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Short communication

Identification of 9,10-anthraquinone contamination during black and green tea processing in Indonesia

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ABSTRACT

Black and green tea are popular owing to their unique flavors and health benefits. However, these teas can be contaminated with anthraquinones, which are associated with health risks in humans. Levels of 9,10-anthraquinone were determined at each step of the manufacturing process for green and black tea using gas chromatography–tandem mass spectrometry. For green tea, anthraquinone was present beginning at the withering process and increased in concentration until the first drying step. Additionally, for black tea, the first drying step resulted in a significant increase in anthraquinone content, whereas the concentration decreased during the final drying step. For black tea, anthraquinone was also detected during withering, although the concentration continued to increase throughout the entire procedure, particularly during drying. Therefore, it can be suggested that the presence of anthraquinone in these teas was influenced by the smoke emitted by the wood fires used to drive the processing machinery.

1. Introduction

Tea from *Camellia sinensis* is a popular beverage worldwide owing to its attractive flavor and the presence of functional compounds that confer health benefits (Carloni et al., 2013). The consumption of tea is associated with improved human health owing to the presence of these bioactive compounds, some of which are antioxidants that have been shown to have anticancer, anti-cardiovascular disease, and anti-bacterial effects (Zhang, Qi, & Mine, 2019). There are two type of tea available commercially. Black tea is made from young leaves with enzymatic oxidation while green tea is made in absence of enzyme oxidation (Jiang et al., 2019).

Recently, the trace contaminant of 9,10-anthraquinone (AQ) has recently been found in teas (Pitoi, Ariyani, Koesmawati, & Yusiasih, 2019) and this chemical may have some negative health effects on consumers. West Sumatra, Indonesia produces both green tea and black tea for local and international markets. For exportation to many developed countries, the tea must fulfill a requirement recently established by the European Union, limiting the AQ content to 0.02 mg/kg. This regulation has negatively affected the export of tea produced in Indonesia. Owing to the potential dangers of this contaminant, it is necessary to reduce the concentration of AQ in tea, not only for exportation but also to ensure safety.

AQ can be determined using QuEChERS method combined with gas chromatography tandem with mass spectrometry (GCMS) (Kitano et al., 2018). QuEChERS, a solvent (acetonitrile) extraction method which can use a step of dispersive solid phase extraction (d-SPE) for clean-up of the extracts prior to GCMS analysis has been found to be effective for AQ determination in teas (Ghani & Ahmad, 2016). Moreover, in MS analysis, the utilization of multiplex reaction monitoring (MRM) in the detection of trace amounts of metabolites has also developed to reduce the detection limit of measurement that would provide reliable results (Syukri et al., 2018).

The occurrence of AQ contamination in green and black tea source remains unclear. Previous study has suggested that AQ originates from the environment in plantations or from the fertilizers used during plant growth (Wang et al., 2018). In addition, Mo, Zhang, Zhu, and Li (2008) have suggested that AQs have been shown to be produced by microorganism activity during the fermentation of Fuzhuan brick tea. Moreover, processing procedures, including machinery used in manufacturing, might also be suggested of affecting the generation of AQ. The processing of green tea consists of five steps: plucking; withering (inactivation of the enzyme); cutting, tearing, and curling; and two drying steps (Donlao & Ogawa, 2019). The processing of black tea is similar; however, withering does not inactivate the enzyme, and the cutting, tearing, and curling step is followed by wet grading, enzyme oxidation, and one drying step (Wu et al., 2019). In this study, it hypothesized that AQ content may be affected by processing steps during

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the production of green and black teas. Therefore, the changes of AQ contents on black and green teas grown and processed in West Sumatra, Indonesia at different processing steps were evaluated.

2. Materials and methods

2.1. Plant material

One bud and three tender leaves tea (*Camellia sinensis cv Assamica*) were collected and processed into green tea and black tea on the Mitra Kerinci tea plantation in West Sumatra, Indonesia in June 2019.

