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Chemical Constituents of *Terminalia chebula*

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Abstract

Despite the revolutionary progress and discoveries, the challenge to combat newly emerged and discovered diseases remains unmet. Natural products are still the prime reservoirs for providing new and novel molecular skeletons as drug candidates and inspiration. *Terminalia chebula* showed promising anti-microbial and anti-viral potentials and about 133 natural products have already been isolated from *T. chebula* and importantly, its richness of variety of metabolic enzymes are biosynthesizing diverse variety of secondary metabolites with attractive novelty and variety, ranging from phenolic derivative to flavonoids and falvins, terpenoids to steroids, alkaloids, tannins and their derivatives and glycosides. Molecular topology and variety in functionalities of in its natural products are important, especially dealing with the variable and adaptive capabilities of viruses and microorganism generated diseases. Importantly, it is useful for natural product chemist to have the updated chemical constituents of *T. chebula*.

Keywords: Terminalia Chebula; Chemical constituents; Nature unique diverse and rich variety

Introduction

Despite of the investments and progress in molecular biology and drug discovery, several infections by pathogenic microbes are major threats for human life. Development of various anti-microbial drugs have meaningfully controlled several pathogenic diseases or made them less destructive. However, emerging resistance against existing antibiotics has made them less effective and is a major threat to the humanity. Therefore the new antibiotics are crucially required on War-footing for the discovery and development of new antibiotics. Prevention of infections, tracking the resistant strains and proper use of antibiotics would be important precautionary measures. Natural products played the fundamental role as traditional herbal medicine and remedies and through advancement in the knowledge and research gave birth to the specific molecular medicines which also enhanced the structural and mechanistic understandings of biochemical entities and processes [1,2]. A major number of drugs have been derived or inspired from natural products [3,4]. Pain relieving properties of the Willow's bark lead to the discovery of acetylsalicylic acid (aspirin) [5]. The first antibiotic, penicillin, was isolated from a mold,

penicillium and terpenoidal anti-cancer taxol was isolated from *Taxus brefolia* [6]. Structural and functional biology and medicinal chemistry have rewarded with revolutionary drugs [7], however, pathogenic micro-organism are major global risk to human health [8]. A progressive microbial resistance is an alarming threat causing community-acquired infections and antibiotic failure. Plants have been and are still the major, rich and diverse source of variety of phytochemicals including potent anti-microbial molecules [9]. Interestingly and presumably, since plant extracts contain many phytochemicals therefore, using plant extracts may pose some difficulty against the development of bacterial resistance as compared to the single molecular drug. Plants have provided anti-AIDS agents as well as anti-cancer taxol and homoharringtonine, benzylisoquinoline, papaverine with high inhibition to the replication of many viruses (e.g. cytomegalovirus, measles and HIV), atropisomeric naphthylisoquinoline dimers, michellamines A, B and C showed potential HIV-1 and HIV-2 on human lymphoblastoid target cell *in vitro*. Low cost to benefit ratio of natural product derived drugs is another attraction [3].

Experimental

Terminalia chebula

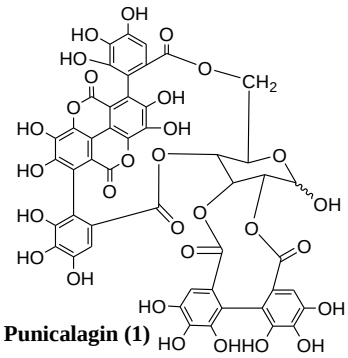
Terminalia chebula is a large sized traditional medicinal plant that is found in Pakistan, India, China and Tibet. It belongs to genus *Terminalia which* contains 250 species widely distributed throughout the tropical areas of the world. Traditionally, it is being used to treat gastrointestinal and urinary tract diseases, fever, cough, diarrhea, wound infections, skin diseases, urinary tract infection and candidiasis [10]. *T. chebula* possesses effective anti-bacterial and anti-viral activity against various bacterial strains [11,12].

Rich, diverse and novel nature molecular engineering (chemical constituents) in *Terminalia chebula*

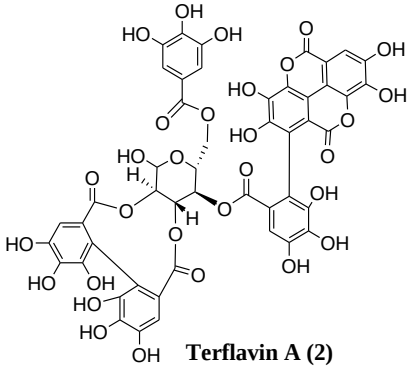
Terminalia chebula habitats in tropical and sub-tropical diverse climatic condition and also is rich with metabolic engineering enzymes, therefore it is extremely rich and diverse precursor for variety of natural products belonging to the various classes with different levels of biosynthesis. Richness, diversity and novelty of tannins and their analogues in *T. chebula* is a complex challenge both in term of isolation and structural modification but is an opportunity for the organic and medicinal chemists to modulate various biological disorders, especially pathogenic diseases because of their structural properties. Opportunity by *T. chebula*, is not limited to the tannins but also is fascinating with the provision of its small bioactive molecules which are very attractive for medicinal chemists and R&D organizations. Additionally, the variety of different classes of bioactive natural products in *T. chebula* further attracts the focused researchers which include, flavonoids, flavins, terpenoids, steroids, various phenols, functionalized aliphatic molecules and their glycosides (FIG. 1-8). Structural diversity and novelty of the nature's molecular engineering in *T. chebula*, especially in terms of in their skeletons, functionalities and linkages, is fascinating and invite focused and applied research to exploit their benefits in the form of new drugs (especially, antibiotics) further studies [13-31]. In addition, we understand and strongly propose that the standardization, efficacy, safety studies and documentations of its herbal products as of urgent prime importance. Focused and guided phytochemical re-investigation would be important and rewarding (TABLES 1 to 7).

