

## Naturally Occurring 1,5-Diarylpentanoids: A Review

Neslihan Celebioglu<sup>1</sup>, Ufuk Ozgen<sup>2</sup> and Hasan Secen<sup>1\*</sup>

<sup>1</sup>Ataturk University, Faculty of Sciences, Department of Chemistry, 25240 Erzurum, Türkiye

<sup>2</sup>Karadeniz Technical University, Faculty of Pharmacy, Department of Pharmacognosy,  
TR-61080 Trabzon, Türkiye

(Received October 17, 2017; Revised November 03, 2017; Accepted November 05, 2017)

**Abstract:** Diarylpentanoids, having Ar-C5-Ar chain, are a small class of natural products, most of which have been discovered in last the decade. This review comprises 20 natural diarylpentanoids, including their isolation, characterization and biological activities.

**Keywords:** Diarylpentanoids; natural product; biological activity. ©2017 ACG Publications. All rights reserved.

### 1. Introduction

Diarylpentanoids are analogs of diarylheptanoids<sup>1-7</sup> found widespread in nature. So far, over 400 Ar-C7-Ar, having diarylheptanoid structure were isolated, reporting their various biological activities such as anticancer, antiemetic, antibacterial, antioxidant and anti-inflammatory. However, diarylpentanoids with Ar-C5-Ar structure are not as widespread as diarylheptanoids in natural sources; only 20 diarylpentanoid compounds have been reported to be isolated (Figure 1, Table 1). This article is the first review on diarylpentanoids, comprising their isolation, characterization and biological activities.

### 2. Natural 1,5-Diarylpentanoids

Daphneolone (**1**) is the most commonly found natural diarylpentanoid, possessing Ar-C5-Ar skeleton and was first isolated from the roots of *Daphne odora* (Thymelaeaceae)<sup>8</sup>. Its structure was elucidated using spectroscopic methods such as <sup>1</sup>H-NMR, UV and MS. Its melting point and specific optical rotation were determined to be 119-120 °C and  $[\alpha]_D^{23} = +10^\circ$  (c 1.1, MeOH), respectively. Later, it was isolated from the roots of *Daphne tangutica*, melting point and specific optical rotation of which were reported to be 118-119 °C and  $[\alpha]_D^{23} = +9^\circ$  (c 1.01, MeOH), respectively<sup>9</sup>.

\* Corresponding author: E-Mail: [hsecen@atauni.edu.tr](mailto:hsecen@atauni.edu.tr)

Daphneolone (**1**) was found in the bark of *Daphne mezereum*<sup>10</sup> using HPLC technique. It was isolated from *Daphne odora* var. *atrocaulis*<sup>11</sup> and aerial parts of *Daphne pedunculata* together with known seven compounds<sup>12</sup>. Compounds (+)-**1**, (+)-**11** ( $[\alpha]_{\text{D}}^{20} = +6.1^\circ$ , *c* 0.12, MeOH), and (+)-**12** ( $[\alpha]_{\text{D}}^{20} = +12.2^\circ$ , *c* 0.13, MeOH) were isolated from tissue culture cells of *Daphne giraldii*, structures of which were elucidated using spectroscopic methods<sup>13</sup>. While **1** and **10** were also isolated from the stems and leaves of *Daphne bholua*<sup>14</sup>, **1** was obtained from the bark and roots of *Daphne retusa*<sup>15</sup>. Moreover, (S,+)-**1** ( $[\alpha]_{\text{D}}^{25} = +4.7^\circ$ , *c* 1.23, MeOH), **6**, **9**, **10** and **18** were isolated from *Daphne acutiloba*. Compounds **1**, **9** and **18** were reported to have anti-HIV activity, among which daphnenin (**18**) showed the highest anti-HIV activity<sup>16</sup>. Compounds **1** and **9** were obtained from EtOAc extract of the aerial part of *Thymelaea lythroides* (Thymelaeaceae)<sup>17</sup>. Daphneolone (**1**) and daphnenone (**10**) were isolated from the flower buds of *Daphne genkwa*<sup>18</sup>. **1** and **9**, isolated from *Daphne acutiloba*, displayed nematocidal activity<sup>19</sup>.

The natural products **2** and **3**, obtained from methanol extract of the rhizomes of *Curcuma domestica* (Zingiberaceae)<sup>20</sup> and *Curcuma longa*<sup>23</sup> were reported to have strong antioxidant and anti-inflammatory activities<sup>20</sup>. Compound **3** was also isolated from *Curcuma longa*<sup>22</sup>. **2** and **8** were isolated from the root tubers of *Curcuma longa*<sup>21</sup>. Moreover, compound **3**, isolated from the rhizomes of *Dioscorea nipponica* (Dioscoreaceae), displayed anti-neuroinflammatory effect<sup>24</sup>. Between **2** and **3** from the methanolic extract (80%) of the rhizomes of *Curcuma xanthorrhiza*, **2** was reported to have inhibitory effect toward NO<sup>25</sup>.

