeysuckle, chrysanthemum, dandelion, liquorice, mulberry leaf and tangerine, respectively were studied, which revealed that TPC and antioxidant activity of candies significantly increased ($p < 0.05$) with increasing heating temperature (> 130 °C) and time (> 40 min) during boiling process. After heating at 140 °C/180 min, TPC increased 2–3 times compared with that of unheated samples, while 2,2′-azino-bis (3-ethylbenzthiazoline-6-sulfonic acid) (ABTS) radical and 2-Diphenyl-1-picrylhydrazyl (DPPH) radical scavenging activity, ferric reducing antioxidant power (FRAP) increased 2–9, 2–5 and 3–6 times, respectively. Contents of the main functional components including rutin, glycyrrhizic and chlorogenic acids and naringin in herb candies decreased 6–85% after heating, while glycyrrhetinic acid content increased by 60% and 157% upon heating at 130 °C and 140 °C/180 min, respectively. Significant enhancements in TPC and antioxidant activities of candies during boiling process might be attributable to Maillard reaction product formation. These results would be helpful to effectively guide the industrial production of Chinese herb candies, and to maintain their health benefits.

Keywords Chinese herb candy · Antioxidant activity · Heating · Functional component · Total phenolic content · Boiling process

Introduction

In recent years, people have become more and more interested in Chinese traditional herbs for their good therapeutic performance and low toxicity in preventing or treating diseases. Epidemiological studies have shown that Chinese traditional herbs possess anti-inflammatory, antiatherosclerotic,

 \boxtimes Jie Chen chenjie@jiangnan.edu.cn \boxtimes Zhiyong He

zyhe@jiangnan.edu.cn

- State Key Laboratory of Food Science and Technology, Jiangnan University, Wuxi, Jiangsu, China
- ² International Joint Laboratory on Food Safety, Jiangnan University, Wuxi, Jiangsu, China
- ³ State Key Laboratory of Dairy Biotechnology, Shanghai Engineering Research Center of Dairy Biotechnology, Dairy Research Institute, Bright Dairy & Food Co., Ltd., Shanghai, China

antitumor, antimutagenic, anticarcinogenic, antibacterial and antiviral activities $[1-4]$ $[1-4]$; these effects could be partly attributed to their antioxidant and free-radical-scavenging activities [[5–](#page-9-2)[9\]](#page-9-3). Nowadays, as environmental pollution is becoming an increasingly serious issue, many people are suffering from respiratory diseases. Therefore, Chinese herbs that have the ability to achieve heat-clearing or treat sore throats such as honeysuckle, chrysanthemum and liquorice have attracted substantial attention. Honeysuckle, chrysanthemum, mulberry leaves and dandelion contain a variety of chemicals such as organic acids, volatile oils, flavonoids, triterpenoid saponins and trace elements, while having the main active ingredient polyphenol $[10, 11]$ $[10, 11]$ $[10, 11]$ $[10, 11]$ $[10, 11]$. According to a study by Jiang et al. [\[12\]](#page-9-6), chlorogenic acid, rutin and quercetin are the main active ingredients contained in honeysuckle, while the quality of honeysuckle has mainly been evaluated by focusing on chlorogenic acid, which is reported to have the ability to inhibit the peroxidation of linoleic acid [[13](#page-9-7)]. The main functional components contained in chrysanthemum are flavonoids, which were reported to possess antimicrobial

activity $[11]$ $[11]$. According to research by He et al. $[14]$ $[14]$, mulberry leaves or leaf-derived extracts exhibit significant hypoglycaemic, hypolipidaemic and antiatherogenic effects on humans and certain animal models, and rutin is their main functional component. Dandelion has the ability to exert antioxidation, anti-inflammatory and analgesic effects, and flavonoids are its main active components, especially quercetin [[15\]](#page-9-9). Liquorice and tangerine are usually used to treat throat diseases, and liquorice is reported to have many pharmacological activities such as antitumor, antimicrobial, antiviral, anti-inflammatory, immunoregulatory and several other activities that contribute to the recovery and protection of the nervous, alimentary, respiratory, endocrine and cardiovascular systems [[16\]](#page-9-10). The main active components in liquorice are flavonoids and triterpenoid saponins.

Since there is substantial overlap between the categories of traditional Chinese herbs and food resources, many traditional Chinese herbs have been used as flavourings, pigments and directly as food [[17,](#page-9-11) [18\]](#page-9-12). For example, liquorice is widely used in the food industry as a sweetener [\[19](#page-9-13)], while honeysuckle and chrysanthemum are often used to produce drinks [[20](#page-9-14)]. Some Chinese herbs are also used in the food industry as antioxidants to prolong the shelf life [[21](#page-9-15)]. In recent years, an increasing number of herbs or herb extracts have also been used to produce candy that can ease diseases of the throat [\[22](#page-9-16)].