TABLE 1. **Chemical constituents of *Terminalia chebula.***

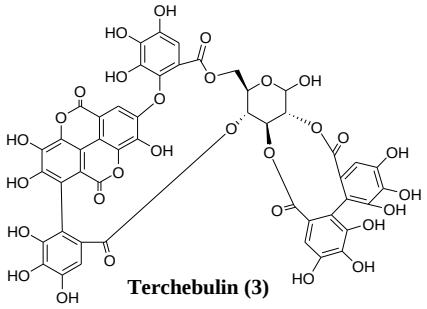
#	Tannins	References
1	Punicalagin ((2, 3-(S)-hexahydroxydiphenoyl-4, 6-(S, S)-gallagyl-D-glucose)	[13]
2	terflavin A	[13]
3	Terchebulin	[13]
4	Terchebin (1, 3, 6-trigalloyl glucose,)	[14]
5	Terflavins B	[13]
6	Terflavin C	[13]
7	Terflavin D	[13]
8	Punicalin	[13]
9	Neo-chebulic acid	[15]
10	X+	[15]
11	1, 6-di-O-galloyl-D-glucose	[16]
12	Gallic acid (3, 4, 5-Trihydroxybenzoic acid)	[16]
13	Casuarinin	[16]
14	Chebulanin	[16]
15	Corilagin	[16]
16	Ellagic acid (2, 3, 7, 8-Tetrahydroxy-chromeno [5, 4, 3-cde]chromene-5, 10-dione)	[16]
17	Chebulagic acid	[16]
18	Chebulinic acid (1, 3, 6-Tri-O-galloyl-2, 4-chebuloyl-β-D-glucopyranoside)	[16]
19	1, 2, 3, 4, 6-penta-O-galloyl-D-glucose	[16]
20	2, 3, 4, 6-tetra-O-galloyl-B-D-glucose	[17]
21	Ethyl gallate (Ethyl 3, 4, 5-trihydroxybenzoate)	[17]
22	Methyl gallate (Methyl-3, 4, 5-trihydroxybenzoate)	[17]
23	Chebulaginic acid	[14]
24	4-O-methylgallic acid	[18]
25	Methyl(S)-flavogallonate	[18]
26	Methyl neochebulagate	[18]
27	Eugenol	[19]
28	Ascorbic acid	[19]
29	Triethyl chebulate	[20]
30	Tannic acid [2, 3-dihydroxy-5-({[(2R,3R,4S,5R,6R)-3, 4, 5, 6-tetrakis({3, 4-dihydroxy-5-[(3, 4, 5-trihydroxyphenyl)carbonyloxy]phenyl}carbonyloxy)oxan-2-yl]methoxy}carbonyl)phenyl 3, 4, 5-trihydroxybenzoate]	[20]
31	2, 4-Chebulyl-beta-D-glucopyranose	[13]



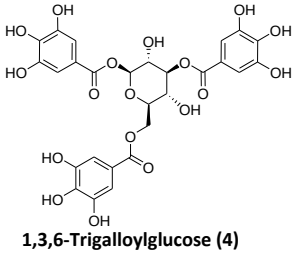
Punicalagin (1)



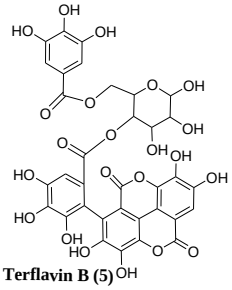
Terflavin A (2)



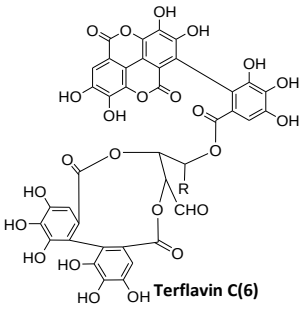
Terchebulin (3)



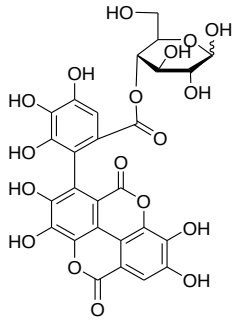
1,3,6-Trigalloylglucose (4)



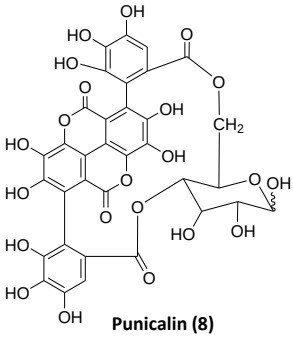
Terflavin B (5)



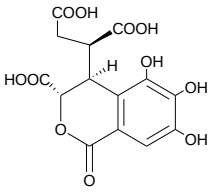
Terflavin C(6)



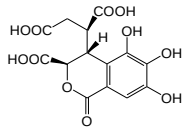
Terflavin D (7)



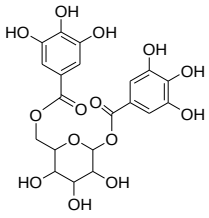
Punicalin (8)



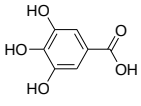
Neo-chebulinic acid (9)



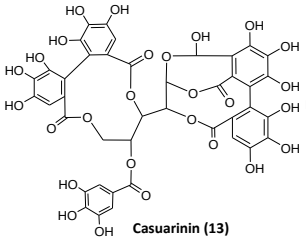
Chebulic acid (10)



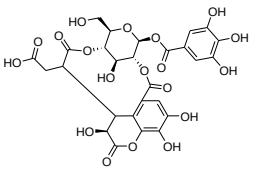
1,6-Di-O-galloyl-B-D-glucose (11)



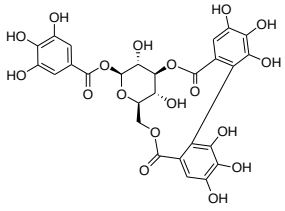
Gallic acid (12)



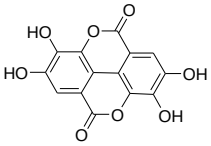
Casuarinin (13)



