In Situ Vinylpyrrole Synthesis. Diels-Alder Reactions with Maleimides to Give Tetrahydroindoles

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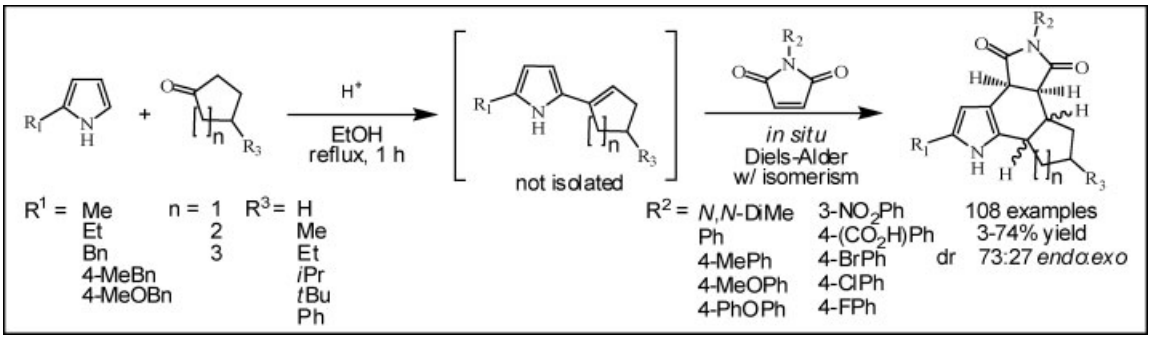

A series of 108 tetrahydroindoles has been prepared by a one-pot synthesis from 2-alkylpyrroles, cyclic ketones, maleimides, and an acid catalyst. A 5-vinylpyrrole is formed by an acid-catalyzed condensation of a 2-alkyl-substituted pyrrole with a ketone, which is subsequently trapped *in situ* by a maleimide in a predominantly *endo*-addition Diels-Alder reaction. Isomerization of the double bond into the pyrrole ring gives a tetrahydroindole with predominant *cis*-fusion of the cycloalkane ring.

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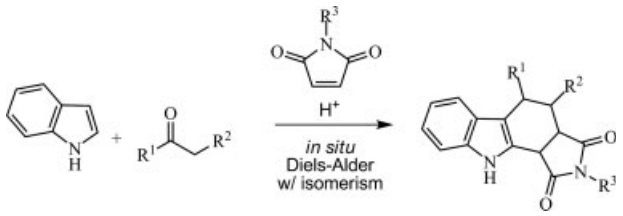
INTRODUCTION

The common occurrence of indole in biologically active compounds [1] highlights the importance of studies on the synthesis of indoles as well as the value of biological testing of indole-containing molecules. Previously, our group has reported the synthesis of 3-vinylindoles, which are trapped *in situ* by a Diels-Alder reaction with various maleimides, a technique which was used to prepare a large variety of tetrahydrocarbazoles [2]. We now wish to report analogous work with cycloadditions of 2-alkyl-substituted vinylpyrroles.

Pyrrole preferentially undergoes electrophilic attack at its 2-position since the most stable resonance structure of the reactive species has its greatest electron density α to the iminium nitrogen. For indole, dearomatization of the fused benzene ring inhibits a similar adjacent placement of charge. Instead, the highest electron-density occurs at the 3-position; thus, indole has favored electrophilic substitution at the 3-position in spite of greater charge separation. Correspondingly, our previous work involved the trapping of 3-vinylindoles produced from condensation of indole with ketones (Scheme 1), whereas in this work, the trapped intermediates are 2-vinylpyrroles. This has bearing on the topology; the products of this work are *e*-side maleimide-fused tetrahydroindoles, whereas the tetrahydroindole component of the products of the vinylindole work is maleimide-fused at the *g*-side.

There are several known examples of 2-vinylpyrroles participating in Diels-Alder reactions [3], including employing as the dienophiles carboxyl-substituted acetylenes [4,5], several acyclic electron-deficient alkenes [5,6], maleic anhydride and/or *N*-phenylmaleimide with *N*-benzenesulfonyl-2-vinylpyrrole [6,7] and methyl 3-nitroacrylate with *N*-*p*-toluenesulfonyl-2-vinylpyrroles [8], tetrachloro- or tetrabromocyclopropene with *N*-*p*-toluenesulfonyl-2-vinylpyrrole [9], *N*-phenylmaleimide with *N*-methyl- and *N*-propanoyloxy-2-vinylpyrrole [6], *N*-H-maleimide with 3-(*N*-alkyl-2-pyrrolyl)acrylates [10] and *N*-alkyl-2-styrylpyrroles [10,11], and one example using various maleimides with both *N*-H and *N*-alkyl-2-vinylpyrroles [12]. Several of these studies report biological activity from this class of compounds, particularly anticancer activity [10–12]. To our knowledge, no prior demonstration of 2-vinylpyrrole formation accompanied by *in situ* trapping with a dienophile exists, a route which avoids the multiple steps involved in synthesizing the vinylpyrrole before the Diels-Alder reaction, affording considerable efficiency over the alternative procedures available for tetrahydroindole formation.

Scheme 1. *In situ* synthesis of tetrahydrocarbazoles from ketones.



RESULTS AND DISCUSSION

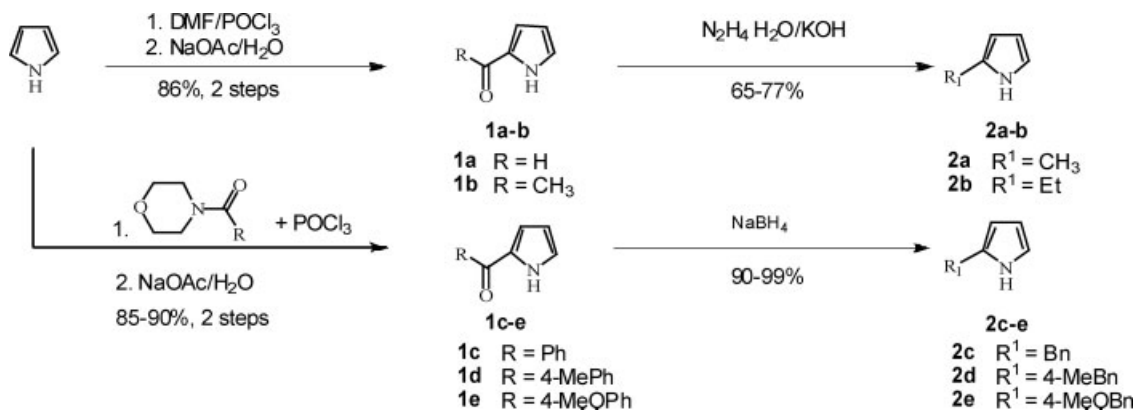
General. Pyrrole is a reactive electron-rich heterocycle which, upon condensation with cyclic ketones, followed by proton-transfer, is believed to form a tertiary alcohol. In the presence of an acid catalyst, the alcohol should readily dehydrate, forming a resonance-stabilized 2-vinylpyrrole. The highly reactive 2-vinylpyrrole is then captured *in situ* by the dienophile. Under acidic conditions, pyrroles are known to form polymers [13]. Tetrameric calix [4] pyrroles are known to form when pyrroles and ketones react in the presence of an acid catalyst [14,15]. When producing vinylpyrroles for *in situ* trapping, we have found that blocking the other 2-position by use of 2-alkyl-substituted pyrroles is useful in preventing formation of complex polymeric mixtures, which generally appeared as dark sticky tars or black powders. Studies using removable blocking groups at the 2-position of pyrroles in the formation of vinylpyrroles for *in situ* trapping and other uses are currently ongoing in our laboratory.

2-Substituted-5-vinylpyrroles were synthesized as outlined in Scheme 2. Pyrrole-2-carboxaldehyde (**1a**) was synthesized *via* Vilsmeier-Haack formylation [16], followed by Wolff-Kishner reduction [17], to give 2-methylpyrrole (**2a**). Wolff-Kishner reduction of commercially available 2-acetylpyrrole (**1b**) produced 2-ethylpyrrole (**2b**) [17]. Vilsmeier-Haack aroylation [18] of pyrrole gave the 2-phenyl (**1c**), 2-(4-methylphenyl) (**1d**), and 2-(4-methoxyphenyl) (**1e**) ketones, which, after sodium borohydride reduction [19], gave the corresponding 2-benzylpyrroles (**2c–e**).