In this context, many functional ingredients in herbs such as polyphenols and flavonoids are thermally sensitive, and the amount of these active components that remains during the boiling process of candy at high temperatures such as up to 140 °C is very important for the health benefits of herb candy products. However, to the best of our knowledge, no research has been performed on the stability of the functional components of herb candies during boiling process.

Therefore, the main objective of this study was to investigate the effect of heat treatment at different temperatures and times on the total phenolic content (TPC), the main functional component content, and the antioxidant activity of herb candies during simulated boiling process with the aim of providing some guidance for widening the application of Chinese herbs in the candy industry.

Materials and methods

Material

Chinese herbal medicines (honeysuckle, chrysanthemum, dandelion, mulberry leaves, liquorice and tangerine) were obtained from Beijing Tong-Ren-Tang drug retail outlet in Wuxi. Standards (chlorogenic acid, rutin, glycyrrhizic acid, glycyrrhetinic acid, naringin) were purchased from J&K Scientific Ltd. (Beijing, China). 2,2-Diphenyl-1-picrylhydrazyl radical (DPPH), 6-hydroxy-2,5,7,8-tetramethylchromane-2-carboxylic acid (Trolox), 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) (ABTS), 2,4,6-tris(2-pyridyl) s-triazine (TPTZ) and gallic acid were obtained from Sigma-Aldrich Inc. (St. Louis, MO, USA). All of the solvents used for HPLC were of HPLC grade and the other reagents used were of analytical grade.

Extraction of antioxidant compounds

The dried Chinese herbs were crushed using a commercial grinder (Purun, Quzhou, China) and then passed through a 60-mesh sieve. Each dried herbal powder (25 g) was extracted with 80% (v/v) ethanol aqueous solution using an ultrasonicator (KUDOS, Shanghai, China) at room temperature for 30 min, with a solid-to-solvent ratio of 1:10. The suspension was filtered through filter paper in a Buchner funnel and the residues were re-extracted twice under the same conditions. The filtrates were pooled and concentrated to 5 ml using a rotary evaporator (EYELA, Shanghai, China) under a vacuum at 45 °C. The extracts were stored at 4 °C for subsequent use.

Simulated boiling process of the candy

The candy was produced as follows: 55 g of granulated sugar and 45 g of glucose syrup were dissolved in 25 ml of water by heating at 70 °C, followed by the addition of 5 ml of herbal extracts and stirring to ensure good mixing. The sugar solution was poured into a pressure-resistant bottle and heated in an oil bath (Jincheng, Jiangsu, China). The temperature was set at 100 °C, 110 °C, 120 °C, 130 °C or 140 °C and the heating time was 10, 20, 30, 40, 60, 120 or 180 min. The heated sugar solutions were cooled down soon and stored at 4 °C for the subsequent experiments.

TPC determination

The values of TPC of sugar solutions containing six herbal extracts subjected to different heating processes were determined using the Folin–Ciocalteu reagent assay, as described by He et al. [[23\]](#page-9-17), with some modifications. The heated sugar syrups (viscous semisolid) were dissolved in water with the ratio of 1:4 (mg/ml) to obtain a dilute sugar solution. Folin–Ciocalteau reagent (1 ml) was mixed well with sugar solution (1 ml) using a vortex machine; after 5 min, 3 ml of a sodium carbonate solution (75%) and 5 ml of distilled water were added to the mixture. The reaction mixture was incubated for 120 min at 25 °C in the dark. The absorbance was then noted at 760 nm using a UV-5300PC spectrophotometer (Metash instruments Co., Ltd., Shanghai, China). The standard calibration curve was prepared using gallic acid; the amount of TPC in each heated sugar solution is expressed as milligrams of gallic acid equivalents (GAE) per gram of candy sample (mg GAE/g).

DPPH free‑radical‑scavenging capacity analysis

The free-radical-scavenging activity of six simulated herb candies against DPPH was analysed in accordance with a method previously reported by Wang et al. [[24\]](#page-9-18) with some modifications. The heated sugar syrups were dissolved in water with the ratio of 1:80 (g/ml) to obtain a dilute sugar solution. A total of 1.5 ml of sugar solution was mixed with 1.5 ml of freshly prepared DPPH solution (0.1 mM); then, the solution was incubated at room temperature for 30 min in the dark. The absorbance at 517 nm was determined using a UV-5300PC spectrophotometer (Metash, Shanghai, China); as a blank, pure ethanol was used instead of the sample. The proportion of DPPH free radicals scavenged for each sample was calculated using the following equation:

DPPH% = $(A_0 - A_1)/A_0$

where A_0 is the absorbance of a DPPH solution without a sample and A_1 is the absorbance of a DPPH solution with a sample.