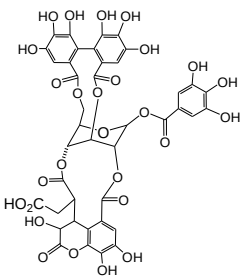
Chebulanin 14



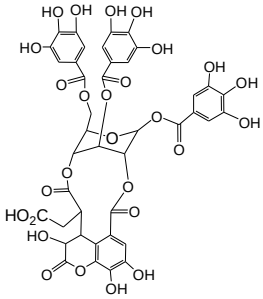
beta-1-O-galloyl-3,6-(R)-hexahydroxydiphenoyl-d-glucose (15)



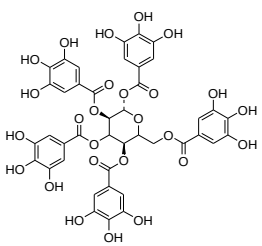
Ellagic acid (16)



Chebulagic acid (17)



Chebulinic acid (18)



1,2,3,4,6-Penta-O-galloyl-B-D-glucose (19)

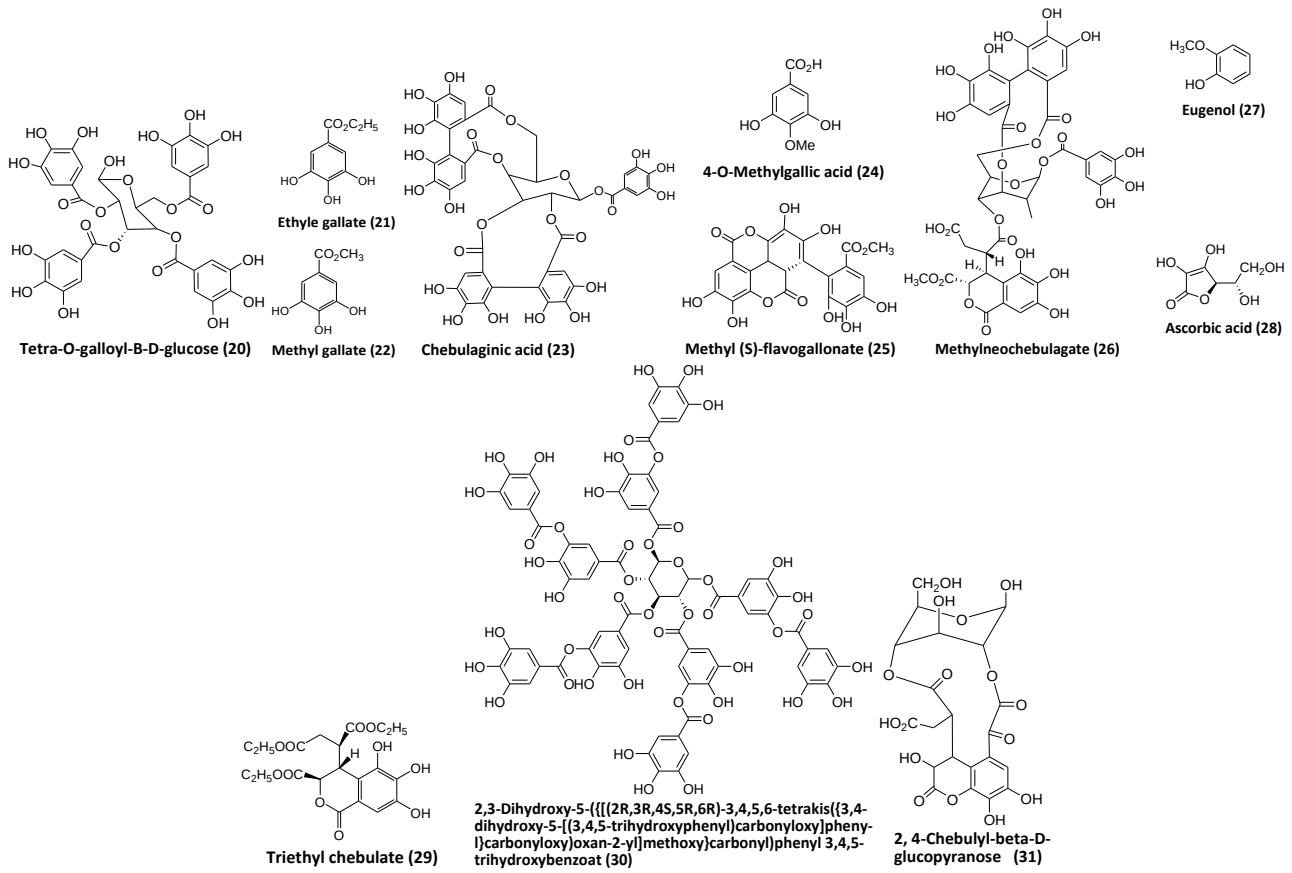
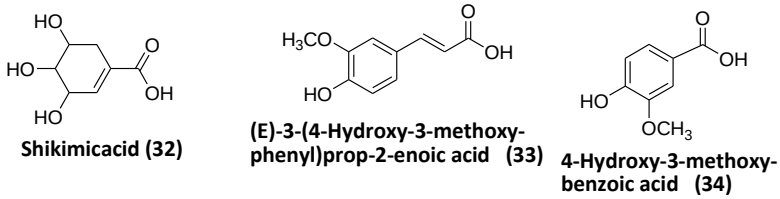


FIG. 1**. Structure of chemical constituents (Tannins) of *Terminalia chebula (1-31).***

TABLE 2**. Phenolic Carboxylic compounds from *Terminalia chebula*.**

#	Phenolic Carboxylic compounds	References
32	Shikimic acid	[20]
33	Ferulic acid	[20]
34	Vanillic acid	[20]
35	p-Coumaric acid	[20]
36	Caffeic acids	[20]
37	Melilotic acid	[21]



