Review

Natural Product Chemistry of Gorgonian Corals of Genus Junceella—Part III

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Abstract: The structures, names, bioactivities, and references of 82 natural products, including 48 new metabolites, purified from the gorgonian corals belonging to the genus Junceella are described in this review. All compounds mentioned in this review were obtained from Junceella fragilis, Junceella gemmacea, Junceella juncea, and Junceella sp., collected from tropical Indo-Pacific Ocean. Some of these compounds exhibited potential biomedical activities.

Keywords: Junceella; gorgonian; briarane; biomedical activity

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1. Introduction

Following previous review articles focused on marine-origin natural products, this review covers
the literature from October 2011 to August 2018, and describes 82 natural products (including 48
new metabolites) from gorgonian corals belonging to the genus _Junceella_ (family Elliselliidae) [1–4].
Extending from previous reviews in 2004 and 2011 [5,6], this review provides structures, names,
bioactivities, and references for all compounds including briarane- and norcembrane-type diterpenoids,
sterol, and nucleosides. Many of these compounds exhibited interesting bioactivities in vitro, which
might indicate a potential for use in biomedical applications. This survey of natural products from
_Junceella_ is presented taxonomically according to species.

2. Junceella

2.1. Junceella Fragilis

Eight 8-hydroxybriaranes, including four new compounds, frajunolides P–S (1–4), and four
known metabolites, umbraculolide A (5) [7,8], juncenolide C (6) [9], junceellonoid A (7) [10],
and juncin R (8) [11], were isolated from _J. fragilis_, collected from the waters of Taiwan [12] (Figure 1).
The structures of briaranes 1–4 were established by spectroscopic methods, and determination of the
absolute configuration of 1 was completed by X-ray diffraction analysis [12]. At a concentration of
10 μg/mL, briaranes 1 and 2 were found to exert inhibitory activities on elastase release (inhibition
rate = 35.6% and 34.1%, respectively) and superoxide anion production (inhibition rate = 32.5
and 28.7%, respectively) by human neutrophils [12].

Figure 1. Structures of frajunolides P–S (1–4), umbraculolide A (5), juncenolide C (6), junceellonoid A
(7), and juncin R (8).

In 2014, _J. fragilis_, collected from the South China Sea, was found to contain 12 new briaranes,
fragilisins A–L (9–20) [13], along with seven known analogues, junceellolides A (21) and B (22) [14],
junceol A (23) [15], junceellonoid D (24) [16,17], fragilide C (25) [18], and frajunolides A (26) [19] and E
(27) [20] (Figure 2) [13]. The structures of briaranes 9–20 were determined by spectroscopic methods.
Briaranes 17–20 were the first iodine-containing briaranes to be isolated. The absolute configuration
of briarane 9 was confirmed by single-crystal X-ray diffraction data [13]. Briaranes 13, 14, 18, 21,
and 24 showed potent antifouling activities against the settlement of barnacle _Balanus amphitrite_ larvae,
with EC₅₀ values of 14.0, 12.6, 11.9, 5.6, and 10.0 μM, respectively [13].

In addition, a new norcembraneoid, fragilide A (28), 16 new briaranes, fragilolides B–Q
(29–44), along with two known briaranes, frajunolides H (45) [20] and N (46) [21], and three known
norcembranoids, scabrolide D (47) [22], sinuleptolide (48) [23], and 5-epi-sinuleptolide (49) [24–26],
were obtained from _J. fragilis_, collected from the inner coral reef around in Hainan Island of
China [27] (Figure 3). The structures of metabolites 28–44 were determined by spectroscopic methods,
including calculated electronic circular dichroism (ECD) data. The structures, including the absolute
configurations of briaranes 37 and 46, were further established by single-crystal X-ray diffraction
analysis using Flack parameter in this study [27]. Compound 28 featured an unprecedented 4,13- and 7,11-fused tetracyclic norcembranoid [27].

Briarane 45 exhibited cytotoxicity toward Hep G2 (human hepatocellular carcinoma), Huh7 (human hepatocellular carcinoma), SMMC-7721 (human papillomavirus-related endocervical adenocarcinoma), A2780 (human ovarian carcinoma), BGC-823 (human gastric adenocarcinoma), HGC-27 (human gastric carcinoma), MGC-803 (human gastric carcinoma), NCI-H1650 (human bronchoalveolar carcinoma), and PA-1 (human ovarian mixed germ cell tumor) cells with IC₅₀ values 0.89, 16.52, 0.61, 1.18, 2.10, 0.61, 1.97, 6.47, and 0.42 μM, respectively. Briaranes 31, 34, 36, 39, 43, and 46 exerted selective inhibitory effects toward hepatitis B and antigen (HBeAg) in a dose of 10 μM, whereas no activity was observed against the expression of hepatitis B surface antigen (HBsAg) [27].

