Proximate and Nutrient Analysis of the Locally Manufactured Herbal Medicines and its Raw Material

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Abstract: Herbal medicines have unique therapeutic properties and therefore, used in rural areas to cure different diseases. Proximate analysis and elemental composition of the locally manufactured formulations from *Hypericum perforatum, Allium sativum, Zingiber officinalis and Valeriana officinalis* were carried out. The heavy metals including Cu, Ni, Zn, Pb, Co, Cd, Fe, and Cr were determined using Atomic Absorption Spectroscopic standard method. Na and Ca was estimated using flame emission spectrophotometer. *Z. officinalis* has highest percentage of carbohydrate, fats, fiber and energy values while in herbal formulations St. John Wort has the highest. In case of micro analysis, St. John Wort Capsules has highest concentrations of Zn while Valerian has highest concentration of Cu, Co, Cd and Fe compared to others, while in medicinal plant species, the content of Cu, Zn, Co and Fe was highest in *V. officinalis.* The level of macronutrients (Ca and Na) was highest in St. John Wort Capsule, *H. perforatum* and *V. officinalis*. However, the concentration of these nutrients in both the medicinal plants and herbal formations were in the optimum level of WHO standards. . [Journal of American Science 2010;6(5):91-96]. (ISSN: 1545-1003).

Keywords: Proximate analysis, herbal formulations, nutrient analysis, Pakistan

1. Introduction

The use of traditional medicines is increasing and getting popularity throughout the developed and developing world (Jia and Zhang, 2005). Herbal medicines as finished labeled medicinal product that contain active ingredients, aerial or underground parts of the plant or other plant material or combinations (Chaudhari, 1996; and Ritch, 2000). About 80% of the people in developing countries relay on traditional medicine for their primary health care (Latif et al., 2004). The worth of herbal product industry is approximately US$ 300 million compared to modern drugs that is US$ 2.5 billions while in recent year it has been gained considerable momentum (Shinwari and Shoukat., 2003; Shinwari et al., 2003; and Shinwari et al., 2006).

Proximate and nutrient analysis of edible fruit and vegetables plays a crucial role in assessing their nutritional significance (Pandey et al., 2006). As various medicinal plant species are also used as food along with their medicinal benefits, evaluating their nutritional significance can help to understand the worth of these plant species (Pandey et al., 2006). As far herbal drug's standardization is concerned, WHO also emphasize on the need and importance of determining proximate and micronutrients analysis. Such herbal formulations must pass through standardization processes (Niranjan and Kanaki, 2008).

Hypericum Perforatum, Valaneria officinalis, Zinigiber officinalis and *Allium sativum,* are the important plant species used in preparation of herbal formulations (st. john wort, garlicare tablet, ginger

capsule, and valerian capsule). The extract of aerial parts of *H. perforatum* is usually gathered during the flowering season or shortly before and has been used in herbal medicines (Barnes et al., 2001). *Hypericum* extracts are marketed not only as herbal medicine but also in the form of dietary supplements (Shinwari et al., 2006). Garlic or *Allium sativum* is medicinally important plant species i.e. helpful in the treatment and prevention of a number of diseases e.g. cancer, coronary heart diseases, obesity, hypercholesterolemia, hypertension and gastrointestinal (Capasso et al., 2003). *Valeriana officinalis* have a broad range of applications such as a tranquillizer for people with hyper-excitability and as a smooth-muscle relaxing agent to treat stomach and intestine cramp (Leoeniewicz et al., 2006). Valerian is also a component of many herbal mixtures, which are widely used to treat sleeping disorders (Bent et al., 2006). Ginger (*Zinigiber officinale*), an important constituent of many herbal formulations, is carminative, pungent, stimulant, used widely for indigestion, stomach ache, malaria and fevers. It is said to be used for abdominal pain, chest congestion, chronic bronchitis, colic and vomiting (Jatoi, et al., 2007). Although, these formulations are providing beneficial effects but without any knowledge of their inorganic constituents.

In the present study, the herbal formulations manufactured by Qarshi Ind. Pvt. Ltd. Pakistan and their raw materials were taken for investigation from the north western part of the NWFP, Pakistan. All these selected plant species based formulations have well documented for their phytochemicals and biological significance but no informations the proximate and elemental data. Keeping in view the importance of the inorganic constituents of the herbal medicines their proximate and elemental analysis were undertaken.

2 Material and Methods

2.1 Sampling

The medicinal plant species were collected from various areas of NWFP Pakistan. The collected plants were packed in the Kraft paper and herbarium sheets were prepared. These plants were identified by a plant taxonomist of Botany Department, Kohat University of Science and Technology, Kohat. The herbal formulations were provided by Qarshi Industries, Pakistan.