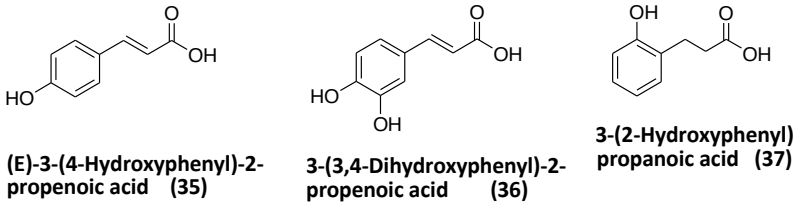


FIG. 2. **Structure of Phenolic Carboxylic Compounds (32-37).**

TABLE 3. **Phenols from *Terminalia chebula.***

No.	Phenols	References
38	Phloroglucinol [benzene-1, 3, 5-triol]	[21]
39	Pyragallol [1, 2, 3-Trihydroxybenzene]	[21]
40	Phenol	[22]

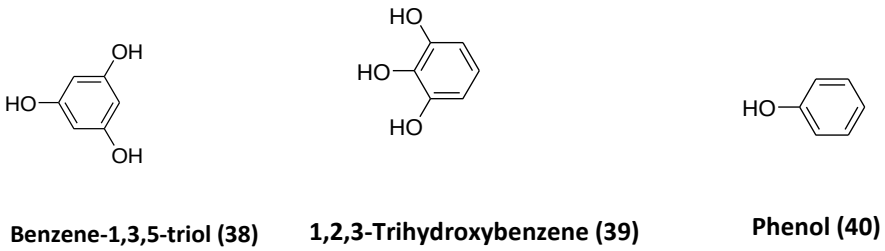
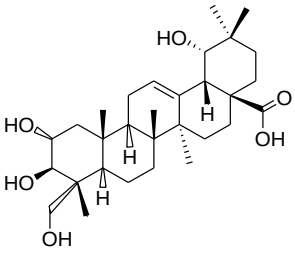


FIG. 3. **Structure of Phenols (39-40).**

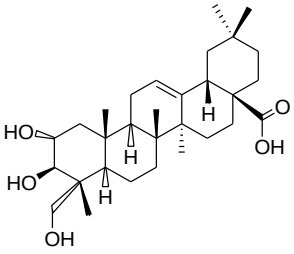
TABLE 4. **Triterpenoids and their Saponins from *Terminalia chebula.***

#	Terpenoids and Triterpene Saponins	Ref.
41	Arjungenin	[18]
42	Arjunolic acid	[18]
43	Arjunic acid	[18]
44	Terminolic acid	[18]
45	Arjunglucoside I	[18]
46	Arjunglucoside II	[18]
47	Arjunetin	[18]
48	Chebuloside II	[18]
49	Bellericoside	[23]
50	Chebuloside I [2α, 3β, 23-Trihydroxy-olean-12-en-28-oic Acid]	[24]
51	2α-Hydroxyursolic acid	[24]
52	2α-Hydroxymicromiric acid	[24]

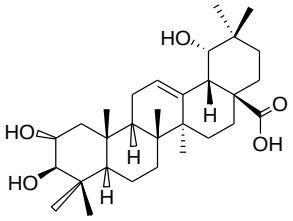
53	Maslinic acid [(4aS, 6aR, 6aS, 6bR, 8aR, 10R, 11R, 12aR, 14bS)-10, 11-dihydroxy-2, 2, 6a, 6b, 9, 9, 12a-heptamethyl-1, 3, 4, 5, 6, 6a, 7, 8, 8a, 10, 11, 12, 13, 14b-tetradecahydropicene-4a-carboxylic acid]	[24]
54	β-caryophyllene	[25]
55	α-Phellandrene[α: 2-Methyl-5-(1-methylethyl)-1, 3-cyclohexadiene]	[25]
56	α-Terpinene	[25]
57	Terpinen-4-ol	[25]
58	Terpinolene	[25]
59	Chebupentol [Olean-12-ene-2, 3, 19, 23, 28-pentol,(2a, 3b, 4a, 19a)]	[25]



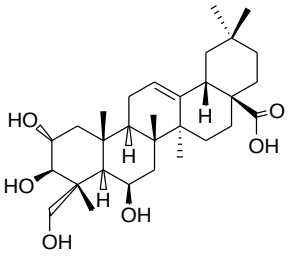
Arjungenin (41)



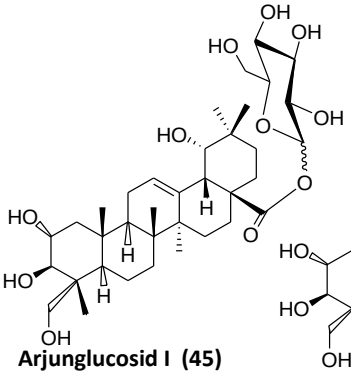
Arjunolic acid (42)



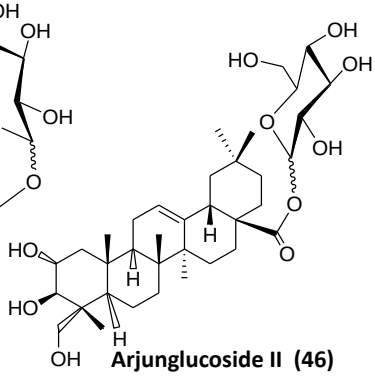
Arjunic acid (43)



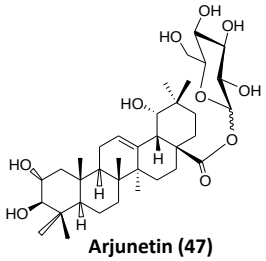
Terminolic acid (44)



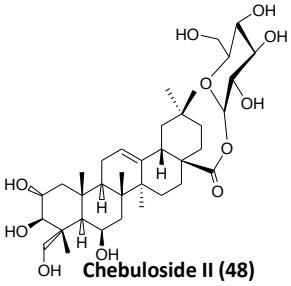
Arjunglucosid I (45)



