Review

Advances in the Propolis Chemical Composition between 2013 and 2018: A Review

Luka Šturm, Nataša Poklar Ulrih

Department of Food Science and Technology, Biotechnical Faculty, University of Ljubljana, Jamnikarjeva 101, Ljubljana 1000, Slovenia

ABSTRACT

Propolis is a lipophilic sticky substance collected by bees that has been used by humans for centuries. Owing to its healing, antioxidant, and other medicinal properties, its chemical composition has been widely studied. Most pharmacological properties of propolis have been attributed to its phenols and terpenes, mainly flavonoids, phenolic acids, and their derivatives. More than 500 components of propolis were known from different parts of the world until 2012. In this article, 305 new constituents of propolis described between 2013 and 2018 are being reviewed, with 19 additional compounds that were discovered between 2011 and 2012, and were excluded from a similar previous review article. Altogether more than 850 compounds were isolated from propolis until 2018.

1. INTRODUCTION

Propolis or sometimes called “bee glue” is a lipophilic, adhesive, gummy, and resins substance collected by different species of bees, including honeybees (e.g. Apis mellifera L.) and stingless bees (e.g. Tetragonisca angustula Illiger), Bees use it to seal holes in their hives, smooth out the internal walls, and protect the entrance against intruders. It also acts as a natural antibiotic to prevent bacterial, viral, or fungal infections within the hive [1–4]. Bees collect the resin from the cracks in the bark and leaf buds of different trees, including different species of poplars, conifers (e.g. pines and cypress), birches, alders, willows, palms, chestnuts, and even trees like eucalyptus, acacia, Clusia spp., and Baccharis dracunculifolia DC. Bees add salivary enzymes to the collected resin, mix it with beeswax and use this partially digested material in their hives [5–9].

The name propolis derives from the Hellenistic ancient Greek meaning “suburb/bee glue” or “defense of the city”, depending on the interpretation [5,10]. The use of propolis dates back to at least 300 BC and has been used by Egyptians, Persians, Greeks, and Romans. It was used mainly as a topical cream for cuts, ulcers, wounds, and other dermatological problems, furthermore it was used for mummification by the Egyptians. However, in medieval times the use of propolis was not very popular. It remained mostly as an alternative herbal medicine, mainly in Eastern Europe, especially in Russia, where it later became known as the “Russian penicillin”. The use of propolis was rediscovered again in the Renaissance with the growing popularity of ancient teachings and medicine. The first scientific researches of propolis began in the 19th century with its distillation and were closely connected with the development of pharmacy. The first major chemical research was conducted at the beginning of the 20th century with its fractionation. The first isolated constituents from propolis were vanillin, cinnamic acid, and cinnamyl alcohol. Even bigger breakthrough happened at the beginning of 1970s with the advances in chromatographic analytical methods, which enabled isolation of newer and newer components from different propolis samples [10]. By the beginning of 21st century, Marcucci [6] and Bankova et al. [11] registered more than 300 constituents in propolis and just between 2000 and 2012, at least 241 new compounds have been isolated from it. Subsequently, the number of constituents grew to over 500 by 2012 and is growing every year as new components are being discovered in propolis from different regions and plant origins [8]. Despite of the progress in pharmacology, the list of preparations and uses of propolis in today’s time is still enormous, mostly because of its antiseptic, bacteriostatic, antibacterial, antymycotic, antiviral, antiprotocoal, antioxidative, spasmyloic, choleric, astringent, anti-inflammatory, anesthetic, antitumor, immunostimulating, cystotatic, hepatoprotective, and other properties [7,12].

Propolis could be typified in several different ways. According to its “gatherers”, two main types of propolis are known, the first being “normal” propolis that are collected by honeybees and the second being so-called geopropolis that are collected by stingless bees, which also add soil to their propolis mixtures [5,13]. According to the plant sources, propolis has been classified into seven main types [14], including poplar propolis, which is the most widespread type of propolis (Europe, North America, non-tropical regions of Asia) [11], Baccharis or Brazil green propolis [15], Clusia or Brazil red propolis [16], eucalyptus propolis [17], Macaranga or Taiwanese green propolis [18], birch propolis [19], and Mediterranean propolis [20]. Meanwhile Graikou et al. [9] also classified propolis into seven...
Phenols, or sometimes referred to as polyphenols, are one of the most numerous and widely distributed groups of substances in the plant Kingdom. They are products of the secondary metabolism of plants. They can range from simple molecules, such as phenolic acids, to highly polymerized compounds, such as tannins. Their most characteristic feature is their aromatic ring and the alcohol (–OH) group associated with it. Phenols are further divided into at least 18 classes: simple phenols, benzoquinones, phenolic acids, acetophenones, phenylacetic acids, hydroxycinnamic acids, phenylpropenes, coumarins and isocoumarins, chromones, naftoquinones, xanthones, stilbenes, anthraquinones, flavonoids, lignans, neolignans, lignins, and condensed tannins. More than 8000 phenolic structures are known, most of them belong to the subclass of flavonoids (5000) [32,33].

Phenols are also the most abundant constituents in propolis, especially in those of poplar origin. On average, they represent around 28 ± 9% of whole mass of poplar type propolis, of which 8 ± 4% are flavones/flavonols and 6 ± 2% are flavonones/dihydroflavonols. The isolated phenols belong to many different classes of compounds, such as flavonoids, lignans, stilbenes, phenylpropanoids (including different acids), and others, among which flavonoids are the most important molecules in propolis [8,11,34]. Ghisalberti [5] mentioned more than 20 isolated phenols in propolis until the year 1979 and until 1987, at least 59 different phenol constituents have been found in propolis samples [24]. The number rapidly grew and Marcucci [6] reported 100 phenol constituents isolated from propolis until the year 1995. From 1995 to 2000, Bankova et al. [11] reported 40 new phenols and from 2000 to 2012, astounding 184 new phenols have been isolated [8]. Additional six were found in 2011 [35] and thirteen in 2012 [36,37], which were previously not included in Huang et al. [8]. Altogether, at least 330 phenols have been isolated in propolis until the year 2012 and despite those numbers, just between 2013 and 2018, 92 flavonoids (including their glycosides) and altogether 218 new phenols were isolated from propolis, which brings the final number of isolated phenols from propolis to at least 548 until 2018.

From all the constituents, phenols (such as flavonoids, lignans, caffeoylquinic acid derivatives, and hydroxycinnamic acid derivatives) and terpenes are also thought to be the main active molecules of propolis from temperate climates, whereas for the tropical regions and also some Mediterranean regions, the predominant active constituents of propolis are phenols, different from those found in poplar propolis (prenylated p-coumaric and cinnamic acids, lignans, stilbenes), and diterpenic acids [1,3,8,9,11,34]. Owing to their abundance and activity, phenols are regarded as the most important constituents of propolis [38,39].

As mentioned, among phenols, flavonoids are the most important propolis constituents, acting as the main biologically active ingredients. They are also used in determining the quality of propolis samples [39]. Walker and Crane [24] reported at least 40 known flavonoids from propolis, whereas Marcucci [6] reported at least 44 in 1995. Bankova et al. [11] mentioned seven newly isolated flavonoids in propolis between 1995 and 2000, but just between 2000 and 2012, an astounding 113 new flavonoids were isolated [8]. Despite high numbers of already isolated flavonoids, 92 (including their glycosides) were discovered in propolis for the first time between 2013 and 2018. According to their chemical structure, isolated flavonoids are classified into 11 subclasses: flavans, isoflavans, flavanones, flavonolones, flavones, isoflavones, isodihydroflavonones, flavonoids, xanthones, chlorogens, and dihydrochalcones, and

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**2. PHENOLS AND FLAVONOIDS**

Phenols, or sometimes referred to as polyphenols, are one of the most numerous and widely distributed groups of substances in the...
neoflavonoids (Figure 1). Besides flavonoids, their glycosides are also being discovered in propolis, although until 2012 they were considered very rare. Only two flavonoid glycosides were isolated from propolis until 2009 and until 2012 their number grew only by one [8,40]. Yet in the past 6 years, 57 flavonoid glycosides were isolated from propolis for the first time, making flavonoid glycosides an important group of compounds in propolis samples. In 2004, some speculations were made that propolis samples could also contain anthocyanidins, although they have yet to be reported in propolis [41].

