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THE DETERMINANT PRINCIPLES OF TASTE OF TERMINALIA CHEBULA RETZ FRUITS - INDIAN TRADITIONAL AYURVEDIC AND SIDDHA CONCEPTS

Ayurveda

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ABSTRACT

The organoleptic character is a determinant of acceptance of medicinal formulations by consumers. Though perception for every individual varies, the motives and health concern are consumer back ground factors for liking. But maintanence of raw drug's desired features in final product is bound to the quality assurance (Good manufacturing practises). Present study categorised phytochemicals responsible for part based taste of Indian *Terminalia chebula* fruits (traditional Ayurvedic and Siddha principles) and discussed influencing factors. If with this accumulating strategy the theoretical biosynthetic path is fixed: marker for maturity stages can be identified; degree of deviation from the wide range of variation in marker or standardization parameters due to influencing factors for the highly susceptible TCF can be predicted which are useful to pass the drug during analytical authentication, for quality control.

KEYWORDS

organoleptic taste, phytochemical marker, Terminalia chebula, Quality

INTRODUCTION:

The organoleptic character (OLC) stands as the first descriptive parameter in the standardisation of both Crude drugs and final products[1]. Taste is the active principle for Indian traditional systems. The subqualities of taste like tactile perceptions flavour add strength to the assessment. *Terminalia chebula* Retz is used in ASU&H (Ayurveda, Siddha Unani & Homeopathy) systems of Indian medicine and Homoeopathy. In the systems of Ayurveda (Bhavaprakasha nighantu of Bhavamishra) and Siddha (Materia medica) the taste is specific to TCF parts and 6 Indian seasonal regimens. So, the marker compounds can also be focussed to lie within the different principle compounds found responsible for the organoleptic character. The TCF kernel is used externally and other parts internally. Further, the seven types of TCFs vary in shape and size where the fruit parts with respective tastes come into role in each stage of maturity (MS) e.g., Phytochemicals of thinny pulp with big seed and normal seeded bulky pulped. With such a broad category of phytochemical (PC) variations, it is time consuming to analyse all the compounds to categorize the seven types. But the OL marker method is hopeful as it embraces both the positive and negative markers and would be definitive in picking out the analytical marker/specific active principle which is useful for standardization of each of the MS extracts[2,3]. Solubility of a compound influences the taste. Though the TC fruits are under the higher influence of the environment[4], the King tree withstands fire and so would be a regenerating tree in the forest in the growing years of global warming, thus saving itself and the medicinal values. Therefore, it is worthy to concentrate on the PC markers of taste of TCFs, in the present study.

RESULT:

TABLE NO 1- TCF TASTE (T) PRINCIPLES

MS	T	Phytochemicals in TCFs[4] found responsible for taste	Important inferences
I	Sweet*	Rich starch grains, maximum trihydroxy chebulagic acids (CLGs[5]), β-d-pyranoses, ferulic, vanillic acids in abundant amounts followed by gallic acid (oat samples[6]), catechins, chebulagic acids, sorbide, senna (mucilaginous sweetish and slightly bitter) calcium[7]. beta-Caryophyllene[8], terpene- sweet and dry)	Induced by tri and di hydroxyls (2H bonds)[9], tannins[10,11] and ellagic acids (EAs) are high. Tannins form insoluble complexes with proteins) [12]. EAs do not contribute to sweet taste- Theoretically tastes of Tannin and EAs compensate with each other and so the taste of CLG gets pronounced.
II	Bitter	Flavonoids, sorbide (hydrogenated D- glucose)→ Isosorbide (dehydrated sorbide)[9, 13,] Coumarins (pleasant sweet odour, bitter taste[14]), chlorogenic acids[15](Any of the phenyl propanoid derivatives like coumaric, caffeic & ferulic acids conjugate with quinic acids) eg., (3-O-caffeoylquinic acid), naringenin, Sennoside, anthraquinones glycosides	Naringenin: bitter marker & can reduce the bitter character of certain flavor formulations[16]; ability to combine with kojic acid[17]. Tannin accumulation in seed[11](? within taste detection threshold)
		4-O-((2', 4'-di-O-galloyl- α -l-rhamnosyl) ellagic acid[18]. Related compound- 4-O-(4''-O-galloyl-α-L rhamnopyranosyl) ellagic acid	Rhamnose at C-2 position exhibit bitterness[19]
III	Astringent	Melilotic acid, marker 1, 2, 3, 4, 6-penta-O-galloyl-D-glucose Ellagitannin(velvety) Eg., (Chebulagic acid, chebulinic acid and corilagin generally)[20,21], terchebulin, Several o – rhamnosyls, glycosilated but galloylated compounds are found. Rutin[22] (quercetin-3-o-rutinoside)8, Kaempferol-3-rutinoside(KR), tannic acids (TA), tannin e.g., terchebin, Polymerization of proanthocyanins (puckering and rough)[23], Luteolin (5,7,3',4'-tetrahydroxyflavone), Quercetin[24], suspected: less pro ACNs and phlobaphenes.	The replacement of the a-L-rhamnosyl residue by a-D- and B-D glucosyl residues influenced solubility and maintanence of bitter taste[17] Bitter to mild astringency- Possiblity of precursor bitter forms to: Arjunosides II (3-O-β-ᴅ-glucosyl-2-deoxy-α-L-rhamnoside of arjunic acid; 2, 3, 4, 6-tetra-O-galloyl-D-glucose[25]. Glycosylation yield velvety smooth dry astringent[23]. Rutinoside is 6-O-(alpha-L-Rhamnopyranosyl)-D-glucopyranose-beta forms tasteless. So rutin and KR, are smooth astringents. TA- wound healer(astringent action)[27] medium sized fruits(maximum yield) - dominant astringent taste [28]
			Anthracenes (astringent action)[26,27,28] Galloylation increases coarse and puckering qualities[23]; Anthraquinones combine with rhamnose through glycosidic linkages [29]. The tannic acids, containing mainly gallotannins, also have high protein precipitation capacities (due to high proline[30]). Ellagitannins have a significantly lower precipitation capacity than the galloylglucoses or gallotannins and the capacity is lower for ellagitannins with fewer galloyl groups[31]. Hydroxylation produces less coarse astringency. Hydroxylated anthraquinones- yellow, orange, and red pigments[32]

