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Evaluation of γ-radiation on green tea odor volatiles

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ABSTRACT

The aim of this study was to evaluate the gamma radiation effects on green tea odor volatiles in green tea at doses of 0, 5, 10, 15 and 20 kGy. The volatile organic compounds were extracted by hydrodistillation and analyzed by GC/MS. The green tea had a large influence on radiation effects, increasing the identified volatiles in relation to control samples. The dose of 10 kGy was responsible to form the majority of new odor compounds following by 5 and 20 kGy. However, the dose of 5 kGy was the dose that degraded the majority of volatiles in non-irradiated samples, following by 20 kGy. The dose of 15 kGy showed has no effect on odor volatiles. The gamma radiation, at dose up to 20 kGy, showed statistically no difference between irradiated and non irradiated green tea on odors compounds.

1. Introduction

Tea is the most consumed beverage in the world after water. Annual production of dried tea leaves is estimated to be 1.8 million tons, providing 40 L of tea beverage per capita worldwide. Compared to oolong and black tea, the consumption of green tea has been increasing during recent years, probably due to the information about valuable health-related properties of this plant (Cheng et al., 2008).

Tea is a significant commercial crop that grows in both tropical and sub-tropical regions, where growing conditions can be very different. Tea is processed from tender shoots of *Camellia sinensis* (L.) O. Kuntze. Quality of tea has three dimensions: physical appearance, infusion and flavor (Rawat and Gulati, 2008).

Green tea, consumed mainly in Japan, China and Korea, is produced when freshly harvested leaves of *C. sinensis* are subjected to withering, and then they are panfried/steamed prior to rolling/shaping and drying (Santana-Rios et al., 2001). Tea consumption in the world has increased because of its health benefits (Kim et al., 2007). Evidence from animal studies indicates that green tea and its catechins retard the development or progression of atherosclerosis in apoE-deficient mice and hypercholesterolemic hamsters. Epidemiological studies have shown an inverse correlation between coronary heart disease risk and green tea consumption in humans (Koo and Noh, 2007).

People enjoy tea for its taste and flavor. Therefore, most studies conducted on tea showed taste and flavor investigations. In the 1970s and 1980s, many researchers analyzed the flavor constituents of various tea extracts (Yanagimoto et al., 2003).

Flavor comprises, principally, taste and aroma. Non-volatile components are generally responsible for the taste, while volatile components give the aroma. In tea, volatile organic compounds (VOC) are present in minimum quantities (0.01% of the total dry weight), but have a high impact on the flavor due to their low threshold value and resulting high odor units. VOC of tea are classified into two groups: group I, consisting mainly of non-terpenoids, which account for the fresh green tea flavor and group II, terpenoids, which impart sweet flowery aroma to tea, viz, monoterpene alcohols such as linalool and geraniol. The presence of group II compounds is highly desirable (Ravichandran and Parthiban, 1998; Rawat et al., 2007).

The past decade had a significant increase in the use of herbal medicine. Despite a great deal of studies on the mycoflora in agricultural products, only a few were concerned with spices and medicinal herbs, which are more and more common in our daily diet and play an important role in the economy, like the green tea (Romagnoli et al., 2007). Nevertheless, some investigators reported the presence of moulds in medicinal plants, herbs and natural drugs that are widely consumed not only as home medicine, but as raw materials for the pharmaceutical industries as well (Aquino et al., 2007; Roy and Chourasia, 1989).

In some kinds of medicinal plants analyzed, the percentage of contamination in packed samples was higher than in those non-packed, this may be caused by other factors such as humidity inside the pack or unsuitable methods of keeping and storing the pack (Abou-Arab et al., 1999).

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Ionizing radiation is one of the most effective means to disinfect dry food ingredients. This treatment can inhibit cellular life division, like microorganisms, and promote a molecular structural modification (Farkas, 2006; Villavicencio et al., 2007). The aim of this study was to evaluate the effects of gamma radiation on odor compounds in green tea.

2. Material and methods

2.1. Samples

The green tea was donated by Herbarium Laboratório Botânico Ltda. (Paraná, Brazil).

2.2. Irradiation

The samples were packed in plastic (polyethylene) bags in the presence of air, sealed and identified with their respective radiation doses. They were irradiated at room temperature in a ^{60}Co source Gammacell 220 (Nordion Ltd., Canada) with a dose rate of 2.38 kGy/h, at doses of 0, 5, 10, 15 and 20 kGy. Harwell Amber 3042 dosimeters were used to measure the radiation dose.