![Figure 1](image)

**Figure 1** Flavonoid groups/classes isolated from different propolis samples around the world.

Among the 92 newly isolated flavonoids, their glycosides (57) are one of the biggest discoveries in the recent years because of their earlier rarity. They were isolated from European (Serbia, United Kingdom, and Portugal) and South American (Brazil) samples. Some other new flavonoid compounds were also isolated from Middle East (Oman and Saudi Arabia), Europe (France and Serbia), Asia (Thailand, Korea, and Fiji), Middle and South America (Ecuador, Argentina, and Brazil), and Africa (Algeria, Congo, Cameroon, and Nigeria). The newly isolated flavonoids are listed in Table 1.

| No. | Chemical name | Geographic location | References |
|-----|---------------|---------------------|------------|
| 1   | Fisetinidol<sup>a</sup> | Oman | Popova et al. [42] |
| 2   | 2,3-trans-3,4-trans Mollissacidin<sup>a</sup> | Oman | Popova et al. [42] |
| 3   | 2,3-trans-3,4-cis Mollissacidin<sup>a</sup> | Oman | Popova et al. [42] |
| 4   | 3,4-Dihydro-2-(3,4-dihydroxyphenyl)-2H-chromene-3,7-diol<sup>a</sup> | Saudi Arabia | Almutairi et al. [43] |
| 5   | 8-[E-phenylprop-2-en-1-on]-5-methoxy-(±)-catechin<sup>a</sup> | France | Boisard et al. [44] |
| 6   | 8-[1-(4′-Hydroxy-3′-methoxyphenyl)prop-2-en-1-yl]-(2S)-pinocembrin<sup>a</sup> | Thailand | Athikomkulchai et al. [45] |
| 7   | 5,4′-Dihydroxy-7,3′-dimethoxyflavanone<sup>a</sup> | Ecuador | Cuesta-Rubio et al. [46] |
| 8   | Mepuberin<sup>b</sup> | Brazil | Casilotto et al. [47] |
| 9   | Pinobanksin 3-(E)-caffeate<sup>a</sup> | Algeria | Piccinelli et al. [48] |
| 10  | 3,5,4′-Trihydroxy-7,3′-dimethoxy flavanol<sup>a</sup> | Ecuador | Cuesta-Rubio et al. [46] |
| 11  | Psiadiarabin<sup>a</sup> | Saudi Arabia | Almutairi et al. [43] |
| 12  | Tangeritin<sup>a</sup> | Serbia | Ristivojević et al. [49] |
| 13  | 5,7-Dihydroxy-6,4′-dimethoxy flavone (pectolinarigenin)<sup>a</sup> | Algeria | Segueni et al. [50] |
| 14  | 6,7-Dihydroxy-7,4′-dimethoxy flavone (ladanein)<sup>a</sup> | Algeria | Segueni et al. [50] |
| 15  | 5,7-Dihydroxy-2-(3,4-dihydroxyphenoxy)-4H-chromen-4-one (2-phenoxychromone)<sup>a</sup> | Brazil | Mitsui et al. [51] |

(Continued)
| No. | Chemical name                                                                 | Geographic location       | References                  |
|-----|------------------------------------------------------------------------------|---------------------------|----------------------------|
| 16  | Pinobanksin-<i>O</i>-hexenoate<sup>●</sup>                                  | Portugal                   | Falçio et al. [52]          |
| 17  | 3,5,3′,4′-Tetrahydroxy-6,7-dimethoxy flavone (eupatolin)<sup>●</sup>          | Ecuador                    | Cuesta-Rubio et al. [46]    |
| 18  | (<i>E</i>-4′-methoxy-4,2′-dihydroxy-3′-(2′,3′,5′-trihydroxy-3′-methylbutyl)-chalcone (Jejuchalcone A)<sup>●</sup> | Korea                     | Shimomura et al. [53]       |
| 19  | (<i>E,E</i>-4,2′-trihydroxy-3′-(7′-hydroxy-3′,7′-dimethyloct-2′,5′-dieneyl)-chalcone (Jejuchalcone B)<sup>●</sup> | Korea                     | Shimomura et al. [53]       |
| 20  | (<i>E</i>-4,2′-trihydroxy-3′-(5′-hydroxy-3′,7′-dimethyloct-2′,6′-dieneyl)-chalcone (Jejuchalcone C)<sup>●</sup> | Korea                     | Shimomura et al. [53]       |
| 21  | (<i>E</i>-4′-methoxy-3′,4′-trihydroxy-2′,2′-dimethylidihydropyranono-(2′,3′)-chalcone (Jejuchalcone D)<sup>●</sup> | Korea                     | Shimomura et al. [53]       |
| 22  | (<i>E</i>-4′-methoxy-3′,4′-dihydroxy-2′-(1′″-hydroxyisopropyl)-dihydrofurano-(2′,3′)-chalcone (Jejuchalcone E)<sup>●</sup> | Korea                     | Shimomura et al. [53]       |
| 23  | (<i>E</i>-4,2′-dihydroxy-2′-methyl-2′-(3′,4′-dihydroxy-4′-methylenpentanyl)-2H-pyranono-(3′,4′)-chalcone (−)-Jejuchalcone F)<sup>●</sup> | Korea                     | Shimomura et al. [53]       |
| 24  | (<i>E</i>-4,2′-dihydroxy-2′-methyl-2′-(3′,4′-dihydroxy-4′-methylenpentanyl)-2H-pyranono-(3′,4′)-chalcone (−)-Jejuchalcone G)<sup>●</sup> | Korea                     | Shimomura et al. [53]       |
| 25  | (−)-(E)-4,2′-dihydroxy-2′-methyl-2′-(3′,4′-dihydroxy-4′-methylenpentanyl)-2H-pyranono-(3′,4′)-chalcone ((−)-Jejuchalcone H)<sup>●</sup> | Korea                     | Shimomura et al. [53]       |
| 26  | Quercetin-3-<i>O</i>-glucuronide<sup>●</sup>                                | Portugal                   | Falçio et al. [52]          |
| 27  | Quercetin-3-<i>O</i>-glucoside<sup>●</sup>                                  | Portugal                   | Falçio et al. [52]          |
| 28  | Kaempferol-3-<i>O</i>-rutinoside<sup>●</sup>                               | Portugal                   | Falçio et al. [52]          |
| 29  | Isorhamnetin-<i>O</i>-pentoside<sup>●</sup>                                | Portugal                   | Falçio et al. [52]          |
| 30  | Quercetin-3-<i>O</i>-harmoside<sup>●</sup>                                 | Portugal                   | Falçio et al. [52]          |
| 31  | Isorhamnetin-<i>O</i>-glucuronide<sup>●</sup>                             | Portugal                   | Falçio et al. [52]          |
| 32  | Kaempferol-<i>β</i>-methyl ether-<i>O</i>-glucoside<sup>●</sup>             | Portugal                   | Falçio et al. [52]          |
| 33  | Isorhamnetin-<i>O</i>-acetylrutinoside<sup>●</sup>                         | Portugal                   | Falçio et al. [52]          |
| 34  | Rhamnetin-<i>O</i>-glucuronide<sup>●</sup>                                | Portugal                   | Falçio et al. [52]          |
| 35  | Quercetin-<i>dimethyl</i> ether-<i>O</i>-rutinoside<sup>●</sup>            | Portugal                   | Falçio et al. [52]          |
| 36  | Quercetin-<i>dimethyl</i> ether-<i>O</i>-glucuronide<sup>●</sup>           | Portugal                   | Falçio et al. [52]          |
| 37  | Kaempferol-<i>O</i>-<i>ρ</i>-coumaroylrhamnoside<sup>●</sup>               | Portugal                   | Falçio et al. [52]          |
| 38  | Caffeic acid 4-<i>O</i>-glucoside<sup>●</sup>                             | Brazil                     | Righi et al. [54]           |
| 39  | Caffeic acid 4-<i>O</i>-arabinoside<sup>●</sup>                           | Brazil                     | Righi et al. [54]           |
| 40  | Caffeic acid 4-<i>O</i>-xyloside<sup>●</sup>                              | Brazil                     | Righi et al. [54]           |
| 41  | Dimethoxy-luteolin-glucoside<sup>●</sup>                                  | Brazil                     | Righi et al. [54]           |
| 42  | Methylkaempferol-<i>O</i>-rutinoside<sup>●</sup>                          | Brazil                     | Righi et al. [54]           |
| 43  | Naringenin-<i>C</i>-glucoside<sup>●</sup>                                | Brazil                     | Righi et al. [54]           |
| 44  | Apigenin-<i>O</i>-rutinoside<sup>●</sup>                                 | Brazil                     | Righi et al. [54]           |
| 45  | Delphinidin arabinoside<sup>●</sup>                                       | Brazil                     | Righi et al. [54]           |
| 46  | Catechin arabinoside<sup>●</sup>                                          | Brazil                     | Righi et al. [54]           |
| 47  | Apigenin-di-<i>C</i>-glucosyl rhamnoside<sup>●</sup>                     | Brazil                     | Righi et al. [54]           |
| 48  | Apigenin-<i>C</i>-harmoside (isomer 1)<sup>●</sup>                       | Brazil                     | Righi et al. [54]           |
| 49  | Apigenin-6,8-<i>D</i>-glucosyl C-<i>Arabinose (isochaftoside)</sup>          | Brazil                     | Righi et al. [54]           |
| 50  | Apigenin-6,8-<i>D</i>-glucosyl C-arabinose (schaftoside)<sup>●</sup>        | Brazil                     | Righi et al. [54]           |
| 51  | Apigenin-6-<i>C</i>-glucosyl-8-<i>C</i>-arabinose (isochaftoside)<sup>●</sup> | Brazil                     | Righi et al. [54]           |
| 52  | Luteolin-<i>O</i>-glucuronide<sup>●</sup>                                | Brazil                     | Righi et al. [54]           |
| 53  | Apigenin-8-C-glucosyl-6-C-arabinose (schaftoside)<sup>●</sup>              | Brazil                     | Righi et al. [54]           |
| 54  | Luteolin-6,8-di-<i>D</i>-glucoside (lucenin-2)<sup>●</sup>                | Brazil                     | Righi et al. [54]           |
| 55  | Apigenin-<i>C</i>-harmoside (isomer 2)<sup>●</sup>                       | Brazil                     | Righi et al. [54]           |
| 56  | Luteolin acetylglicoside<sup>●</sup>                                      | Brazil                     | Righi et al. [54]           |
| 57  | Chrysoeriol-<i>C</i>-glucoside<sup>●</sup>                                | Brazil                     | Righi et al. [54]           |
| 58  | Dimethoxy naringenin-diglicoside<sup>●</sup>                             | Brazil                     | Righi et al. [54]           |
| 59  | Apigenin-di-<i>O</i>-glucoside<sup>●</sup>                                | Brazil                     | Righi et al. [54]           |
| 60  | Quercetin-<i>O</i>-arabinoside<sup>●</sup>                               | Brazil                     | Righi et al. [54]           |
| 61  | Isorhamnetin-glucoside<sup>●</sup>                                       | Brazil                     | Righi et al. [54]           |
| 62  | Apigenin-<i>O</i>-glucuronide<sup>●</sup>                                | Brazil                     | Righi et al. [54]           |
| 63  | Naringenin-4′-<i>O</i>-<i>β</i>-glucopyranoside<sup>●</sup>               | Brazil                     | Da Silva et al. [55]        |
| 64  | Myricetin-3-<i>O</i>-<i>β</i>-glucopyranoside<sup>●</sup>                 | Brazil                     | Da Silva et al. [55]        |
| 65  | Chrysin glycoside formate adduct<sup>●</sup>                             | United Kingdom             | Saleh et al. [56]           |
| 66  | Galangin glycoside<sup>●</sup>                                            | United Kingdom             | Saleh et al. [56]           |