2.2 Proximate Analysis

Proximate analysis including moisture, ash, crude fiber, fats, carbohydrates and proteins were determined of both formulations and their respective raw material using AOAC (1990). The moisture contents was determined by oven dehydration method at 105 ^{0}C for 5hr using MC (%) = $\frac{W_0}{W_i}\times 100$ formula. Total ash was determined by weighing the furnace incinerated residue at 550 ^{0}C for 12hrs. The formula for calculating the ash in percent is $\frac{M_a}{M_s}\times 100$. Crude fats were determined using petroleum ether as extracting solvent in soxhlet apparatus. The percentage crude fats were calculated by $CF\ (\%)=\frac{M_{ex}}{M_s}\times 100$. The crude fibers of the samples were estimated by treating moisture and fats free material with dilute acidic solution followed by dilute base particularly NaOH. After base treatment the residue was filtered and washed with hot water and then ignited. The loss in weight was calculated from the ash left after incineration in the furnace by $\frac{W_2-W_3}{W_1}\times 100$. The crude protein was determined using micro Kjeldahl method. Percentage carbohydrate was calculated by 100 – (percentage of ash + percentage of moisture + percentage of fat + percentage of protein). All these methods were adopted, with little modification, from AOAC (1990) and Awan & Salim (1997).

2.3 Elemental Analysis

The samples were digested by a mixture of concentrated nitric acid and perchloric acid mixed in 1:1 v/v ratio. The heavy metals including Cu, Ni, Zn, Pb, Co, Cd, Fe, and Cr were determined using Atomic Absorption Spectroscopic standard method. Na and Ca was estimated using flame emission spectrophotometer.

3. Results and Discussions

3.1 Weights of the products

The weight variation among 10 tablets of the selected four herbal formulations is presented in Table 1. The data reveals that there was large difference from tablet to tablet as indicated by their corresponding standard deviation but comparatively ginger tablets shows less variation.

Table 1. Variation in the average weight of a single

S.No.	Product name	Weight
1	St. John Wort	424.89 ± 1.93
2	GarliCare Tablet	573.84 ±1.98
3	Ginger Capsule	625.85 ± 1.74
4	Valerian Capsule	426.13±1.23

± = standard deviation; Means of ten Tables

3.2 Analysis of herbal product

The proximate data for all formulations is tabulated in Table 2 and their respective raw material in the Table 3. The moisture contents was noted highest in Ginger and Valerian capsules i.e. 7.26±0.09 and 7.14±0.05% respectively. The fats contents were found highest in St. John Wort and Valerian tablets i.e. 4.98±0.01 and 4.0±0.03 respectively (Table 2). Looking at the results of carbohydrates, it was highest in St. John Wort Capsule, and (79.37±0.09 and 76.41±0.25). Comparing the crud fiber, it was higher in St. John Wort Capsule and Valerian Tablets (14.33±0.24 and 18.60±0.22 respectively). The protein contents of St. John Wort Capsule and Ginger Tablets was 6.66±0.04 and 8.60±1.0 respectively. The details of other proximate parameter are given Table 2.

The moisture, fats, crude protein, carbohydrates and ash concentrations of the ginger tablets are comparable (6.9%, 8.6%, 6.4%, 72.40% and 5.7%) to the value reported in the Encyclopedia of Chemical Technology (1980).

The data reveals that the formulations based on the selected raw materials are not the rich sources of lipid and can be administered to patients in whom high fats contents is a risk factor. The level of dietary fibre is low in garlicare tablets and high in valerian capsule. This trend is persistent in its respective raw materials, generally the fiber content are high as compared to other plant leaves and seeds in the cited literature (Elegbede, 1998). Although high fiber contents increases digestibility, but on the other hand high level of fibers in the diet can produce intestinal irritation, which ultimately decrease nutrients utilization (Oyenuga and Fetuga, 1975). It can be deduced that the fiber contents of these formulation are mild in concentration and have more beneficial effects rather than hazardous. The ash contents of St. John Wort capsule and Garlicare tablets are comparatively low. It is evident that these formulations besides its targeted objectives offer good source of carbohydrates, and can produce energy, thus meeting the increased demand of energy especially in aged population

3.3 Medicinal plant species

The moisture contents of the *Allium sativum* was found 67.66±0.18 and *Valeriana officinalis* was 6.82±0.09. The fats contents of the *Allium sativum* and *Valeriana officinalis* was found 2.43±0.11 and 4.14±0.09% respectively. The carbohydrate contents of the *Allium sativum* and *Valeriana officinalis* was found 14.98±0.06 and 67.52±0.07 respectively. The fibre contents of the *Allium sativum* was found 2.43±0.07 and *Valeriana officinalis* was 16.78±0.09. The ash contents of the *Allium sativum* and *Valeriana officinalis* was 1.73±0.16 and 17.10±0.08 respectively. The calorific value of the *Allium sativum* and *Valeriana officinalis* was estimated 134.60±0.65 and 324.95±0.52 respectively.