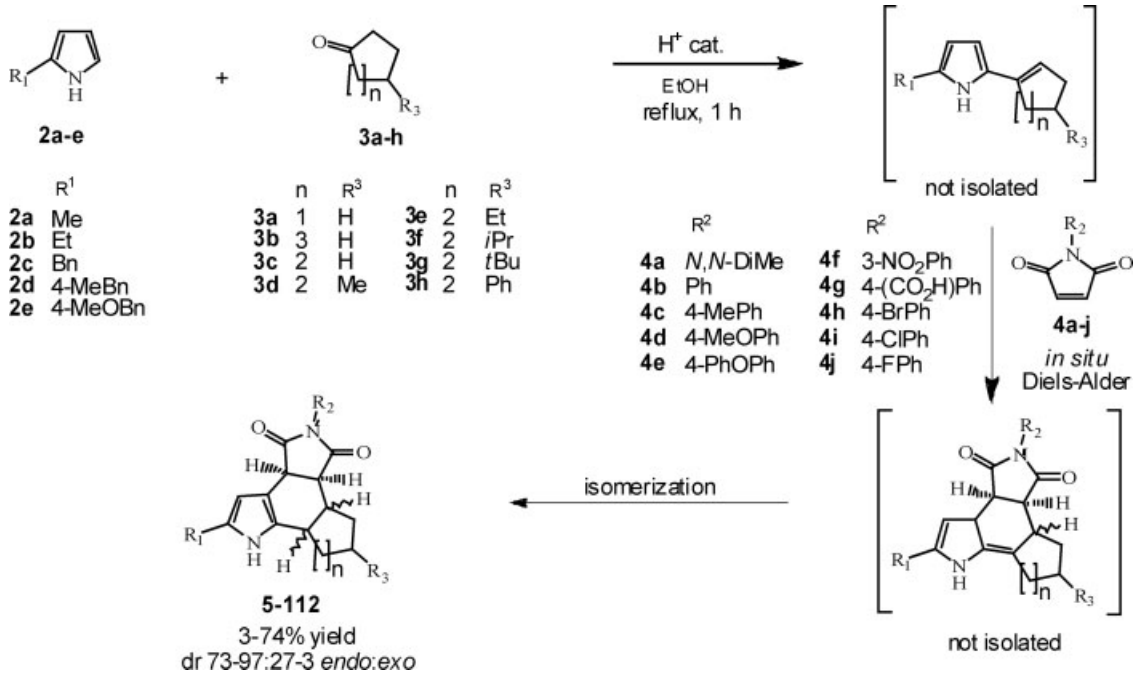
Condensation of **2a–e** with cyclopentanone (**3a**), variously 4-substituted-cyclohexanones (**3c–h**), or cycloheptanone (**3b**), gave the corresponding vinylpyrroles. These acted as electron-rich dienes for normal electron-demand Diels-Alder reactions, which occurred *in situ* with various substituted maleimides (**4a–j**, Scheme 3). The unrearranged form of the Diels-Alder adduct was not isolated. Instead, spontaneous isomerization of the double bond into the five-membered ring gave aromatized tetrahydroindoles (**5-112**, Table 1). *cis*-Fusion of the cycloalkyl ring involves less strain, but, since isomerism to the pyrrole is likely irreversible, thermodynamic equilibration may not determine the type of ring-fusion. Orbital symmetry considerations forbid suprafacial 1,3-hydride shifts and antarafacial 1,3-hydride shifts are geometrically difficult [20]; therefore, the isomerism probably takes place through acid catalysis. A proton should approach preferentially from the less sterically hindered face, the face opposite to the maleimide fusion and the same face from which protons 3b-H and 6a-H protrude (in the Experimental, this face is always designated ''α''). This face of hydrogen delivery gives *cis*-fusion of the cycloalkyl ring with a *syn* relationship between all four of the protons on the cyclohexene ring.

The ^{1}H NMR data of **5-112** show mixtures of isomers, which were usually isolated by precipitation from the crude ethanolic reaction mixture, possibly influencing the reported distribution of isomers because of solubility differences. Both *endo*- and *exo*-Diels-Alder additions are possible, and *cis*- or *trans*-fusion gives the possibility of four isomers, *endo*-addition with *cis*-fusion (**En-c**), *endo*-addition with *trans*-fusion (**En-t**), *exo*-addition with *cis*-fusion (**Ex-c**), and *exo*-addition with *trans*-fusion (**Ex-t**, Fig. 1). Between one and four isomers are recognizable in each spectrum, corresponding to these stereoisomeric products. Smaller minor isomer peaks are

Scheme 2. Synthesis of 2-substituted pyrroles.



Scheme 3. *In situ* synthesis of tetrahydrocarbazoles fromcyclic ketones.



visible next to or overlapping the peaks belonging to the major isomer, particularly for protons 1-H, 3b-H, 6a-H, 6b-H, and the proton α to the point of cycloalkane ring-fusion to the pyrrole ring, labeled 9a-H, 10a-H, or 11a-H, the numbers depending on which sized cyclic ketone, **3a**, **3b**, or **3c**–**h**, was used.

In some products derived from the 4-substituted-cyclohexanones **3d**–**h**, additional isomerism is observed due to the stereogenic center at position 8 (see Fig. 1 for numbering). This is supported by the observation that the ratio of the integrated areas of proton peaks belonging to the alkyl substituents at position 8 is generally not equal to the ratio of *endo*/*exo*-addition *cis*/*trans*-fusion isomers present in the mixture determined from the integrated areas of protons 1-H, 3b-H, and 6a-H. Since our major concern in analyzing the ^{1}H NMR data is the diastereoselectivity of the Diels-Alder reaction and subsequent isomerization, it is the distribution of the four isomers **En-c**, **En-t**, **Ex-c**, and **Ex-t** that is reported in Table 1 and in the Experimental section, and it is these four isomers to which the text refers in subsequent discussion.

^{1}H NMR, nuclear Overhauser effect (NOE), and computational analyses. For all products, two isomers are present in greater quantity than the other two, corresponding to the expected *endo*-addition Diels-Alder products. At a minimum, in the isolated products, *endo*-addition is preferred over *exo*-addition in a 73:27 diastereomeric ratio, and at a maximum in a 97:3 diastereomeric ratio (in which no **Ex-c** and **Ex-t** isomers are visible by ^{1}H NMR). The *endo*-addition preference in Diels-Alder reactions is commonly explained by a favorable secondary orbital interaction that occurs in the transition state when the molecular orbitals of the carbonyls of the imide dienophile overlap with the developing molecular orbital from the diene, an interaction not present with an *exo*-approach. Although both stepwise and concerted mechanisms are theoretically possible to give tetrahydroindoles **5**-**112** [21], the stereochemical relationships found below in the major isomers are consistent with that expected for an *endo*-addition; therefore, a concerted reaction pathway is likely. To verify the *endo*-addition preference, and to confirm that *cis*-fusion is predominant, NOE experiments were performed on nine representative tetrahydroindoles, compounds **13**, **20**, **26**, **47**, **55**, **61**, **89**, **84**, and **103**.

Consistent NOE interactions were observed between the 3b-H and 6b-H protons of the two major isomers of each of these products, giving evidence that they arise from *endo*-addition. To determine whether *cis*- or *trans*-fusion occurred in a particular *endo*-addition isomer, NOE experiments must compare interactions of the protons at the points of the cycloalkane ring-fusion. In *trans*-fused products, the distance between the protons should be greater, giving a weaker NOE interaction. For careful comparison of the relative strength of these interactions, a reference NOE interaction of consistent strength should be present in each experiment. Because the distance between the proton α to the point of cycloalkane ring-fusion to the pyrrole ring and the 6a-H

Table 1

Summary of *in situ* cycloaddition results; see structures in Fig. 1.