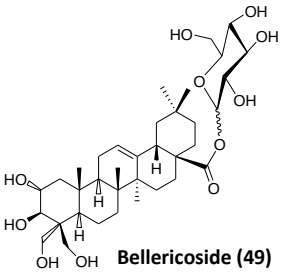
Arjunglucoside II (46)



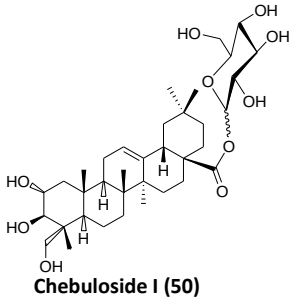
Arjunetin (47)



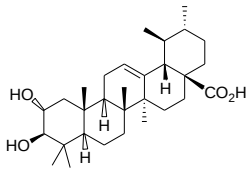
Chebuloside II (48)



Bellericoside (49)



Chebuloside I (50)



(1S,2R,4aS,6aR,6aS,6bR,8aR,10R,11R,12aR,14bS)-10,11-Dihydroxy-1,2,6a,6b,9,9,12a-heptamethyl-2,3,4,5,6,6a,7,8,8a,10,11,12,13,14b-tetradecahydro-1H-picene-4a-carboxylic acid (51)

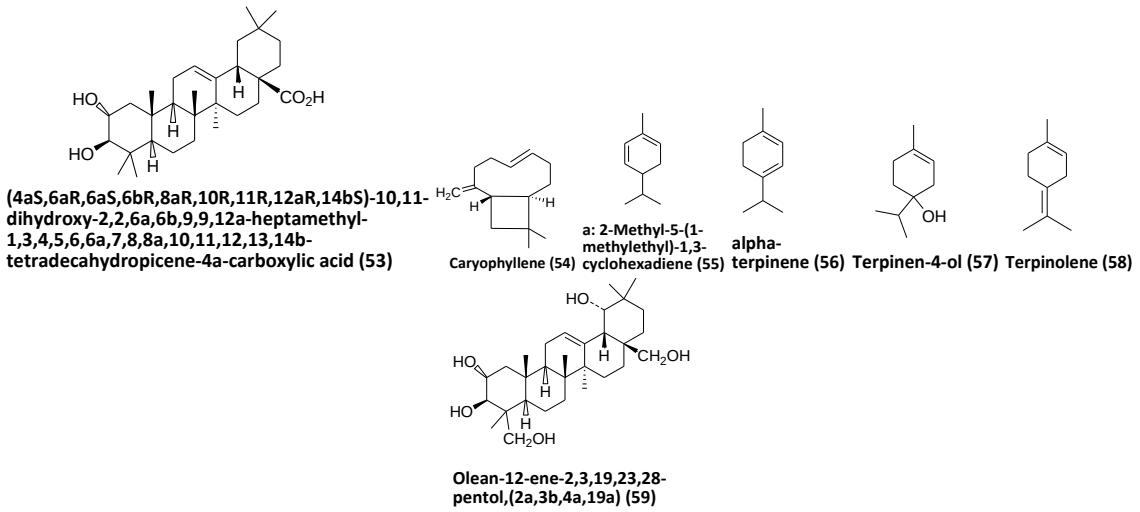


FIG. 4. **Structure of Terpenoids and Triterpene Saponins (41-59).**

TABLE 5**. Flavonoids isolated from *Terminalia chebula.***

#	Flavonoids	References
60	Rutin	[25]
61	Quercetin	[25]
62	Luteolin [2-(3, 4-Dihydroxyphenyl)-5, 7-dihydroxy-4-chromenone]	[26]
63	Isoquercetin [2-(3, 4-Dihydroxyphenyl)-5, 7-dihydroxy-3-[(2S, 3R, 4S, 5S, 6R)-3, 4, 5-Trihydroxy-6-(hydroxymethyl)oxan-2-yl]oxychromen-4-one]	[26]
64	3'-Methoxy quercetin	[13]
65	3, 4-Dimethoxy quercetin [5, 7-Dihydroxy-2-(3-hydroxy-4-methoxyphenyl)-3-Methoxy-4H-chromen-4-one]	[13]
66	Pelargonidin	[13]