(Continued)
Table 1  Flavonoids identified in propolis for the first time since 2011—Continued

| No. | Chemical name                                      | Geographic location | References          |
|-----|---------------------------------------------------|---------------------|---------------------|
| 67  | 7′-Methoxy-5′-hydroxy-8′-C-flavone rhamnosidea    | Brazil              | Coelho et al. [57]  |
| 68  | Acacetin-di-C-acetyl dirhamnosideb                | Brazil              | Coelho et al. [57]  |
| 69  | Apigenin-di-C-malonyl glucoside dihexoside (isomer 1)k,c | Brazil              | Coelho et al. [57]  |
| 70  | Apigenin-di-C-malonyl glucoside dihexoside (isomer 2)k,c | Brazil              | Coelho et al. [57]  |
| 71  | Apigenin-di-C-malonyl trihexosidec               | Brazil              | Coelho et al. [57]  |
| 72  | Acacetin-di-C-malonyl trihexosidec               | Brazil              | Coelho et al. [57]  |
| 73  | Apigenin-di-C-malonyl trihexoside (isomer 2)k,c   | Brazil              | Coelho et al. [57]  |
| 74  | Acacetin-8-C-arabinoside-7-O-rhamnosideb         | Brazil              | Coelho et al. [57]  |
| 75  | Apigenin-di-C-malonyl trihexoside (isomer 3)k,c   | Brazil              | Coelho et al. [57]  |
| 76  | Catechin rhamnosideb                             | Brazil              | Coelho et al. [57]  |
| 77  | Chrysin-8-C-rhamnoside-7-O-rhamnosideb           | Brazil              | Coelho et al. [57]  |
| 78  | Luteolin-8-C-caffeoyl rhamnosideb               | Brazil              | Coelho et al. [57]  |
| 79  | Caffeoylquinic acid-O-arabinosideb               | Brazil              | Coelho et al. [57]  |
| 80  | Apigenin-7-O-glucoside (apigenin)b               | Serbia              | Ristivojević et al. [49] |
| 81  | Apigenin 8-C-xylloside-6-C-glucoside (vicenin 3)b | Brazil              | Cisilotto et al. [47] |
| 82  | Apigenin 6-C-xylloside-8-C-glucoside (vicenin 1)b | Brazil              | Cisilotto et al. [47] |

Prenylated flavonoids

| No. | Chemical name                                      | Geographic location | References          |
|-----|---------------------------------------------------|---------------------|---------------------|
| 83  | 7′-O-methyl-8′-prenylnaringeninb                 | Oman                | Popova et al. [42]  |
| 84  | 3′,8′-Diprenylnaringeninc                        | Oman                | Popova et al. [42]  |
| 85  | 8′-Prenyl-5,7′-dihydroxy-3′-(3-hydroxy-3-methylbutyl)-4′-methoxyflavononea | Oman                | Popova et al. [42]  |
| 86  | Lonchocarpol Aa                                  | Congo and Cameroon  | Papachroni et al. [58] |
| 87  | 6,8-Diprenyl-eriodictyloia                       | Congo               | Papachroni et al. [58] |
| 88  | 6,8-Diprenyl-aromadendrinb                      | Cameroon            | Papachroni et al. [58] |
| 89  | Lespedezaflavonaneabc                           | Cameroon            | Papachroni et al. [58] |
| 90  | Glyasperin Aa                                   | Fiji                | Trusheva et al. [59] |
| 91  | 8′-Prenylnaringeninb                            | Nigeria             | Omar et al. [60]    |
| 92  | 6′-Prenylnaringeninb                            | Nigeria             | Omar et al. [60]    |

The molecular structure of the compound is not completely defined. *Constituent isolated from the honeybee propolis (from the genus *Apis* sp.). †Constituent isolated from the stingless bee propolis (from genera *Scautothrigona* sp. or *Melipona* sp.). ‡Newly discovered compound. Compounds already mentioned in Huang et al. [8] are excluded.

