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Research Article

Formulation and comparative analysis of polyherbal cough syrup prepared with different palmyrah sweeteners

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Abstract: The purpose of this study was to examine the phytochemical and nutritional analyses of polyherbal cough syrup that had been manufactured and was made with palmyra sweetener and a decoction that contained coarse dried herbs. Medicinal plants were collected, air-dried, and then used to prepare water and ethanol extracts. Among both extracts, water extract was selected for further study because it contained a high amount of components such as ash, protein, and sugar than ethanol extract. Three different polyherbal syrup was prepared with decoction by using sweeteners such as white sugar (TI), palmyrah jaggery (TII), and sugar candy (TIII). Physicochemical analysis of TII showed a low amount of water content [21.7($\pm$1.01)] (v/v %) and water activity (a_w) [0.627($\pm$0.001)]. In addition that it showed significantly higher amounts of nutritional content such as total ash [6.5($\pm$0.35)], total sugar [39.7($\pm$1.08)], crude protein [3.0($\pm$0.2)], and crude fat [0.9($\pm$0.1)] g/100g also mineral content as calcium [415.7($\pm$1.2)], magnesium [264.1($\pm$0.7)], sodium [595.45($\pm$4.8)], potassium [1747.2($\pm$11.7)], phosphorous [359.9($\pm$5.5)] and iron [141.4($\pm$1.1)] (mg/100g) when compared with other syrup as TI and TIII. Therefore cough syrup prepared with decoction and palmyrah jaggery was selected for the studies as stability and standardization.

Keywords: Polyherbal cough syrup, Decoction, Hot water, Palmyrah sweeteners.

1. Introduction

Cough is one of the symptoms of various respiratory disorders, including asthma, bronchitis, pneumonia, and others, about which there has been increasing global awareness in recent years. People are hesitant to use prescription medications of natural origin instead of synthetic ones because of the adverse effects. Additionally, Sri Lanka has certain naturally occurring medicinal herbs. The manufacture of polyherbal syrup made use of Palmyrah resources, including sweets like jaggery and sugar candy, due to their medicinal and curative properties as well as their health-improving ingredients.

In Sri Lanka, Ayurveda and Siddha are like medicine containing wealthy traditional constituents. These traditional schemes are more effective but do not have proper standardization. The development of these types of traditional medicines with safety, efficacy, and quality will help preserve the traditional heritage and give good motivation for the usage of accepted natural foodstuffs in healthcare (Soni et al., 2013). Since further analysis is also necessary to assess quality assurance, ensure uniformity of dosage, check stability, and detect adulterated or non-adulterated drugs and contaminants. Therefore, evaluating the quality of these raw materials is necessary. In addition, it is essential to maintain the stability and reproducibility of a particular extract to ensure fixed effectiveness through acceptable levels of active compounds. This procedure can be done using chemical or biological analysis (Vlietinck et al., 2009).

In Siddha medicine, there are different formulations of cough syrup. To the best of our knowledge, there are no previous studies done in cough syrup making with different Palmyrah sweetening agents. We hypothesize that the polyherbal syrup formulated using our country's natural medicinal plant materials and supplementary sweeteners from Palmyrah containing hypoglycemic effect is better for intake and cost-effective and more environmentally friendly for human health care. Therefore, the objective of this study was to compare the phytochemical and nutritional analysis of produced polyherbal cough syrup prepared with palmyra sweetener and decoction, which contained dried herbal coarse powders.

2. Methodology

2.1. Collection of plants

Patpadakam (*Mollugo cerviana*), milaku (*Piper nigrum*), Sukku (*Zingiber Officinale*), Thippili (*Piper longum*) and white sugar were purchased from a local market, and Adathodai (*Justicia adhatoda*), Vaddakkathari (*Solanum surattense*), paipudol (*Trichosanthes cucumerina*), Thuthuvalai (*Solanum trilobatum*), Iyanku (*Azima tetracantha*), Kandankathari (*Solanum xanthocarpum*), and Thulasi (*Ocimum tenuiflorum*) were collected from the local field in Jaffna kaithady area (located between 9°38'59.99" N Latitude, 81°09'60.00" E Longitude). Palmyrah Jaggery and sugar candy were obtained from the Chavachacheri Palm Development cooperative Society, Jaffna, Sri Lanka.