No.	R^1	R^2	n	R^3	Yield %	Ratio of isomers			
						En-c:	En-t:	Ex-c[a]:	Ex-t
5	Me	Ph	1	H	62	3.4	1.0	0.1	
6	Me	4-MePh	1	H	42	3.9	1.0		
7	Me	4-*i*PrPh	1	H	50	7.0	1.0		
8	Me	4-MeOPh	1	H	46	5.4	1.0		
9	Me	4-PhOPh	1	H	52	7.6	1.0		
10	Me	3-NO_2Ph	1	H	35	9.0	1.0		
11	Me	4-(CO_2H)Ph	1	H	20	8.9	1.0	0.7	
12	Me	4-BrPh	1	H	63	4.2	1.0		
13	Me	4-ClPh	1	H	65	2.5	1.0		
14	Me	4-FPh	1	H	59	11.1	1.0		
15	Me	*N*,*N*-DiMe	2	H	45	19.2	1.0		
16	Me	*N*,*N*-DiMe	2	Et	49	8.4	1.0	0.2	
17	Me	*N*,*N*-DiMe	2	*i*Pr	42	1.0			
18	Me	*N*,*N*-DiMe	2	*t*Bu	52	2.4	1.0	0.1	
19	Me	*N*,*N*-DiMe	2	Ph	48	1.0			
20	Me	Ph	2	H	60	12.5	1.0		
21	Me	Ph	2	Me	48	1.8	1.0	0.3	
22	Me	Ph	2	Et	37	5.6	1.0	0.1	
23	Me	Ph	2	*i*Pr	39	5.0	1.0		
24	Me	Ph	2	*t*Bu	38	8.3	1.0	0.2	
25	Me	Ph	2	Ph	43	3.8	1.0		
26	Me	4-MePh	2	H	42	1.0	1.6		
27	Me	4-MePh	2	Me	37	1.1	1.0	0.1	
28	Me	4-MePh	2	Et	38	2.1	1.0		
29	Me	4-MePh	2	*i*Pr	41	3.2	1.0		
30	Me	4-MePh	2	*t*Bu	27	1.0	12.4	0.6	
31	Me	4-MePh	2	Ph	41	11.9	1.0		
32	Me	4-MeOPh	2	H	34	3.5	1.0		
33	Me	4-MeOPh	2	Me	61	2.3	1.0	0.3	
34	Me	4-MeOPh	2	Et	36	4.3	1.0	0.1	
35	Me	4-MeOPh	2	*i*Pr	74	2.9	1.0	0.3	0.3
36	Me	4-MeOPh	2	*t*Bu	35	4.4	1.0	0.3	
37	Me	4-MeOPh	2	Ph	57	4.7	1.0	0.8	
38	Me	4-PhOPh	2	H	44	5.0	1.0		
39	Me	4-PhOPh	2	Me	52	1.2	1.0	0.1	
40	Me	4-PhOPh	2	Et	46	1.7	1.0	0.3	
41	Me	4-PhOPh	2	*i*Pr	42	2.1	1.0	0.3	
42	Me	4-PhOPh	2	*t*Bu	30	4.8	1.0	0.9	
43	Me	4-PhOPh	2	Ph	48	5.4	1.0	1.0	0.7
44	Me	3-NO_2Ph	2	H	40	2.8	1.0		
45	Me	3-NO_2Ph	2	Me	44	3.7	1.0		
46	Me	3-NO_2Ph	2	Et	41	1.0	3.3		
47	Me	3-NO_2Ph	2	*i*Pr	30	2.8	1.0	0.2	
48	Me	3-NO_2Ph	2	*t*Bu	31	2.1	1.0		
49	Me	3-NO_2Ph	2	Ph	40	4.2	1.0	0.6	
50	Me	4-(CO_2H)Ph	2	H	31	1.5	1.0		
51	Me	4-(CO_2H)Ph	2	Me	31	1.6	1.0		
52	Me	4-(CO_2H)Ph	2	Et	31	3.7	1.0	0.1	
53	Me	4-(CO_2H)Ph	2	*i*Pr	30	5.3	1.0	0.2	
54	Me	4-(CO_2H)Ph	2	Ph	46	4.3	1.0		
55	Me	4-BrPh	2	H	41	1.8	1.0		
56	Me	4-BrPh	2	Me	49	3.6	1.0	0.2	
57	Me	4-BrPh	2	Et	47	3.0	1.0	0.3	0.3
58	Me	4-BrPh	2	*i*Pr	41	1.8	1.0	0.3	
59	Me	4-BrPh	2	*t*Bu	31	3.3	1.0	0.5	
60	Me	4-BrPh	2	Ph	43	2.2	1.0	0.6	0.1
61	Me	4-FPh	2	H	45	1.0	1.8		
62	Me	4-FPh	2	Me	38	1.6	1.0	0.2	

(*Continued*)

Table 1. (*Continued*)

						Ratio of isomers			
No.	R^1	R^2	*n*	R^3	Yield %	En-c:	En-t:	Ex-c[a]:	Ex-t
63	Me	4-FPh	2	Et	44	1.0	2.3	0.6	
64	Me	4-FPh	2	*i*Pr	41	1.9	1.0	0.2	
65	Me	4-FPh	2	*t*Bu	34	1.6	1.0	0.3	0.2
66	Me	4-FPh	2	Ph	49	8.0	1.0	0.5	0.4
67	Me	Ph	3	H	21	3.4	1.0	0.9	
68	Me	4-*i*PrPh	3	H	3	3.0	1.0		
69	Me	4-MeOPh	3	H	11	1.9	1.0		
70	Me	3-NO_2Ph	3	H	24	4.1	1.0	0.7	
71	Me	4-ClPh	3	H	17	2.8	1.0		
72	Et	*N*,*N*-DiMe	2	H	28	8.5	1.0		
73	Et	*N*,*N*-DiMe	2	Et	31	4.1	1.0		
74	Et	*N*,*N*-DiMe	2	*t*Bu	23	14.0	1.0		
75	Et	Ph	2	H	48	1.4	1.0		
76	Et	Ph	2	Et	35	1.0	1.2	0.2	0.1
77	Et	Ph	2	*t*Bu	27	4.4	1.0	0.3	
78	Et	4-MeOPh	2	H	41	1.0	5.6		
79	Et	4-MeOPh	2	Et	36	6.5	1.0	0.3	
80	Et	4-MeOPh	2	*t*Bu	28	2.1	1.0	0.1	
81	Bn	*N*,*N*-DiMe	2	H	35	3.2	1.0		
82	Bn	*N*,*N*-DiMe	2	Et	29	5.3	1.0	0.3	
83	Bn	*N*,*N*-DiMe	2	*t*Bu	25	1.0			
84	Bn	Ph	2	H	56	3.6	1.0		
85	Bn	Ph	2	Et	36	1.0	1.7	0.6	0.4
86	Bn	Ph	2	*i*Pr	61	1.0	4.1	0.7	
87	Bn	Ph	2	*t*Bu	39	3.0	1.0	0.5	0.3
88	Bn	Ph	2	Ph	63	1.0	2.8		
89	Bn	4-MeOPh	2	H	59	3.0	1.0	0.3	
90	Bn	4-MeOPh	2	Et	36	2.5	1.0	0.3	0.2
91	Bn	4-MeOPh	2	*i*Pr	63	1.0	3.8	0.8	
92	Bn	4-MeOPh	2	*t*Bu	24	24.0	1.0	0.3	0.2
93	Bn	4-MeOPh	2	Ph	57	1.0	3.2	0.5	
94	4-MeBn	Ph	2	H	64	1.0	1.6		
95	4-MeBn	Ph	2	*i*Pr	61	1.0	2.7	0.8	
96	4-MeBn	Ph	2	Ph	64	1.0	5.0		
97	4-MeBn	4-MeOPh	2	H	65	1.0	1.9		
98	4-MeBn	4-MeOPh	2	*i*Pr	57	1.0	3.4	0.9	
99	4-MeBn	4-MeOPh	2	Ph	62	1.0	3.0		
100	4-MeOBn	*N*,*N*-DiMe	2	H	24	3.8	1.0		
101	4-MeOBn	*N*,*N*-DiMe	2	Et	22	4.0	1.0	0.2	
102	4-MeOBn	*N*,*N*-DiMe	2	*t*Bu	21	1.0			
103	4-MeOBn	Ph	2	H	60	1.0	1.8		
104	4-MeOBn	Ph	2	Et	32	1.1	1.0	0.3	0.3
105	4-MeOBn	Ph	2	*i*Pr	51	1.0	4.5	0.9	
106	4-MeOBn	Ph	2	*t*Bu	29	5.2	1.0	0.6	
107	4-MeOBn	Ph	2	Ph	61	1.0	5.2		
108	4-MeOBn	4-MeOPh	2	H	42	1.2	1.0		
109	4-MeOBn	4-MeOPh	2	Et	29	1.9	1.0	0.2	
110	4-MeOBn	4-MeOPh	2	*i*Pr	53	1.0	3.8	0.6	
111	4-MeOBn	4-MeOPh	2	*t*Bu	22	2.7	1.0	0.7	0.3
112	4-MeOBn	4-MeOPh	2	Ph	59	1.0	8.1		

[a] Ex-c is assumed to be the major *exo*-addition product.

proton should be relatively constant for the *cis*- and *trans*-fused products, NOE interactions between these two protons were used as the reference.