Among other 126 isolated “non-flavonoid” phenols, compounds from stilbenes and phenolic acids groups were the most abundant. Five of the isolated phenols were found in 2011 [35], whereas one phenolic acid ester [36] and twelve phenylpropanoids [37] were isolated in 2012. All of them are included in this review as they were excluded from the previous review article [8]. Otherwise, phenols were isolated from propolis from Europe (Italy, Portugal, Serbia, and United Kingdom), South and Middle America (Chile, Honduras, Brazil, and Argentina), Africa (Egypt, Nigeria, Ghana, Algeria, and Cameroon), Asia (Thailand, Fiji, China, and Malaysia), Middle East (Saudi Arabia), Oceania (Australia), and North America (United States and Mexico). The phenols are listed in Table 2.

Table 2  Phenolic compounds identified in propolis for the first time since 2011

| No. | Chemical name                                      | Geographic location | References          |
|-----|---------------------------------------------------|---------------------|---------------------|
| 93  | Caffeoyl glycerolb                                | Serbia              | Ristivojević et al. [49] |
| 94  | Tricoumaroyl glycerolb                           | Serbia              | Ristivojević et al. [49] |
| 95  | Coumaroyl feruloyl glycerol (isomer 1)b           | Serbia              | Ristivojević et al. [49] |
| 96  | Coumaroyl feruloyl glycerol (isomer 2)b           | Serbia              | Ristivojević et al. [49] |
| 97  | Dicaffeoyl coumaroyl glycerolb                   | Serbia              | Ristivojević et al. [49] |
| 98  | Dicaffeoyl feruloyl glycerolb                    | Serbia              | Ristivojević et al. [49] |

Phenolic acid esters

| No. | Chemical name                                      | Geographic location | References          |
|-----|---------------------------------------------------|---------------------|---------------------|
| 99  | (E)-cinnamyl-(Z)-cinnamatea                        | Honduras            | Lotti et al. [36]    |
| 100 | Decyl caffeatea                                   | Egypt               | El-Hady et al. [61]  |
| 101 | Caffeic acid phenacyl estera                       | United Kingdom      | Saleh et al. [56]    |
| 102 | Caffeic acid sesquiterpene ester (isomer 1)b       | United Kingdom      | Saleh et al. [56]    |
| 103 | Caffeic acid sesquiterpene ester                  | United Kingdom      | Saleh et al. [56]    |
| 104 | Methylgalangin hydroxypropionyl ester             | United Kingdom      | Saleh et al. [56]    |
| 105 | Caffeic acid monoterpene(geranyl) ester            | United Kingdom      | Saleh et al. [56]    |
| 106 | Methyl methylene dioxy kaempferol hexanoyl ester (isomer 1)b | United Kingdom      | Saleh et al. [56]    |
| 107 | Methyl methylene dioxy kaempferol hexanoyl ester (isomer 2)b | United Kingdom      | Saleh et al. [56]    |
| 108 | Caffeic acid sesquiterpene ester (isomer 2)b       | United Kingdom      | Saleh et al. [56]    |

Xanthones

| No. | Chemical name                                      | Geographic location | References          |
|-----|---------------------------------------------------|---------------------|---------------------|
| 109 | α-Mangostinb                                      | Thailand            | Sanpa et al. [4]    |

(Continued)
Table 2  Phenolic compounds identified in propolis for the first time since 2011—Continued

| No. | Chemical name                                      | Geographic location | References                  |
|-----|----------------------------------------------------|---------------------|-----------------------------|
| 110 | γ-Mangostin<sup>a</sup>                           | Thailand            | Sanpa et al. [4]            |
| 111 | Mangostin<sup>a</sup>                             | Thailand            | Sanpa et al. [4]            |
| 112 | 8-Deoxygartanin<sup>b</sup>                       | Thailand            | Sanpa et al. [4]            |
| 113 | Gartanin<sup>b</sup>                              | Thailand            | Sanpa et al. [4]            |
| 114 | Garcinone B<sup>b</sup>                           | Thailand            | Sanpa et al. [4]            |
| 115 | Furofuran lignan methylpinoresinol<sup>b</sup>     | Thailand            | Sanpa et al. [4]            |
| 116 | 2-Acetyl-1-feruloyl-3-cafeoylglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 117 | (+)-2-Acetyl-1-cafeoyl-3-cinnamoylglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 118 | (−)-2-Acetyl-1-cafeoyl-3-cinnamoylglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 119 | (+)-2-Acetyl-1-(E)-coumaroyl-3-(E)-cinnamoylglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 120 | (−)-2-Acetyl-1-(E)-feruloyl-3-(E)-cinnamoylglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 121 | (−)-2-Acetyl-1-(E)-feruloyl-3-(E)-cinnamoylglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 122 | 2-Acetyl-1,3-dicinnamoylglycerol<sup>c</sup>      | China               | Shi et al. [37]             |
| 123 | (−)-2-Acetyl-1-(E)-cinnamoyl-3-(3′(ζ),16″)-dihydroxy-palmitoyleglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 124 | 2-Acetyl-1,3-dicaffeoylglycerol<sup>c</sup>        | China               | Shi et al. [37]             |
| 125 | 2-Acetyl-1-feruloyl-3-coumaroylglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 126 | 2-Acetyl-1-feruloyl-3-coumaroylglycerol<sup>c</sup> | China               | Shi et al. [37]             |
| 127 | 2-Acetyl-1,3-diferuloylglycerol<sup>c</sup>       | China               | Shi et al. [37]             |
| 128 | 6-O-p-coumaroyl-D-galactopyranose<sup>c</sup>     | Brazil              | De Souza et al. [13]       |
| 129 | 6-O-cinnamoyl-1-O-p-coumaroyl-β-D-glucopyranose<sup>b</sup> | Brazil              | De Souza et al. [13]       |
| 130 | Dicoumaroyl glycerol<sup>b</sup>                  | United Kingdom      | Saleh et al. [56]           |
| 131 | Acetyl coumaroyl glycerol<sup>b</sup>             | United Kingdom      | Taddeo et al. [62]          |
| 132 | Boropic acid<sup>b</sup>                         | Italy               | Taddeo et al. [62]          |
| 133 | 4′-Geranylxyferulic acid<sup>b</sup>              | Italy               | Taddeo et al. [62]          |
| 134 | 7-Isopentenyloxycoumarin<sup>b</sup>              | Italy               | Taddeo et al. [62]          |
| 135 | Aurapten<sup>b</sup>                             | Italy               | Taddeo et al. [62]          |
| 136 | Scopolin<sup>a</sup>                             | Algeria             | Soltani et al. [31]        |

Phenylpropanoid glycosides

| 137 | Torachrysone-O-hexose<sup>c</sup>                | Malaysia            | Zhao et al. [63]           |
| 138 | Torachrysone-O-(acetyl)-hexose<sup>c</sup>       | Malaysia            | Zhao et al. [63]           |
| 139 | Torachrysone-O-(galloyl)-hexose<sup>c</sup>      | Malaysia            | Zhao et al. [63]           |
| 140 | Gallic acid-hexose<sup>c</sup>                    | Malaysia            | Zhao et al. [63]           |

Stilbenes

| 141 | (E)-4-(3-methyl-2-butene-1-yl)-3,4,5-trihydroxy-3′-methoxystilbene<sup>c</sup> | Australia           | Duke et al. [64]           |
| 142 | (E)-2-(3-methyl-2-butene-1-yl)-3,4,5-trihydroxy-stilbene (2-prenylresveratrol)<sup>a</sup> | Australia           | Duke et al. [64]           |
| 143 | (E)-2,4-bis-(3-methyl-2-butene-1-yl)-3,3′,4,′5-tetrahydroxy-stilbene<sup>a</sup> | Australia           | Duke et al. [64]           |
| 144 | (E)-2-(3-methyl-2-butene-1-yl)-3-(3-methyl-2-butenoyloxy)-3′,4,′5-trihydroxy-stilbene<sup>a</sup> | Australia           | Duke et al. [64]           |
| 145 | (E)-2,6-bis-(3-methyl-2-butene-1-yl)-3,3′,5,′5′-tetrahydroxy-stilbene<sup>a</sup> | Australia           | Duke et al. [64]           |
| 146 | (E)-2,6-bis-(3-methyl-2-butene-1-yl)-3′,4′,5′-trihydroxy-3′′-methoxystilbene<sup>a</sup> | Australia           | Duke et al. [64]           |
| 147 | (E)-5-(2-(8-hydroxy-2-methyl-2′-(4-methylpent-3-en-1-yl)-2H-chromen-6-yl)vinyl)-2-(3-methylbut-2-en-1-yl)benzene-1,3-diol<sup>a</sup> | Ghana               | Almutairi et al. [65]     |
| 148 | 5′-(E)-5,5-di-dihydroxystyryl)·3·((E)-3,7-dimethylocta-2,6-dien-1-yl)benzene-1,2-diol<sup>a</sup> | Ghana               | Almutairi et al. [65]     |
| 149 | Schweinfurthin C<sup>c</sup>                      | Nigeria             | Zhang et al. [56]          |
| 150 | Mappain<sup>c</sup>                              | Nigeria             | Zhang et al. [56]          |
| 151 | Geranyl stilbenoid<sup>c</sup>                   | Nigeria             | Zhang et al. [56]          |
| 152 | Solomomin B<sup>d</sup>                          | Fiji                | Trusheva et al. [59]      |
| 153 | Solomomin C<sup>d</sup>                          | Fiji                | Trusheva et al. [59]      |