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2.2. Study Location

Plant extracts and decoction were prepared at Unit Siddha medicine, the University of Jaffna, and analysis of decoction and cough syrup was carried out at Palmyrah Research Institute.

2.3. Determination of moisture content

The course powder of each plant material was used to determine the water content using the hot air oven drying method (AOAC, 2000).

2.4. Method of preparation of extracts

The aqueous extract was prepared with dried powder mixed with (1:16 ratio) water, and the mixture was kept at 90oC until the total volume became one-eighth of the original volume. The mixture was cooled and filtered, and used for the analysis. Then ethanol extract was prepared with dried powder (50g) and extracted with 250 ml of 95% ethanol in soxhlet apparatus, evaporated, dissolved in distilled water, and used for the analysis.

2.5. Quality evaluation of plant extracts

Total ash and acid insoluble ash content were determined using a sample obtained from moisture determination by incineration and then burning in a muffle furnace at 550°C for 5 h. Cooled in a desiccator and weighed (AOAC, 2000). Ash and acid insoluble ash content were calculated and also Reducing sugar content (DNS method (Miller, 1959)), total sugar content (DNS method(Miller, 1959)), protein (BCA assay (Smith et al., 1985)), and total phenolic content (Folin-Ciocalteu assay (Maurya and Singh, 2010)) were determined for plant extracts.

2.6. Phytochemical screening

Aqueous and ethanol extracts of each plant were screened for the existence of phytochemical components by carrying out the standard methods such as Molisch's test, Fehling's test for reducing sugar, Ninhydrin test for protein, lead acetate test for flavonoids, foam test for saponins, ferric chloride test for phenols, Salkowski's test phytosterols, Mayer's test, and Wagner's test for Alkaloids.

2.7. Preparation of decoction

The eleven dried coarsely powdered plant materials were mixed with water in 1:16 ratio and the mixture was boiled until the total volume became one-eighth of the original volume (Kumar et al., 2019). The mixture was cooled and filtered. The filtrate decoction was taken to prepare the final poly herbal cough syrup.

2.8. Formulation of polyherbal cough syrup

Formulations of three polyherbal cough syrups were prepared by concentrating the decoction with different sweeteners such as white sugar, Palmyrah jaggery, and sugar candy (15%) until the internal temperature became 105^{o}C. Table 1 shows the different treatments of polyherbal syrup.

Table 1: Different treatments of polyherbal syrup

	Added Sweetener	Treatment
1	White sugar	TI
2	Palmyrah jaggery	TII
3	Sugar candy	TIII

2.9. Physicochemical analysis

pH: pH was measured by using a digital pH meter (Sension PH 31-Spain) at room temperature (28 $\pm$ 2 ºC) in a clean beaker containing (25 mL) cough syrup sample.

Water activity: A water activity meter (Novisina) was used to measure the water activity of the cough syrup sample at a temperature of 28 $\pm$ 2 ºC. The syrup sample was measured, and the sample was placed in the water activity meter holder. When the sample reading was stable, measurement was noted.

Total soluble solids: The total soluble solids as Brix of the decoction and syrup were analyzed using a refractometer at a temperature of 28 $\pm$ 2 ºC (Atago - smart 1 Germany).

Determination of acidity: The acidity of the decoction and syrup was determined as citric acid (%w/w) by titrating the sample against 0.1 N NaOH using phenolphthalein as an indicator and is expressed as citric acid (SLS: 729:1985 (Sri Lanka Standard, 1985)).

2.10. Nutritional analysis

The moisture content of the decoction and syrup was determined by the hot air oven drying method using 5g weight at 105°C until a constant weight was obtained. Then moisture content was determined. Ash content was determined by incineration of the dried sample obtained from moisture determination in a muffle furnace at 550°C for 5h. Crude protein content was calculated by converting the nitrogen content, determined by Kjeldahl's method (6.25xN). The fat extractions were performed by modification of the method of Bligh and Dyer as described by AOAC (2000) with petroleum ether (Boling point 40-60°C, 70mL) in the solvent extractor.