ABTS free‑radical‑scavenging activity analysis

The free-radical-scavenging activity of herb candies against ABTS was measured following the method described by He et al. [\[23\]](#page-9-17) with some modifications. The heated sugar syrups were dissolved in water with the ratio of 1:80 (g/ml) to obtain a dilute sugar solution. A total of 0.1 ml of sugar solution was mixed with 3.9 ml of ABTS solution and incubated in the dark at room temperature for 10 min, after which the absorbance was read at 734 nm using a UV-5300PC spectrophotometer (Metash, Shanghai, China). The standard calibration curve was prepared using Trolox standard solutions of known concentrations (100, 200, 300, 400, 500, 600 and 800 mM), and the free-radical-scavenging capacity against ABTS is expressed as micromoles of Trolox equivalents (TE) per gram of sample (mM TE/g).

Ferric reducing antioxidant power (FRAP) determination

The FRAP assay was performed in accordance with the method described by He et al. [[25](#page-9-19)] with modifications. The heated sugar syrups were dissolved in water with the ratio of 1:80 (g/ml) to obtain a dilute sugar solution. A total of 0.2 ml of sugar solution and 0.6 ml of distilled water were mixed with 6 ml of FRAP solution, and incubated at 35 °C for 10 min in the dark. The absorbance of the mixture at 593 nm was then determined using a UV-5300PC spectrophotometer (Metash, Shanghai, China). The standard calibration curve

was prepared in accordance with the method of determining the free-radical-scavenging activity against ABTS.

HPLC analysis

The analysis of functional compounds was carried out according to the method described by Chen et al. [\[26](#page-9-20)] with slight modifications. Before HPLC analysis, a total of 1 g of sugar solution containing herbal extracts was dissolved in 5 ml of distilled water, then 1 ml of sugar solution (200 mg/ ml) was mixed with 4 ml of absolute ethanol, and incubated at room temperature for 12 h to remove protein, sugar and other components. Then, the mixture was filtered using a 0.45-µm membrane filter.

A CBM-20A HPLC system (Shimadzu, Japan) was used for the analysis of functional compounds. The separation was achieved using an X-Bridge C18 column (250×4.6 mm, 5 µm; Waters Corporation). For the analysis of functional compounds, elution was performed using mobile phase A (0.1% phosphoric acid) and mobile phase B (acetone), and samples (10 µl) were eluted at a flow rate of 1.0 ml/min. The gradient conditions were as follows: 10–25% solvent B (0–5 min), 25–35% solvent B (5–15 min), 35–39% solvent B (15–25 min), 39–60% solvent B (25–35 min), 60–80% solvent B (35–40 min), 80–90% solvent B (40–50 min), 90–100% solvent B (50–60 min), 100–10% solvent B (60–61 min) and 10–10% solvent B (61–65 min). The identification of functional compounds was based on matching their retention times with those of pure standards. The quantitation of functional compounds was based on the linear regression equations of each standard and expressed as microgram or milligram of standards per gram of sample $(\mu g/g \text{ or } mg/g).$

Statistical analysis

Candy samples were analysed in duplicate, and the measurements were performed in triplicate, except for the HPLC analysis. The data are expressed as means \pm standard deviations of triplicate measurements ($n=2\times3$). One-way analysis of variance was conducted to evaluate the significance of differences using Statistix 9 software, and $p < 0.05$ was considered statistically significant.

Results and discussion

TPC of six herb candies during boiling process

The levels of TPC of six herb candies from different heating processes are shown in Fig. [1.](#page-3-0) The findings suggest that the effect of heating on the TPC of different herb candies varied among the species. As shown in Fig. [1](#page-3-0)a, the TPC **Fig. 1** The total phenolic content in six simulated Chinese herb candies from different heating processes. **a** Honeysuckle, **b** chrysanthemum, **c** mulberry leaves, **d** dandelion, **e** licorice, **f** tangerine