2.2. Reagents

AQ standard was obtained from Supleco®, Sigma–Aldrich, Germany. The stock solution was prepared in toluene and working solutions were prepared in high performance liquid chromatography (HPLC) grade acetonitrile that purchased from Smart lab, Indonesia. Acetic acid, acetone and HPLC grade water were also purchased from Smart lab, Indonesia. Anhydrous Magnesium sulphate (MgSO4), Sodium Acetate (CH_3COONa) and Sodium chloride (NaCl) were purchased from Merck, Germany.

2.3. Green tea processing

Leaves were picked and withered on a rotary panner (RP) (RP-TEHA Bandung, Indonesia) at 90–100 °C for 5 min until the moisture content was reduced to 60–70%. Leaves were then rolled in an open top roller (OTR) (OTR-TEHA, Bandung, Indonesia) for 15 min and dried in an endless chain pressure (ECP) dryer (ECP-TEHA, Bandung, Indonesia) at 110–135 °C for 25 min until the moisture content was 30%. A second drying process used a ball tea (BT) machine (BT-TEHA, Bandung, Indonesia) set at approximately 80 °C, reducing the moisture content to 5%. This process is illustrated in Fig. S1. The output samples from RP, OTR, and ECP were then dried to 5% moisture content in an oven at 80 °C before analysis. Withering with RP deactivated the enzymes, reduced the water content, and made the leaves limp, thus facilitating the rolling process. The OTR reduced the tea leaves to smaller sizes by curling them and removed intracellular fluids, including pectin. The first drying occurred in the ECP, which concentrated the remaining intracellular fluids and reduced the water content to 30%. The BT dried the tea and curled the leaf fragments further (Anggraini, 2018).

2.4. Black tea processing

For black tea, the shoots were picked, withered in a withering trough (WT) (WT-TEHA, Bandung, Indonesia) for 15 h, and rolled using the OTR. Then, the material was wet sorted with a double Indian ballbreaker net sorter (DIBN-TEHA, Bandung, Indonesia), in which wet fermentation occurred, and the size of the leaf fragments was made uniform to allow for even enzymatic oxidation. This occurred over 90–110 min and oxidized catechin to theaflavin and thearubigin. Finally, the leaves were dried to a moisture content of 5% in a two-stage dryer (TSD) (TSD-TEHA, Bandung, Indonesia). This processing is illustrated in Fig. S2. The outputs of WT, OTR, wet sorted, and enzyme oxidation processes were dried to 5% moisture content in an oven at 80 °C before analysis. Withering of black tea was performed at a lower temperature (ambient temperature) than withering of green tea because there was no destruction of enzymes, and then enzyme had to be kept active to metabolize the catechins and derivatives. For black tea, withering only concentrated the intracellular fluids and made the leaves limp to facilitate further processing. The function of the OTR was the same for both black and green teas.

The TSD used for black tea and the RP and ECP used in processing green tea in this study all used wood as fuel. The BT used electric elements. To gauge the heat stability of AQ in the processed teas, samples of the green tea from BT and the black tea from TSD were heated for 30 min at 80 °C, 90 °C, or 100 °C in an oven.