2.11. Mineral analysis

Sodium, potassium (flame photometry method), phosphorous spectrophotometry AOAC (2000), iron (1,10-phenanthroline method[9.), calcium, magnesium (EDTA titration method (Vogel et al., 1989)).

2.12. Statistical analysis

The results obtained from each treatment with three replicates were subjected to analysis of variance by com-

Table 3: Quantitative analysis of extracts

Plant Name	Extract	Total ash (g/100g)	Acid insoluble ash (g/100g)	Reducing sugar (g/100g)	Total sugar (g/100g)	Protein (mg/100g)	Total Phenolic Content (mg/100g)
Patpadakam	Water	1.42(±0.06)	0.25(±0.01)	1.48(±0.01)	1.90(±0.01)	29.1(±0.5)	3.40(±0.99)
	Ethanol	0.48(±0.01)	0.07(±0.01)	1.16(±0.01)	1.71(±0.01)	37.7(±0.6)	4.86(±0.01)
Milaku	Water	2.31(±0.29)	0.01(±0.01)	0.54(±0.01)	1.08(±0.02)	32.0(±0.4)	3.75(±0.06)
	Ethanol	0.38(±0.01)	0.00(±0.01)	0.05(±0.01)	0.05(±0.01)	5.1(±0.1)	0.56(±0.01)
Adathodai	Water	5.92(±0.03)	0.05(±0.01)	0.78(±0.01)	1.50(±0.01)	61.7(±0.2)	8.35(±0.02)
	Ethanol	3.12(±0.11)	0.07(±0.01)	0.42(±0.01)	0.92(±0.01)	10.8(±0.0)	2.11(±0.01)
Vaddakkathari	Water	3.36(±0.05)	0.18(±0.01)	0.53(±0.01)	0.59(±0.01)	27.0(±0.2)	3.42(±0.02)
	Ethanol	1.32(±0.06)	0.13(±0.01)	0.11(±0.01)	0.22(±0.02)	7.6(±0.4)	1.13(±0.01)
Paipudol	Water	2.46(±0.04)	0.15(±0.01)	0.24(±0.01)	0.46(±0.01)	11.0(±0.1)	1.56(±0.01)
	Ethanol	0.36(±0.02)	0.01(±0.01)	0.11(±0.01)	0.13(±0.01)	10.3(±0.1)	0.31(±0.01)
Sukku	Water	3.51(±0.06)	0.11(±0.01)	1.23(±0.01)	0.98(±0.01)	36.2(±0.8)	3.29(±0.04)
	Ethanol	0.37(±0.01)	0.02(±0.01)	1.63(±0.01)	1.55(±0.01)	64.5(±0.5)	4.88(±0.15)
Thuthuvalai	Water	5.28(±0.02)	0.31(±0.01)	2.18(±0.05)	2.93(±0.03)	49.1(±0.5)	4.84(±0.08)
	Ethanol	3.27(±0.01)	0.25(±0.01)	2.22(±0.63)	3.72(±0.01)	41.0(±0.5)	3.87(±0.02)
Iyanku	Water	7.10(±0.09)	0.05(±0.01)	0.16(±0.01)	0.76(±0.04)	19.4(±0.5)	2.27(±0.1)
	Ethanol	4.71(±0.03)	0.13(±0.01)	1.22(±0.03)	1.30(±0.02)	15.0(±0.5)	1.28(±0.02)
Kandankathari	Water	3.32(±0.04)	0.27(±0.01)	0.32(±0.01)	0.59(±0.02)	24.0(±0.5)	2.69(±0.02)
	Ethanol	2.61(±0.03)	0.23(±0.01)	0.06(±0.01)	0.57(±0.01)	7.8(±0.1)	0.82(±0.01)
Thulai	Water	3.75(±0.08)	0.14(±0.01)	1.53(±0.03)	1.65(±0.02)	50.1(±0.1)	3.27(±0.04)
	Ethanol	1.49(±0.01)	0.09(±0.01)	0.91(±0.01)	0.76(±0.01)	46.2(±0.6)	3.52(±0.02)
Thippili	Water	2.75(±0.07)	0.18(±0.01)	2.67(±0.01)	3.88(±0.01)	46.7(±0.1)	3.98(±0.01)
	Ehanol	0.08(±0.01)	0.03(±0.01)	0.04(±0.01)	0.13(±0.02)	2.5(±0.1)	0.02(±0.01)