Lignans

| 154 | Meso- (rel 7S,8S),(R,8′R)-3,4,5′-tetrahydroxy-7,7′-epoxy lignan<sup>a</sup> | Argentina           | Agüero et al. [35]        |
| 155 | (7S,8S),(R,8′S)-3,4,5′-tetrahydroxy-4-methoxy-7,7′-epoxy lignan<sup>a</sup> | Argentina           | Agüero et al. [35]        |

Phenolic acids

| 156 | Caffeic acid derivative 1<sup>c</sup>             | Portugal            | Falcão et al. [52]        |
| 157 | Caffeic acid derivative 1 (isomer)<sup>c</sup>   | Portugal            | Falcão et al. [52]        |
| 158 | Caffeic acid derivative 2<sup>c</sup>             | Portugal            | Falcão et al. [52]        |
| 159 | Ferulic acid derivative<sup>c</sup>              | Portugal            | Falcão et al. [52]        |
| 160 | Sandaracopimaric acid<sup>d</sup>                | Saudi Arabia        | Jer et al. [67]           |
| 161 | (E)-3-hydroxy-1,7-diphenyleth-1-ene-5-acetate<sup>e</sup> | Chile               | Nina et al. [68]          |
| 162 | (E)-5-hydroxy-1,7-diphenyleth-1-ene-3-acetate<sup>e</sup> | Chile               | Nina et al. [68]          |
Table 2  Phenolic compounds identified in propolis for the first time since 2011—Continued

| No. | Chemical name                                                                 | Geographic location | References                  |
|-----|-------------------------------------------------------------------------------|---------------------|-----------------------------|
| 163 | Caffeic acid hextrieneoate<sup>a</sup>                                         | United Kingdom      | Saleh et al. [56]           |
| 164 | Benzoyl dihydroxyphenylpropionic acid<sup>b</sup>                             | United Kingdom      | Saleh et al. [56]           |
| 165 | Benzoyl hydroxyphenylacetic acid<sup>b</sup>                                  | United Kingdom      | Saleh et al. [56]           |
| 166 | Hydroxy phenyl acetyl dihydroxyphenylacetic acid<sup>c</sup>                  | United Kingdom      | Saleh et al. [56]           |
| 167 | Pinobanksin phenyl propionate (isomer 1)<sup>d</sup>                          | United Kingdom      | Saleh et al. [56]           |
| 168 | Dimethyl pinocembrin benzoate<sup>e</sup>                                     | United Kingdom      | Saleh et al. [56]           |
| 169 | Pentenoyl hydroxyphenylpropionic acid<sup>c</sup>                             | United Kingdom      | Saleh et al. [56]           |
| 170 | Pinobanksin phenyl propionate (isomer 2)<sup>d</sup>                          | United Kingdom      | Saleh et al. [56]           |
| 171 | Pinobanksin benzoate<sup>e</sup>                                              | United Kingdom      | Saleh et al. [56]           |
| 172 | Pinobanksin phenyl propionate (isomer 3)<sup>d</sup>                          | United Kingdom      | Saleh et al. [56]           |
| 173 | Methyl pinobanksin acetate<sup>e</sup>                                        | United Kingdom      | Saleh et al. [56]           |
| 174 | Pinobanksin caffeate<sup>e</sup>                                               | United Kingdom      | Saleh et al. [56]           |
| 175 | Caffeoyl(dimethyl pinocembrin<sup>e</sup>                                       | United Kingdom      | Saleh et al. [56]           |
| 176 | Methyl chrysin acetate derivative<sup>e</sup>                                 | United Kingdom      | Saleh et al. [56]           |
| 177 | Pinobanksin dimethyl cinnamate<sup>e</sup>                                     | United Kingdom      | Saleh et al. [56]           |
| 178 | (4R,5R,9R,10R)-13-hydroxypodocarp-8(14)-en-19-oic acid<sup>b</sup>             | Brazil              | Cisilotto et al. [47]       |

Other phenols

| No. | Chemical name                                                                 | Geographic location | References                  |
|-----|-------------------------------------------------------------------------------|---------------------|-----------------------------|
| 179 | Nordihydroguaiaretic acid<sup>a</sup>                                         | Argentina           | Agüero et al. [35]          |
| 180 | 3'-Methyl-nordihydroguaiaretic acid<sup>a</sup>                               | Argentina           | Agüero et al. [35]          |
| 181 | 4'-Methyl-nordihydroguaiaretic acid<sup>a</sup>                               | Argentina           | Agüero et al. [35]          |
| 182 | (E)-cinnamyl-(E)-cinnamylidenate<sup>a</sup>                                 | Thailand            | Athikomkulchai et al. [45]  |
| 183 | Kaempferol-dimethyl ether<sup>e</sup>                                          | Portugal            | Falcão et al. [52]          |
| 184 | 5-Hexadecylresorcinol<sup>e</sup>                                              | Cameroon            | Kardar et al. [69]          |
| 185 | 5-(10Z-pentadecenyl)-resorcinol<sup>e</sup>                                   | Cameroon            | Kardar et al. [69]          |
| 186 | 5-(12Z-heptadecenyl)-resorcinol<sup>e</sup>                                   | Cameroon            | Kardar et al. [69]          |
| 187 | 5-(14Z-heptadecenyl)-resorcinol<sup>e</sup>                                   | Cameroon            | Kardar et al. [69]          |
| 188 | 5-(14Z-nonadecenyl)-resorcinol<sup>e</sup>                                    | Cameroon            | Kardar et al. [69]          |
| 189 | 3-Undecyl phenol<sup>e</sup>                                                   | Cameroon            | Kardar et al. [69]          |
| 190 | 3-Tetradecylyphenol<sup>e</sup>                                                | Cameroon            | Kardar et al. [69]          |
| 191 | 3-Pentadecylyphenol<sup>e</sup>                                                | Cameroon            | Kardar et al. [69]          |
| 192 | 3-Hexadecylyphenol<sup>e</sup>                                                 | Cameroon            | Kardar et al. [69]          |
| 193 | 3-Heptadecylyphenol<sup>e</sup>                                                | Cameroon            | Kardar et al. [69]          |
| 194 | 3-Nonadecylyphenol<sup>e</sup>                                                 | Cameroon            | Kardar et al. [69]          |
| 195 | 3-(10Z-pentadecenyl)-phenol<sup>e</sup>                                        | Cameroon            | Kardar et al. [69]          |
| 196 | 3-(12Z-pentadecenyl)-phenol<sup>e</sup>                                       | Cameroon            | Kardar et al. [69]          |
| 197 | 3-(8Z-heptadecenyl)-phenol<sup>e</sup>                                        | Cameroon            | Kardar et al. [69]          |
| 198 | 3-(12Z-heptadecenyl)-phenol<sup>e</sup>                                       | Cameroon            | Kardar et al. [69]          |
| 199 | 3-(14Z-heptadecenyl)-phenol<sup>e</sup>                                       | Cameroon            | Kardar et al. [69]          |
| 200 | 3-(13Z-nonadecenyl)-phenol<sup>e</sup>                                         | Cameroon            | Kardar et al. [69]          |
| 201 | 3-(14Z-nonadecenyl)-phenol<sup>e</sup>                                        | Cameroon            | Kardar et al. [69]          |
| 202 | Deperoxidized derivative of phuketione C<sup>a</sup>                           | Cameroon            | Almutairi et al. [65]       |
| 203 | 1.3-Dihydroxy-5-heptadecylenzene<sup>a</sup>                                 | Egypt               | El-Hady et al. [61]         |
| 204 | 1.3-Dihydroxy-5-heptadecylenzene (C17:0) derivative<sup>a</sup>              | Egypt               | El-Hady et al. [61]         |
| 205 | 1,3-Dihydroxy-5-heptadecylenzene (C19:1) derivative<sup>a</sup>              | Egypt               | El-Hady et al. [61]         |
| 206 | (E)-4-(3'-ethoxypropyl-1'-enylphenol (Ethyl p-coumaroyl ether)<sup>a</sup>    | United States       | Sarka et al. [70]           |
| 207 | Coumaric acid cinnamyl ether<sup>e</sup>                                       | United Kingdom      | Saleh et al. [56]           |
| 208 | Dimethyl kaempferol phenethyl ether<sup>e</sup>                               | United Kingdom      | Saleh et al. [56]           |
| 209 | Dihydroxy propionyl pinocembrin methyl ether<sup>e</sup>                      | United Kingdom      | Saleh et al. [56]           |
| 210 | Pinocembrin methyl ether (isomer 1)<sup>e</sup>                               | United Kingdom      | Saleh et al. [56]           |
| 211 | Dimethyl galangin phenacetl ether<sup>e</sup>                                 | United Kingdom      | Saleh et al. [56]           |
| 212 | Pinocembrin methyl ether (isomer 2)<sup>e</sup>                               | United Kingdom      | Saleh et al. [56]           |
| 213 | Hexadecenoil dimethyl pinobanksin<sup>e</sup>                                | United Kingdom      | Saleh et al. [56]           |
| 214 | Pinobanksin-5-methylthether-3-O-propanoate<sup>a</sup>                        | Mexico              | Allday et al. [71]          |
| 215 | Pinobanksin-5-methylthether-3-O-butryate<sup>a</sup>                         | Mexico              | Allday et al. [71]          |
| 216 | Tetragocarbone A<sup>a</sup>                                                   | Australia           | Nishimura et al. [72]       |
| 217 | Tetragocarbone B<sup>a</sup>                                                   | Australia           | Nishimura et al. [72]       |
| 218 | 3-(2-Hydroxy-4-methoxybenzyl)-6-methoxy-2,3-dihydrobenzofuran (Riverinol)<sup>a</sup> | Nigeria             | Omar et al. [60]            |