IV	Pungent	Quinones8, Hydroxycinnamic acid derivatives(HCAs) detected in fruit coat - Caffeic and chlorogenic acids) 33Eugenol (pungent) and ferulic acid (FA) are the related compounds to HCAs in TCFs.	FA - strong inducer of the heat shock response. HCAs also contribute to radioprotective activity34 and proved in Aqueous extracts35
V	Sour	Acetic acid and other Organic acids: Chebulic acid- Coumarin conjugated with Gallic acid form the chebulin; Gallic acid (GA): present with Initial mild sour and long-lasting sweet pleasant after taste 36; may also elicit astringent sensations (Otero-Losada, 1999)37	GA: absent in young fruit and present only in traces in the mature fruit38; does not combine with protein39 and therefore no astringent taste but used as remote astringent in internal haemorrhage.
VI	All	Most/ all of the above along with Pro anthocyanins (pro-ACNs) and ACNs	The shikkimate to acetate or malonate pathway40 completed

* The sweet principles should either start accumulating (in ovular/ kernal area) or represent the seed of mature fruit or coalesce with the II MS as in Ayurveda, the seeds are documented to be sweet/bitter[41,42].

Table No 2- Markers of TCFs common to other medicinal plants

API reference standard (RS)[2]	Medicinal plant (part used)	Family
Catechin	*Albizia lebbeck* (L.) Benth (stem bark)	Mimosaceae
Pterostilbene	*Pterocarpus marsupium Roxb.* (heart wood)	Fabaceae
Sennoside A&B	*Cassia senna* L. (dried leaflets)	Caesalpiniaceae
Markers[1]		
Naringin	Citrus fruits	Rutaceae
	Vitis vinifera L.[9, 43] *(Fruit peel)*	Vitaceae
Eugenol	*Ocimum tenuiflorum* L. (dried leaf)	Lamiaceae
Chrysophenol (Anthraquinone)	*Rheum emodi* Wall (Rhizome)	Polygonaceae
Quercetin-3-glucoside	*Hibiscus rosa-sinensis* Linn (Flower)	Malvaceae
Rutin	*Melia azedirecta* Linn (Leaves)	Meliaceae
Quercetin-3'-O-glucoside and quercetin[†]	*Abelmoschus manihot* (Linn.) Medicus (Flowers)	Malvaceae
Luteolin-7-glucoside[‡]	Cymbopogon citratus (DC. ex Nees) Stapf. (whole plant)	Poaceae

†Gunasingh and Nagarajan, 1980; Matouschek and Stahl, 1991
‡Xianyin Lai et al, 2017

DISCUSSION:

Taste and active principle: The presence of smooth astringents o-Glycosyl group in bitter flavonoid astringents has enhanced anti stress activity[44]. eg., Quercetion-3-o-rutinoside, Sennoside B-O-glycoside. Astringents[22] are blood purifiers and induce vata humour which is responsible for free movements. Bitters potentiate knowledge[*] (*agreement with referred findings)..