2.5. Determination of AQ content in dried tea

A QuEChERS approach was used for sample preparation. The extraction of AQ was performed according to the methods described by Kitano et al. (2018) and European Commission DG-SANTE (2017). One gram of tea from each stage of processing was weighed in a 50-mL polypropylene centrifuge tube (Eppendorf, Madrid, Spain). Then, 10 mL HPLC-grade water was added, and the mixture was vortexed vigorously. To create a calibration curve, a standard working solution of AQ in acetonitrile was added to the hydrated dried tea sample at concentrations of 10, 20, 50, 100, and 200 μg/kg. The mixture was left standing for 30 min to allow AQ to interact with the tea before extraction. Acetonitrile (10 mL) containing 1% acetic acid was added, and the mixture was vortexed or shaken by hand for 1 min. The mixture was placed in a laboratory rotator (Fisherbrand, Thomas Scientific, Swedesboro, USA) for 30 min. Then, 4 g anhydrous $MgSO_4$, 1 g anhydrous CH_3COONa, and 1 g anhydrous NaCl were added, and the mixture was shaken immediately by hand for 1 min. The mixture was then placed in the laboratory rotator for 5 min and centrifuged (Kubota 1720, Osaka, Japan) at 8000 rpm for 5 min. Subsequently, 5 mL of the supernatant was placed in a 15-mL polypropylene centrifuge tube containing 150 mg primary secondary amine, 150 mg graphitized carbon black, and 900 mg $CaCl_2$; the sample was vortexed for 5 min and then centrifuged (Kubota 1720, Osaka, Japan) at 8000 rpm for 5 min. The supernatant (2.5 mL) was transferred to a 15-mL glass tube and dried by evaporation using a nitrogen evaporator at 40 °C (Reacti-Vap, Thomas Scientific, Swedesboro, USA). The resulting product was dissolved in 1 mL acetone, placed in an ultrasonic bath for 1 min, and vortexed for 1 min. After precipitating the polymer matrix, the solution was left as it was for 10 min, 1 mL of supernatant was transferred into a 2 mL of vial while 1 ng of internal standard triphenyl phosphate (TPP) was added. Finally, the mixture was filtered with a 0.45-μm polytetrafluoroethylene syringe filter for analysis by GC–MS/MS.

2.6. Analysis of AQ in steeped tea

Five grams of dried tea was placed in a laboratory tea tasting cup (equipped with a lid and a row of small holes for straining the tea). Then, 150 mL hot water (80 °C, 90 °C, or 100 °C) was added, and the cup covered and left for 5 min. Ten milliliters of the steeped tea was placed into a 50-mL polypropylene centrifuge tube (Eppendorf, Madrid, Spain). AQ was then extracted in same way as the previous samples of hydrated dried tea.

The concentrations of AQ were determined by intrapolation of the relative peak areas for AQ to internal standard peak area in the sample on the spiked calibration curve. The calculated dilution factor of 15 was used for AQ calculation in steeped tea. Blank water samples were also analyzed under the same extraction and chromatographic conditions to assess field contamination of samples and to ensure that equipment cleaning and measurement techniques were adequate. Each analysis was carried out at least in triplicate.

2.7. GC–MS/MS analysis

A Shimadzu GCMS-TQ8030 (Shimadzu Corporation, Kyoto, Japan) was used for determination of AQ residue. The instrument is equipped with Shimadzu AOC-20i autosampler and split/splitless injector in the split mode at 260 °C. HP-1 MS fused silica column, 30 m length × 0.25 mm i.d. × 0.25 μ film thickness, sourced from Agilent Technologies, Santa Clara, CA, USA was used. Oven temperature program was 150 °C for 1 min, raised to 300 °C at 15 °C then held for 2 min. Injected volume was 1 μL. The MS interface was heated to 280 °C, and the ion source was heated to 250 °C. MS was conducted in electron

ionization (EI) mode using an acceleration voltage of 70 eV and was operated in MRM mode using argon at 200 kPa as carrier gas. The MS detector was run in the scan mode (from 50 to 500 *m/z*) where the transitions of MRM in this measurement were set as 208/180 and 208/152 for AQ, and 326/171 for TPP.

2.8. Statistical analysis

Statistical analyses were performed using SPSS V.16 software for Windows (SPSS, Chicago, USA). One-way analysis of variance (ANOVA) procedure was used for statistical analysis. The results were reported significant at the 5% level ($P < 0.05$).

3. Results and discussion

3.1. AQ content during green tea processing

Teas contain AQ derivatives as contaminants (Shang et al., 2019). AQ is a phenolic compound and a member of the quinone family (Fouillaud, Caro, Venkatachalam, Grondin, & Dufossé, 2018). The manufacturing process of green tea uses an RP for enzyme inactivation; an OTR to roll, cut, and tear the leaves; an ECP for the first drying; and a BT for the final drying and shaping of the green tea leaves. The AQ levels (or concentrations) determined in the samples is shown in Fig. 1. The AQ contents of dried samples were measured after each step of green tea processing and found to increase during processing. Significant differences in AQ content were observed for green tea at the OTR, ECP, and BT steps. Notably, no AQ was found in fresh shoots; however, after withering, the contaminant was present in small amounts, and the amount of AQ continued to increase after rolling using the OTR. The OTR did not use heating itself; however, when the tea was applied to the RP, the shoots of tea were moved to the OTR using a conveyor located in the green tea processing room. This room contained abundant amounts of smoke because the RP engine used firewood as a heat source; this smoke was thought to be the source of the contaminant.