Moreover, the norcembranoids 28 and 47–49 were assayed for their potential inhibitory effects against nitric oxide (NO) production induced by lipopolysaccharides (LPS) (large molecules consisting of lipids and polysaccharide composed of O-antigen joined by chemical bonds), in RAW264.7 cells, and these four compounds exhibited the inhibitory activities with 27.8%, 43.5%, 56.0%, and 57.9% inhibition, respectively, at a dose of 100 μM [27]. In order to explore the mechanism of these NO inhibitors, the expression of the antioxidant response element (ARE) mediated luciferase and NF-κB was evaluated. Norcembranoids 48 and 49 showed the effects against NF-κB by the inhibitory rates of 25.1% and 28.6% in a dose of 50 μM, respectively. Significant induction of luciferase was observed as the dose of 50 μM for 48 and 49 with 3.8 and 5.6 folds more than that of blank control [27]. The antioxidant capacity of 28 and 47–49 were evaluated by the modified 2,2′-azino-bis-(3-ethylbenothiazoline-6-sulfonic acid) radical cation decolorization assay [27].

Figure 2. Structures of fragilisinins A–L (9–20), junceellolides A (21) and B (22), junceol A (23), junceellonoid D (24), fragilide C (25), and frajunolides A (26) and E (27).
In 2017, Cheng et al. reported the occurrence of four pairs of chlorinated briarane derivatives, including five new metabolites, 3-deacetylpraelolide (50), 13-α-acetoxy-3-deacetylpraelolide (51), 13-α-acetoxy-2-deacetylpraelolide (52), 13-α-acetoxy-3-deacetyljuncellin (53), 13-α-acetoxy-2-deacetyljuncellin (54), along with three known metabolites, fragilide J (55) [28], 3-deacetyljuncellin (56), and 2-deacetyljuncellin (57) [29], from J. fragilis, collected off the inner coral reef in Hainan Island, China (Figure 4), although briaranes 56 and 57 were obtained as a pair of inseparable mixture [30].

The structures of briaranes 50–54 were elucidated by spectroscopic methods in association with chemical conversion. The absolute configurations of briaranes 50 and 55 were further determined by acetylation of these two compounds to yield the same crystal product and analyses of X-ray crystal data of this compound by Flack parameter further confirmed the absolute configurations of briaranes 50 and 55 [30], although briaranes 56 and 57 existed in an inseparable mixture in CHCl3 at room temperature. Lowering the temperature to 4 °C resulted in the generation of a crystal, while the
X-ray diffraction analysis using Flack parameter determined the crystal product to be in accordance with briarane 56. Each pair of the isomers (50/55, 51/52, 53/54, and 56/57) featured by dynamical interconversion through as acetyl migration in 1,2-diol, which was postulated to be generated under the formation of cyclic orthoacetate intermediated. In the mixture of briaranes 56 and 57, increasing temperature gradients resulted in the variation of 56/57 ratio, while the ratio of 56/57 varied from 1:1 to 2:3 at 50 °C [30]. The mixtures of 50/55, 51/52, 53/54, and 56/57 were tested for their inhibitory effects against NO production induced by LPS in RAW264.7 cells and these four components displayed inhibitory activities against NO production with the inhibition rates of 39.4%, 46.4%, 42.7%, and 36.3%, respectively, at a concentration of 50 µM [30].

Two new briaranes, fragilides K (58) and L (59), along with five known chlorinated briaranes, gemmacolides V (60) and X (61) [31], praelolide (62) [7,14,16,32–37], and juncins P (63) [35] and ZI (64) [11], were obtained from a Formosan J. fragilis [38] (Figure 5).

Based on spectroscopic methods, the structures of briaranes 58 and 59 were elucidated and the cyclohexane rings in 58 and 59 were found to exist in chair and twist boat conformation, respectively. At a concentration of 10 µM, briaranes 59, 61, and 64 showed anti-inflammatory activity against the expression of pro-inflammatory protein inducible nitric oxide synthase (iNOS) to 49.13%, 36.22%, and 43.33%, respectively, and briaranes 60 and 61 elicited reduction of the pro-inflammatory protein cyclooxygenase-2 (COX-2) to 47.49% and 43.64%, respectively [38].

2.2. Junceella Gemmacea

Four new briaranes, juncellolides M–P (65–68), along with seven known briaranes, juncellolides A–D (21,22,69,70) [14] (the structures of briaranes 21 and 22, please see Figure 2), juncellin A (= juncellin) (71) [7,14,16,34–37,39,40], praelolide (62) [7,14,16,32–37], and juncin ZI (64) [11] (the structures of briaranes 62 and 64, please see Figure 5) were isolated from J. gemmacea, collected from the South China Sea [41] (Figure 6). The structures, including the absolute configurations,
of new briaranes 65–68 were deduced on the basis of spectroscopic analyses, particularly with ECD experiments.