<sup>a</sup>The molecular structure of the compound is not completely defined. <sup>b</sup> Constituent isolated from the honeybee propolis (from the genus Apis sp.). <sup>c</sup> Constituent isolated from the stingless bee propolis (from genera Scaptotrigona sp., Melipona sp., Tetragonula sp., Trigona sp., Tetrigona sp., or Heterotrigona sp.). <sup>d</sup> Newly discovered compound. Compounds already mentioned in Huang et al. [8] are excluded.
2.1. Terpenoids

Terpenes and terpenoids are the biggest and most diverse group of secondary plant metabolites, which include more than 25,000 compounds. They are molecules composed from one or more isoprene (C5) units. Term terpene refers to a hydrocarbon molecule, whereas term terpenoid refers to hydrocarbon molecule that has been modified (e.g., addition of oxygen). Terpenes are further divided into seven classes: hemiterpenes (C5), monoterpenes (C10), sesquiterpenes (C15), diterpenes (C20), triterpenes (C30), tetraterpenes (C40), and polyterpenes (C45 or more) [73]. They are the second biggest and most important group of compounds and also the most abundant volatile components of propolis [74]. As mentioned before, they are one of the main biologically active substances in propolis and they play a major role in determining its quality. Terpenes were found mainly in tropical propolis, being rarer in poplar propolis type, yet some of them were also isolated from the Mediterranean propolis. Sesquiterpenes are the main group of terpenes found in propolis and are further divided into acyclic, monocyclic, dicyclic, and tricyclic sesquiterpenes. Other important terpenes from propolis are monoterpenes, triterpenes, and diterpenes, latter being the most important terpene from the pharmacological point of view [8,20,40,75].

Walker and Crane [24] mentioned 18 isolated terpenoids from propolis and Marcucci [6] added another 11. Between 2000 and 2012 Huang et al. [8] reported 58 terpenoids isolated from propolis for the first time, whereas between 2013 and 2018 another 46 were reported. In total, at least 133 terpenes were isolated from propolis until 2018. Terpenes isolated between 2013 and 2018 were found in propolis samples from Africa (Cameroon, Algeria, Egypt, and Nigeria), Asia (Malaysia and Thailand), South America (Chile, Brazil, and Bolivia), Middle East (Saudi Arabia), and Oceania (Australia and Pitcairn Island). Otherwise, most of the newly isolated terpenoids after 2013 belong to the group of triterpenes. Newly isolated terpenoids are listed in Table 3.

![Terpenoid groups/classes isolated from different propolis samples around the world.](Image)

### Table 3 Terpenoids identified in propolis for the first time since 2011

| No. | Chemical name                           | Geographic location | References                  |
|-----|-----------------------------------------|---------------------|-----------------------------|
| 219 | 1,8-Terpineol<sup>a</sup>               | Cameroon            | Papachroni et al. [58]      |
| 220 | β-Panasinsene<sup>a</sup>               | Malaysia            | Tuan et al. [76]            |
| 221 | Cistadiol<sup>a</sup>                   | Algeria             | Piccinelli et al. [48]      |
| 222 | 18-Hydroxy-cis-clerodan-3-ene-15-oic acid<sup>a</sup> | Algeria | Piccinelli et al. [48] |
| 223 | Propsiadin ((ent)-2-oxo-kaur-16-en-6,18-diol)<sup>a</sup> | Saudi Arabia | Almutairi et al. [43] |
| 224 | Psidin<sup>a</sup>                      | Saudi Arabia        | Almutairi et al. [43]       |
| 225 | Poilaneic acid<sup>a</sup>              | Chile               | Nina et al. [68]            |
| 226 | 7,8,18-Trihydroxy serrulat-14-ene<sup>a</sup> | Australia | Aminimoghadamfarouj and Nematollahi [75] |
| 227 | 5,18-Epoxy serrulat-14-en-7,8-dione<sup>a</sup> | Australia | Aminimoghadamfarouj and Nematollahi [75] |
| 228 | (18R)-5,18-epoxy serrulat-14-en-8,18-diol<sup>a</sup> | Australia | Aminimoghadamfarouj and Nematollahi [75] |
| 229 | rel-(5S,6S,8R,9R,10S,18R,19S)-18,19-epoxy-2-oxoclerodan-3,12(E),14-triene-6,18,19-triol 18,19-diacetate 6-benzoate<sup>a</sup> | Brazil | Tazawa et al. [77] |
| 230 | Abietinal<sup>b</sup>                   | Pitcairn Island     | Georgieva et al. [78]       |
| 231 | 3β-Acetoxy-19(29)-taraxasten-20α-ol<sup>a</sup> | Saudi Arabia | Jerz et al. [67] |
| 232 | Pseudotaraxasterol-3β-O-acetate<sup>a</sup> | Saudi Arabia | Jerz et al. [67] |
| 233 | β-Sitosterol<sup>a</sup>               | Saudi Arabia        | Odiba et al. [79]           |
| 234 | 25-Cyclopropyl-3β-hydroxysyr-12-ene<sup>a</sup> | Cameroon | Sakava et al. [80] |
| 235 | Cycloart-3β-hydroxy-12,25(26)-diene<sup>a</sup> | Cameroon | Sakava et al. [80] |
| 236 | Lup-20(29)-en-3β-oate<sup>a</sup>      | Cameroon            | Sakava et al. [80]          |
| 237 | Olean-12-en-3β,28-diol (erythrodiol)<sup>a</sup> | Cameroon | Sakava et al. [80] |
| 238 | 3β-Teraxasterol-acetate<sup>a</sup>     | Cameroon            | Papachroni et al. [58]      |

(Continued)
2.2. Fatty Acids

Fatty acids are one of the “waxy” nonpolar parts of propolis, and Heinen and Linskens [83] were one of the first researchers who isolated fatty acids (ranging from C_{18} to C_{40}) from propolis. Until 2018, there were many more found in propolis and some authors reported them as long as C_{36} [84]. Despite the fact that fatty acids were discovered in propolis relatively soon, most of the authors do not mention them in their articles. They can be present in propolis as glycosides, free fatty acids, different types of esters, or others [56,84]. There are many different types of fatty acids found in propolis: saturated, monounsaturated, polyunsaturated, and even omega-3 and omega-6 fatty acids [85]. As they are not among the most widely reported compounds in propolis, between 2013 and 2018 only two authors reported fatty acids isolated from propolis for the first time. Among those reported, 13 were free fatty acids and 4 were fatty acid glycosides. All of them were from European samples (Bulgaria and United Kingdom). Details can be found in Table 4.