2.3. Extraction of VOC by hydrodistillation

100 g of green tea plus 10 μL (1 μg/μL ethanol) of timol (standard) was placed in a round-bottom flask with 700 mL of boiling distilled water. The contents were distilled for 60 min to collect the volatile oil. The essential oil was separated from the aqueous phase by dichloromethane (10 mL) for 60 min, dried over pure nitrogen flow, re-suspended with 1 mL of dichloromethane and stored in a freezer (−4 °C) (Ravichandran and Parthiban, 1998).

2.4. Gas chromatography–mass spectrometry (GC–MS) analysis

The concentrated extracts were analyzed in a HP 5973 MSD detector coupled to a HP 6980 GC. An HP-5 MS column (30 m/0.25 mm id, film thickness 0.25 μm) was used with helium as a carrier gas. The injector temperature was 200 °C in splitless mode.

The GC oven temperature was programmed to hold at 50 °C for 0.5 min and then to increase to 250 °C at 5 °C/min, finally holding at 250 °C for 0.5 min. Column flow rate was 1 mL/min. Ion source temperature was 230 °C and the interface temperature was set at 200 °C. The MS was scanned at 70 eV over 30–550 amu. Sample injection volume was 1.0 μL.

2.5. Statistical analysis

In this study, the numerical observations of volatiles' behaviour with respect to radiation doses cannot be represented by precisely specified values, making information vague or fuzzy. The principal component analysis (PCA) is a technique whose main objective is to obtain a small number of linear combinations (called principal components) of a set of variables that have the most possible information contained in the original variables through the assignment of a different "role" to each value of an interval in contrast to the interval valued data approach in which each value has a uniform importance (Giordani and Kiers, 2004). PCA was performed using the Win-DAS software (Kemsley, 1998).

3. Results and discussion

A total of 124 volatiles were identified in green tea. Of these 124 compounds, the irradiation formed 66.12% (n=82) of new compounds (not founded in non irradiated sample). From total of volatiles identified, only 45 compounds (36.29%) have some kind of odor (Table 1). The odor description of all compounds founded in this paper (irradiated and non irradiated samples) is described in literature.

Almost the half of compounds identified with odor, 48.9% (n=22), were formed after irradiation. The dose of 10 kGy was responsible to form the majority (n=11) of these new VOC; however, four compounds ((+)-aromadendrene; 2,6-dimethylnaphthalene; 4-decalactone and *p*-toluol) were not resistant when a dose of 15 kGy was used and one compound (benzyl benzoate) was degraded with 20 kGy.

The dose of 5 kGy was the second dose that formed more VOC with odor (n=9) and only the 4-vinylguaiacol was degraded when applied 10 kGy. The dose of 20 kGy formed the compounds butanoic acid and trans-2-decenal and the dose of 15 kGy not formed any VOC with odor.

Regarding the aromas found naturally (not irradiated samples), majority of the VOCs are stable in all different radiation doses used. Only four compounds (1-tetradecanal, cis-geraniol, octadecanal and cis-linalool oxide) were degraded with 5 kGy and the caryophyllene oxide was degraded with 20 kGy.

Most of the compounds (n=19) found naturally in the green tea in this work has some kind of pleasant odor. The same profile has been reported in other studies (Hattori et al., 2005; Yamaguichi and Shibamoto, 1981). Among the pleasant odors are the flavors of fruit (3,5-octadien-2-one and β-damascone), sweet (2-undecenal, octanoic acid and cis-geraniol), green (nonanoic acid), mint (α-terpineol, β-cyclocitral and methyl salicylate), herb/wood (α-cadinol, caryophyllene oxide and safranal), honey (phenethyl alcohol) and flower (1-tetradecanal, geranylacetone, linalool, nerolidol, cis-linalool oxide and geraniol).

Although the dose of 10 kGy is the dose that created more odor compounds, only p-toluol has an unpleasant odor (medicine/smoke). Among the pleasant odors that the dose of 10 kGy created are the aromas of wood ((+)-aromadendrene), green ((E)-2-nonenal), grass (2,6-dimethylnaphthalene), fruit/flower (4-decalactone, acetophenone, β-ionone, β-damascenone and farnesol), sweet (hexanoic acid) and herb (benzyl benzoate).