Compounds **4** and **5** from the roots<sup>26,28</sup> and rhizomes<sup>27</sup> of *Stellera chamaejasme* (Thymelaeaceae), showed contact activity<sup>26</sup>, good level of anti-feedant activity<sup>26</sup> and high bioactivity against aphids<sup>28</sup>. While **4**, **5**, **6**, (+)-**14** ( $[\alpha]_{\text{D}}^{20} = +30.1^\circ$  (*c* 0.41, CHCl<sub>3</sub>)) and (±)-**15** were isolated from *Diplomorpha ganpi* (Thymelaeaceae), (-)-**13** ( $[\alpha]_{\text{D}}^{20} = -31.4^\circ$ , *c* 0.74, CHCl<sub>3</sub>) was isolated from *Diplomorpha canescens* as a new compound<sup>29</sup>. Moreover, **4**, **6** and (-)-**14** were obtained from the stem barks of *Wikstroemia coriacea* (Thymelaeaceae)<sup>30</sup>. Compound (-)-**14**, named as coriaceol was isolated from the stem bark of *Wikstroemia coriacea*. Although its exact stereochemistry could not be determined due to the decomposition at C-2, its specific optical rotation of  $[\alpha]_{\text{D}}^{25} = -12^\circ$  (*c* 0.02, CHCl<sub>3</sub>) might be attributed to be the enantiomer of (+)-**14**<sup>30</sup>. The known compounds **4**, **5**, **6** and (-)-*erythro*-**20** were isolated from *Diplomorpha sikokiana*<sup>31</sup>.

**6** ( $[\alpha]_{\text{D}}^{18} = +4^\circ$  (*c* 0.275, MeOH), *S* configuration) from the roots of *Stellera chamaejasme*<sup>32,33,42</sup> exhibited immunomodulatory and antitumor activities<sup>32</sup> and toxic activity against *Peries rapae*<sup>33</sup>. **6** was also isolated from *Euphorbia altotibetica* (Euphorbiaceae)<sup>34</sup>.

A new diarylpentanoid **7**, named kinsenone, having (*E*, *Z*) configurations, from *Anoectochilus formosanus* (Orchidaceae), had strong antioxidant activity<sup>35</sup>.

Compound **8** which is a diastereomer of **7**, possessing (*E*, *E*) configurations was purified from *Curcuma longa*. It was reported to have antiviral activity against influenza viruses<sup>36</sup>.

Natural products **9** and **10** were isolated from *Daphne odora* var. *marginata*. **10** was reported to exhibit cytotoxic activity against human tumor cell lines K562, A549, MCF-7, LOVO and HepG2<sup>37</sup>. Compound **9** was also isolated from the root barks of *Daphne giraldii*<sup>38</sup>, which was given different names of daphneone and daphnolon.

Compounds **16** and **17**, having specific optical rotations of  $[\alpha]_{\text{D}}^{16} = -51.4^\circ$  (*c* 0.40, MeOH) (*S*) and  $[\alpha]_{\text{D}}^{16} = -26.8^\circ$  (*c* 0.05, MeOH) (*R*), respectively, and **9** and **10**, isolated from the roots and leaves of *Daphne giraldii*, were reported to display cytotoxic activities against human melanoma A375-S2 tumor cells. Their structures were elucidated using spectroscopic methods such as <sup>1</sup>H-NMR, <sup>13</sup>C-NMR, two-dimensional NMR, UV and MS<sup>39</sup>.

Compound **19** (Artamenone) was isolated from the stem and root barks of *Artabotrys modestus* ssp. *macranthus* (Annonaceae)<sup>40</sup> and fruits of *Lycium barbarum* (Solanaceae)<sup>41</sup>.

Compound (-)-*erythro*-**20** ( $[\alpha]_{\text{D}} = -19.2$  (*c* 0.52, MeOH); mp. 91-92 °C), a unique example of the natural 1,5-diarylpentanoids containing two OH groups in the C5 chain, was first isolated from the wood of *Flindersia laevis* (Rutaceae)<sup>43</sup>. Later, (-)-*erythro*-**20** was isolated from *Wikstroemia sikokiana* and its absolute configuration was established as **1(S)** and **3(S)** after a series of chemical synthesis<sup>44</sup>.

