

Chemical composition and antioxidant studies of underutilized part of mangosteen (*Garcinia mangostana* L.) fruit

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ARTICLE INFO

Received on: 05/04/2019
Accepted on: 20/05/2019
Available online: 03/08/2019

Key words:

Mangosteen's peel, antiradical, underutilized part, *in vitro* antioxidants, xanthones, DPPH.

ABSTRACT

Mangosteen (*Garcinia mangostana* L) is one of Indonesian fruit with export commodity due to its sweet-sour and pleasant taste. The pulp of this fruit is frequently consumed freshly, while the seed and peel are removed and become a waste. The chemical components contained in mangosteen's seed and peel, especially xanthones, have been reported as antioxidants either *in vitro* or *in vivo*. Several traditional medicine products used the extracts of mangosteen as one of its components; therefore, the characterization of mangosteen extracts through identification of its active components is very important. This review article highlighted the updates on the characterization and antioxidant activities of mangosteen's seed and peel to prove that the wastes of mangosteen fruit could be advantageous to be developed as functional food as antioxidants. Several databases have been used during performing this review, including PubMed, Scopus, Biological abstracts, chemical abstracts, and Google Scholar.

INTRODUCTION

There are some evidences and are supported by scientific publications that phytochemicals contained in fruits are believed to play a beneficial role in the prevention and the treatment of degenerative diseases. Currently, with the jargon "back to nature," there is a great interest in the potential health benefits of exotic fruits due to their antioxidant content and bioactive compounds. Among the exotic fruit commonly consumed is mangosteen or known with "Buah Manggis" in Indonesia (Aramwit *et al.*, 2010; Hait-Darshan *et al.*, 2009). Mangosteen with scientific name of *Garcinia mangostana* L. is a tropical queen of fruits, belonging to the family of Clusiaceae commonly found in throughout India and Southeast Asia, such as Indonesia, Malaysia, Myanmar, Philippines, Sri Lanka, and Thailand (Ji *et al.*, 2017). For Indonesia,

mangosteen has emerged as one of the export commodities of fruits because of its sweet-sour and pleasant taste (Muchtaridi *et al.*, 2017). Mangosteen is cultivated, especially as a source of its highly palatable fruit, which consisted of pulp, contained in a dark purple rind. This fruit is commonly consumed freshly, as a consequence, the peel and seed of mangosteen fruit resulted high amount of wastes. Therefore, some scientists try to take the advantage of the underutilized part (peel and seed) of mangosteen fruit as one of herbal components (Ovalle-Magallanes *et al.*, 2017).

Nowadays, the use of underutilized part of mangosteen, especially peel and seed have increased exponentially, as seen by the huge number of herbal products available in herbal markets containing mangosteen peel, such as Mastin^(R) and SidoMuncul SARI KULIT MANGGIS^(R) (Indonesia), Mangosteen Xango (Malaysia), as well as Mangosteen pericarp Acne Cream^(R) and Mangosteen powder^(R) (Thailand) (Limphapayom *et al.*, 2017). Some modern pharmaceutical formulas containing mangosteen peel extract have been developed by some pharmaceutical formulators, including the encapsulation of nanoemulsions of mangosteen peel extract intended as topical formulation (Mulia *et al.*, 2018a). The encapsulation of

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mangosteen extract in nanoemulsions is promising strategy to utilize the active compound contained, particularly α -mangostin. [Mulia et al. \(2018b\)](#) developed nanoemulgel mangosteen extract in virgin coconut oil for topical formulation. The developed nanoemulgels exhibited better penetration ability than its nanoemulsion. [Pratiwi et al. \(2017\)](#) developed self-nanoemulsifying drug delivery system (SNEDDS) with enhanced solubilization of ethanol extract from mangosteen peel intended for treatment of topical gangrene foot. Previously, SNEDDS from ethyl acetate extract of mangosteen peel intended for the improvement of diffusion and pre-cut absorption was previously developed ([Pratiwi et al., 2016](#)).