of honeysuckle candy was almost unaffected upon heating at 100 °C and 110 °C; however, it increased from 2.40 mg GAE/g for an unheated sample to 3.94 and 5.16 mg GAE/g after heating at 130 °C and 140 °C for 180 min, respectively. Similarly to that of honeysuckle, the TPC of chrysanthemum candy changed little upon heating at 100 °C. However, the increase was significant when the temperature was higher; upon heating at 120–140 °C for 180 min, the TPC increased 2.4–3.4 times. As for candy samples containing mulberry leaves, upon heating at 100 °C, 110 °C and 120 °C for less than 120 min, TPC changed non-significantly, while it increased 1.4–2.3 times and 2.1–2.9 times after heating at 130 °C and 140 °C for over 60 min, respectively. As shown in Fig. [1d](#page-3-0), the TPC in dandelion candy changed non-significantly after heating at 100–140 °C for less than 40 min, whereas it increased 1–3 times when the heating time was 60–180 min. The TPC of liquorice candy did not change significantly upon heating at 100 °C for 10–180 min, as well as at 110 °C, 120 °C, 130 °C and 140 °C for 10–40 min, whereas it was almost three times higher than the unheated level upon heating at 140 °C for 180 min, as shown in Fig. [1e](#page-3-0). As for tangerine candy, the change of TPC was not significant, but it peaked upon heating at 140 °C for 180 min.

Some authors reported that conventional heating such as boiling, heat and moisture treatment caused the decline in TPC of samples due to the heat degradation of phenolic

compounds [[27,](#page-9-21) [28\]](#page-9-22). On the other hand, in agreement with our present findings, some other studies also revealed that thermal processing, such as roasting and microwave heating, can increase the TPC of nuts [[29\]](#page-9-23), citrus peel [\[30\]](#page-9-24) and grape seeds [\[31](#page-9-25)]. They suggested that the higher TPC value after heat treatment might have been due to the release of bound phenolic compounds and the formation of intermediate Maillard reaction products (MRP). Many antioxidant phenolic compounds in plants are generally present in covalently bound forms. Chinese herbs contain a large number of phenolic components in free form as well as in bound form [\[32\]](#page-9-26). However, in our present study, the ethanol extracts of herbs but not Chinese herbs were applied in the simulated heat processing of herb candies, thus the phenolic compounds contained in the herb extracts were mainly in free form. Therefore, the increase of TPC of herb candies after heat treatment might only be due to the formation of intermediate MRP. During the heating process, reactions between amine groups of amino acids or proteins and carbonyl groups of sugars or carbohydrates can take place. These reactions produce a large number of intermediate MRP, including volatile compounds, polyphenols, peptide polymers and brown complex pigments (melanoidins). Some of the MRP with a reductone-type structure or phenol-like complexes can interfere with the determination of TPC by Folin–Ciocalteu assay, leading to higher absorbance in this assay and the overestimation of the phenolic content [[33](#page-9-27)]. Given that Chinese herbal extracts contain proteins and are heated in sugar solution, the formation of MRP is expected.

Antioxidant activities

Antioxidant activities are influenced by many factors, and one single method cannot describe this fully; therefore, it is necessary to perform more than one type of antioxidant capacity measurement to take into account the various mechanisms of antioxidant action [[34\]](#page-9-28). In the present study, three different methods were applied to describe the antioxidant activities of herbs and their trends of change.

Free‑radical‑scavenging capacity against DPPH

The free-radical-scavenging activity against DPPH of candy samples containing herb extracts from different heating processes is shown in Fig. [2.](#page-5-0) The results indicate that DPPH scavenging activity of herb candies was increased by heating; however, for most of the samples, it did not increase significantly upon heating for less than 40 min. As for candies containing honeysuckle and mulberry leaves, the changes of DPPH scavenging activity were not significant upon heating at a lower temperature, as shown in Fig. [2a](#page-5-0), c; however, heating at a higher temperature increased the DPPH scavenging activity 1–2 times. As for chrysanthemum sample, the DPPH

scavenging activity was increased 1–3 times by heat treatment; it increased significantly upon heating for more than 40 min, and reached its maximum upon heating for 180 min, as shown in Fig. [2](#page-5-0)b. The DPPH scavenging activity of dandelion sample under different heating conditions is shown in Fig. [2](#page-5-0)d. Heating at 100 °C only resulted in slightly higher levels of DPPH scavenging activity compared with that of an unheated sample. However, when the temperature was higher, it increased significantly after heating for 40 min. For liquorice candy, upon heating at 100 °C, 110 °C, 120 °C and 130 °C, the DPPH scavenging activity was almost stable for heating periods shorter than 60 min and then increased significantly with increasing heating time. However, upon heating at 140 °C for 30 min, the DPPH scavenging activity increased. Dandelion candy had the smallest increase in DPPH scavenging activity, whereas liquorice sample had the largest; heat treatment increased the DPPH scavenging activity of dandelion and liquorice candy by 1–1.6 times and 2–5 times, respectively. The DPPH scavenging activity of tangerine sample is shown in Fig. [2f](#page-5-0); it was increased 1–3 times by heating. Its changes at 100 °C, 110 °C, 120 °C and 130 °C were almost the same, whereas upon heating at 140 °C, the change of DPPH scavenging activity between 40 and 60 min was the most significant, with an increase from 30.43 to 50.84%.