Number of chlorine atoms in each of the TCF part is a simple measure to reveal the sweetness[9]. Berberine chloride (API reference standard for Berberis aristata & bitter stomachic) expressed astringent activity. Theoretically bitterness of berberine[45] is reduced. Chlorides increase solubility. Chlorine presence in TCFs is not documented as per online search. Addition of NaCl and $CaCl_2$ effected sennoside concentration in negative and positive ways, respectively and the two samples of water treatment with TCF has both reduced and increased the hardness. As the probability is 1/9 with the chemical compounds involved in hardness, TCFs are in close association with chlorides. 2- 5 times calcium% increase between the III and V stages[20] enhanced the sennoside content and reduced astringent taste principles[46]. So the form of chlorides in TCFs is essential to be structured out. Magnesium regulates both and showed 1.5 times increase in these stages. Miraculin effect is the perception of acidic taste as sweet by raising the intracellular calcium[47*].Naringenin is difficult to absorb on oral ingestion[48] and sparingly soluble[2].The elicitation of anthraquinones and narigenin association with rhamnose in TCFs can reveal the shunt in pathways. This would provide route for further assessment of taste and markers of each MS under influencers. Likewise, Melilotic acid, *p*-coumaric and vanillic acids were present only in the intermediate and mature fruits[38].

Anthraquinone astringency in TCFs: absent/present as poor quantity of quinones; increased in big sized fruits[49]; absent in fresh plant parts; present only after drying[50]; Quinones alone detected in intermediate and mature stages; M-hydroxy quinones are water soluble compounds; emerge with the production of fat in plant organs. The presence of high proteins and fats in TCFs is evident from the research done in unripe TCFs at Himachal Pradesh (lower temperature and less rainfall than Sikkim)[20,51,52.] The contribution of colour by hydroxylated anthraquinones sets to unripe to ripe MS and reduction in astringency in the next MS. The anthraquinones (if present) would have precipitated with protein as an astringent and formed insoluble complex in the young MS under low temperature. The protein content of TCFs is more than that of the common apple (Barthaker & Arnold, 1991). The effect of drying on hydroxylated forms and protein annealing may explain the reason for their presence only in dried TCFs. Generally, phenolic hydroxyls constitute a total of 11% in TCFs[20]. Terpenoids and Sennoside content of youngest senna leaves are found higher than the mature leaves[53]*i.e., the formation of glycosylated AQs is likely to happen during late III and late IV MS drying and is to be correlated to the decrease with increase in area and age[*28]

Increase of the astringent quercetins through out maturity[38] (implies sustained Tannin biosynthetic pathway Figure 1)[54] and phenolic, anthraquinone, flavonoid enhancement by soil sucrose[55] correlated (table 3)[4]. The influence of rainfall is evident from the research done at Sikkim and Himachal Pradesh and understood in two ways. 1. Unripe to ripening MS: Minerals (K, Mg, Mn) increased with reduction of protein, fat, minerals (Zn, Cu) concentration. 2. Rainfall vs carbohydrate% (the substrate to the shikkimate pathway commence with the glycolysis): the rise in the concentration of tissue carbohydrates, ash content and moisture content is more than 25% in higher rainfall area. Protein reduction and emergence of anthraquinones is applicable to both ways[56]. Tannin also decreased between mid September and November end harvests[20*]

TCF luteolin extraction showed: dry weight more than gallic acid; less DPPH radical scavenging activity; the inhibition of the autoxidation of methyl linoleate activity of TCF extract > tannic acid > gallic acid > galloyl-free chebulinic acid > ellagic acid and chebulinic acid luteolin is equivalent to that of ethyl gallate & standard α-tocopherol and more than that of activity of whole. From this, it is also inferred that the galloylation (hydrolysed derivative) woven soft astringency improved the DPPH and decreased activity of later[57]. But, luteolin of TCF is not yet considered among the marker of any category.

Maturity may decrease the chain lengths of astringents and condensed tannins (CT) like Pro ACNs and phlobabhenes[*] after sugar rise. ACN (mature TCF) cut the terminal of condensed tannin (CT epicatechin) which reduces astringency. (In wine, aging increases sugar but perceived bitterness may due to other factors in the media).

The bulky angles (ridge) indicate an extension from the intercarpellary septum or bitter[4] calyx (II MS). But since it is the first part of the sour petiole [58], ridges have high relevance with sour taste). Ascorbic acid reduction between above harvests in Himachal correlated with sourness which is not specific in the VI MS (table 4)[*4].

In mammalian cells, a protein ionotropic receptor 7aIR7a works in concert with other proteins in taste receptor cells to detect sour tasted acetic acid[59]. The sugar solubilises ACNs. Anthocyanin glycosides in red wine do not contribute to the perceived astringency or bitterness (Vidal et al., 2004a). The additions of sugar reduced the bitter taste also.

Active principles can be common to medicinal plants but marker compounds should be specific to a particular drug/ atleast the species. So the update in table no 2 may help gain accuracy. In this regard, the associated families with constituents of TCFs can be consulted[60].

CONCLUSION:

This study on taste correlated well with previous studies and would be useful to elucidate the biosynthetic pathway conversions in TCFs, to predict and identify the marker of 6 MS of TCFs (e.g., Ellagi tannins in bitterness area should be low) and is helpful to segregate the 7 types based on OLC. Such studies on initial descriptors help to assure the quality.

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