In the cyclohexanone-derived products, the ratio of the strength of the NOE interaction for the **En-c** isomer between the 10a-H and 6b-H protons (**a** in Fig. 2) to the 10a-H and 6a-H protons (**b**) should appear as markedly less than the ratio for the **En-t** isomer between the 10a-H and 6b-H protons (**a**′) to the 10a-H and 6a-H protons (**b**′). Restating using the labels of Figure 2, **a** is less than **a**′, and **b** is approximately equal to **b**′; therefore, **a**:**b** is less than **a**′:**b**′. For the two predominant isomers

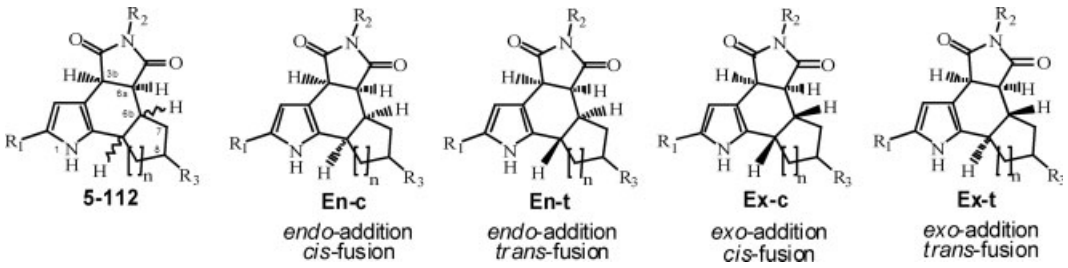


Figure 1. Stereochemistry of the tetrahydroindoles.

in the cyclohexanone-derived products, it was always observed that for one isomer the interaction between the 10a-H and 6b-H protons relative to that between the 10a-H and 6a-H protons was roughly one-third stronger (**En-c**) than for the other (**En-t**). This relationship was also observed for the cyclopentanone-derived product **13**. Thus, NOE evidence supports the assertion that the two most prevalent isomers are **En-c** and **En-t**. Unfortunately, in no ^{1}H NMR spectrum of the cycloheptanone-derived products were protons at position 11a sufficiently free from overlap to allow accurate observation and comparison of the NOE interactions.

To support the bond-length relationships used to analyze the results of the NOE experiments, a general simplified structure was used to perform computational analysis at the RHF/STO-6G level for the *endo*-addition cyclopentane, cyclohexane, and cycloheptane *cis*- and *trans*-fused products. In these simplified structures, the tetrahydroindole had a phenyl group at the 5-position and was unsubstituted at the 2-position. Calculations indicate that in the **En-c** isomer, the ratio of the distance between the proton at the point of the cycloalkane ring fusion α to the pyrrole and the 6b-H proton should differ significantly from the ratio of the distance between these protons in the **En-t** isomer. The computational models indicate that this ratio in the **En-t** isomer is 72.4, 69.2, and 69.5% of the ratio for the **En-c** isomer for the cyclopentane-, cyclohexane-, and cycloheptane-fused products, respectively.

In all nine of the representative NOE experiments performed, the ^{1}H NMR peak of the 1-H proton of the **En-c** isomer always appeared upfield from the peak corresponding to the 1-H proton of the **En-t** isomer. This consistent relationship made identifying the number of products having **En-t** as the major isomer a relatively simple process of inspecting the two predominant 1-H peaks in each spectrum; products with **En-t** as the major isomer display the unique signature of having their major 1-H peak farthest downfield. As expected, **En-c** is usually the major isomer. Out of 108 products, only 23 (21%) had **En-t** as the major isomer.

Based on the observation that there are a maximum of four isomers present, and the common general observations of minor *exo*-addition Diels-Alder products in the literature [22], it seems reasonable to assume that the minor peaks appearing in the ^{1}H NMR spectra indicate *exo*-addition products. Sufficient steric bulk of substituents on the ketone or maleimide may cancel out favorable secondary orbital interactions and allow some *exo*-approach Diels-Alder products. The two minor isomers were not present in sufficient concentration in any sample, nor were the 6b-H protons sufficiently resolved to perform NOE studies to confirm these assertions, or to check whether *cis*- or *trans*-fusion is predominant among the *exo*-addition isomers. Separation of *endo*- and *exo*-addition isomers was not achieved by chromatography nor by crystallization, which prevented analysis of individual isomers.

Diastereotopism of the protons on the methylene unit of a benzyl group is sometimes observed as second-order doublets. The 3bα-H proton appears as a doublet of doublets; COSY experiments indicate that the 3bα-H proton is coupled not only to the 6a-H proton but also to the proton at the point of cycloalkane ring-fusion α to the pyrrole ring (which would be the 10a-H proton in the cyclohexanone case), with a coupling constant of ~2.0 Hz [8,23]. In the 2-methyl compounds **5**-**71**, the 2-methyl group often appears as a doublet of doublets; COSY experiments indicate that this is due in part to ~0.9 Hz coupling with the 3-H proton [24]. COSY experiments suggest that the 2-methyl group is also sometimes coupled with the 1-H proton at ~0.9 Hz, though to the best of our knowledge, this type of coupling has no literature precedent.

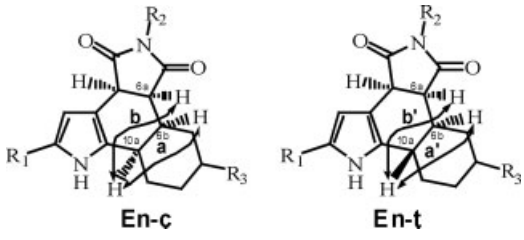


Figure 2. NOE interactions.

Biological activity. By participating in the Developmental Therapeutics Program at the National Cancer Institute (NCI), we submitted 32 representative compounds to the NCI for a one-dose three-human tumor cell line prescreen: compounds **20**, **22**, **24**, **32**, **34**, **72**, **73**, **74**, **75**, **76**, **77**, **78**, **79**, **80**, **81**, **82**, **83**, **84**, **85**, **87**, **89**, **90**, **92**, **100**, **101**, **102**, **103**, **104**, **106**, **108**, **109**, and **111**. Of these, seven compounds, **79**, **101**, **103**, **104**, **106**, **108**, and **109**, were judged by the NCI to have activity sufficient to justify screening with 60 human-tumor cell lines at five concentrations with 10-fold dilutions, from 1×10^{-4}M to 1×10^{-8}M. Of these seven compounds, compounds **103**, **106**, **108**, and **109**, were found to have high levels of activity against many of the 60 different cell lines tested. Compound **103** was most active against non-small cell lung cancer EKVX, with an IC_{50} of 113.2 μg/mL. Compound **109** was most active against colon cancer KM12, with an IC_{50} of 80.9 μg/mL. Compounds **106** and **108** were found to be active against several different cell-lines and were the best performing of the 32 compounds. Compound **106** had its highest activities against melanoma SK-MEL-5, colon cancer KM12, and breast cancer MDA-MB-435, with IC_{50} values of 62.5, 73.5, and 113.8 μg/mL, respectively. Compound **108** was most active against colon cancer HCT-15, with an IC_{50} value of 18.3 μg/mL.

CONCLUSIONS

In summary, a series of 108 novel tetrahydroindoles has been prepared *via* a Diels-Alder reaction of maleimides with 5-alkyl-2-vinylpyrroles formed *in situ* from an acid-catalyzed condensation between 2-alkylpyrroles and cyclic ketones. This one-pot method of tetrahydroindole synthesis is convenient and offers a fair-yielding and highly convergent synthetic route toward substituted indoles with good diastereoselectivity for the **En-c** isomer. Further extensions of this general methodology are currently underway in our laboratory.

EXPERIMENTAL

General. Solvents and reagents were purchased and used as received. Flash chromatography was performed using 230–450 mesh silica gel. TLC analyses were performed on plastic-backed plates precoated with 0.2 mm silica with F_{254} indicator. Infrared spectra were recorded on a 4000 FT-IR spectrometer; only the most intense and/or diagnostic peaks are reported. High-resolution mass spectra were recorded with a time-of-flight instrument using electrospray ionization with PEG as an internal calibrant. For NMR spectra, chemical shifts (δ) were referenced to the solvent. The abbreviations for splitting include: s, singlet; d, doublet; t, triplet; q, quartet; m, multiplet; br, broad. ^{13}C NMR spectra were proton decoupled. Melting points are uncallibrated. Elemental analyses were performed by M-H-W Laboratories, Phoenix, AZ.