Traditionally, underutilized form of mangosteen (peel and seed) in the form of infusions and decoctions has been used to treat infections of skin, urinary tract, and gastrointestinal, and has been believed to act as laxative, anti-scorbutic, and anti-fever agent ([Ovalle-Magallanes et al., 2017](#)). Besides, peel and seed of mangosteen have been reported for the treatment of diarrhea, abdominal pain, dysentery, suppuration, wound infection, and chronic ulcer ([Cui et al., 2010](#); [Gorinstein et al., 2011](#); [Suksamram et al., 2006](#)) and to treat inflammatory and immunological related-diseases, such as acne, food allergies, and arthritis ([Wang et al., 2017](#)). These activities indeed need to be correlated with the chemical composition contained in mangosteen's seed and peel ([Genovese et al., 2016](#)).

METHODS

During performing this review, we used several databases, including Scopus, PubMed, and Google Scholar to identify and to download the abstracts, reports, and research papers related to chemical composition, characterization, and antioxidant activities of mangosteen's peel and mangosteen's seed. The keywords used during searching of information was (antioxidant + mangosteen peel or mangosteen seed + chemical composition + *in vitro* + *in vivo*) in the month of February–March 2019.

CHEMICAL CONSTITUENTS OF MANGOSTEEN'S PEEL

The peel of mangosteen fruit has been reported to contain some phenolic compounds, such as tannins, flavonoids, xanthenes, and other bioactive substances which support the medicinal properties ([Pothitirat et al., 2009](#)). Some chemical compounds or secondary metabolites have been isolated from Mangosteen rind which includes xanthenes (a class of polyphenolic compounds with a characteristic tricyclic aromatic ring system) or xanthen-9H-ones. Of the 40 xanthenes present in the pericarp of the fruit, the most abundant xanthenes found are α -mangostin, β -mangostin, and γ -mangostin ([Chen et al., 2008](#); [Pedraza-Chaverri et al., 2009](#); [Zarena and Sankar, 2011](#)). Mangostins (α -, β - and γ -) are the most frequently studied. They have a unique chemical structure with a tricyclic aromatic system carrying isoprene, methoxyl and hydroxyl groups ([Fig. 1](#)). Some biological activities either *in vitro* or *in vivo* are correlated with the contents of these mangostins, including antibacterial, antifungal, antimalarial, anticarcinogenic, and antiatherogenic activities ([Obolskiy et al., 2009](#)).

The other and new xanthenes are found, namely, isogarcinol ([Chen et al., 2017](#)), garcinone E, 9-hydroxycalabaxanthone, 8-deoxygartanin, and gartanin 1,3,6,7-tetrahydroxy-2,8-(3-methyl-2-butenyl), [1,3,6-trihydroxy-

7-methoxy-2,8-(3-methyl-2-butenyl)xanthone], 1,3,7-Trihydroxy-2,8-di-(3-methylbut-2-enyl)xanthon, 1,7-dihydroxy-3-methoxy-2-(3-methylbut-2-enyl)xanthon ([Wittenauer et al., 2012](#)), mangostanaxanthenes V and VI, mangostanaxanthone IV, garcimangosone D, aromadendrin-8-C- β -D-glucopyranoside, 1,2,4,5-tetrahydroxybenzene, 2,4,3'-trihydroxybenzophenone-6-O- β -glucopyranoside, maclurin-6-O- β -D-glucopyranoside, and 2,4,6,3',5'-pentahydroxybenzophenone ([Mohamed et al., 2017](#)), and epicatechin ([Yu et al., 2007](#)). In addition, [Jung et al. \(2006\)](#) have isolated two highly oxygenated prenylated xanthenes, namely, 8-hydroxycudraxanthone G and mangostingone or [7-methoxy-2-(3-methyl-2-butenyl)-8-(3-methyl-2-oxo-3-butenyl)-1,3,6-trihydroxyxanthone, along with cudraxanthone G, 8-deoxygartanin, and garcimangosone B.

The composition of chemical constituents depending on several factors such as maturity, locations of cultivation, and extracting solvents. [Pothitirat et al. \(2009\)](#) evaluated the chemical compositions of active compounds (α -mangostin, total phenolics, flavonoids, and tannin contents) in mangosteen peel at two stages of maturity (young and mature). The young and mature of ethanolic (95%) extracts of mangosteen rind contained α -mangostin of 8.07 ± 0.11 and 13.63 ± 0.06 %wt/wt of extract, respectively. The mature of mangosteen also have the higher contents of phenolics, flavonoids, and tannin significantly than the young ones.