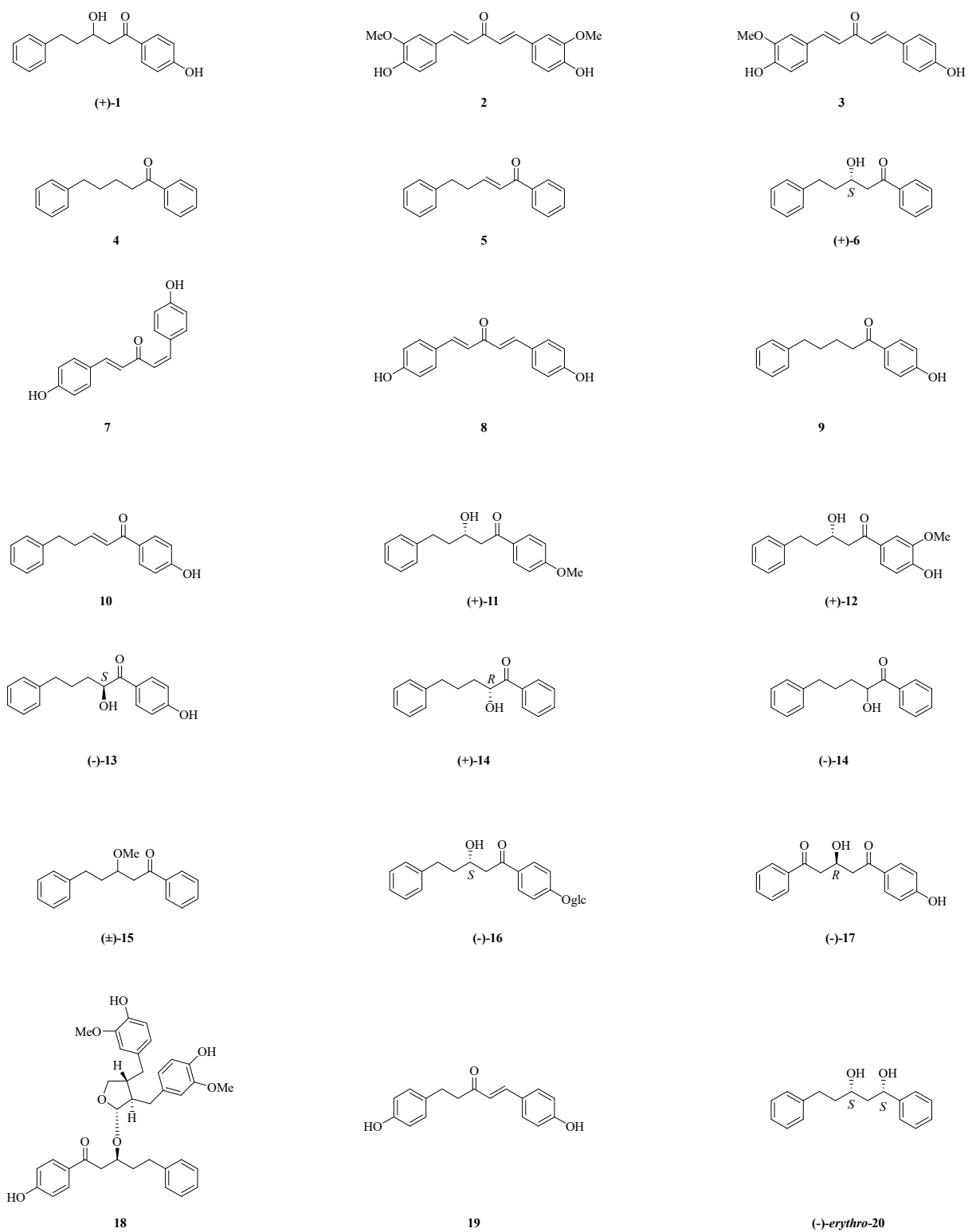


Figure 1. Diarylpentanoid compounds isolated from nature

**Table 1.** Diarylpentanoids of structures, isolations and the reported biological activities

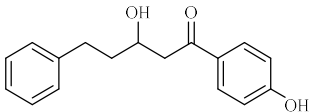
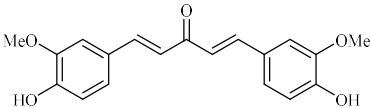
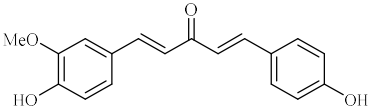
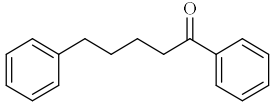
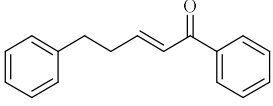
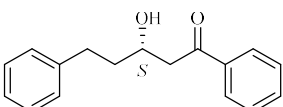
Natural Product	Plant Source	Bioactivity	Year/Ref.
 (+)-1	<i>Daphne odora</i>		1974, [8]
	<i>Daphne tangutica</i>		1982, [9]
	<i>Daphne mezereum</i>		1990, [10]
	<i>Daphne odora</i> var. <i>atrocaulis</i>		2005, [11]
	<i>Daphne pedunculata</i>		2008, [12]
	<i>Daphne giraldii</i>		2009, [13]
	<i>Daphne bholua</i>		2009, [14]
	<i>Daphne retusa</i>		2011, [15]
	<i>Daphne acutiloba</i>		2012, [16]
	<i>Tyhmelaea lythroides</i>		2013, [17]
<i>Daphne genkwa</i>		2013, [18]	
<i>Daphne acutiloba</i>		Nematicidal	2015, [19]
 2	<i>Curcuma domestica</i>	Antioxidant, antiinflammatory	1993, [20]
	<i>Curcuma longa</i>		2008, [21]
	<i>Curcuma longa</i>	Inhibitory effect on NO	2011, [23]
	<i>Curcuma xanthorrhiza</i>		2014, [25]