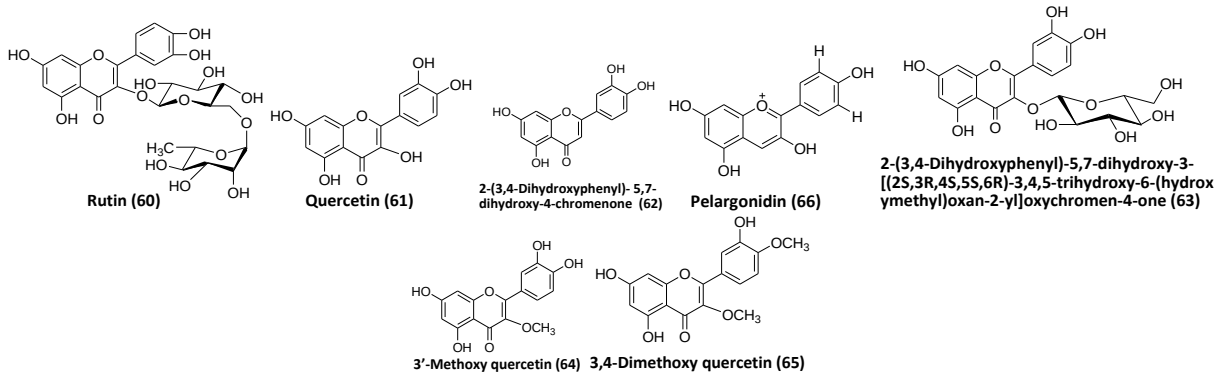


FIG. 5. **Structure of Flavonoids (60-66).**

TABLE 6. **Sterols isolated from *Terminalia chebula.***

#	Sterols	References
67	β-Sitosterol	[19]
68	Daucosterol	[27]

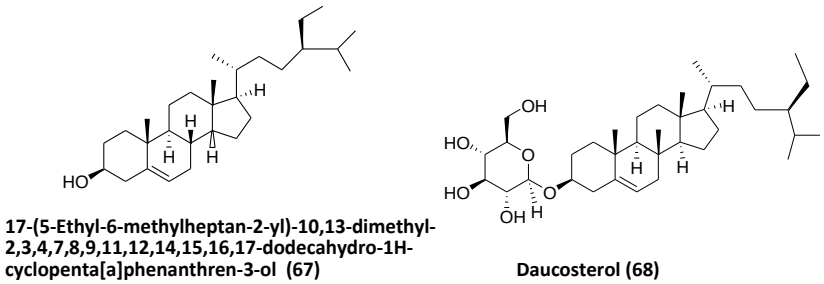


FIG. 6. **Structures of Sterols (67,68).**

TABLE 7. **Miscellaneous compounds from *Terminalia chebula.***

No.	Miscellaneous compounds	References
69	Behenic acid [Docosanoic acid]	[25]
70	Stearic acid [Octadecanoic acid]	[25]
71	Palmitic acid [hexadecanoic acid]	[14,22]
72	Oleic acid [(9Z)-Octadec-9-enoic acid]	[14,22]
73	Arachidic acid [icosanoic acid]	[14,22]
74	Linoleic acid [9Z, 12Z)-9, 12-Octadecadienoic acid]	[14,22]
75	12-Hydroxyoctadec-cis-9-enoic acid (ricinoleic acid)	[21]

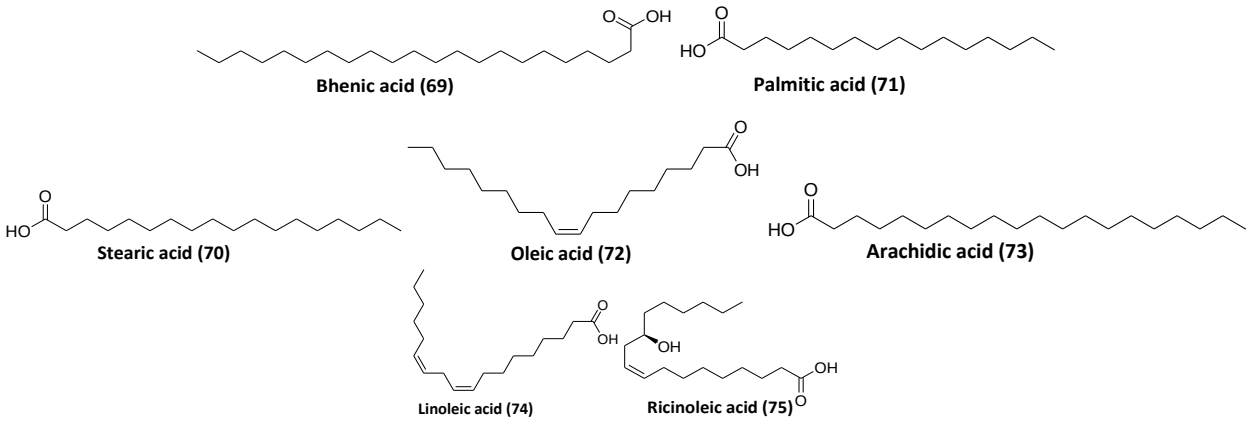
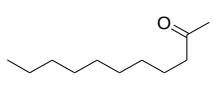


FIG. 7. **Structure of Miscellaneous compounds present in *Terminalia chebula* (69-75).**

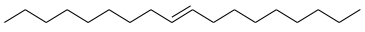
TABLE 8. **Compounds from *Terminalia chebula* fruit.**

No.	Compound isolate from *Terminalia chebula* fruit	Ref.
76	2-Undecanone	[22]
77	Cyclododecane	[22]
78	9-Octadecene	[22]
79	Hexadecane	[22]
80	Cylohexane	[22]
81	8-Pentadecanone	[22]
82	9-Eicosene	[22]
83	Triacontane	[22]
84	Tetradecane	[22]
85	Oxirane	[22]
86	1, 16-Hexadecanediol	[22]
87	Heptylcyclohexane	[22]
88	10-Nonadecanone	[22]
89	Phthalic acid	[22]
90	Tritetracontane	[22]
91	9-Heptadecanone	[22]
92	Tetratetracontane	[22]
93	Linoleic acid ethyl ester	[22]
94	9-Octadecenoic acid ethyl ester	[22]
95	9, 12, 15-Octadecatrienoic acid	[22]
96	1-Tricosene	[22]

97	1, 19-Eicosadiene	[22]
98	Heptafluorobutyric acid	[22]
99	1-Octanol	[22]
100	1-Decanol	[22]
101	Cyclooctacosane	[22]
102	1H-Indene	[22]
103	Hexacosyl pentafluoropropionate	[22]
104	Octatriacontyl pentafluoroprppionate	[22]
105	Tetratriacontane	[22]
106	1, 2-benzenedicarboxylic acid	[22]
107	Ibogamin-9(17H)-ol [(9α)-12-Methoxy-16, 17-didehydro-9, 17-dihydroibogamin-9-ol]	[22]
108	9-Tricosene	[22]
109	Tetratriacontyl heptafluorobutyrate	[22]
110	Dotricontyl heptafluorobutyrate	[22]
111	Tetracosanoic acid [Lignoceric acid]	[22]
112	Pentatriacontane	[22]
113	Eicosyl trifluoroacetate	[22]
114	Squalene	[22]
115	Tetracosyl heptafluorobutyrate	[22]
116	Tetratriacontyl heptafluorobutyrate	[22]
118	Heptafluorobutyric acid	[22]
119	Sulfurous acid	[22]
120	Octacosanoic acid	[22]
121	Vitamin E	[22]
122	Tetracosyl heptafluorobutyrate	[22]
123	Hexacosanoic acid	[22]
124	Octatriacontyl pentafluoropropionate	[22]
125	Triacontanoic acid [Melissic acid]	[22, 25]
126	Tricosyl pentafluoropropionate	[22]
127	Acetic acid	[22]
128	Heptacosanoic acid	[22]
129	Tetratriacontyl pentafluoropropionate	[22]
130	Tetracosanoate	[22,28,29]
131	Kaempferol-3-rutinoside	[22,30]
132	Ethanedioic acid	[22,31]