[Muchtaridi et al. \(2017\)](#) have evaluated the levels of α -mangostin, γ -mangostin, and gartanin in ethanolic (70%) extracts of Mangosteen rind from four different locations in Indonesia (Bogor, Purwakarta, Subang, and Tasikmalaya). The analysis showed that the levels of α -mangostin, γ -mangostin, and gartanin are 13.87%, 8.28% and 10.44%, respectively, from Bogor; 10.07%, 6.33%, and 8.76% from Purwakarta; 10.88%, 6.01%, 8.08% from Subang and 8.53%, 6.07%, 17.28% from Tasikmalaya, respectively.

[Kusmayadi et al. \(2018\)](#) have evaluated the effects of extracting solvents, namely, methanol, ethanol, acetone, hexane, ethyl acetate, acetic acid, and aquadest used during extraction of mangosteen peel at different times (24, 36, and 48 hours). The results showed that acetone was the best solvent used for xanthone extraction with time extraction of 36 hours which is not significantly different at 48 hours ($p < 0.05$). [Ghasemzadeh et al. \(2018\)](#) also investigated the effects of solvents for extraction of α -mangostin mangosteen peel extract along with extraction time, microwave power, and solvent percentage using experimental design approach of response surface methodology. The ethyl acetate was the best solvents capable of extracting the highest concentration of α -mangostin, followed by dichloromethane, ethanol, and water. The extraction condition also affected the antioxidant activities *in vitro* of mangosteen peel. [Samuagam et al. \(2013\)](#) have optimized the extraction condition (ethanol percentage, time extraction, and temperature) and the optimum conditions to get maximum yield of extract were 60% ethanol concentration for 60 minutes at 25°C. Using this condition, the mangosteen peel extract revealed IC_{50} value of 19.75 μ g/mL (DPPH radical scavenging assay), 54.66% nitric oxide scavenging activity, and 79.94% β -carotene bleaching (BCB) assay.

ANTIOXIDANT ACTIVITIES

Several methods have been used for the evaluation of antioxidant activities *in vitro*, including radical scavenging