The dose of 5 kGy was the only dose that formed only pleasant odors, creating the aromas of clove/curry (4-vinylguaiacol), flower (phyton, farnesylacetone and phytol), sweet (phenylacetaldehyde and phenylmethanol), almond (benzaldehyde), mint (geranial) and fruit (spathulenol), unlike the dose of 20 kGy that formed only unpleasant aromas. The compounds butanoic acid (rancid aroma) and trans-2-decadal (tallow aroma) appeared due the interaction of this dose with lipids causing the oxidation of the essential oils and other lipids that are present in this kind of tea.

Even if the doses of 10 and 20 kGy form unpleasant odors, it does not mean that these compounds have a negative impact on the tea flavor. Compounds with unpleasant odors were also found in the control sample as (E,E)-2,4-heptadienal (fried odor), (Z)-2-decenal (tallow odor) and 2,4-decadienal (fried/fat odor). The same theory is true for compounds with pleasant odors formed after the irradiation, meaning they do not better the aroma of tea. For this, a sensory analysis should be performed to verify that the quantity of these compounds formed by radiation, pleasant or unpleasant, has a directly interference on consumers perception.

However, Furgeri et al. (2007) studyied the sensorial analysis in maté (*Ilexis paraguariensis*), irradiated at doses up to 10 kGy, demonstrated that there was no difference between the irradiated and non-irradiated samples. Salum et al. (2007) showed in their sensorial analysis that the participants had difficulties to define among the differences in odor between the control and irradiated cinnamon and nut meg.

Table 1
Volatile organic compounds with odors identified in green tea at different radiation doses.

#CAS	RT (min)	Compounds	Doses (kGy)					Odor	Reference[a]
			0	5	10	15	20		
100-52-7	6.53	Benzaldehyde		x	x	x	x	Almond, caramel	Schieberle and Grosch, 1987
142-62-1	7.51	Hexanoic acid			x	x	x	Sweet	Adedeji et al., 1991
107-92-6	7.60	Butanoic acid					x	Rancid, cheese	Adedeji et al., 1991
4313-03-5	7.72	(E,E)-2,4-Heptadienal	x	x	x	x	x	Fried	Ullrich and Grosch, 1988
100-51-6	8.48	Phenylmethanol		x	x	x	x	Sweet, flower	Blank et al., 1989
122-78-1	8.68	Phenylacetaldehyde		x	x	x	x	Honey, sweet	Schieberle and Grosch, 1987
98-86-2	9.29	Acetophenone			x	x	x	Flower, almond	Guichard et al., 1995
106-44-5	9.70	p-Toluol			x			Medicine, smoke	Blank et al., 1989
38284-27-4	9.85	3,5-Octadien-2-one	x	x	x	x	x	Fruit, mushroom	Ullrich and Grosch, 1988
5989-33-3	9.96	cis-Linalool oxide	x					Flower	Chung et al., 1993
78-70-6	10.32	Linalool	x	x	x	x	x	Flower, lavender	Schieberle and Grosch, 1998
60-12-8	10.65	Phenethyl alcohol	x	x	x	x	x	Honey, spice, rose	Blank et al., 1989
18829-56-6	11.96	(E)-2-Nonenal			x	x	x	Cucumber, green	Ullrich and Grosch, 1987
124-07-2	12.67	Octanoic acid	x	x	x	x	x	Sweat, cheese	Adedeji et al., 1991
98-55-5	12.82	α-Terpineol	x	x	x	x	x	Anise, mint	Berger et al., 1989
119-36-8	12.90	Methyl Salicylate	x	x	x	x	x	Peppermint	Berger et al., 1989
116-26-7	13.05	Safranal	x	x	x	x	x	Herb, sweet	Jorgensen et al., 2000
432-25-7	13.63	β-Cyclocitral	x	x	x	x	x	Mint	Berger et al., 1989
106-25-2	13.85	cis-Geraniol	x					Sweet	Chung et al., 1993
106-24-1	14.59	Geraniol	x	x	x	x	x	Rose, geranium	Berger et al., 1989
2497-25-8	14.77	(Z)-2-Decenal	x	x	x	x	x	Tallow	Schnermann and Schieberle, 1997
141-27-5	15.02	Geranial		x	x	x	x	Lemon, mint	Schieberle and Grosch, 1998
112-05-0	15.51	Nonanoic acid	x	x	x	x	x	Green	Jirovetz et al., 2002
7786-61-0	16.14	4-Vinylguaiacol		x				Clove, curry	Blank et al., 1989
2363-88-4	16.25	2,4-Decadienal	x	x	x	x	x	Fried, fat	Ullrich and Grosch, 1987
706-14-9	17.41	4-Decalactone			x			Peach	Berger et al., 1989
3913-81-3	17.45	trans-2-Decenal					x	Tallow	Gasser and Grosch, 1990
2463-77-6	17.47	2-Undecenal	x	x	x	x	x	Sweet	Gasser and Grosch, 1990
23726-93-4	18.01	β-Damascenona			x	x	x	Apple, rose, honey	Guth and Grosch, 1991
581-42-0	18.76	2,6-Dimethyl Naphthalene			x			Grass	Le Guen et al., 2000
23726-92-3	19.66	β-Damascone	x	x	x	x	x	Apple	Fuhrmann and Grosch, 2002
3796-70-1	19.77	Geranylacetone	x	x	x	x	x	Magnolia, green	Mau et al., 2003
14901-07-6	19.84	β-Ionone			x	x	x	Violet, raspberry	Berger et al., 1989
638-66-4	20.17	Octadecanal	x					Oil	Choi, 2003
7212-44-4	22.39	Nerolidol	x	x	x	x	x	Wood, flower	Chung et al., 1993
6750-60-3	22.70	Spathulenol		x	x	x	x	Herb, fruit	Jirovetz et al., 2002
481-34-5	24.43	α-Cadinol	x	x	x	x	x	Herb, wood	Chung et al., 1993
1139-30-6	24.86	Caryophyllene oxide	x	x	x	x		Herb, sweet, spice	Chung et al., 1993
489-39-4	25.36	(+)-Aromadendrene			x			Wood	Chung et al., 1993
4602-84-0	25.89	Farnesol			x	x	x	Muguet	Berger et al., 1989
124-25-4	25.99	1-Tetradecanal	x					Flower	Chisholm et al., 2003
120-51-4	26.75	Benzyl benzoate			x	x		Balsamic, herb	Adedeji et al., 1991
502-69-2	28.45	Phytone		x	x	x	x	Jasmine	Mau et al., 2003
762-29-8	29.90	Farnesylacetone		x	x	x	x	Flower, ether	Mau et al., 2003
150-86-7	33.54	Phytol		x	x	x	x	Flower	Chung et al., 1993