Table 3 | Terpenoids identified in propolis for the first time since 2011—Continued

| No. | Chemical name | Geographic location | References |
|-----|---------------|---------------------|------------|
| 239 | Taraxasterol acetate<sup>a</sup> | Cameroon | Papachroni et al. [58] |
| 240 | 3α-Hydroxy-olean-12-en-30-ol<sup>a</sup> | Cameroon | Papachroni et al. [58] |
| 241 | Bacchara-12,21-dien-3β-ol<sup>a</sup> | Cameroon | Papachroni et al. [58] |
| 242 | Betulinaldehyde<sup>a</sup> | Cameroon | Papachroni et al. [58] |
| 243 | 4,4-Dimethyl-3-oxocholest-5-en-7-one<sup>a</sup> | Egypt | El-Hady et al. [61] |
| 244 | 3-Oxo-cycloart-24-ene-21,26-diol-26-acetate<sup>a</sup> | Thailand | Sanpa et al. [4] |
| 245 | Dipterocarpos<sup>a</sup> | Thailand | Sanpa et al. [4] |
| 246 | 3-O-acetyl ursolic acid<sup>a</sup> | Thailand | Sanpa et al. [4] |
| 247 | Ocotillone I<sup>a</sup> | Thailand | Sanpa et al. [4] |
| 248 | Ocotillone II<sup>a</sup> | Thailand | Sanpa et al. [4] |
| 249 | Cabralealactone (isomer 1)<sup>b</sup> | Thailand | Sanpa et al. [4] |
| 250 | Cabralealactone (isomer 2)<sup>b</sup> | Thailand | Sanpa et al. [4] |
| 251 | Ursolic aldehyde<sup>a</sup> | Thailand | Sanpa et al. [4] |
| 252 | Oleanolic aldehyde<sup>a</sup> | Thailand | Sanpa et al. [4] |
| 253 | Cycloart-24-en-3β,26-diol<sup>b</sup> | Bolivia | Nina et al. [81] |
| 254 | Cycloart-24-en-3-one<sup>b</sup> | Bolivia | Nina et al. [81] |
| 255 | 24(E)-cycloart-24-en-26-ol-3-one<sup>b</sup> | Bolivia | Nina et al. [81] |
| 256 | Mangiferonic acid methyl ester<sup>b</sup> | Bolivia | Nina et al. [81] |
| 257 | Lup(20,29)-en-3-one<sup>b</sup> | Bolivia | Nina et al. [81] |
| 258 | Methyl-3β,27-dihydroxycycloart-24-en-26-oate<sup>a</sup> | Cameroon | Tallia et al. [81] |
| 259 | 20-Hydroxy-24-dammaren-3-one<sup>b</sup> | Malaysia | Zhao et al. [63] |
| 260 | 3-Oxo-cycloart-24-E-en-21,26-diol-21,26-diacetate<sup>a</sup> | Pitcairn Island | Georgieva et al. [78] |
| 261 | 3-Oxo-cycloart-24-E-en-21,26-diol-21-acetate<sup>b</sup> | Pitcairn Island | Georgieva et al. [78] |
| 262 | 3-Oxo-cycloart-24-E-en-21,26-diol-21-acetate<sup>b</sup> | Pitcairn Island | Georgieva et al. [78] |
| 263 | 3-Oxo-cycloart-24-E-en-26-al<sup>b</sup> | Pitcairn Island | Georgieva et al. [78] |

| Compounds already mentioned in Huang et al. [8]. Despite the fact that fatty acids were discovered in propolis relatively soon, most of the authors do not mention them in their articles. They can be present in propolis as glycosides, free fatty acids, different types of esters, or others [56,84]. There are many different types of fatty acids found in propolis: saturated, monounsaturated, polyunsaturated, and even omega-3 and omega-6 fatty acids [85]. As they are not among the most widely reported compounds in propolis, between 2013 and 2018 only two authors reported fatty acids isolated from propolis for the first time. Among those reported, 13 were free fatty acids and 4 were fatty acid glycosides. All of them were from European samples (Bulgaria and United Kingdom). Details can be found in Table 4.

Table 4 | Fatty acids and their glycosides identified in propolis for the first time since 2011

| No. | Chemical name | Geographic location | References |
|-----|---------------|---------------------|------------|
| 265 | 9-Oxo-(E)-12(Z)-octadecadienoic acid<sup>a</sup> | Bulgaria | Bilikova et al. [86] |
| 266 | Dihydroxylinolenic acid<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 267 | Dihydroxylinolenic acid (isomer 1)<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 268 | Dihydroxy eicosenic acid<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 269 | Hydroxylinolenic acid (isomer 1)<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 270 | Dihydroxy docosahexanoic acid<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 271 | Hydroxylinolenic acid (isomer 2)<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 272 | Dihydroxylinolenic acid (isomer 2)<sup>b</sup> | United Kingdom | Saleh et al. [56] |
| 273 | Hydroxylinoleic acid<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 274 | Hydroxyheptadecanoic acid acetate<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 275 | Hydroxydocosapentaenoic acid<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 276 | Dihydroxylinolenic acid (isomer 3)<sup>a</sup> | United Kingdom | Saleh et al. [56] |
| 277 | Hydroxydocosahexanoic acid<sup>a</sup> | United Kingdom | Saleh et al. [56] |

1. The molecular structure of the compound is not completely defined. 2. Constituent isolated from the honeybee propolis (from the genus Apis sp.). 3. Constituent isolated from the stingless bee propolis (from genera Tetragonula sp., Tetrigona sp., or Heterotrigona sp.). 4. Newly discovered compound. Compounds already mentioned in Huang et al. [8] are excluded.
2.3. Alcohols

Propolis, among other things, also contains different types of aliphatic compounds, such as simple alcohols, fatty alcohols, sugar alcohols, sterols, and others [8,24,84]. Between 2013 and 2018, two new alcohols were isolated from propolis samples from Africa (Cameroon) and Middle East (Oman). Table 5 includes only alcohols that were not included in the previous tables.

2.4. Alkaloids and their Derivatives

One of the most surprising discoveries regarding propolis in the recent years is definitely the discovery of alkaloids and their derivatives in propolis samples. Neither alkaloids nor nitrogenous compounds (except some vitamins from only a few samples) as such were reported from propolis before 2011–2012 [8,87]. To the best of our knowledge, alkaloids and their derivatives were first isolated from propolis in 2015 [57], when they were isolated from Brazilian propolis. They were later reported again, when they were isolated from Algerian propolis in 2017 [31] and from Brazilian propolis in 2018 [47]. Altogether 16 alkaloids and 5 alkaloid derivatives were isolated from propolis samples from two different countries. Specifics are listed in Table 6.

2.5. Other Compounds

Researchers also reported some new compounds in propolis that do not belong to any of the previously mentioned groups but were still isolated from propolis for the first time. Nineteen new compounds were isolated between 2013 and 2018, with the addition of one compound isolated in 2011 [35], which was not included in the review article by Huang et al. [8]. The newly isolated compounds were found in propolis from South America (Argentina), Africa (Algeria), and Europe (United Kingdom). Specifics are listed in Table 7.