plete randomized design (CRD). The significant difference among the treatment was tested in the least Significant Difference (LSD) at a 5% level using Minitab 19 software.

3. Results and discussions

Collected plant materials were prepared to free from soil and other contaminants and preserved by using shade drying that utilizes solar energy as a heating source. Plant materials were kept under shade in a room with good ventilation and without direct sunlight exposure. Table 2 shows the moisture content of plant materials.

Table 2: Moisture content

No.	Plant Name	Loss on drying (%)
1	Patpadakam	10.4(±0.10)
2	Milaku	11.3(±0.03)
3	Adathodai	13.7(±0.10)
4	Vaddakkathari	12.3(±0.07)
5	Paipudol	12.2(±0.08)
6	Sukku	11.4(±0.10)
7	Thuthuvalai	13.6(±0.08)
8	Iyanku	15.4(±0.08)
9	Kandankathari	10.3(±0.10)
10	Thulai	13.1(±0.07)
11	Thippili	12.0(±0.09)

Sharma, Chen, and Lan (Sharma et al., 2009) reported that, during shade-drying practice, solar energy is used for heating the ventilated air before exposure to the plant materials. The cost of the energy required for the drying process influences the cost and quality of the product; therefore shade drying method was selected. Nowadays, shade drying is still practiced due to its low cost and high-quality dehydrated products in rural areas or small businesses (Janjai and Bala, 2012). The dried plant material was ground to coarse powder with the help of a motor and pestle.

3.1. Determination of moisture content of plant materials

Total ash value helps us to determine the mineral content of the extracts; among both extracts, the water extract of all plants (see Table 3) showed higher ash content than the ethanol extract, besides indicating the presence of impurities and adulteration in the plant samples (Jarald, 2007). Acid insoluble ash indicates that plant extract was contaminated with sand materials such as silicon (Mukherjee, 2003).

All plants' reducing and total sugar content showed a higher value for water extract than ethanol extract except sukku thuthuvalai and iyanku. Likewise, water extracts of all plant extracts showed higher amounts of protein and total phenol content when compared with ethanol extract, except patpadakam and sukku.

Phytochemical screening

Therapeutic activities vary due to the phytochemicals or plant chemicals present in the plant material (Omoregie and Osagie, 2012) and responsible for their defensive health activities such as antioxidants (polyphenols), antibacterial (saponins), and anti-cancer

Table 4: Phytochemical analysis of extracts

No.	Plant Name	Extract	Test for Phenols	Test for Tannins	Test for Flavonoids	Test for Saponins	Test for Ninhydrin	Test for Carbohydrate	Test for Reducing Sugar	Test for Alkaloid Mayer's	Test for Alkaloid Wagner's	Test for Phytol sterols
1	Patpadakam	Water	*	*	-	*	*	*	*	-	-	*
		Ethanol	*	*	-	*	*	*	*	-	*	*
2	Milaku	Water	*	*	-	-	*	*	*	-	*	*
		Ethanol	*	*	-	-	-	*	*	*	*	*
3	Adathodai	Water	*	*	-	-	*	*	*	-	-	*
		Ethanol	*	*	-	-	*	*	*	-	*	*
4	Vaddakkathari	Water	*	*	-	-	*	*	*	-	-	*
		Ethanol	*	*	-	*	-	*	*	-	-	*
5	Paipudol	Water	*	*	-	-	*	*	*	-	-	*
		Ethanol	*	*	-	*	-	*	*	-	-	*
6	Sukku	Water	*	*	-	*	*	*	*	-	-	*
		Ethanol	*	*	-	*	*	*	*	*	*	*
7	Thuthuvalai	Water	*	*	-	-	*	*	*	*	*	*
		Ethanol	*	*	-	-	*	*	*	*	*	*
8	Iyanku	Water	-	-	-	-	*	*	*	-	-	-
		Ethanol	-	-	-	-	*	*	*	-	-	-
9	Kandankathari	Water	*	*	-	*	*	*	*	*	*	*
		Ethanol	-	-	*	-	-	*	*	*	*	*
10	Thulai	Water	*	*	-	*	*	*	*	*	-	*
		Ethanol	*	*	-	-	-	*	*	*	-	-
11	Thippili	Water	*	*	-	*	*	*	*	*	*	*
		Ethanol	-	-	-	-	-	*	*	*	*	-