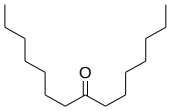

2-Undecanone (76)

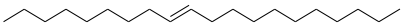

Cyclododecane (77)

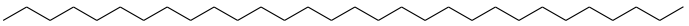

9-Octadecene (78)

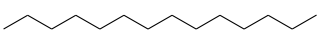
Hexadecane (79)


Cyclohexane (80)

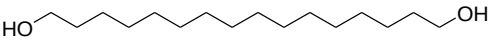

8-Pentadecanone (81)

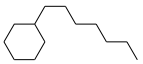

9-Eicosene (82)

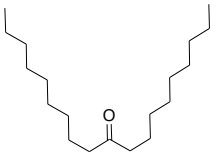

Triacontane (83)

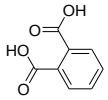

Tetradecane (84)


Oxirane (85)

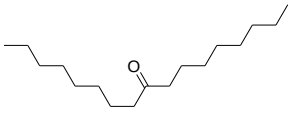

1,16-Hexadecanediol (86)

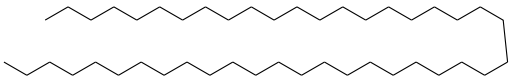

Heptylcyclohexane (87)

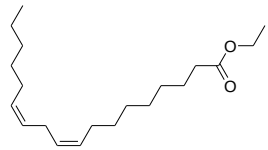

10-Nonadecanone (88)

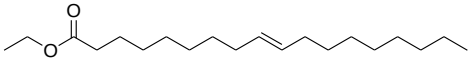

Phthalic acid (89)

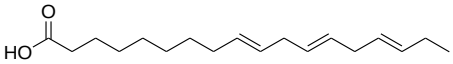
Tritetracontane (90)


9-Heptadecanone (91)

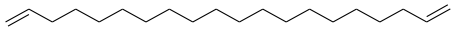

Tetratetracontane (92)

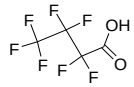

Ethyl linoleate (93)

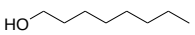

9-Octadecenoic acid ethyl ester (94)

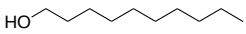

9,12,15-Octadecatrienoic acid (95)

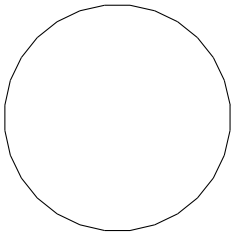

1-Tricosene (96)


1,19-Eicosadiene (97)

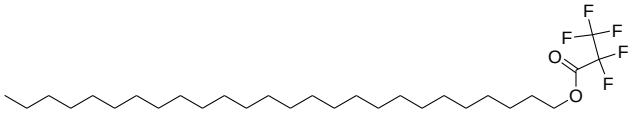

Heptafluorobutyric acid (98)


1-Octanol (99)

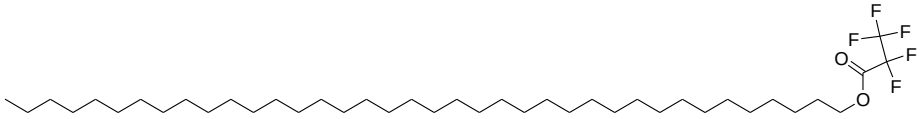

1-Decanol (100)


Cyclooctacosane (101)


1H-Indene (102)

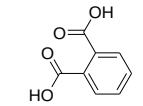


Hexacosyl pentafluoropropionate (103)

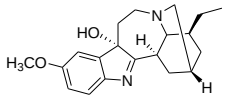


Octatriacontyl pentafluoroprppionate (104)

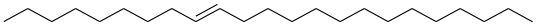
Tetratriacontane (105)



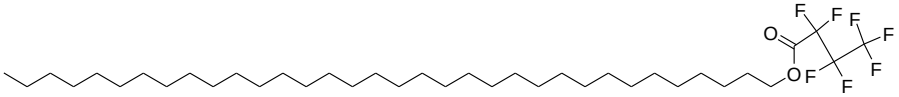
1,2-benzenedicarbo-xylic acid (106)



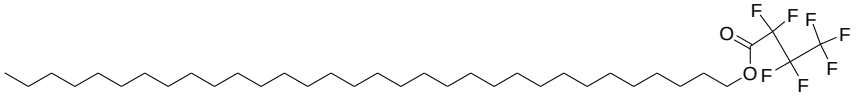
(9a)-12-Methoxy-16,17-didehydro-9,17-dihydroibogamin-9-ol (107)



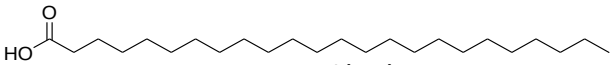
9-Tricosene (108)



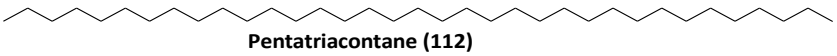
Tetratriacontyl heptafluorobutyrate (109)



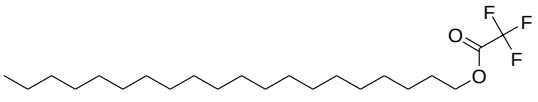
Dotricontyl heptafluorobutyrate (110)



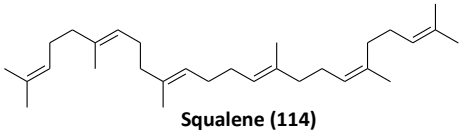
Tetracosanoic acid (111)