^{1}H NMR analyses. All the Diels-Alder products are identified such that both *endo*- and *exo*-addition products have their protons 3b-H and 6a-H in the α-orientation, as shown in Figure 1. Major and minor isomers are identified when possible with the abbreviations maj and min. The ratio of products is given such that the most prevalent minor isomer is 1.0 for easy readability of the *cis*:*trans* or *trans*:*cis* ratio of the *endo*-addition isomers. When the orientation of a proton is unclear, the orientation is omitted from the identification. Insufficient resolution or peak overlap sometimes leads to the labeling of a splitting pattern as ''apparent'' (app.), which is used when there are discrepancies between the splitting of the same proton of several isomers in a single ^{1}H NMR spectrum, or when it is certain that coupling from a particular proton occurs but is not visible.

Most of the protons of the fused cycloalkane rings appear upfield as multiplets. For compounds with more than one isomer present, it would be confusing and nonintuitive to label the integration of these multiplets with multiplicity that varies depending on the number of isomers present in the mixture. Therefore, when the peaks of all the isomers overlap into a single peak, the integration is designated as 1H. When it is clear that the protons of several but not all isomers overlap in a particular peak, this multiplicity is indicated with an integration larger than 1H. When it is not clear whether multiple peaks overlap, the integration reflects the number of protons which are thought to be definitely in the peak. Thus, sometimes, fewer isomers are identified for a particular proton than there are isomers present in the mixture, because it is not clear where the peak(s) from one isomer occurs.

With some protons, the peaks belonging to the various isomers overlap. In these cases, sometimes the peak is identified as it would be if there was a single isomer present, omitting the designation maj and min, and also omitting the designation α or β if the orientation is unknown or mixed. These designations are only omitted when it is clear, which isomers overlap into a single identified peak, and when it is clear, the protons are of mixed or unknown orientation. Overlap of signals from a proton with multiple orientations occurs most frequently with protons at the 6b-position in compounds with more than one minor isomer present. In the case of a compound with three isomers present, with the peak from 6bαmaj-H distinct but the peaks from the 6bαmin-H and 6bβmin-H protons overlapping into one, the overlapped peak is labeled 6b-H and is assigned an integration of two. This situation also occurs with peaks belonging to protons α to the point of cycloalkane ring fusion to the pyrrole ring in compounds with more than one isomer present.

General reaction conditions. Method A: A solution of the pyrrole (3.00 mmol), the cyclohexanone (4.00 mmol), and the maleimide (4.00 mmol) was heated to reflux in ethanol (5.0 mL). Hydrochloric acid (0.20 mL, 37% aqueous solution) was added to the hot solution, causing it to turn red-brown in color. The solution was refluxed for 1 h. In most cases, slow precipitation of the *in situ* product was observed throughout this time. After the mixture had cooled to rt, the precipitate was vacuum-filtered, washed with ethanol (5.0 mL), and reprecipitated from ethanol (5.0 mL). In cases where no precipitate was observed during reflux, which occurred particularly when 4-*tert*-butylcyclohexanone and/or 4-methoxyphenylmaleimide were used, the desired product was isolated by flash chromatography on silica gel using ethyl acetate:hexane as the eluent.

Method B: Hydrochloric acid (0.10 mL, 37% aqueous solution) was added to a solution of the pyrrole (5.00 mmol), the

cyclic ketone (6.50–9.82 mmol), and the maleimide (4.80 mmol) in ethanol (15.0 mL), and the resulting solution was refluxed with stirring for 1–6 h, as determined by TLC. As the solution was allowed to cool to rt, a precipitate developed, which was vacuum-filtered. Purification to give the desired product was accomplished in one of several ways: (1) washing with diethyl ether (5–20 mL) and/or ethanol (5–20 mL), (2) reprecipitation from ethanol (15–20 mL) and/or diethyl ether (15–20 mL) and then, if necessary, washing with diethyl ether (5–20 mL), (3) purified using flash chromatography on silica gel, or (4) a combination of the above techniques, as noted.

Compounds 5 through 112. ***2-Methyl-5-phenyl-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (5).*** Method B with **3a** (800 mg, 9.50 mmol), 2-h reflux, ethanol wash (10 mL), and then a diethyl ether wash (10 mL) gave **5** (950 mg, 62%) as a colorless solid, a mixture of three isomers (maj:min:min = 3.4:1.0:0.1): mp 260–262°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.55 (bs, 1H, 1min-H), 8.22 (bs, 1H, 1min-H), 7.64 (bs, 1H, 1maj-H), 7.36–7.52 (m, 3H, Ph), 7.26–7.31 (m, 2H, Ph), 6.11 (dd, *J* = 2.6, 1.1 Hz, 1H, 3maj-H), 6.03–6.05 (m, 1H, 3min-H), 5.76 (app. d, *J* = 3.0 Hz, 1H, 3min-H), 4.02 (dd, *J* = 8.3, 1.7 Hz, 1H, 3bαmin-H), 4.01 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.63 (dd, *J* = 8.9, 6.2 Hz, 1H, 6aαmin-H), 3.55 (dd, *J* = 8.4, 6.0 Hz, 1H, 6aαmaj-H), 3.20–3.27 (m, 1H, 9aαmaj-H), 3.10–3.16 (m, 1H, 9aβmin-H), 2.75–2.88 (m, 1H, 6b-H), 2.29 (s, 3H, 2-CH_3), 1.88–2.08 (m, 2H, cyclopent.), 1.57–1.75 (m, 3H, cyclopent.), 1.38–1.49 (m, 1H, cyclopent.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.48 (bs, 1H, 1min-H), 10.29 (bs, 1H, 1maj-H), 7.38–7.54 (m, 3H, Ph), 7.19–7.25 (m, 2H, Ph), 5.76 (dd, *J* = 2.3, 1.1 Hz, 1H, 3maj-H), 5.57 (dd, *J* = 2.3, 1.1 Hz, 1H, 3min-H), 4.18 (app. d, *J* = 8.7 Hz, 1H, 3bαmin-H), 4.05 (dd, *J* = 8.3, 2.0 Hz, 1H, 3bαmaj-H), 3.53 (dd, *J* = 8.3, 5.9 Hz, 1H, 6aαmin-H), 3.48 (dd, *J* = 8.3, 5.9 Hz, 1H, 6aαmaj-H), 3.07–3.14 (m, 1H, 9aαmaj-H), 2.99–3.04 (m, 1H, 9aβmin-H), 2.56–2.66 (m, 1H, 6b-H), 2.04–2.18 (m, 1H, cyclopent.), 2.15 (s, 3H, 2-CH_3), 1.77–1.94 (m, 1H, cyclopent.), 1.36–1.61 (m, 3H, cyclopent.), 1.15–1.30 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.4, 177.0, 132.2, 131.0, 129.3, 129.2, 128.5, 128.1, 127.8, 126.6, 115.5, 109.3, 108.7, 105.6, 104.1, 41.9, 41.7, 38.5, 37.2, 36.5, 31.1, 30.5, 24.9, 22.4, 21.9, 13.3; IR (thin film, cm^{-1}) 3397(bs), 3059(m), 2934(m), 2857(m), 1775(s), 1695(s), 1498, 1391(m), 1189(m), 1170(m); HRMS *m*/*z* ($M + Na^+$) calcd 343.1418, found 343.1417. Anal. Calcd for $C_{20}H_{20}N_2O_2$: C, 74.98; H, 6.29; N, 8.74. Found: C, 75.20; H, 6.16; N, 8.90.