*Present, ¯Absent

protect cells against free radical damage (Dua et al., 2013), anti-periodic, anti-spasmodic, anti-inflammatory, anti-arthritic, anti-allergic, and anti-diabetic properties (Chopra et al., 1956; Chopra et al., 1994; Zhao et al.,1991).

3.2. Qualitative analysis of extracts

Phytochemical analysis showed the existence of pharmaceutically bioactive components in all the plants. Water extracts of all plants were positive for ninhydrin test but not presented in ethanol extract of milaku, vaddakkathari, paipudol, kandankathathari, thulasi and thippili. Both extracts of all the plant extracts were detected for carbohydrate and reducing sugars. Both water and ethanol extracts of patpadakam and sukku showed positive results for saponin test that of negative for milaku, adathodai, thuthuvalia, Iyanku and thippli. Alkaloids were not detected in both extracts of vaddakkathari, iyanku and paipudol, while milaku, thuthuvalai, kandankaththari, thulasi and thippili showed positive for both extracts (see Table 4).

The eleven dried raw materials were coarsely powdered. The dried powder and water were mixed with a 1: 16 ratio and boiled until the total volume became one-eighth of the original volume. The mixture was cooled and filtered. The decoction used for the syrup formulation was evaluated for their physicochemical parameters to ensure that they meet the standards (Sri Lanka Standard, 1985). Consequently, which was used for the determination of pH, total soluble solids, total acidity, total ash, total sugar, and water content (see Table 5).

Table 5: Evaluation of decoction

No.	Parameter	Results
1	Water content (v/v%)	95.89(±0.04)
2	Water activity (aw)	0.743(±0.001)
3	pH	6.81(±0.07)
4	Total soluble solids (oBrix)	3.95(±0.15)
5	Acidity as acetic acid (v/w %)	0.022(±0.003)
6	Protein (g/100g)	0.55(±0.01)
7	Total sugar (g/100g)	3.5(±0.01)
8	Total ash (g/100g)	1.34(±0.11)

Table 6: Physicochemical analysis

No.	Parameter	**Treatment I** (TI)	**Treatment II** (TII)	**Treatment III** (TII)
1	Water content (w/w)	$21.3(\pm1.36)^{a}$	$21.7(\pm1.01)^{ab}$	$23.5(\pm0.97)^{b}$
2	Water activity (aw)	$0.631(\pm0.001)^{b}$	$0.627(\pm0.001)^{c}$	$0.634(\pm0.001)^{a}$
3	pH	$6.8(\pm0.04)^{a}$	$6.6(\pm0.02)^{b}$	$6.7(\pm0.05)^{a}$
4	Total soluble solids (obrix)	$67.3(\pm0.8)^{a}$	$66.3(\pm0.7)^{a}$	$66.5(\pm1.0)^{a}$
5	Acidity as acetic acid (v/w)	$0.13(\pm0.04)^{a}$	$0.14(\pm0.04)^{a}$	$0.12(\pm0.02)^{a}$

Each values in the table is represented as mean± SD (n=3). Values in the same raw followed by different letter (a - c) are significantly different ($p < 0.05$).

