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Herbal Chemo-Prospecting for New Phytomedicines

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Abstract

Plant biodiversity which is bestowed with immense chemical diversity has been the richest source of drugs. Chemical composition and molecular structure studies greatly enable possibility of new molecules for drug formulation. Thus, newer approaches like chemoprofiling using sophisticated analytical techniques and bioactivity determinations are needed that can result into new biomolecules. *Valeriana* species with cosmopolitan distribution are the source of important phytomedicines for curing nervous unrest and emotional troubles. These possess variety of bioactive sesquiterpenoids and valepotriates. *Nepeta* species are known for the presence of different isomeric nepetalactones, irododial derivatives and structurally related compounds. *Tanacetum nubigenum* exists in two chemically distinct chemotypes.

Introduction

Biodiversity has been source of numerous bioactive lead molecules for drug discovery in the changing scenario of pharmaceutical industry. Some important anticancer, antimalarial and antihypertensive drugs have their roots in traditional herbal remedies. In today's world, we find a renewed interest in "green" chemistry and the sustainable use of natural products for drug development. Herbal materials, in addition to traditional remedies, also contribute to nutraceuticals and cosmeceuticals. Natural products, the greatest treasures of nature, are continuous and unending source of potential lead molecules for pharmaceutical industry. WHO reported about a decade ago that 80% of the world's inhabitants continue to rely mainly on traditional systems for their health care. Modern research in drug discovery from medicinal plants involves a multifaceted approach combining phytochemical, bioprospecting, pharmacological and analytical techniques. Modern sophisticated chromatography-spectroscopy hyphenated techniques viz. GC-MS/MS, LC-MS/MS, GC-FTIR, LC-NMR, ICP-MS etc have contributed in the standardization, validation, trace analysis and study of marker compounds of herbal medicines. Traditional medicine involves knowledge, skills and practices based on theories and experiences indigenous to different cultures. It is convenient source of health care to millions being accessible and affordable for many living in remote areas. In recent years, paradigm shift has caused multidrug-multitarget concept. Interestingly, Ayurvedic and traditional medicines seem to support this approach. Bioassay-guided fractionation has been useful in phytochemical-pharmacological standardization or 'fingerprinting' of medicinal plant extracts in test animal or human systems. Chemoprofiling is establishing a characteristic chemical pattern for the herbal materials. The quality control and validation of herbal drugs has been difficult because of variation in chemical profile of their chemical constituents. For a better evaluation of the plant material we need to measure a set of compounds and resulting bioactivity rather than one single compound. We have been actively engaged in chemoprofiling of the plant materials collected from different parts of Himalayan region. Chemical variation is one phenomenon commonly observed which complicates standardization and validation of herbal materials.

Results and Discussion

Certain herbal species show drastic qualitative changes in secondary metabolites within the same species and thus exist as distinct chemotypes consequently affecting their bioefficacy and ultimately their pharmaceutical potential. This difference is caused by intrinsic (genetic) and extrinsic (altitude, soil, sun, moisture etc) factors. During our investigations on chemistry and bioactivy of Himalayan medicinal/ aromatic plants, we noticed distinct chemotypes within certain species some of which have been used in traditional system of medicine such as *Nepeta, Valeriana* and *Tanacetum* species among others. Some observations on their biologically active iridoid constituents are being presented here. *Nepeta* species are used as diuretic, diaphoretic, antispasmodic, antiasthmatic, febrifuge and sedative agents [1]. The diastereomeric nepetalactones 1 and other iridoid constituents are found to be responsible for the diverse biological activities viz. feline attractant, canine attractant, and insect repellant and arthropod defense activity. Iridoids and iridoid glycosides have also been reported

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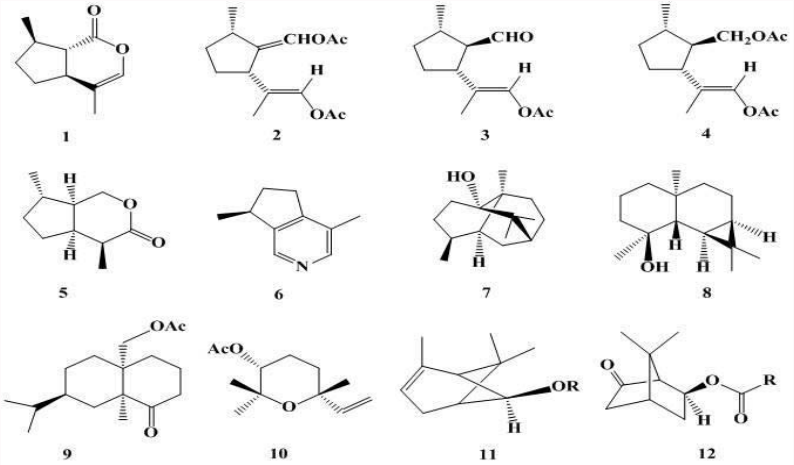