2-Methyl-5-(4-methylphenyl)-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (6). Method B with **3a** (800 mg, 9.50 mmol), 3.5-h reflux, reprecipitation from ethanol (15 mL), and then a diethyl ether wash (10 mL) gave **6** (670 mg, 42%) as a colorless solid, a mixture of two isomers (maj:min = 3.9:1.0): mp 214–216°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.23 (bs, 1H, 1min-H), 7.65 (bs, 1H, 1maj-H), 7.27 (d, *J* = 7.8 Hz, 2H, Ph), 7.16 (d, *J* = 7.8 Hz, 2H, Ph), 6.11 (dd, *J* = 2.6, 1.1 Hz, 1H, 3maj-H), 5.75 (dd, *J* = 2.6, 0.75 Hz, 1H, 3min-H), 3.99 (dd, *J* = 8.6, 2.0 Hz, 1H, 3bα-H), 3.62 (dd, *J* = 8.9, 6.2 Hz, 1H, 6aαmin-H), 3.53 (dd, *J* = 8.3, 6.2 Hz, 1H, 6aαmaj-H), 3.19–3.26 (m, 1H, 9aαmaj-H), 3.10–3.16 (m, 1H, 9aβmaj-H), 2.73–2.88 (m, 1H, 6bα-H), 2.40 (s, 3H, 4′-CH_3 min), 2.39 (s, 3H, 4′-CH_3 maj), 2.28 (s, 3H, 2-CH_3), 1.88–2.07 (m, 2H, cyclopent.), 1.50–1.75 (m, 3H, cyclopent.), 1.25–1.49 (m, 1H, cyclopent.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.47 (d, *J* = 2.1 Hz, 1H, 1min-H), 10.28 (d, *J* = 1.8 Hz, 1H, 1maj-H), 7.28 (d, *J* = 7.8 Hz, 2H, Ph), 7.08 (d, *J* = 8.4 Hz, 2H, Ph), 5.75 (dd, *J* = 2.3, 1.1 Hz, 1H, 3maj-H), 5.57 (dd, *J* = 2.4, 0.6 Hz, 1H, 3min-H), 4.15 (dd, *J* = 7.2, 1.2 Hz, 1H, 3bαmin-H), 4.02 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.51 (dd, *J* = 8.3, 5.9 Hz, 1H, 6aαmin-H), 3.46 (dd, *J* = 8.4, 6.0 Hz, 1H, 6aαmaj-H), 3.07–3.13 (m, 1H, 9aαmaj-H), 2.98–3.04 (m, 1H, 9aβmin-H), 2.52–2.65 (m, 1H, 6bα-H), 2.35 (s, 3H, 4′-CH_3 maj), 2.34 (s, 3H, 4′-CH_3 min), 2.02–2.18 (m, 1H, cyclopent.), 2.15 (s, 3H, 2-CH_3), 1.77–1.89 (m, 1H, cyclopent.), 1.34–1.60 (m, 3H, cyclopent.), 1.14–1.29 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.6, 177.1, 138.6, 130.0, 129.9, 129.5, 128.0, 127.8, 126.4, 108.7, 105.6, 104.1, 41.9, 41.8, 41.2, 38.5, 37.2, 36.5, 31.4, 30.5, 24.9, 24.5, 22.4, 21.9, 21.3, 13.3; IR (thin film, cm^{-1}) 3381(bs), 2948(m), 2871(m), 2366(w), 1775(w), 1706(s), 1514(m), 1383(m), 1194(m), 1179(m), 1162(m); HRMS *m*/*z* ($M + Na^+$) calcd 357.1574, found 357.1572. Anal. Calcd for $C_{21}H_{22}N_2O_2$: C, 75.42; H, 6.63; N, 8.38. Found: C, 75.38; H, 6.58; N, 8.55.

5-(4-Isopropylphenyl)-2-methyl-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (7). Method B with **3a** (800 mg, 9.50 mmol), 4-h reflux, and then a diethyl ether wash (20 mL) gave **7** (760 mg, 50%) as a colorless solid, a mixture of two isomers (maj:min = 7.0:1.0): mp 199–201°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.30 (bs, 1H, 1min-H), 7.69 (bs, 1H, 1maj-H), 7.32 (d, *J* = 8.4 Hz, 2H, Ph), 7.19 (d, *J* = 8.4 Hz, 2H, Ph), 6.11 (d, *J* = 1.8 Hz, 1H, 3maj-H), 5.75 (d, *J* = 2.1 Hz, 1H, 3min-H), 4.01 (dd, *J* = 8.3, 1.7 Hz, 1H, 3bαmin-H), 3.99 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.62 (dd, *J* = 8.9, 6.2 Hz, 1H, 6aαmin-H), 3.53 (dd, *J* = 8.4, 5.7 Hz, 1H, 6aαmaj-H), 3.19–3.25 (m, 1H, 9aαmaj-H), 3.10–3.16 (m, 1H, 9aβmin-H), 2.96 (septet, *J* = 6.9 Hz, 1H, C*H*(CH_3)$_2$ min), 2.95 (septet, *J* = 6.9 Hz, 1H, C*H*(CH_3)$_2$ maj), 2.75–2.88 (m, 1H, 6bα-H), 2.28 (s, 3H, 2-CH_3), 1.88–2.09 (m, 2H, cyclopent.), 1.33–1.74 (m, 4H, cyclopent.), 1.271 (d, *J* = 6.9 Hz, 1H, CH(C*H*$_3$)$_2$ min), 1.269 (d, *J* = 6.6 Hz, 6H, CH(C*H*$_3$)$_2$ maj); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.48 (d, *J* = 2.1 Hz, 1H, 1min-H), 10.28 (bs, *J* = 1.8 Hz, 1H, 1maj-H), 7.35 (d, *J* = 8.4 Hz, 2H, Ph), 7.12 (d, *J* = 8.4 Hz, 2H, Ph), 5.76 (d, *J* = 1.2 Hz, 1H, 3maj-H), 5.57 (d, *J* = 1.5 Hz, 1H, 3min-H), 4.16 (app. d, *J* = 8.4 Hz, 1H, 3bαmin-H), 4.03 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.52 (dd, *J* = 8.3, 5.9 Hz, 1H, 6aαmin-H), 3.46 (dd, *J* = 8.4, 6.0 Hz, 1H, 6aαmaj-H), 3.03–3.14 (m, 1H, 9aαmaj-H), 2.97–3.04 (m, 1H, 9aβmin-H), 2.94 (septet, *J* = 6.9 Hz, 1H, C*H*(CH_3)$_2$ min), 2.93 (septet, *J* = 6.9 Hz, 1H, C*H*(CH_3)$_2$ maj), 2.53–2.65 (m, 1H, 6bα-H), 2.03–2.19 (m, 1H, cyclopent.), 2.15 (s, 3H, 2-CH_3), 1.77–1.91 (m, 1H, cyclopent.), 1.35–1.61 (m, 4H, cyclopent.), 1.23 (d, *J* = 6.9 Hz, 6H, CH(C*H*$_3$)$_2$ min), 1.22 (d, *J* = 6.9 Hz, 6H, CH(C*H*$_3$)$_2$ maj); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.6, 177.2, 149.3, 129.7, 128.0, 127.9, 127.3, 126.3, 108.7, 105.6, 41.9, 41.7, 38.5, 36.5, 34.0, 30.5, 24.9, 24.0, 22.4, 13.3; IR (thin film, cm^{-1}) 3378(bs), 2961(m), 2872(m), 1774(w), 1701(s), 1515(m), 1384(m), 1182(m), 1162(m); HRMS *m*/*z* ($M + Na^+$) calcd 385.1887, found 385.1886. Anal. Calcd for $C_{23}H_{26}N_2O_2$: C, 76.21; H, 7.23; N, 7.73. Found: C, 76.18; H, 7.41; N, 7.51.

5-(4-Methoxyphenyl)-2-methyl-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (8). Method B with **3a** (800 mg, 9.50 mmol), 1.5-h reflux, and then reprecipitation from ethanol (15 mL) gave **8** (850 mg, 46%) as a

colorless solid, a mixture of two isomers (maj:min = 5.4:1.0): mp 213–215°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.25 (bs, 1H, 1min-H), 7.69 (bs, 1H, 1maj-H), 7.15–7.23 (m, 2H, Ph), 6.95–7.02 (m, 2H, Ph), 6.11 (d, J = 1.5 Hz, 1H, 3maj-H), 5.75 (d, J = 2.1 Hz, 1H, 3min-H), 3.99 (dd, J = 8.4, 1.8 Hz, 1H, 3bα-H), 3.84 (s, 3H, OCH_3 min), 3.83 (s, 3H, OCH_3 maj), 3.61 (dd, J = 8.7, 6.3 Hz, 1H, 6aαmin-H), 3.53 (dd, J = 8.6, 5.9 Hz, 1H, 6aαmaj-H), 3.18–3.25 (m, 1H, 9aαmaj-H), 3.09–3.16 (m, 1H, 9aβmin-H), 2.74–2.87 (m, 1H, 6b-H), 2.28 (s, 3H, 2-CH_3), 1.88–2.07 (m, 2H, cyclopent.), 1.26–1.70 (m, 4H, cyclopent.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.46 (bs, 1H, 1min-H), 10.27 (bs, 1H, 1maj-H), 7.12 (d, J = 9.0 Hz, 2H, Ph), 7.02 (d, J = 9.0 Hz, 2H, Ph), 5.74–5.77 (m, 1H, 3maj-H), 5.55–5.58 (m, 1H, 3min-H), 4.12–4.16 (m, 1H, 3bαmin-H), 4.01 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.78 (s, 3H, OCH_3), 3.45–3.52 (m, overlapped, 1H, 6aαmin-H), 3.45 (dd, J = 8.3, 5.9 Hz, 1H, 6aαmaj-H), 3.06–3.13 (m, 1H, 9aαmaj-H), 2.98–3.04 (m, 1H, 9aβmin-H), 2.52–2.65 (m, 1H, 6bα-H), 1.99–2.18 (m, 1H, cyclopent.), 2.15 (s, 3H, 2-CH_3), 1.78–1.96 (m, 1H, cyclopent.), 1.32–1.62 (m, 3H, cyclopent.), 1.12–1.28 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.7, 177.3, 159.5, 128.0, 127.8, 124.8, 114.7, 114.6, 108.7, 105.6, 55.6, 41.9, 41.7, 38.5, 37.2, 36.5, 30.5, 31.5, 24.9, 22.3, 22.0, 13.3; IR (KBr, cm^{-1}) 3384(bs), 2869(m), 1773(w), 1704(s), 1697(bs), 1515(s), 1391(m), 1252(m), 1176(m); HRMS m/z (M + Na^+) calcd 373.1523, found 373.1528. Anal. Calcd for $C_{21}H_{22}N_2O_3$: C, 71.98; H, 6.33; N, 7.99. Found: C, 72.12; H, 6.51; N, 7.82.