Free‑radical‑scavenging capacity against ABTS

As shown in Fig. [3,](#page-6-0) ABTS scavenging activity was also increased by heating. It can be concluded that the ABTS scavenging activity of the six herb candies did not change significantly upon heating at a lower temperature or for a shorter time. As for honeysuckle sample, upon heating at 100 °C, 110 °C and 120 °C, the change was not significant; however, upon heating at 140 °C, the ABTS scavenging activity increased significantly for heating periods longer than 60 min, and the change of the free-radical-scavenging activity in chrysanthemum sample was almost the same as that in honeysuckle sample, as shown in Fig. [3](#page-6-0)a, b. After heating, the ABTS scavenging activities of honeysuckle and chrysanthemum candy increased 1–3 times. Upon heating at 100 °C, 110 °C and 120 °C, the ABTS scavenging activity of candy containing mulberry leaves was almost stable, it only increased 1.1–1.4 times, upon heating at 130 °C for 180 min and 140 °C for 120 and 180 min, the ABTS scavenging in heated samples was almost 5–7 times higher than that in unheated samples, respectively. As for dandelion sample, heating at 100 °C and 110 °C only resulted in a slight increase in ABTS scavenging activity, but significant increases occurred upon heating at 120 °C, 130 °C and 140 °C after 120 min, as shown in Fig. [3d](#page-6-0). Dandelion sample had the most significant increase in ABTS scavenging activity, it increased 2–9 times after heating. Figure [3](#page-6-0)e shows

Fig. 2 DPPH free radical scavenging activities of six herb candies from different heating processes. **a** Honeysuckle, **b** chrysanthemum, **c** mulberry leaves, **d** dandelion, **e** licorice, **f** tangerine

that the change of ABTS scavenging in liquorice candy was similar to that in honeysuckle sample, heating increased the ABTS scavenging ability of liquorice sample 1–5 times. As for tangerine candy, it was associated with the smallest increases, namely heating only led to slight increases in ABTS scavenging activity of 1.1–1.5 times.

Ferric reducing antioxidant power (FRAP)

The FRAP of six herb candies from different heating processes is shown in Fig. [4](#page-7-0). This figure reveals that heating can also increase their FRAP. The same as ABTS scavenging activity, upon heating at a lower temperature or for a shorter time, the FRAP in herb candies only changed slightly. For honeysuckle candy, the smallest increase in FRAP occurred upon heating at 130 °C and 140 °C, after 120 min, FRAP increased significantly, and the change of FRAP in chrysanthemum candy sample was similar to that in honeysuckle. Heat treatment increased the FRAP of honeysuckle and chrysanthemum candies 1.2–2.5 times and 1.3–2.6 times, respectively. As shown in Fig. [4](#page-7-0)c, upon heating at 100 °C, 110 °C and 120 °C, the change of FRAP in candy containing mulberry leaves was not significant, however, it increased significantly upon heating at 130 °C and 140 °C after 60 min, and reached its maximum at 140 °C for 180 min. After heating, the FRAP of mulberry leaves samples increased 1–5 times. Similar findings were also made for dandelion sample, the FRAP in dandelion **Fig. 3** ABTS scavenging activities of six herb candies from different heating process. **a** Honeysuckle, **b** chrysanthemum, **c** mulberry leaves, **d** dandelion, **e** licorice, **f** tangerine

candy also increased significantly upon heating at 120 °C for 180 min, as shown in Fig. [4d](#page-7-0). The FRAP of dandelion sample increased 1–6 times after heating, which was the most significant. The FRAP of liquorice candy is shown in Fig. [4](#page-7-0)e, upon heating at 100 °C and 110 °C, the change of FRAP was slight, but upon heating at 130 °C and 140 °C, it increased significantly after 60 min. As for tangerine sample, upon heating at 140 °C, the FRAP increased significantly after 40 min, and heating increased the FRAP of liquorice and tangerine samples 1.2–3.1 times and 1.3–2.7 times, respectively. The changes of FRAP in mulberry leaves and dandelion samples were the most significant, and these FRAP levels upon heating at 140 °C for 180 min were almost 5 and 6 times higher than that in unheated samples, respectively.