 3	<i>Curcuma domestica</i>	Antioxidant, antiinflammatory	1993, [20]
	<i>Curcuma longa</i>		2009, [22]
	<i>Curcuma longa</i>	Anti-neuroinflammatory	2011, [23]
	<i>Dioscorea nipponica</i>		2013, [24]
	<i>Curcuma xanthorrhiza</i>		2014, [25]
 4	<i>Stellera chamaejasme</i>	Contact activity, anti-feedant activity	2001, [26]
	<i>Stellera chamaejasme</i>		2001, [27]
	<i>Stellera chamaejasme</i>	Aphicide	2002, [28]
	<i>Diplomorpha ganpi</i>		2012, [29]
	<i>Wikstroemia coriacea</i>		2013, [30]
	<i>Diplomorpha sikokiana</i>		2016, [31]
 5	<i>Stellera chamaejasme</i>	Contact activity, anti-feedant activity	2001, [26]
	<i>Stellera chamaejasme</i>		2001, [27]
	<i>Stellera chamaejasme</i>	Aphicide	2002, [28]
	<i>Diplomorpha ganpi</i>		2012, [29]
	<i>Diplomorpha sikokiana</i>		2016, [31]
 6	<i>Stellera chamaejasme</i>	Immunomodulatory, antitumor activity	2001, [32]
	<i>Stellera chamaejasme</i>	Toxic activity	2008, [33]
	<i>Diplomorpha ganpi</i>		2012, [29]
	<i>Daphne acutiloba</i>		2012, [16]
	<i>Euphorbia altotibetica</i>		2012, [34]
	<i>Wikstroemia coriacea</i>		2013, [30]
	<i>Diplomorpha sikokiana</i>		2016, [31]
<i>Stellera chamaejasme</i>		2015, [42]	

Table 1 Continued...