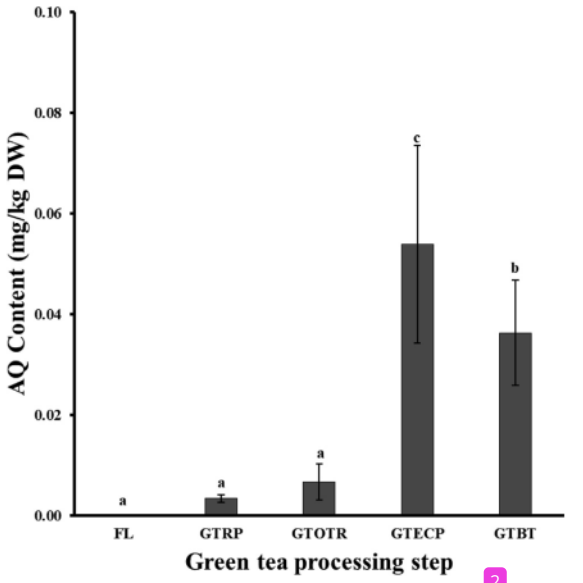

Fig. 1. AQ contents during different green tea processing steps (FL: fresh leaves, GTRP: green tea rotary panner, GTOTR: green tea open top roller, GTECP: green tea endless chain pressure, GTBT: green tea ball tea). The data marked different letters showed significant ($P < 0.05$) difference among different treatment.

As processing proceeded, the tea that had been dried using the ECP exhibited the highest AQ content (greater than 0.05 mg/kg dry weight [DW]). However, after the second drying using the BT, the AQ content decreased. The BT used for this step was a closed dryer, which isolated the tea from the smoke in the processing room. Based on the lack of AQ in fresh leaves, it can be assumed that the source of the contaminant was not the growing process.

Because the AQ content was roughly proportional to the time the tea was exposed to the smoke in the processing room, it can be suggested that the source of contamination was likely the smoke from the wood fires driving the machinery. During green tea processing, including RP for withering and ECP for first drying using firewood, the production space was filled with smoke because there was no air flow to remove the smoke produced from wood burning. The AQ content decreased after the second drying probably because AQ is not heat stable (Fouillaud et al., 2018; Mcdougall, Dobson, & Jordan-Mahi, 2010; Yen & Chung, 1999) and the BT operated at a high temperature for 12 h, leading to the breakdown of this chemical.

3.2. AQ content during black tea processing

Levels (or concentrations) of AQ determined (found or measured) in black tea samples is shown in Fig. 2. Significant differences were observed in AQ contents between fresh and processed black tea. The AQ was not detected in the fresh leaves and it significantly increased after withering through process. Moreover, the highest of AQ was detected after the final drying of black tea production. During processing, the smoke in the room may also have been the source of AQ in black tea. Indeed, the drying process of black tea with the TSD used firewood as a heat source. After enzymatic oxidation, the AQ content increased to 0.08 mg/kg DW, which was four times the limit established by the European Union (0.02 mg/kg). Both green and black teas were processed in a smoky environment, and both showed AQ contamination levels above the allowed standard.

Naeher, Brauer, Lipsett, and Smith (2007) showed that biomass combustion causes the production of many pollutants, including AQ.