![Structures of briaranes](image)

Figure 6. Structures of junceellolides M–P (65–68), junceellolides C (69) and D (70), and junceellin A (71).

2.3. Junceella Juncea

Five 8-hydroxybriaranes, including a new briarane, (15S*,2S*,8S*,9S*,10S*,11R*,12R*,14S*,17R*)-11,20-epoxy-14-(3-methylbutanoyl)-2,9,12-triacetoxy-8-hydroxybriar-5(16)-en-18,7-olide (72) along with four known metabolites, gemmacolides A (73) and B (74) [42,43], and juncins H (75) [44] and K (76) [45], were isolated from J. juncea collected from Tuticorin coast of the Indian Ocean (Figure 7) [46]. The structure of briarane 72 was established by spectroscopic data and 72 was found to exhibit activities against the fungi Aspergillus niger, Candida albicans, and Penicillium notatum. Briaranes 73 and 74 displayed activities against the bacteria Bacillus pumilis and Escherichia coli. While the briaranes 75 and 76 showed activities against B. subtilis, B. pumilis, Proteus vulgaris, and E. coli [46].

![Structures of briaranes](image)

Figure 7. Structures of (15S*,2S*,8S*,9S*,10S*,11R*,12R*,14S*,17R*)-11,20-epoxy-14-(3-methylbutanoyl)-2,9,12-triacetoxy-8-hydroxybriar-5(16)-en-18,7-olide (72), gemmacolides A (73) and B (74), and juncins H (75) and K (76).

Furthermore, Chang et al. isolated three new briaranes, juncenlides M–O (77–79), from J. juncea, collected in the waters of Taiwan [47] (Figure 8). Structures of new briaranes 77–79 were established by spectroscopic methods. Briaranes 78 and 79 showed inhibitory activities against the release of elastase and 79 also exhibited inhibitory activity against the generation of superoxide anon [47].
3. Conclusions

Marine natural products currently under clinical trials are limited. Based on the potential medical use and complex structures, it is very difficult to obtain enough material for further studies by chemical methods. How to make the best use of aquaculture technology to enhance in captivity mass production of marine organisms and the compounds of this type were suggested originally synthesized from the marine invertebrates originally studied is an important direction of marine natural product research.

2.4. Junceella sp.

Three known briaranes, junceellolide A (21) [14], praelolide (62) [7,14,16,32–37], and junceellin A (71) [7,14,16,34–37,39,40], (the structures of briaranes 21, 62, and 71, please see Figure 2, Figure 5, and Figure 6, respectively), and a known sterol, 5α,8α-epidioxy-24(δ)-methylcholesta-6,22-dien-3β-ol (80) [48] (Figure 9), were obtained from the ethanol extract of a gorgonian coral identified as Junceella sp., collected off the Vietnam Thu Island in May 2010 [49]. However, by assuming that enantiomeric series for sterols, the configuration at C-24 in 80 should be assigned as 5*-form on the basis of the 13C NMR chemical shift of C-24 and C-28 [50]. Furthermore, two nucleosides, deoxyadenosine (81) and deoxythymidine (82) [51,52], were obtained from aqueous solution of this specimen. Structures of all isolates were established using spectroscopic data (Figure 9) [49].

In the cytotoxic activity test, briaranes 71 and sterol 80 exhibited weak cytotoxicity toward the THP-1 (human acute monocytic leukemia) tumor cells with IC50 values 55.4 and 130 μM, respectively. Sterol 80 also possessed weak clonogenic activity with INCC50 values 53.3 and 62 μM toward THP-1. Moreover, sterol 80 produced an inhibition zone 12 mm in diameter against Bacillus subtilis. Briarane 62 inhibited weakly Candida albicans. Briaranes 21 and 71 and sterol 80 inhibited weakly Vibrio parahaemolyticus [49].

3. Conclusions

The natural products obtained from gorgonian corals belonging to the genus Junceella complied in this review indicated that the terpenoid derivatives, particularly briarane-type diterpenoids, are the major components of the natural products isolated. Of the 82 metabolites, 75 compounds (91.5%) are briarane-type diterpenoids. Of the briaranes, 50 compounds are halogenated (50/75 = 66.7%). Briarane-type natural products are a large family of natural products that are only isolated from marine organisms and the compounds of this type were suggested originally synthesized from the 3,8-cyclization of cembranoids by the host corals and not by their zooxanthellae [37,53,54]. Briarane-type diterpenoids continue to attract attention owing to their complex structures and potential biomedical activities.

Studies on the novel substances for biomedical use from the marine invertebrates originally distributed in the Indo-Pacific Ocean will play an important role in natural product research [55]. Marine natural products currently under clinical trials are limited. Based on the potential medical use and complex structures, it is very difficult to obtain enough material for further studies by
chemical methods. How to make the best use of aquaculture technology to enhance in captivity mass production of raw materials needed for extraction of biomedical use marine natural compounds is very important in the future [56].

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