In the present study, heating led to higher DPPH and ABTS scavenging activities and higher FRAP compared to those of the unheated control. In some previous studies, the conclusion that different thermal treatments can significantly increase antioxidant activities was also drawn. According to a study by Zou et al. [\[35\]](#page-9-29), ethanol and acetone extracts of roasted wheat germ showed higher scavenging activities than those of raw wheat germ. In addition, in a study by Jeong et al. [\[36](#page-10-0)], roasting increased the radical-scavenging activity of sesame meal extracts. Moreover, Yamaguchi et al. [[37\]](#page-10-1) indicated that cooking can increase the radicalscavenging activity of many vegetables as polyphenols and ascorbate oxidases are inactivated by the high temperature. Similar to the TPC variation above described, the increasing antioxidant activity after heat treatment could be due to the **Fig. 4** The FRAP of six herb candies from different heating process. **a** Honeysuckle, **b** chrysanthemum, **c** mulberry leaves, **d** dandelion, **e** licorice, **f** tangerine

formation of MRPs during heat processing. It was reported that some of MRP also exhibit high antioxidant activity [\[38](#page-10-2)].

Main functional compounds

According to a study by Jiang et al. [[12](#page-9-6)], chlorogenic acid and rutin were chosen as the main functional components of honeysuckle. Similarly, chlorogenic acid and rutin, chlorogenic acid, rutin, glycyrrhizic and glycyrrhetinic acids and naringin were chosen as the main active components of chrysanthemum, dandelion, mulberry leaves, liquorice and tangerine, respectively [\[14](#page-9-8), [39](#page-10-3)[–42](#page-10-4)].

The contents of functional compounds in six Chinese herb candies are shown in Fig. [5.](#page-8-0) It can be concluded that heat treatment can decrease the contents of functional compounds, except for glycyrrhetinic acid in liquorice candy. Upon heating for 180 min at different temperatures, the levels of functional components in herb candies were reduced by different levels, ranging from 8 to 80%. As shown in Fig. [5a](#page-8-0), b, the content of chlorogenic acid decreased 10.8–67.2% upon heating for more than 40 min, and as for rutin in honeysuckle candy, the content did not decrease significantly upon heating at 100 °C, 110 °C, 120 °C and 130 °C for 30 min, when heated at 140 °C, it decreased significantly with increasing heating time (up to 81.6% for 140 °C/180 min). The content of chlorogenic acid in chrysanthemum candy decreased steadily with an increased duration of heating, and the content of rutin decreased significantly upon heating for more than 40 min, and as heating at 140 °C/180 min they decreased 81.8% and

Fig. 5 The content of functional component in six herb candies from different heating processes. **a** Chlorogenic acid in honeysuckle candy, **b** rutin in honeysuckle candy, **c** chlorogenic acid in chrysanthemum candy, **d** rutin in chrysanthemum candy, **e** rutin in mulberry leaves

candy, **f** chlorogenic acid in dandelion candy, **g** glycyrrhizic acid in licorice candy, **h** glycyrrhetinic acid in licorice candy, **i** naringin in tangerine candy

79.3%, respectively. Figure [5](#page-8-0)e shows the levels of rutin in mulberry leaves samples, when the heating temperature was lower than 140 °C, the content of rutin did not decrease significantly before 30 min, but it decreased 64.1–82.3% with increasing heating time upon heating at 140 °C. The content of chlorogenic acid in dandelion candy decreased about 3.4–84.1% after heating, as shown in Fig. [5f](#page-8-0). The levels of functional components in liquorice candy are shown in Fig. [5](#page-8-0)g, h. The content of glycyrrhizic acid did not decrease significantly upon heating at 100 °C, 110 °C and 120 \degree C, however, upon heating at 130 \degree C for longer than 120 min and heating at 140 °C, the content of glycyrrhizic acid decreased about 29.3–63.7%. As for glycyrrhetinic acid, its content was almost stable when the heating temperature was lower than 130 °C, however, upon heating at 130 °C and 140 °C, it increased significantly for heating periods longer than 60 min, when heating at 130 °C and 140 °C for 180 min, glycyrrhetinic acid content increased by 60% and 157%, respectively, which may have been due to the transformation of glycyrrhizic acid, as glycyrrhizic acid can be converted into glycyrrhetinic acid in a high temperature and acidic environment [\[43](#page-10-5)].

According to Zou et al. [[35](#page-9-29)], the content of individual phenolic components in roasted wheat germ decreased with increasing roasting time, except for ferullic acid. The content of ferullic acid increased upon roasting for 10 min and then decreased significantly when the roasting time was increased to 20 min. The same phenomena were also revealed by other studies. Şahin et al. [[33\]](#page-9-27) studied the effect of different roasting processes on carob powder and indicated that the levels of phenolic compounds decreased after 75 min of roasting at temperatures of 135 °C, 150 °C and 165 °C. However, in some previous studies, heat treatment led to an enhancement of the content of individual functional components in green tea [\[44\]](#page-10-6) and citrus peel [\[45](#page-10-7)]. In the present study, the content of individual functional components in all herb candies was decreased with increasing heating time, except for glycyrrhetinic acid. This difference may have been due to the different materials and processing conditions.