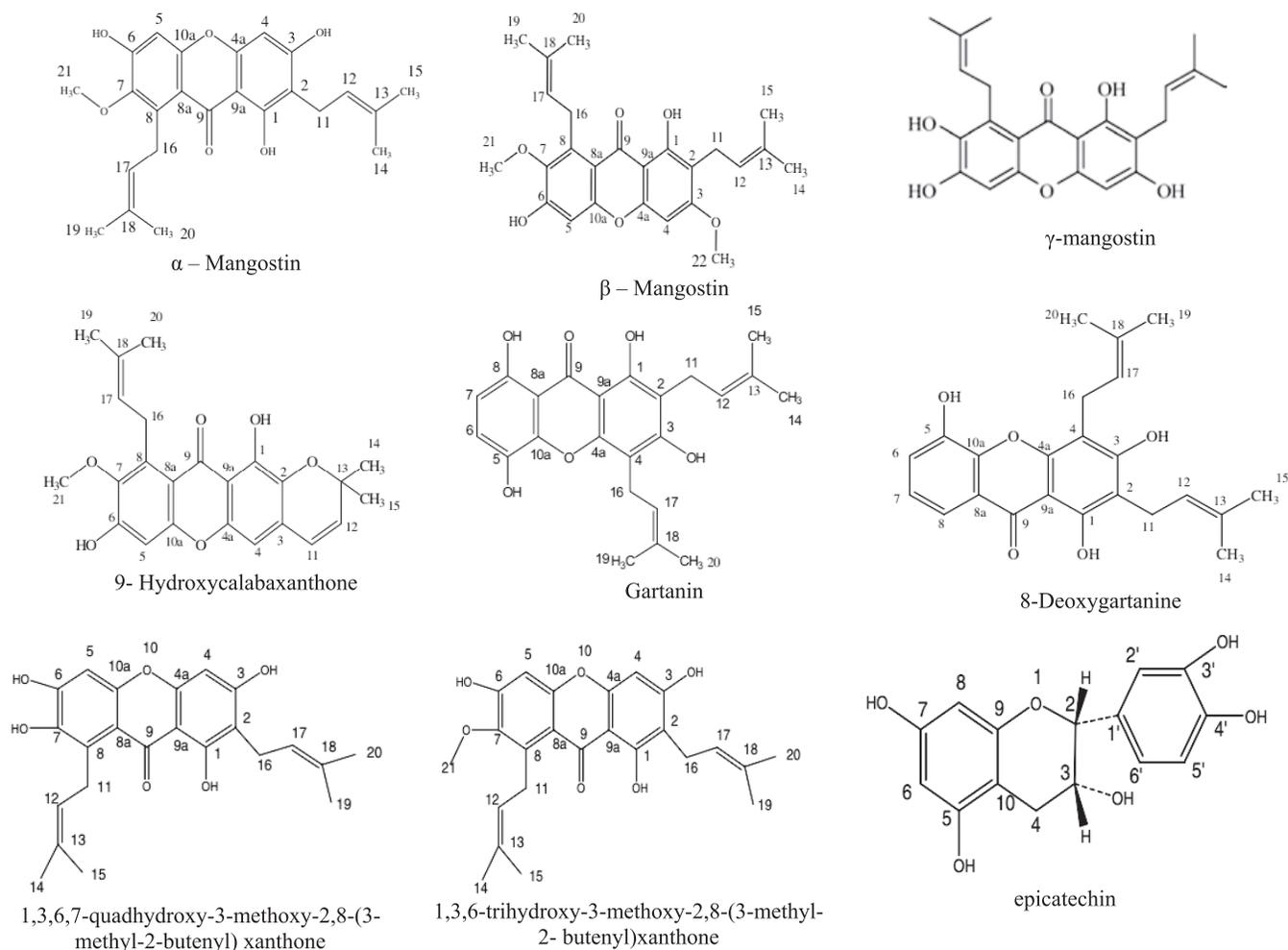


Figure 1. Some chemical compounds isolated from rind and seed of mangosteen fruit.

activities using radicals of 1,1-diphenyl-2-picrylhydrazyl (DPPH) and 2,2-azinobis(3-ethylbenzothiazoline-6-sulfonic acid) diammonium salt (ABTS) cation, superoxide anion ($O_2^{\cdot-}$), nitric oxide ($\cdot NO$), peroxyxynitrite ($ONOO^{\cdot}$), and hydroxyl radical ($\cdot OH$), the deoxyribose assay, oxygen radical absorbance capacity (ORAC), chelating activity using ferrous ion (Fe^{2+}), ferric reducing antioxidant power (FRAP), phosphomolybdenum method, *cytochrome c* reducing capacity, ferric thiocyanate method, and thiobarbituric acid assay (Suttirak and Manurakchinakorn, 2012).

Radical scavenging activities using DPPH radical

Among radicals used for modeling, the antioxidant activities of samples *in vitro*, DPPH and ABTS radicals are the most reported in scientific literature. The scavenging activity of DPPH radical measures the capability of evaluated samples to donate hydrogen radicals to capture DPPH radical ($DPPH^{\cdot}$). The color changes of DPPH solution from a deep purple to a light yellow as indicated by decreased absorbance at 515–517 nm (Surveswaran *et al.*, 2007). Phenolics and flavonoids are typical compounds capable of donating radical hydrogens (Yu *et al.*, 2007). Tjahjani *et al.* (2014) have evaluated DPPH radical scavenging activities of ethanolic extracts (96% and 70%) and fractions of hexane, ethyl acetate, butanol, and water of mangosteen peel. The antioxidant

activities were expressed with inhibition concentration of 50% of radical (IC_{50}). The lower IC_{50} indicated the more active of evaluated samples. Hexane fraction showed the most active antiradical with IC_{50} of $3.62 \pm 0.04 \mu g/ml$, followed by 70% ethanol extract, 96% ethanol extract, and ethyl acetate extract with IC_{50} values of 6.56 ± 0.31 , 7.48 ± 0.19 , and $13.29 \pm 0.12 \mu g/ml$, respectively.