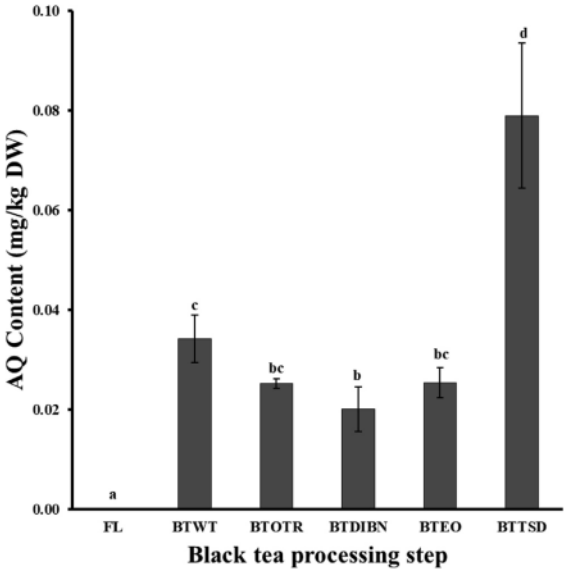

Fig. 2. AQ contents during different black tea processing steps (FL: fresh leaves, BTWT: black tea withering through, BTOTR: black tea open top roller, BTDIBN: black tea double Indian breaker netsortieder, BTEO: black tea enzyme oxidation, BTTSD: black tea two stage dryer). The data marked different letters showed significant ($P < 0.05$) difference among different treatment.

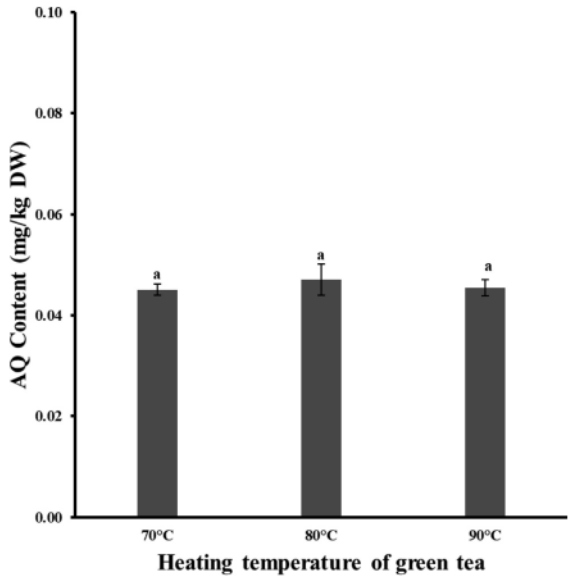

Fig. 3. AQ contents after additional heating of green tea (BT output). The data marked same letter showed not significant ($P < 0.05$) difference among different treatment.

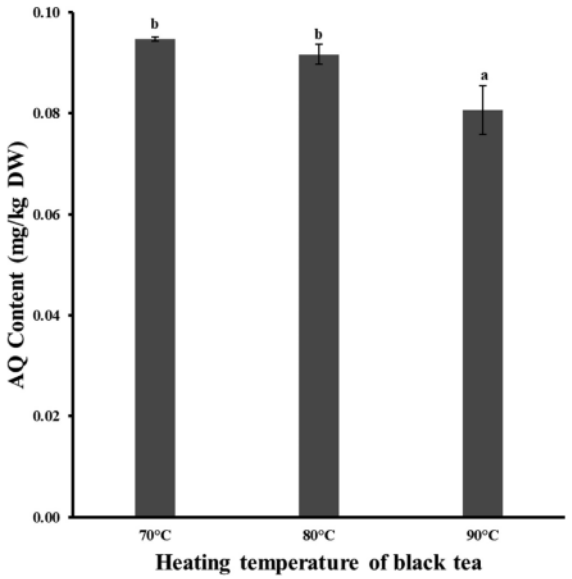

Fig. 4. AQ contents after additional heating of black tea (TSD output). The data marked different letters showed significant ($P < 0.05$) difference among different treatment.

During combustion, the pyrolysis reaction breaks the polymers, producing a variety of substances. AQ has also been detected in barbecue samples during smoking, and the content increases following exposure to high smoke densities (Zastrow, Schwind, Schwagele, & Speer, 2019). *Cinnamomum* sp., which is also used as a source of firewood for processing machinery, has also been reported to produce AQ (Kumar, Kumari, & Mishra, 2019).