Conclusion

In summary, the effects of heating on the TPC and antioxidant activities of six simulated Chinese herb candies during boiling process were studied for the first time. Heat treatment had different effects on TPC, antioxidant activities, and content of major individual functional components of herb candy. In general, thermal processing could increase the TPC of six herb candies, especially those containing chrysanthemum and liquorice, whereas the TPC of tangerine candy exhibited the smallest increase. The increases of TPC are due to the formation of MRP, thus enhancing their antioxidant activities during processing of herb candy. However, heat treatment significantly decreased the content of individual functional components because of their thermal decomposition. Among these functional components, chlorogenic acid and rutin are more heat-sensitive than the others; in particular, chlorogenic acid content in dandelion candy was reduced by 83% after heating at 140 °C for 180 min, while glycyrrhetinic acid content in liquorice candy increased significantly due to the decomposition of glycyrrhizic acid into glycyrrhetinic acid. Further study is required to identify the MRP formed during the boiling process and how these compounds can influence the antioxidant activities of herb candies.

Funding This research was supported financially by the National Natural Science Foundation of China (No. 31771978), the National First-class Discipline Program of Food Science and Technology (No. JUFSTR20180201) and the Innovation and Exploration Fund of State Key Laboratory of Food Science and Technology, Jiangnan University (No. SKLF-ZZB-201801).

Compliance with ethical standards

Conflict of interest The authors report no conflicts of interest.