2-Methyl-5-(4-phenoxyphenyl)-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (9). Method B with **3a** (800 mg, 9.50 mmol), 3-h reflux, reprecipitation from ethanol (15 mL), and then a diethyl ether wash (15 mL) gave **9** (1130 mg, 52%) as a colorless solid, a mixture of two isomers (maj:min = 7.6:1.0): mp 227–228°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.22 (bs, 1H, 1min-H), 7.65 (bs, 1H, 1maj-H), 7.34–7.41 (m, 2H, Ph), 7.13–7.28 (m, 3H, Ph), 7.04–7.10 (m, 4H, Ph), 6.11 (dd, J = 2.4, 0.9 Hz, 1H, 3maj-H), 5.76 (app. d, J = 2.4 Hz, 1H, 3min-H), 4.00 (dd, J = 8.4, 1.8 Hz, 1H, 3bα-H), 3.84 (dd, J = 8.4, 6.0 Hz, 1H, 6aαmaj-H), 3.63 (dd, J = 8.6, 6.2 Hz, 1H, 6aαmin-H), 3.20–3.27 (m, 1H, 9aαmaj-H), 3.13–3.17 (m, 1H, 9aβmin-H), 2.75–2.88 (m, 1H, 6bα-H), 2.29 (s, 3H, 2-CH_3), 1.88–2.08 (m, 2H, cyclopent.), 1.53–1.73 (m, 3H, cyclopent.), 1.30–1.48 (m, 1H, cyclopent.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.48 (d, J = 2.4 Hz, 1H, 1min-H), 10.28 (d, J = 2.1 Hz, 1H, 1maj-H), 7.40–7.47 (m, 2H, Ph), 7.16–7.26 (m, 3H, Ph), 7.07–7.11 (m, 4H, Ph), 5.76 (dd, J = 2.1, 0.9 Hz, 1H, 3maj-H), 5.57 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H), 4.17 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.03 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.52 (dd, J = 8.3, 5.9 Hz, 1H, 6aαmin-H), 3.47 (dd, J = 8.3, 5.9 Hz, 1H, 6aαmaj-H), 3.07–3.13 (m, 1H, 9aαmaj-H), 2.98–3.04 (m, 1H, 9aβmin-H), 2.54–2.65 (m, 1H, 6bα-H), 2.15 (s, 3H, 2-CH_3), 2.02–2.15 (m, 1H, cyclopent.), 1.76–1.90 (m, 1H, cyclopent.), 1.35–1.61 (m, 3H, cyclopent.), 1.13–1.29 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.5, 177.1, 157.5, 156.5, 130.0, 128.1, 128.0, 127.8, 126.9, 124.0, 119.6, 118.9, 108.6, 105.6, 41.9, 41.7, 38.5, 37.2, 36.5, 30.5, 24.9, 22.4, 13.3; IR (thin film, cm^{-1}) 3381(bs), 2950(m), 2872(m), 2365(w), 2343(w), 1775(w), 1706(s), 1590(w), 1507(m), 1489(m), 1385(m), 1240(m), 1163(m); HRMS m/z (M + Na^+) calcd 435.1680, found 435.1682. Anal. Calcd for $C_{26}H_{24}N_2O_3$: C, 75.71; H, 5.86; N, 6.79. Found: C, 75.86; H, 5.73; N, 6.76.

4-(2-Methyl-4,6-dioxo-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]-5-indolyl) benzoic acid (10) Method B with **3a** (800 mg, 9.50 mmol), 1.5-h reflux, reprecipitation from ethanol (15 mL), and then a diethyl ether wash (10 mL) gave **10** (600 mg, 35%) as a colorless solid, a mixture of two isomers (maj:min = 9.0:1.0): mp 262–264°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 13.10 (bs, 1H, CO_2H), 10.49 (d, J = 3.0 Hz, 1H, 1min-H), 10.29 (d, J = 3.0 Hz, 1H, 1maj-H), 8.05 (d, J = 8.4 Hz, 2H, Ph), 7.39 (d, J = 8.7 Hz, 2H, Ph), 5.76 (d, J = 1.5 Hz, 1H, 3maj-H), 5.58 (d, J = 1.8 Hz, 1H, 3min-H), 4.21 (app. d, J = 8.7 Hz, 1H, 3aαmin-H), 4.04 (dd, J = 8.3, 1.7 Hz, 1H, 3aαmaj-H), 3.56 (dd, J = 8.1, 5.4 Hz, 1H, 6aαmin-H), 3.51 (dd, J = 8.3, 5.9 Hz, 1H, 6aαmaj-H), 3.08–3.15 (m, 1H, 9aαmaj-H), 3.00–3.05 (m, 1H, 9aβmin-H), 2.56–2.67 (m, 1H, 6bα-H), 2.15 (s, 3H, 2-CH_3), 2.02–2.15 (m, 1H, cyclopent.), 1.77–1.90 (m, 1H, cyclopent.), 1.33–1.64 (m, 3H, cyclopent.), 1.15–1.27 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.4, 177.3, 167.9, 157.2, 136.8, 130.9, 130.6, 127.6, 127.4, 127.1, 108.0, 105.2, 42.1, 41.7, 38.5, 36.8, 31.2, 30.4, 25.1, 22.4, 13.4; IR (thin film, cm^{-1}) 3394(bs), 2910(m), 1773(w),1696(s), 1515(w), 1391(m), 1289(m), 1172(m); HRMS m/z (M + Na^+) calcd for $C_{21}H_{20}N_2O_4$: 387.1316, found 387.1302.

2-Methyl-5-(3-nitrophenyl)-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (11). Method B with **3a** (800 mg, 9.50 mmol), 4-h reflux and then purification with column chromatography (CH_2Cl_2) gave **11** (350 mg, 20%) as a yellow solid, a mixture of three isomers (maj:min:min = 8.9:1.0:0.7): mp 212–216°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.24–8.28 (m, 3H, Ph, Ph, 1min-H), 7.63–7.74 (m, 3H, Ph, 1maj-H), 6.10 (dd, J = 2.6, 1.1 Hz, 1H, 3maj-H), 6.03 (dd, J = 2.9, 1.1 Hz, 1H, 3min-H), 5.77 (dd, J = 2.6, 1.1 Hz, 1H, 3min-H), 4.20 (dd, J = 8.1, 1.8 Hz, 1H, 3bαmin-H), 4.05 (dd, J = 8.4, 2.1 Hz, 1H, 3bαmaj-H), 3.68 (dd, J = 8.6, 6.2 Hz, 1H, 6aαmin-H), 3.63 (dd, J = 8.1, 4.2 Hz, 1H, 6aαmin-H), 3.60 (dd, J = 8.6, 5.9 Hz, 1H, 6aαmaj-H), 3.21–3.29 (m, 1H, 9aαmaj-H), 3.10–3.19 (m, 1H, 9amin-H), 2.79–2.89 (m, 1H, 6b-H), 2.30 (dd, J = 0.8 Hz, 3H, 2-CH_3), 1.90–2.05 (m, 2H, cyclopent.), 1.58–1.74 (m, 3H, cyclopent.), 1.34–1.49 (m, 1H, cyclopent.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.52 (d, J = 1.2 Hz, 1H, 1min-H), 10.46 (d, J = 1.8 Hz, 1H, 1min-H), 10.32 (d, J = 1.8 Hz, 1H, 1maj-H), 8.25–8.32 (m, 1H, Ph), 8.15–8.17 (m, 1H, Ph), 7.73–7.86 (m, 2H, Ph), 5.78 (d, J = 1.5 Hz, 1H, 3maj-H), 5.72 (d, J = 1.8 Hz, 1H, 3min-H), 5.58 (d, J = 1.8 Hz, 1H, 3min-H), 4.22 (app. d, J = 8.1 Hz, 1H, 3bαmin-H), 4.09 (dd, J = 8.4, 2.1 Hz, 1H, 3bαmaj-H), 4.03 (dd, J = 8.3, 1.7 Hz, 1H, 3bαmin-H), 3.72 (dd, J = 4.7, 8.0 Hz, 1H, 6aαmin-H), 3.60 (dd, J = 8.3, 5.6 Hz, 1H, 6aαmin-H), 3.54 (dd, J = 8.1, 6.0 Hz, 1H, 6aαmaj-H), 3.09–3.15 (m, 1H, 9aαmaj-H), 2.99–3.06 (m, 1H, 9amin-H), 2.58–2.67 (m, 1H, 6b-H), 2.03–2.17 (m, 1H, cyclopent.), 2.15 (s, 3H, 2-CH_3), 1.74–1.91 (m, 1H, cyclopent.), 1.37–1.64 (m, 3H, cyclopent.), 1.15–1.33 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.6, 176.3, 133.2, 132.3, 130.0, 128.3, 127.8, 123.1, 121.7, 108.1, 105.5, 42.0, 41.6, 38.6, 36.5, 30.5, 25.0, 22.4, 13.3; IR (thin film, cm^{-1}) 3388(bs), 2953(m), 2926(m), 1779(w),1712(s), 1532(s), 1376(m), 1349(m), 1159(m); HRMS m/z (M + Na^+) calcd for $C_{20}H_{19}N_3O_4$: 388.1269, found 388.1258.