[a] The references in this table correspond to the papers that describe the compounds odors.

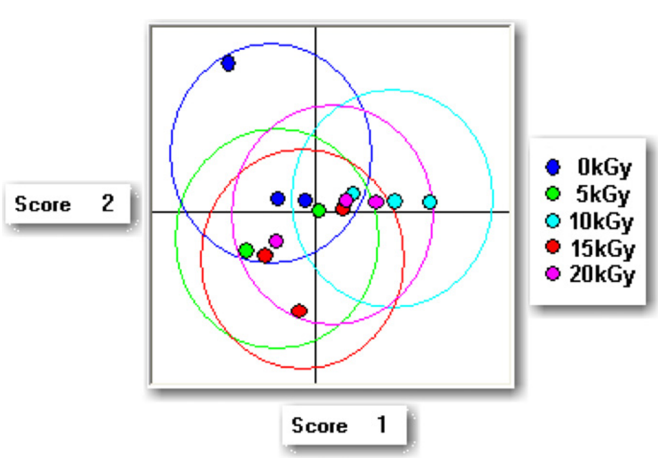

Fig. 1. Principal components analysis from green tea irradiated with different doses.

The PCA showed a slight deviation from the control sample for the other doses. However, this variation cannot be considered since there was a difference between the radiation doses and control samples (Fig. 1).

Similar results were described by Seo et al. (2007), using the maximum dose of 20 kGy, demonstrating that the profile of volatiles' composition in *Angelica gigas* (a medicinal plant), statistically, did not change with irradiation.

Jo et al. (2003) irradiated green tea leaves extracts at doses up to 20 kGy and showed that the extract had enhanced its color proportionally to the increased dose, but there was no difference between irradiated and non-irradiated leaves as to their physiological activity.

4. Conclusion

The gamma radiation showed it has a direct influence on volatile compounds, increasing VOC quantity, degrading few compounds and can producing even pleased as unpleased odors that are not found naturally in green tea, although no statistical difference among irradiated and non-irradiated samples was found. Sensorial analysis will be performed to understand if new compounds are formed and the degraded volatiles have some influence to consumer perception.

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