It is essential to quantify the level of toxic trace elements in medicines directly derived from the herbal source and used as it is, due to its deleterious effects upon human health. All the four formulations were analyzed for heavy metal and essential mineral contents. The concentration of Cu, Ni, Zn, Pb, Co, Cd, Fe, and Cr in St. John Wort was 25.4, <0.006, 78.2, <0.015, 2.6, <0.0008, 1020.4 and <0.003 ppm respectively. The Ca and Na in St. John Wort was 192 and 14.84 ppm respectively.

3.4 Nutrient Analysis

3.4.1 Micro-Nutrient in Herbal Products

The amount of Cu, Ni, Zn, Pb, Co, Cd, Fe, and Cr in Garlicare tablets were 12.2, 5.8, 38.2, <0.015, <0.009, <0.0008, 142 and <0.003 ppm respectively. The amount of mineral including Ca and Na in the Garlicare tablets is 64.77 and 7.78 ppm respectively. The quantity of Cu, Ni, Zn, Pb, Co, Cd, Fe, and Cr in Ginger Capsules were 19, 9.4, 52.6, 13.6, <0.009, 0.0008, 226.8 and <0.003 ppm respectively. The quantity of Na and Ca in the Ginger capsule are 5.88 and 74.62 ppm respectively. The concentration of Cu, Ni, Zn, Pb, Co, Cd, Fe, and Cr in Valerian Capsule were 58, <0.006, 55, <0.015, 9.8, 0.2, 1681.8, and <0.003 ppm respectively.

In all four samples the chromium (Cr) concentration was very low, while the Cd concentration is also low except in valerian capsule with value of 0.2 ppm. Cadmium concentration of 0.2 ppm is also not in the limits set by WHO because the daily intake of 0.06–0.07 mg/day is permissible (FAO, 1993). The level of lead (Pb) is also below the permissible level in St. John wort capsule, garlicare and valerian tablets but higher in the ginger tablets with recorded value of 13.6 ppm. This is exceptionally high because the WHO acceptable daily intake of Pb

for adults was 0.21-0.25 mg/day (FAO, 1993). It is well reported in the literature that Pb has pronounced ill effects on the central nervous system especially in children (FAO, 1993).

The range of Ni obtained in this study was lower than 0.05–5 mg/kg reported for plant foods by the National Academy of Sciences in case St. John wort and valerian tablet where it is <0.006; in case of

Table 2. Proximate Analytical Data of the Selected Herbal formulations (in percentage)

Product Name	Moisture	Crude Fat	Carbohydrate	Fibre	Protein	Ash	Energy Value
St. John Wort	4.11±0.09	4.98±0.01	79.37±0.09	14.33±0.24	6.66±0.04	4.89±0.05	389.30±0.68
GarliCare Tablet	2.75±0.12	3.53±0.23	70.24±0.17	1.07±0.03	4.34±0.09	19.15±0.18	330.10±2.2
Ginger Capsule	7.26±0.09	3.60±0.13	76.41±0.25	4.24±0.12	8.60±1.0	4.13±0.06	372.40±0.84
Valerian Capsule	7.14±0.05	4.00±0.03	69.46±0.13	18.60±0.22	5.14±0.02	14.27±0.15	334.39±0.76

Table 3. Proximate Analytical Data of the Raw material used in the selected Medicines

Species Name	Moisture	Fats	Carbohydrate	Fibre	Proteins	Ash	Energy Value
H. perforatum	8.31±0.06	5.06±0.08	72.2±0.09	13.0±0.00	9.54±0.16	4.54±0.014	374.09±0.06
A. sativum	67.66±0.18	2.43±0.11	14.98±0.06	2.43±0.07	13.2±0.04	1.73±0.16	134.60±0.65
Z. officinalis	9.21±0.07	7.30±0.07	72.36±0.04	16.36±0.03	7.27±0.07	4.83±0.07	380.3±0.06
V. officinalis	6.82±0.09	4.14±0.09	67.52±0.07	16.78±0.09	4.39±0.09	17.10±0.08	324.95±0.52