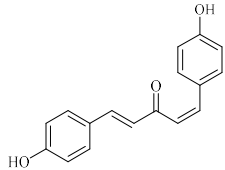
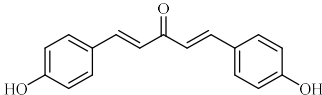
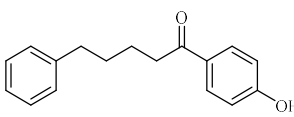
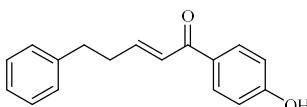
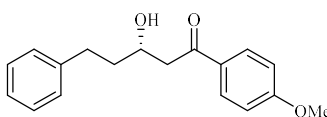
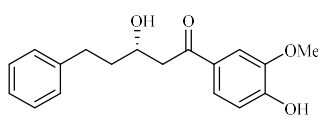
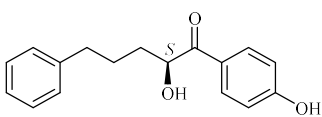
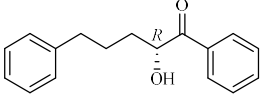
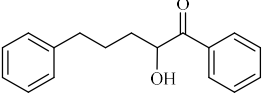
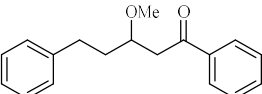
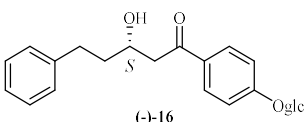
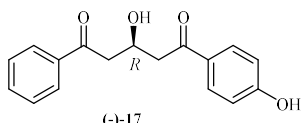
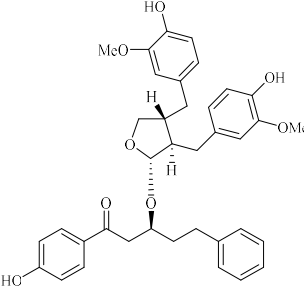
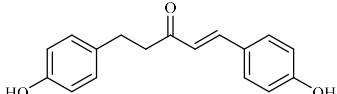
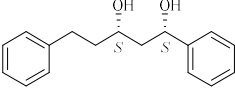
Natural Product	Plant Source	Bioactivity	Year/Ref.
 <p>7</p>	<i>Anoectochilus formosanus</i>	Antioxidant	2002, [35]
 <p>8</p>	<i>Curcuma longa</i> <i>Curcuma longa</i>	Antiviral	2008, [21] 2012, [36]
 <p>9</p>	<i>Daphne odora</i> var. <i>marginata</i> <i>Daphne giraldii</i> <i>Daphne giraldii</i> <i>Daphne acutiloba</i> <i>Tyhmelaea lythroides</i> <i>Daphne acutiloba</i>	Cytotoxicity    Nematicidal	2006, [37] 2006, [38] 2012, [39] 2012, [16] 2013, [17] 2015, [19]
 <p>10</p>	<i>Daphne odora</i> <i>Daphne bholua</i> <i>Daphne acutiloba</i> <i>Daphne giraldii</i> <i>Daphne genkwa</i>	Cytotoxicity  Cytotoxicity	2006, [37] 2009, [14] 2012, [16] 2012, [39] 2013, [18]
 <p>(+)-11</p>	<i>Daphne giraldii</i>		2009, [13]
 <p>(+)-12</p>	<i>Daphne giraldii</i>		2009, [13]
 <p>(-)-13</p>	<i>Diplomorpha canescens</i>		2012, [29]

Table 1 Continued...

Natural Product	Plant Source	Bioactivity	Year/Ref.
 <p>(+)-14</p>	<i>Diplomorpha ganpi</i>		2012, [29]
 <p>(-)-14</p>	<i>Wikstroemia coriacea</i>		2013, [30]
 <p>(+)-15</p>	<i>Diplomorpha ganpi</i>		2012, [29]
 <p>(-)-16</p>	<i>Daphne giraldii</i>	Cytotoxicity	2012, [39]
 <p>(-)-17</p>	<i>Daphne giraldii</i>	Cytotoxicity	2012, [39]
 <p>18</p>	<i>Daphne acutiloba</i>	Anti-HIV	2012, [16]
 <p>19</p>	<i>Artabotrys modestus</i> ssp. <i>macranthus</i> <i>Lycium barbarum</i>		2013, [40] 2014, [41]
 <p>(-)-erythro-20</p>	<i>Flindersia laevicarpa</i> <i>Wikstroemia sikokiana</i> <i>Diplomorpha sikokiana</i>		1962, [43] 1987, [44] 2016, [31]

### 3. Conclusion

This review surveyed the structures and biological activities of diarylpenantoids, which are rarely found compounds in nature. So far, only 20 natural diarylpenantoid compounds, having Ar-C5-Ar, were isolated.

In the light of phytochemical studies, we can say that diarylpenantoids have been found in the genus *Daphne*, *Tyhmelaea*, *Curcuma*, *Stellera*, *Diplomorphai*, *Dioscorea*, *Wikstroemia*, *Anoectochilus*, *Euphorbia*, *Artabotrys* and *Lycium* and the families Thymelaeaceae, Zingiberaceae, Dioscoreaceae, Euphorbiaceae, Orchidaceae, Annonaceae, Solanaceae, Rutaceae.

We think that naturally occurring 1,5-diarylpenantoids can be models to develop biologically active drug-like compounds and further studies may discover new aspects of these compounds.

**Acknowledgements:** The reported review is a part of PhD dissertation of Neslihan Celebioglu. We are thankful Dr. Necla Oztaskin and Umut Duaci for their kind help.

### ORCID

Neslihan Celebioglu: [0000-0003-1882-0720](https://orcid.org/0000-0003-1882-0720)

Ufuk Ozgen: [0000-0001-9839-6717](https://orcid.org/0000-0001-9839-6717)

Hasan Secen: [0000-0002-5388-6111](https://orcid.org/0000-0002-5388-6111)

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