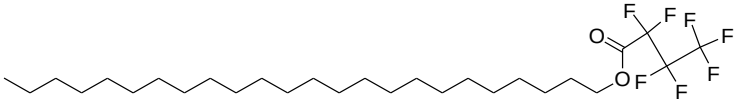
Pentatriacontane (112)



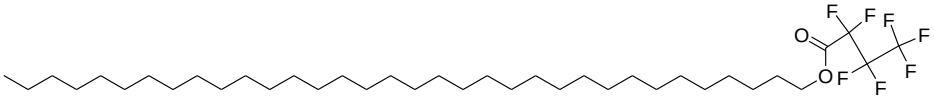
Eicosyl trifluoroacetate (113)



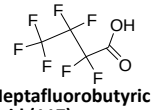
Squalene (114)



Tetracosyl heptaflourobutyrate (115)

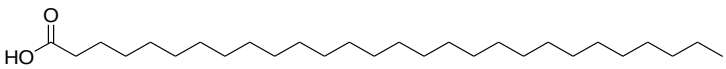


Tetratriacontyl heptafluorobutyrate (116)



Heptafluorobutyric acid (117)

Sulfurous acid (118)



Octacosanoic acid (119)

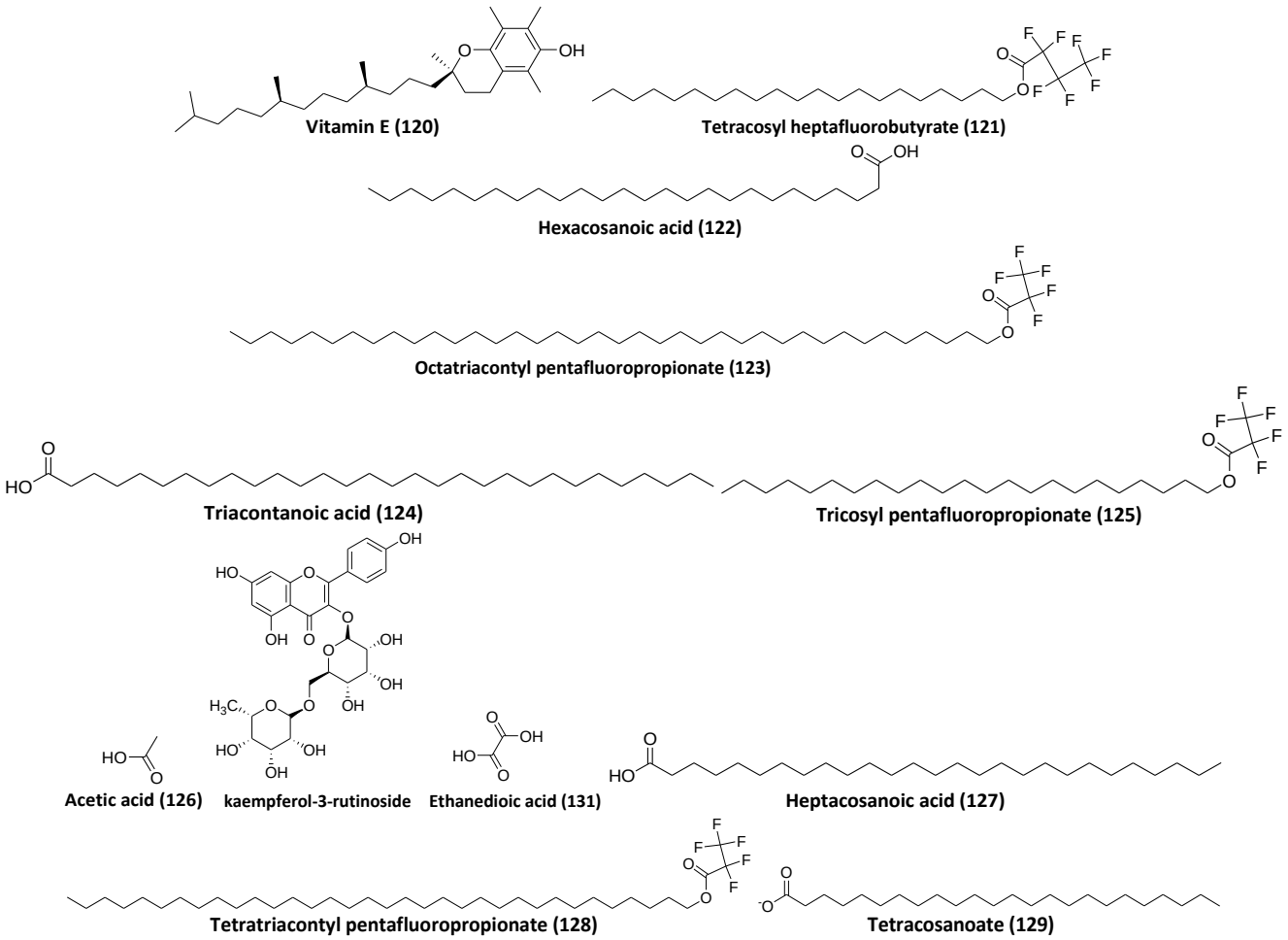


FIG. 8. **Structures of Compounds isolated from *Terminalia chebula* fruit (76-132).**

Results, Discussion and Conclusion

In light of role of nature engineered secondary metabolites in pharmaceuticals, the structural and functional diversity and novelty of natural products of *Terminalia chebula* along with the rich and diverse biosynthesis, together with their wide range of therapeutic applications and importantly its valuable anti-pathogenic biological properties coupled with the burnings needs for new antibiotics, we conclude and propose:

1. Focused and guided phytochemical re-investigations on the *Terminalia chebula* and its herbal products,
2. Structure activity relation (SAR) further medicinal chemistry to explore the drug potentials of already known bioactive molecules of *Terminalia chebula*, especially anti-microbial agents.

Further studies for the standardization, formulation, documentation, efficacy and safety of traditional herbal medicinal products (THMPs) of *Terminalia chebula* . Hence, it is crucial to be shared with the relevant scientists.

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