Table 4. Nutrient concentration (ppm) of the preparations

Product Name	Cu	Ni	Zn	Pb	Co	Cd	Fe	Cr
St. John Wort Capsules	25.4	<0.006	78.2	<0.015	2.6	<0.0008	1020.4	<0.003
GarliCare Tablet	12.2	5.8	38.2	<0.015	<0.009	<0.0008	142	<0.003
Ginger Tablet	19	9.4	52.6	13.6	<0.009	<0.0008	226.8	<0.003
Valerian Tablet	58	<0.006	55	<0.015	9.8	0.2	1681.8	<0.003

garlicare it not very high but the value in terms of ginger tablets is very alarming of 9.4 ppm (WHO, 1998; and Pizzaro et al., 1999). Globally the dietary intake of copper (Cu) in healthy non occupational exposed population vary between 0.9-2.2 mg/day (Pizzaro et al., 1999).

Table 5. Mineral contents (ppm) of the Preparation

Specie Name	Cu	Ni	Zn	Pb	Co	Cd	Fe	Cr
H. perforatum	25.4	<0.006	78.2	<0.015	2.6	<0.0008	1020.4	<0.003
A. sativum	14.4	<0.006	52.8	<0.015	<0.009	1.4	165.8	<0.003
Z. officinalis	36.8	<0.006	54.4	5.6	2	<0.0008	518.2	<0.003
V. officinalis	57.4	48.4	177.2	13.6	19	<0.0008	26690	<0.003

The level of copper in all formulations is very high than the acceptable range set by WHO of 2-5 mg intake per day (WHO, 1998; Cantilli, et al., 1994). It has been reported that Cu consumption in excess of 3 ppm of drinking water result in nausea and other adverse effects on the gastrointestinal tract (GIT) (Pizzaro, et al., 1999). The levels of Zn found in the formulations are also not in line with the WHO values of 2-5 mg intake per day. There is no documented evidence of adverse health effects from the intake of Zn normally found in various diets consumed world wide. But chronic zinc ingestion i.e. 300mg/day for six weeks cause suppression of the immune system and decrease in high density lipoproteins (Cantilli et al., 1994).

3.4.2 Macro-Nutrients in Herbal Drugs

The mineral contents comprising sodium and Calcium found in the valerian capsule was 8.942 and 67.1ppm respectively. The recommended daily intake level of iron (Fe) outlined by WHO is about 10-30 mg/day (Cantilli et al., 1994). Keeping in view the recommended level, all of the samples shows high iron contents but comparatively in garlicare and ginger tablets is low, although not very low to be recommended for consumption. The acute chronic dose of iron in infants has been estimated to approximately 20mg/kg and the lethal dose of about 200-300 mg/kg body weight. The chronic iron overloads result in hepatomegaly, cardiac disease and liver chirrosis (Weber, 1988). The level of Ca lay down by WHO is 450-1200 mg/day, which in agreement to the one found in formulations as indicated in the Table 5.

Table 6. Micro Nutrients in Medicinal Plants (ppm)

Product Name	Ca	Na
St. John Wort Capsule	192	14.84
GarliCare Tablet	64.77	7.78
Ginger Tablet	74.62	5.88
Valerian Tablet	67.1	8.942

The highest concentration of micronutrients e.g Cu, Ni, Zn, Pb, Co and Fe were found in *V. officinalis* followed by *H. perforatum* as shown in the Table. 6.

Table 7. Macro Nutrients in medicinal plants (ppm)

Specie Name	Ca	Na
H. perforatum	192	14.84
A. sativum	40.31	8.44
Z. officinalis	11.96	9.33
V. officinalis	176.9	16.32

The Ca concentration in *H. perforatum* was found the highest among all the medicinal plants having 192 ppm concentration followed by *V. officinalis*, *A. sativum*, and *Z. officinalis* has the concentration of 176.9, 40.31 and 11.96 ppm (Table 7). In case of Na, *V. officinalis* has the highest concentration of 16.32 ppm followed by *H. perforatum*, which has a concentration of 14.84 ppm (Table 7).

The findings of the proximate contents of most of the species analyzed in present study were almost complying with the previous reports (Odhav et al., 2007; and Odebunmi et al., 2009). The difference in the analysis might be attributed to the conditions on which the plant species is harvested along with environmental parameters (Nordeide et al., 1996).

Acknowledgements

The authors wish to thank the Higher Education Commission, Government of Pakistan, for providing financial support for the current study under the National Research Program for Universities (NRPU).

References

1. Jia W, Zhang L. Challenges and Opportunities in the Chinese Herbal Drug Industry. 2005; Pp 229-250
2. Chaudhari RD. Herbal drug industry, Eastern Publisher, New Delhi.1996; 1, 498-499.