Table 7: Nutritional analysis

No.	**Parameter** (g/100g)	**Treatment** I (TI)	**Treatment II** (TII)	**Treatment III** (TII)
1	Total ash	$5.0(\pm0.56)^{b}$	$6.5(\pm0.35)^{a}$	$5.1(\pm0.54)^{b}$
2	Total sugar	$38.2(\pm1.07)^{b}$	$39.7(\pm1.08)^{a}$	$39.2(\pm1.0)^{ab}$
3	Crude protein	$2.7(\pm0.1)^{b}$	$3.0(\pm0.2)^{a}$	$2.8(\pm0.1)^{b}$
4	Crude fat	$0.6(\pm0.1)^{b}$	$0.9(\pm0.1)^{a}$	$0.4(\pm0.1)^{c}$

Each values in the table is represented as mean± SD (n=3). Values in the same raw followed by different letter (a - c) are significantly different ($p < 0.05$).

Table 8: Mineral analysis

No.	**Minerals** (mg/100g)	**Treatment I** (TI)	**Treatment II** (TII)	**Treatment III** (TII)
1	Calcium	$306.3(\pm1.2)^{a}$	$415.7(\pm1.2)^{a}$	$414.2(\pm1.2)^{b}$
2	Magnesium	$250.2(\pm0.7)^{b}$	$264.1(\pm0.7)^{a}$	$230.9(\pm0.6)^{c}$
3	Sodium	$341.7(\pm1.9)^{b}$	$595.45(\pm4.8)^{a}$	$339.5(\pm2.1)^{b}$
4	Potassium	$1406.9(\pm2.5)^{b}$	$1747.2(\pm11.7)^{a}$	$1412.3(\pm5.5)^{b}$
5	Phosphorous	$269.7(\pm3.5)^{b}$	$359.9(\pm5.5)^{a}$	$245.8(\pm3.5)^{c}$
6	Iron	$135.8(\pm3.1)^{b}$	$141.4(\pm1.1)^{a}$	$144.4(\pm1.5)^{a}$

Each values in the table is represented as mean± SD (n=3). Values in the same raw followed by different letter (a - c) are significantly different ($p < 0.05$).

3.3. Analysis of polyherbal cough syrup

The analytical techniques used for the physicochemical evaluation of polyherbal syrup formulations allow the manufacturers to regular quality standards and specifications of the products; thereby, they can easily find marketing approval from regulatory authorities for therapeutic efficiency, safety, and shelf-life for their herbal products. The water content in the syrup affects the uniformity of the cough syrup, which is significantly higher for the white sugar (TI) added syrup than sugar candy added syrup (TIII). There were no significant differences with jaggery syrup (TII). Water activity was significantly higher in sugar candy-added syrup than in sugar-added syrup. While that was less for jaggery-added syrup, which means water which is favorable for the microorganism, was low in TII, and also, which treatment showed a significantly lower pH value. There were no significant differences between all syrup in total soluble solids and total acidity (see Table 6). Plant materials used for the decoction preparation contained nutritional characteristics such as sugar, protein, ash, and others. The cough syrup TII showed the highest amount significantly in total ash [6.5(±0.35)], total sugar [39.7(±1.08)], crude protein [3.0(±0.2)], and crude fat [0.9(±0.1)] when compared with TIII and TI. In comparison, there were no significant differences between TII and TI in total ash, total sugar, and crude protein (see Table 7). The mineral content of the diets is more important to regulate enzymes and hormones. However, all the analyzed minerals, such as calcium, magnesium, sodium, potassium, phosphorous, and iron, showed significantly higher amounts in TII than in TIII and TI. Calcium and phosphorous were significantly higher in TI compared with TII, while there were no significant differences in sodium and potassium. There were no significant differences in iron content between TII and TIII, while that of lower in TI (see Table 8).

4. Conclusion

Polyherbal cough syrups were formulated with decoction, prepared with hot water extraction, and made with three different sweeteners: white sugar, palmyrah jaggery, and sugar candy. Among the syrup, palmyrah jaggery added syrup showed significantly higher amounts of nutritional characteristics such as ash, sugars, protein, and fat, which contained more minerals such as calcium, magnesium, sodium, potassium, phosphorous, and iron. Therefore jaggery-added cough syrup (TII) was selected for the stability study.

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