Palakawong *et al.* (2010) have evaluated the 50% ethanolic extracts of peel, leaves, and bark of mangosteen. Among these samples, the peel extract showed the highest antiradical activities using DPPH radicals with IC_{50} of 5.94 $\mu g/ml$, followed by bark 6.46 and leaves 9.44 $\mu g/ml$. However, the antioxidant of evaluated extracts was lower than that of vitamin C used as a positive control with IC_{50} of 4.30 $\mu g/ml$. This study obtained IC_{50} values less than those reported by Weecharangsan *et al.* (2006) using the same extraction methods, in which 50% of ethanolic extract of mangosteen peel was of 30.7 $\mu g/ml$. The difference in IC_{50} values could be explained that the used mangosteen samples were in the different maturity stage, in which Palakawong *et al.* (2010) used mangosteen in maturity stage of 3, while Weecharangsan *et al.* (2006) used the maturity stage of 5 or 6. Supiyanti *et al.* (2010) also reported that IC_{50} of ethanolic extract of mangosteen peel was about 8.56 $\mu g/ml$, while vitamin C as positive control had IC_{50} of 3.37 $\mu g/ml$.

The antiradical scavenging activity of mangosteen peel extracts expressed by %inhibition has been used for comparative studies of extracting solvents (methanol, ethanol, acetone, and aqueous). At the same concentration, aqueous extract exhibited the highest radical scavenging activity compared methanol, ethanol, and acetone with %inhibition of $67.45 \pm 1.05\%$, $18.81 \pm 1.44\%$, $46.97 \pm 0.29\%$, and $9.19 \pm 1.77\%$, respectively (Kamaludin *et al.*, 2016). DPPH radical assay was also used to compare 80% ethanol extract of peel and pulp of yellow mangosteen (*Garcinia tinctoria*), and the results showed that mangosteen peel had higher antiradical assay (IC_{50} of 48.8 $\mu\text{g/ml}$) than that of pulp (IC_{50} of 153.2 $\mu\text{g/ml}$). This result corresponds to the levels of total phenolic contents present in peel and pulp of mangosteen fruit (Arazo *et al.*, 2011).

Mangosteen peel extracts (ethanol 7%) has been formulated as oral solution dosage forms and its antioxidant properties have been assessed using DPPH radical scavenging activity. Xanthenes, the active compounds contained in mangosteen peel, are not soluble in water, therefore some co-solvents, namely, polyethylene glycol (PEG) 400-glycerol (20–20, 20–40, 40–20, 40–40) are optimized intended to improve its solubility. Oral solution with the composition of co-solvents of PEG 400-glycerol (40:40) has the highest DPPH radical scavenging activity with IC_{50} of 24.81 $\mu\text{g/ml}$ (Sumarny *et al.*, 2014). The IC_{50} obtained was indeed lower than that of its extract due to the addition of co-solvents in the formula with no activity as radical scavenger.

ABTS radical scavenging activities

ABTS (2,2'-azinobis(3-ethylbenzothiazoline-6-sulfonic acid)) radical scavenging method was used to confirm the results obtained from DPPH radical scavenging since both methods are based on a similar antioxidant mechanism and both radicals used were soluble in polar solvents (methanol and ethanol). This assay measures the ability of antioxidants to scavenge stable radical cation ABTS + having blue-green color with maximum absorption at 734 nm which decreases in its intensity due to the presence of antioxidants (Zhong and Shahidi, 2015). The ABTS scavenging activity of mangosteen's peel and seed on free radical ABTS was compared with amount of trolox (standard), as a consequence, ABTS radical scavenging activity was expressed as trolox equivalent antioxidant capacity (TEAC). Okonogi *et al.* (2007) evaluated ABTS radical scavenging of 95% ethanol extract of mangosteen peel, and the results showed that extract had TEAC of 3.00 ± 0.016 (mM/mg), higher than the same extract of Banana, Coconut, Dragon fruit, Passion fruit, and Long-gong with TEAC values of 1.80 ± 0.038 , 1.53 ± 0.044 , 0.685 ± 0.001 , 0.591 ± 0.008 , and 0.207 ± 0.002 mM/mg extract, respectively. The same results (TEAC value of 3.00 ± 0.016 mM/mg) were also obtained by Tachakittirungrod *et al.* (2007). In addition, Surveswaran *et al.* (2007) reported that TEAC value of ethanol extract of mangosteen peel was 3.91 mM/mg, comparable to that reported by Okonogi *et al.* (2007). The TEAC values of ethyl acetate and acetone extracts of mangosteen peel were also reported, i.e., 3.821 and 3.815 $\mu\text{M/ml}$ (Zarena and Sankar, 2009). Xanthenes (mangostins, garcinone-E, methoxy-bmangostin, garcimangosone A, garcimangosone B, garcimangosone C) present in mangosteen were reported to be responsible for this antioxidant activity.