Figure 1: These results suggest that unless extensive phytochemical investigations are done, the herbal materials can not be standardized. Also, close research collaboration among traditional folk-lore practicener, pharmacists, phytochemists, clinicians and analytical scientists can produce new, safer and bioactive natural products for drug development. The chemical diversity among medicinal plants helps in sustainable use as pharmaceuticals and flavour chemicals.

from *Nepeta* species including three new iridodial derivatives *viz.* iridodial β-monoenol acetate 2, dihydroiridodial diacetate 3, and iridodial dienol diacetate 4 from *N. leucophylla* [2]. Several di- and tri-terpenoids ursolic acid and oleanolic acid and phenolic constituents coleon U-12 methyl ether and dehydro coleon U-12 methyl ether have been isolated from *Nepeta leucophylla* and *N. elliptica* [3-5]. A highly antifungal compound, actinidine **6** has been isolated from *N. clarkei* Benth [6]. Pregeijerene and isoiridomyrmecin 5 were isolated from essential oil of *N. govaniana* [7]. *Valeriana* species (Valerianaceae) with global distribution are the source of important phytomedicines for curing nervous unrest, emotional troubles (as tranquillizer/ sedative), epilepsy and insanity [8]. It continues even today to be a safe sedative/ hypnotic choice for patients with mild to moderate insomnia. *Valeriana* species possess variety of sesquiterpenoid flavour chemicals and valepotriates possessing unique structural features. The activity of Valerian herbs is largely due to combination of nonvolatile valeporiates and sesquiterpenoids constituents. Some valepotriates exhibit significant cytotoxic activity. Our investigations have indicated the existence of chemically distinct chemotypes within certain *Valeriana* species. *V. wallichii* DC (*V. jatamansi*) is an important herb used in Indian system of medicine. Our investigations established three chemotypes in terms of volatile flavor chemicals [8,9]. Chemotype-I possessed patchouli alcohol (40.2%, **7**) in contrast to chemotype-II which possessed maaliol (64.3%, **8**) while the third was marked by the presence of kanokonyl acetate (42.4%, **9**) [8,9]. Notably, the marker constituents of one chemotype of *V. wallichii* were completely absent in other chemotypes. Furthermore, these three chemotypes grow in separate areas and were not noticed as mixed population in the natural habitat. Thus, three chemotypes, though morphologically indistinguishable, may be identified by their characteristic major terpenoid constituents. As expected, the differences were also reflected in the antidepressant activity and analgesic action in experimental animal models [11-14]. Iridoids representing a group of cyclopentan-pyran monoterpenoid secondary metabolites, the non-glycosidic iridoids or valepotriates are unique feature of the family Valerianaceae. The valerian preparations are often used as mild sedative and sleep promoting agent or a substitute for synthetic sedatives in the treatment of nervous excitation and anxiety-induced sleep disturbances.

Tanacetum species have been used for the prevention of migraine, headache, arthritis, fevers, muscle tension and pain. *T. nubigenum*, a high altitude flavouring species, is used locally to cure fever in the form of decoction. Our studies established two distinct chemotypes [14,15]. Chemotype-I is marked by presence of *cis*-chrysanthenol and its acetate, isobutyrate, methyl butyrate angelate and tiglate derivatives in addition to exo-6-propionyloxy camphor and 4 other related higher camphor esters in its essential oil while the chemotype-II is characterized by the presence of an entirely different constituents, (3R, 6R)-linalool oxide and its acetate and dioxaspiro acetylenic compounds as its major flavouring compounds.

These results suggest that unless extensive phytochemical investigations are done, the herbal materials cannot be standardized. Also, close research collaboration among traditional folk-lore practicener, pharmacists, phytochemists, clinicians and analytical scientists can produce new, safer and bioactive natural products for drug development. The chemical diversity among medicinal plants helps in sustainable use as pharmaceuticals and flavour chemicals.

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