3.3. AQ contents in processed green tea and black tea after heating

The influence of heating to 70 °C, 80 °C, and 90 °C on AQ contents in green and black teas was then evaluated (Figs. 3 and 4). Importantly, heating had no effect on the AQ contents of green tea, potentially because the heating duration may have been insufficient for breaking down AQ. However, in black tea, heating significantly affected AQ content. Indeed, heating at 90 °C reduced the AQ content of black tea by 25%. Thus, AQ reacted differently to changes in temperature in the two teas. It is possible that the interaction between smoke and the different compounds in green and black teas could produce AQs with different degrees of heat stability. Previous studies have shown that the heat stability of phenolic compounds, including AQ, is low and that these compounds can be degraded after only 10 min at 70 °C, 80 °C, 100 °C, or 180 °C (Mcdougall et al., 2010; Yen & Chung, 1999); dimeric AQ derivatives are decomposed to monomeric anthrone glycosides. Further studies are required to elucidate the reason for these differences among green and black teas.

3.4. AQ content in green tea and black tea after steeping

AQ could be ingested by drinking tea. Therefore, the AQ content of teas steeped were next evaluated. The five minutes step time is referring to the optimum time to reach 80% releases of its cathechin on common steeping tea production (Vastag, 1998). AQ in steeped tea was extracted by solid-liquid extraction by water and then was subsequent extracted by organic solvent using QuEChERS approach. The AQ contents in both green and black tea are shown in Table 1. Both teas showed very low AQ contents. However, significant differences were observed for different steeping temperatures in both green and black teas. Thus, the steeping process appeared to have diluted the AQ content owing to the large amount of water used, and AQ did not seem to dissolve well in the water. Indeed, AQ is not as soluble in water as it is in chloroform, glycerol, and sulfuric acid (20%) (Cao & Zhao, 2011). Because Indonesians generally drink their tea very sweet and often do not strain out the leaves, it would be interesting to see whether the addition of sugar during the steeping process or allowing the leaves to remain in the tea longer could increase AQ contents.

4. Conclusion

AQ which was found in black and green tea has produced during processing in a tea manufacturing plant. In green tea, AQ started accumulating during the drying process using an RP and then increased continuously throughout the process until the first drying; AQ content finally decreased slightly during the second drying process. In black tea, the increase continued throughout the entire process. A simple subsequent heating step slightly reduced AQ levels in the processed black tea but not in the green tea. Thus, our findings supported that the main source of AQ contamination was smoke from the processing equipment. Although measurements of AQ contents in beverages produced from the two products suggested that the AQ levels present in the drink were unlikely to pose any health risks, tea processing will need to be modified for the export market. Reducing smoke levels in the rooms used for processing could be the simplest step and could be achieved either by improving ventilation or exhaust systems or by the use of cleaner heat sources to drive the machinery. This remediation could also have a positive impact on the workers who spend long hours in tea factory buildings.

CRediT authorship contribution statement

Tuty Anggraini: Conceptualization, Data curation, Funding acquisition, Investigation, Methodology, Writing - original draft, Writing - review & editing. **Neswati:** Project administration. **Ririn Fatma Nanda:** Software. **Daimon Syukri:** Formal analysis, Validation,

Table 1
AQ contents in green and black tea (final processing step) brew at different temperatures.

	70 °C		80 °C		90 °C	
	AQ content ± SD (mg/kg DW)	% RSD	AQ content ± SD (mg/kg DW)	% RSD	AQ content ± SD (mg/kg DW)	% RSD
Green tea	$0.002 \pm 6.49 \times 10^{-7a}$	1.12	$0.003 \pm 6.36 \times 10^{-5a}$	1.28	$0.003 \pm 3.83 \times 10^{-5b}$	2.18
Black tea	$0.002 \pm 3.34 \times 10^{-5a}$	5.02	$0.003 \pm 3.53 \times 10^{-5a}$	5.06	$0.005 \pm 5.06 \times 10^{-5b}$	3.97

Note: The data marked different letters showed significant ($P < 0.05$) difference among different treatment. SD: standard deviation. RSD: relative standard deviation.

Writing - review & editing.

Declaration of Competing Interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2020.127092.

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