References

1. S.Q. Liu, Z.B. Tan, P.T. Li, X.L. Gao, Y. Zeng, S.L. Wang, J. Pharmaceut. Biomed. **121**, 56–62 (2016)

- 2. C.K. Firempong, H.Y. Zhang, J.J. Zhang, Y. Wang, X. Cao, E. Omari-Siaw, S.S. Tong, J.N. Yu, X.M. Xu, Eur. J. Integr. Med. **7**, 365–371 (2015)
- 3. C.C. Han, J.Y. Guo, Inflammation **35**, 913–919 (2012)
- 4. H.F. Yang, X.L. Chen, C.M. Jiang, K.W. He, Y.Y. Hu, J. Vet. Res. **61**, 405–410 (2017)
- 5. G.R. Schinella, H.A. Tournier, J.M. Prieto, dB.P. Mordujovich, J.L. Ríos, Life Sci. **70**, 1023–1033 (2002)
- 6. P. Chumark, P. Khunawat, Y. Sanvarinda, S. Phomchirasilp, N.P. Morales, L. Phivthong-Ngam, P. Ratanachamnong, S. Srisawat, K.U.S. Pongrapeeporn, J. Ethnopharmacol. **116**, 439–446 (2008)
- 7. D.J. Moura, M.F. Richter, J.M. Boeira, J.A.P. Henriques, J. Saffi, Mutagenesis **22**, 293–302 (2007)
- 8. N.L.M. Nasir, N.E. Kamsani, N. Mohtarrudin, F. Othman, S.F.M. Tohid, Z.A. Zakaria, Pharm. Biol. **55**, 2102–2109 (2017)
- 9. Y.X. Li, Y.B. Liu, A.Q. Ma, Y. Bao, M. Wang, Z.L. Sun, Food Sci. Biotechnol. **26**, 1675–1683 (2017)
- 10. X. Chen, L. Hu, X. Su, L. Kong, M. Ye, H. Zou, J. Pharmaceut. Biomed. **40**, 559–570 (2006)
- 11. S. Zhu, Y. Yang, H. Yu, Y. Ying, G. Zou, J. Ethnopharmacol. **96**, 151–158 (2005)
- 12. L.F. Jiang, G.M. Zhou, Y.Y. Li, J. Liq. Chromatogr. Relat. Technol. **34**, 1473–1487 (2011)
- 13. M. Ohnishi, H. Morishita, H. Iwahashi, S. Toda, Y. Shirataki, M. Kimura, K. Ryo, Phytochemistry **36**, 579–583 (1994)
- 14. J. He, Y. Feng, H.Z. Ouyang, B. Yu, Y.X. Chang, G.X. Pan, G.Y. Dong, T. Wang, X.M. Gao, J. Pharmaceut. Biomed. **84**, 189–195 (2013)
- 15. K. Schutz, R.A. Carle, J. Ethnopharmacol. **107**, 313–323 (2006)
- 16. R. Yang, L.Q. Wang, B.C. Yuan, Y. Liu, Planta Med. **81**, 1654– 1669 (2015)
- 17. J.X. Wang, X.H. Xiao, G.K. Li, J. Chromatogr. A **1198**, 45–53 (2008)
- 18. J. Zhao, Y. Deng, S.P. Li, TRAC Trend. Anal. Chem. **96**, 138–150 (2017)
- 19. D. Komes, A. Belscak-Cvitanovic, S. Juric, A. Busic, A. Vojvodic, K. Durgo, Int. J. Food Sci. Nutr. **67**, 53–66 (2016)
- 20. Z. Wang, M.N. Clifford, P. Sharp, Food Chem. **108**, 369–373 (2008)
- 21. H.X. Luo, S.H. Lin, F.Z. Ren, L.P. Wu, L.S. Chen, Y. Sun, J. Food Protect. **70**, 1440–1445 (2007)
- 22. J. Yu, Z. Xuan, Y. Ruan, H. Zhang, K. Shi, Y. Guo, China J. Chin. Mater. Med. **40**, 351–355 (2015)
- 23. Z.Y. He, Y.D. Tao, M.M. Zeng, S. Zhang, G.J. Tao, F. Qin, J. Chen, Food Chem. **200**, 107–116 (2016)
- 24. T. Wang, N. Guo, S.X. Wang, P. Kou, C.L. Zhao, Y.J. Fu, Food Bioprod. Process. **108**, 69–80 (2018)
- 25. Z.Y. He, B. Yuan, M.M. Zeng, G.J. Tao, J. Chen, Food Chem. **175**, 457–464 (2015)
- 26. H.X. Chen, W.J. Liu, Med. Plant 5, 57–58, 64 (2014)
- 27. G. Rocchetti, L. Lucini, G. Chiodelli, G. Giuberti, D. Montesano, F. Masoero, M. Trevisan, Food Res. Int. **100**, 69–77 (2017)
- 28. T. Beta, T. Hwang, Food Chem. **246**, 58–64 (2018)
- 29. M.M. Win, A. Abdul-Hamid, B.S. Baharin, F. Anwar, N. Saari, Eur. Food Res. Technol. **233**, 599–608 (2011)
- 30. S.M. Jeong, S.Y. Kim, D.R. Kim, S.C. Jo, K.C. Nam, D.U. Ahn, C.L. Seung, J. Agric. Food Chem. **52**, 3389–3393 (2004)
- 31. S.Y. Kim, S.M. Jeong, W.P. Park, K.C. Nam, D.U. Ahn, S.C. Lee, Food Chem. **97**, 472–479 (2006)
- 32. H. Peleg, M. Naim, R.L. Rouseff, U. Zehavi, J. Sci. Food Agric. **57**, 417–426 (1991)
- 33. H. Şahin, A. Topuz, M. Pischetsrieder, F. Özdemir, Eur. Food Res. Technol. **230**, 155–161 (2009)
- 34. S.P. Wong, L.P. Leong, K. Jhw, Food Chem. **99**, 775–783 (2006)
- 35. Y. Zou, M. Yang, G. Zhang, H. He, T. Yang, J. Am. Oil Chem. Soc. **92**, 1302–1312 (2015)
- 36. S.M. Jeong, S.Y. Kim, D.R. Kim, K.C. Nam, D.U. Ahn, S.C. Lee, J Food Sci. 69, C377–C381 (2004)
- 37. T. Yamaguchi, M. Katsuda, Y. Oda, J. Terao, K. Kanazawa, S. Oshima, T. Inakuma, Y. Ishiguro, H. Takamura, T. Matoba, Food Sci. Technol. Int. **9**, 79–83 (2003)
- 38. K. Brudzynski, D. Miotto, Food Chem. **127**, 1023–1030 (2011)
- 39. O. Kenny, T.J. Smyth, C.M. Hewage, N.P. Brunton, Int. J. Food Sci. Technol. **50**, 766–773 (2015)
- 40. D. Liu, L. Wei, D. Zhu, M. Geng, W. Zhou, T. Yang, J. Plant Nutr. Soil Sci. **173**, 268–274 (2010)
- 41. E. Luengo, I. Álvarez, J. Raso, Innov. Food Sci. Emerg. Technol. **17**, 79–84 (2013)
- 42. R.M. Wang, C. Lin, J.L. Liu, F. Yu, J.P. Gao, X.J. Pan, Chin. J Chem. Eng. **20**, 152–157 (2012)
- 43. D.H. Kim, S.W. Lee, M.J. Han, Biol. Pharm. Bull. **22**, 320–322 (1999)
- 44. L.F. Wang, D.M. Kim, C.Y. Lee, J. Agric. Food Chem. **48**, 4227– 4232 (2000)
- 45. G. Xu, X. Ye, J. Chen, D. Liu, J. Agric. Food Chem. **55**, 330–335 (2007)