Peroxynitrite scavenging activity

Peroxynitrite (ONOO⁻) scavenging activity is analyzed by monitoring the oxidation of dihydrorhodamine 123

(DHR 123). In this procedure, DHR 123 is oxidized by native ONOO⁻ and ONOO⁻-derived from the peroxynitrite donor 3-morpholinonydnonimine hydrochloride (SIN-1). The oxidized DHR 123 is then evaluated by the luminescence spectrophotometer using excitation wavelength of 480 nm and emission wavelength of 530 nm. The peroxynitrite scavenging activity could be related to the fluorescence intensities of oxidized DHR 123 (Zou *et al.*, 2005). The radical activity was expressed as IC_{50} values. Jung *et al.* (2006) have evaluated 14 compounds isolated from mangosteen peel. Among these compounds, smeaxanthone A, γ -mangostin, and gartanin showed the highest peroxynitrite radical scavenging activities with IC_{50} values of 2.2, 8.0, and 9.1 μM , respectively. As positive control, DL-penicillamine had IC_{50} value of 3.1 μM .

Ferric reducing power

Ferric reducing antioxidant power (FRAP) measures the reducing power of samples. FRAP assay is relied on the ability of the antioxidant to reduce Fe^{3+} to Fe^{2+} in the presence of TPTZ (2,4,6-tripyridyl-s-triazine) resulted an intense blue color of Fe^{2+} -TPTZ complex with an absorption maximum at 593 nm (Yang and Zhai, 2010). Azima *et al.* (2014) have evaluated the reducing power of mangosteen peel extracts. The sample preparation was carried out by stirring samples in 100 mM citrate buffer (pH 3.0) in the ration 1:4 for 10 minutes at 100°C. The filtrate were collected and evaporated by using rotary evaporator at 60°C and 114 mbar. The mangosteen peel extract had the highest FRAP with FRAP value of 79.37 ± 0.77 A mM/g Trolox equivalent antioxidant capacity (TEAC) compared to guava peel extract (FRAP value of 25.66 ± 1.40 mM/g TEAC) and *Clitoria ternatea* extract (13.32 ± 0.28 mM/g TEAC). This antioxidant activity correlated with the contents of antocyanin.

In vivo antioxidant activity of 80% ethanolic extract of mangosteen has been evaluated by Samuagam *et al.* (2015). In this study, rats were treated with mangosteen peel extracts for 14 and 30 days with dose of 100 mg/kg/day, orally, and the liver was taken for antioxidant assays. For positive control, vitamin E was used. The results showed that rats given with mangosteen's peel extract exhibited the significant increase ($p < 0.05$) of enzymatic antioxidants (catalase, superoxide dismutase, glutathione reductase, and lipid peroxidation levels) compared with no treated-rats group (control group).

CONCLUSION

Mangosteen peel can be considered as the wastes due to the consumption of mangosteen fruit. With the potentiality of underutilized part of fruit as antioxidant, some scientists have explored the possibility mangosteen peel as functional food or food component with beneficial effects on human health. Mangosteen peel contained high amount of phenolic compounds, such as mangostin and gartanin, which are believed to be responsible for antioxidant activities. Mangosteen peel has been reported to have antioxidant activities either *in vitro* or *in vivo*, having potential to be developed as food antioxidants.

ACKNOWLEDGMENTS

We would like to thank the Consortium of World Class Research University for financial support through Program Penelitian Kolaborasi Indonesia 2019 with contract number 2053/UN1.PIII/DIT-LIT/LT/2019.

COMPETING INTERESTS

The authors declare that they have no competing interests.

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How to cite this article:

Rohman A, Rafi M, Alam G, Muchtaridi M, Windarsih A. Chemical composition and antioxidant studies of underutilized part of mangosteen (*Garcinia mangostana* L.) fruit. *J Appl Pharm Sci*, 2019; 9(08):047–052.