Besides compounds mentioned above, there might be some that were not included in this review, either because their structures were not determined [47,60,66], because authors did not pay enough attention to their novelty and they were not specifically labelled as new [13,31,47,53,63,64,67,88], or simply because they were overlooked. In conclusion, actual number of compounds isolated in the recent years could be even higher.

Table 5 | Alcohols and related compounds identified in propolis for the first time since 2011

| No. | Chemical name | Geographic location | References |
|-----|---------------|---------------------|------------|
| 282 | Pinitol<sup>b</sup> | Oman                | Popova et al. [42] |
| 283 | 1<sup>-</sup>O-eicosanyl glycerol<sup>4</sup> | Cameroon            | Talla et al. [82] |

<sup>a</sup>Constituent isolated from the honeybee propolis (from the genus *Apis* sp.).<sup>b</sup>Constituent isolated from the stingless bee propolis (from genera *Scaptotrigona* sp. or *Melipona* sp.). Compounds already mentioned in Huang et al. [8] are excluded.

Table 6 | Alkaloids and their derivatives identified in propolis for the first time since 2011

| No. | Chemical name | Geographic location | References |
|-----|---------------|---------------------|------------|
| 284 | 7(3-Methoxy-2-methylbutyryl)-9-echimidinylretronecine derivative (1)<sup>6</sup> | Brazil             | Coelho et al. [57] |
| 285 | 7(3-Methoxy-2-methylbutyryl)-9-echimidinylretronecine derivative (2)<sup>6</sup> | Brazil             | Coelho et al. [57] |
| 286 | Pagicerine<sup>c</sup> | Algeria            | Soltani et al. [31] |
| 287 | Demecolcine<sup>c</sup> | Algeria            | Soltani et al. [31] |
| 288 | Papaverine<sup>c</sup> | Algeria            | Soltani et al. [31] |
| 289 | Aspidospermidine<sup>c</sup> | Algeria            | Soltani et al. [31] |
| 290 | Morphinan-6-one-2-ol<sup>c</sup> | Algeria            | Soltani et al. [31] |
| 291 | Thebaine<sup>c</sup> | Algeria            | Soltani et al. [31] |
| 292 | N,O-dimethyl stephine<sup>c</sup> | Algeria            | Soltani et al. [31] |
| 293 | Morpholine<sup>c</sup> | Algeria            | Soltani et al. [31] |
| 294 | Lelobanoline<sup>c</sup> | Brazil             | Cisilotto et al. [47] |
| 295 | 2-[6-(2-Hydroxy-propyl)-1-methyl-[2]-piperidyl]-1-phenylethanone<sup>c</sup> | Brazil             | Cisilotto et al. [47] |
| 296 | Norlobelanidine<sup>c</sup> | Brazil             | Cisilotto et al. [47] |
| 297 | Norlobeline<sup>b</sup> | Brazil             | Cisilotto et al. [47] |
| 298 | Lobeline<sup>b</sup> | Brazil             | Cisilotto et al. [47] |
| 299 | Lobelanidine<sup>b</sup> | Brazil             | Cisilotto et al. [47] |

<sup>a</sup>The molecular structure of the compound is not completely defined.<sup>b</sup>Constituent isolated from the honeybee propolis (from the genus *Apis* sp.).<sup>c</sup>Constituent isolated from the stingless bee propolis (from genera *Scaptotrigona* sp. or *Melipona* sp.). Compounds already mentioned in Huang et al. [8] are excluded.
Table 7 Compounds, not belonging to any previously mentioned groups, identified in propolis for the first time since 2011

| No. | Chemical name                                                                 | Geographic location | References                      |
|-----|-------------------------------------------------------------------------------|---------------------|---------------------------------|
| 305 | 4-[4-(4-Hydroxy-phenyl)-2,3-dimethyl-butyl]-benzene-1,2-diol                  | Argentina           | Agüero et al. [35]              |
| 306 | Ethoxy sulfonate<sup>a</sup>                                                  | United Kingdom      | Saleh et al. [56]               |
| 307 | 3,4,5-Triphenylpyrazole                                                      | Algeria             | Soltani et al. [31]             |
| 308 | 3-(4-Methoxyphenyl)benzof[<i>f</i>]<i>quinazoline</i>                          | Algeria             | Soltani et al. [31]             |
| 309 | 2-(4-Methoxyphenyl)-4-[(2-propyn-1-yl)thio]<i>quinazoline</i>                | Algeria             | Soltani et al. [31]             |
| 310 | 4-Aminobenzo[<i>g</i>]<i>quinazoline</i>                                      | Algeria             | Soltani et al. [31]             |
| 311 | 5-(4-Diethylaminobenzylidene)<i>rhdanine</i>                                | Algeria             | Soltani et al. [31]             |
| 312 | Carbamazepine<sup>a</sup>                                                    | Algeria             | Soltani et al. [31]             |
| 313 | 1-(3H-imidazol-4-yl)-ethanone<sup>a</sup>                                    | Algeria             | Soltani et al. [31]             |
| 314 | Nifenazone<sup>a</sup>                                                        | Algeria             | Soltani et al. [31]             |
| 315 | Podoflox<sup>a</sup>                                                          | Algeria             | Soltani et al. [31]             |
| 316 | Brallobarbital<sup>a</sup>                                                    | Algeria             | Soltani et al. [31]             |
| 317 | Cyclobarbital<sup>a</sup>                                                     | Algeria             | Soltani et al. [31]             |
| 318 | 6,7,8-Trimethoxy-<i>isoquinoline</i><sup>a</sup>                             | Algeria             | Soltani et al. [31]             |
| 319 | 1-<i>Butyl</i>-<i>isoquinoline</i><sup>a</sup>                               | Algeria             | Soltani et al. [31]             |
| 320 | 1-(Phenylthioxomethyl)-2,5-pyrrolidinedione<sup>a</sup>                      | Algeria             | Soltani et al. [31]             |
| 321 | 2-(4-Methoxyphenyl)-2-methyl-1,3-dioxolane<sup>a</sup>                        | Algeria             | Soltani et al. [31]             |
| 322 | 1′H-cholesta-3,5-dieno-[3,4-<i>b</i>]<i>indol</i>                             | Algeria             | Soltani et al. [31]             |
| 323 | 3-(3,4-Dimethoxyphenyl)-6-nitro-coumarin<sup>a</sup>                          | Algeria             | Soltani et al. [31]             |
| 324 | 10-Butyl-3,7-dinitro-10H-<i>phenothiazine</i><sup>a</sup>                     | Algeria             | Soltani et al. [31]             |

<sup>a</sup>The molecular structure of the compound is not completely defined. <sup>*</sup>Constituent isolated from the honeybee propolis (from the genus <i>Apis</i> sp.). Newly discovered compound. Compounds already mentioned in Huang et al. [8] are excluded.

3. CONCLUSION

Until 2000 at least 300 compounds were reported from propolis [6,11] and Huang et al. [8] reported another 241 between 2000 and 2012. Despite these numbers, just between 2013 and 2018 at least 305 compounds were isolated from propolis for the first time, including the first isolation of alkaloids. This number excludes 19 compounds isolated between 2011 and 2012, which were excluded from the previously mentioned review by Huang et al. [8] and were thus included in this article, bringing the total number to 324. Altogether, until 2018 more than 850 compounds are reported from propolis.

Compounds included in this article were isolated from 6 different continents and from 29 different countries (including the 19 added compounds isolated in 2011–2012 mentioned above). New compounds were isolated on more than one occasion from propolis of some countries, most often from Brazil (6×). Most of the compounds belong to the groups of flavonoids (92), phenols (126), and terpenes (46), whereas fatty acids (17), alcohols (2), alkaloids (21), and other compounds (20) represent a minor fraction. Despite the fact that propolis has been intensely studied for at least 30–40 years, new discoveries are being made on a yearly basis and it is not yet known how many more will be discovered in the upcoming years.

CONFLICTS OF INTEREST

The authors declare they have no conflicts of interest.

AUTHOR CONTRIBUTIONS

The informations were gathered and the bulk of the article was written by Luka Šturm, while the critical revision and final approval were done by dr. Nataša Poklar Ulrih, whom also submitted the article.

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