3. Ritch R. tential role for Ginkgo biloba extract in the treatment of glaucoma, Med. Hypothesis 2000; 54 (2): 221-235
4. Latif A, Ahmad H, Begum S, Hussian SK, Adnan M. Medicinal and other economic plants as substitute to forest logging in Miandam and Sulatanr Valley-Swat. Proc. of international workshop on conservation and sustainable use of medicinal and aromatic plants in Pakistan. WWF-Pakistan. 2004.
5. Shinwari ZK, Shoukat M. Mid-Term Consultancy Report for NTFPs of Palas Valley. PCDP, Kohistan 2003.
6. Shinwari ZK, Watanabe T, Rehman M, Youshikawa T. A pictorial guide to Medicinal Plants of Pakistan. Kohat University of Science & Technology, Pakistan. 2006; p. 247
7. Niranjan RM, Kanaki S. Phytochemical Standardization of Herbal Drugs and Polyherbal Formulations. Bioactive Molecules and Medicinal Plants. 2008; Pp. 349-369.
8. Pandey M, Abidi AB, Singh S, Singh RP. Nutritional Evaluation of Leafy Vegetable Paratha *J. Hum. Ecol* 2006; 19 (2): 155-156.
9. Barnes J, Anderson LA, Phillipson JD. *J. Pharm Pharmacol* 2001; 53: 583-600.
10. Capasso F, Gaginella TS, Grandolini G,Izzo AA. A Quick Reference of Herbal Medicine, Springer-Verlag, *Phytotherapy* 2003.
11. Leoeniewicz A, Jaworska K and yrnicki W. Macro- and micronutrients and their bioavailability in Polish herbal medicaments. *Food Chemistry* 2006; 99 (4): 670-679.
12. Bent S, Padula A, Moore D, Patterson M and Mehling W. Valerian for sleep: a systematic review and metaanalysis. *Am. J. Medicine* 2006; 119 (12): 1005-1012.
13. Jatoi SA, Kikuchi A, Gilani SA and Watanabe KN . Phytochemical, Pharmacological and Ethnobotanical studies in Mango Ginger (Curcuma amada Roxb; Zingiberaceae). Phytotherapy Research 2007; 21: 507-516.
14. AOAC. *Official Methods of Analysis.* 15th Edn. Association of Official Analytical Chemists Washington, DC, USA. 1990
15. Awan JA, Salim UR. Food analysis manual, Vet Ag publication 1997; 5: 2-7.
16. Elegbede JA, Legumes. In Nutritional quality of plant foods. Osagie AU, Eka OU (Eds). Post Harvest Research Unit, University of Benin 1998; pp. 53-83.
17. Oyenuga VA, Fetuga BL. Some aspects of the biochemistry and nutritive value of the watermelon seed (Citrullus vulgaris schrad) *J. Sci. Food Agric* 1975; 26: 843-846.
18. FAO/WHO. Evaluation of Certain Food Additives and Contaminants. WHO Technical Report series 837. Geneva: FAO/WHO 1993.
19. WHO. Quality control methods for medicinal plant materials. World Health Organization, Geneva. ISBN 92 4 154510 0 (QV 766) 1998.
20. Pizzaro F, Olivares M, Uauy R, Contreras P, Rebelo A, and Gidi V. Environ. Health Perspect 1999; 107 (2): 117-121.
21. Cantilli, R, Abernathy CO, Donohue JM, Mertz W, Olin S. Risk Assessment of essential trace elements. Washington, DC. ILSI Press 1994; Pp. 113-125.
22. Weber G. HPLC with electrochemical detection of metalflavonoid-complexes isolated from food. Chromatographia 1988; 26: 133–138.
23. Odhav B, Beekrum S, Akula U, and Baijnath H. Preliminary assessment of nutritional value of traditional leafy vegetables in KwaZulu-Natal, South Africa. *J. Food Comp. Anal* 2007; 20: 430-435.
24. Odebunmi, EO, Oluwaniyi OO, Awolola GV and Adediji OD. Proximate and nutritional Composition of kola nut (*Colanitida*), bitter cola (*Garcinia cola*) and alligator pepper (*Afromomum melegueta*). *Af. J. Biotech.* 2009; 8 (2): 308-310.
a. Nordeide, MB, Hatloy A, Folling M, Lied E, and Oshaug A. Nutrient composition and nutritional importance of green leaves and wild food resources in an agricultural district, Koutiala, in southern Mali. *Int. J. Food Sci. Nut.* 1996; 47: 455-478.

12/1/2009