5-(4-Bromophenyl)-2-methyl-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (12). Method B with **3a** (800 mg, 9.50 mmol), 1.5-h reflux, and then

reprecipitation from ethanol (15 mL) gave **12** (1250 mg, 63%) as a colorless solid, a mixture of two isomers (maj:min = 4.2:1.0): mp 266–268°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.20 (bs, 1H, 1min-H), 7.63 (bs, overlapped, 1H, 1maj-H), 7.60 (d, J = 8.7 Hz, 2H, Ph), 7.20 (d, J = 8.7 Hz, 2H, Ph), 6.10 (dd, J = 2.6, 1.1 Hz, 1H, 3maj-H), 5.75 (dd, J = 2.9, 0.8 Hz, 1H, 3min-H), 4.00 (dd, J = 8.6, 1.5 Hz, 1H, 3bα-H), 3.63 (dd, J = 8.6, 6.2 Hz, 1H, 6aαmin-H), 3.54 (dd, J = 8.4, 6.0 Hz, 1H, 6aαmaj-H), 3.20–3.27 (m, 1H, 9aαmaj-H), 3.10–3.16 (m, 1H, 9aβmin-H), 2.74–2.88 (m, 1H, 6bα-H), 2.29 (s, 3H, 2-CH_3), 1.87–2.08 (m, 2H, cyclopent.), 1.52–1.72 (m, 3H, cyclopent.), 1.23–1.48 (m, 1H, cyclopent.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.49 (d, J = 2.4 Hz, 1H, 1min-H), 10.29 (d, J = 1.8 Hz, 1H, 1maj-H), 7.70 (d, J = 8.4 Hz, 2H, Ph), 7.20 (d, J = 8.7 Hz, 2H, Ph), 5.75 (d, J = 2.3, 0.75 Hz, 1H, 3maj-H), 5.57 (d, J = 2.4, 0.6 Hz, 1H, 3min-H), 4.17 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.04 (dd, J = 8.1, 1.8 Hz, 1H, 3bαmaj-H), 3.54 (dd, J = 8.4, 5.7 Hz, 1H, 6aαmin-H), 3.48 (dd, J = 8.3, 5.9 Hz, 1H, 6aαmaj-H), 3.07–3.13 (m, 1H, 9aαmaj-H), 2.98–3.04 (m, 1H, 9aβmin-H), 2.53–2.65 (m, 1H, 6b-H), 2.02–2.18 (m, 1H, cyclopent.), 2.15 (s, 3H, 2-CH_3), 1.77–1.89 (m, 1H, cyclopent.), 1.33–1.61 (m, 3H, cyclopent.), 1.13–1.25 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.5, 132.5, 132.4, 128.0, 127.8, 105.6, 41.9, 41.6, 38.5, 36.5, 24.9, 22.4, 13.3; IR (thin film, cm^{-1}) 3396(bs), 2872(m), 2364(m), 1774(w), 1697(s), 1490(m), 1387(m), 1177(m), 1167(m); HRMS m/z (M + Na^+) calcd 421.0523, found 421.0519. Anal. Calcd for $C_{20}H_{19}BrN_2O_2$: C, 60.16; H, 4.80; N, 7.02. Found: C, 60.25; H, 4.98; N, 7.14.

5-(4-Chlorophenyl)-2-methyl-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (13). Method B with **3a** (800 mg, 9.50 mmol), 2-h reflux, reprecipitation from ethanol (10 mL), and then a diethyl ether wash (10 mL) gave **13** (1100 mg, 65%) as a colorless solid, a mixture of two isomers (maj:min = 2.5:1.0): mp 257–260°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.22 (bs, 1H, 1min-H), 7.67 (bs, 1H, 1maj-H), 7.44 (d, J = 8.7 Hz, 2H, Ph), 7.26 (d, J = 9.0 Hz, 2H, Ph), 6.10 (dd, J = 2.6, 1.1 Hz, 1H, 3maj-H), 5.76 (dd, J = 2.7, 0.9 Hz, 1H, 3min-H), 4.01 (dd, J = 8.7, 2.1 Hz, 1H, 3bαmin-H), 4.00 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.62 (dd, J = 8.9, 6.2 Hz, 1H, 6aαmin-H), 3.54 (dd, J = 8.4, 5.7 Hz, 1H, 6aαmaj-H), 3.20–3.26 (m, 1H, 9aαmaj-H), 3.10–3.16 (m, 1H, 9aβmin-H), 2.72–2.87 (m, 1H, 6bα-H), 2.29 (s, 3H, 2-CH_3), 1.87–2.07 (m, 2H, cyclopent.), 1.52–1.75 (m, 3H, cyclopent.), 1.22–1.49 (m, 1H, cyclopent.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.49 (d, J = 1.8 Hz, 1H, 1min-H), 10.29 (d, J = 2.7 Hz, 1H, 1maj-H), 7.57 (d, J = 8.7 Hz, 2H, Ph), 7.27 (d, J = 8.7 Hz, 2H, Ph), 5.76 (d, J = 2.1 Hz, 1H, 3maj-H), 5.57 (d, J = 1.8 Hz, 1H, 3min-H), 4.17 (app. d, J = 8.1 Hz, 1H, 3bαmin-H), 4.04 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.54 (dd, J = 8.3, 5.9 Hz, 1H, 6aαmin-H), 3.49 (dd, J = 8.3, 5.9 Hz, 1H, 6aαmaj-H), 3.07–3.14 (m, 1H, 9aαmaj-H), 2.98–3.04 (m, 1H, 9aβmin-H), 2.53–2.65 (m, 1H, 6bα-H), 2.02–2.17 (m, 1H, cyclopent.), 2.15 (s, 3H, 2-CH_3), 1.76–1.93 (m, 1H, cyclopent.), 1.35–1.62 (m, 3H, cyclopent.), 1.12–1.28 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.1, 176.7, 134.3, 130.6, 129.6, 129.4, 128.1, 127.8, 121.2, 115.8, 108.4, 105.6, 104.1, 41.6, 41.1, 38.5, 37.2, 36.5, 31.4, 31.1, 30.5, 24.9, 24.5, 22.4, 21.9, 13.3; IR (thin film, cm^{-1}) 3398(bs), 2929(m), 1774(w), 1696(s), 1494(m), 1391(m), 1177(m), 1168(m); HRMS m/z (M + Na^+) calcd 377.1028, found 377.1023. Anal. Calcd for $C_{20}H_{19}ClN_2O_2$: C, 67.70; H, 5.40; N, 7.89. Found: C, 67.81; H, 5.35; N, 8.07.

5-(4-Fluorophenyl)-2-methyl-3b,6a,6b,7,8,9,9a-heptahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indole-4,6-dione (14). Method B with **3a** (800 mg, 9.50 mmol), 2-h reflux, reprecipitation from ethanol (20 mL), and then a diethyl ether wash (10 mL) gave **14** (950 mg, 59%) as a colorless solid, a mixture of two isomers (maj:min = 11.1:1.0): mp 230–232°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.24 (bs, 1H, 1min-H), 7.69 (bs, 1H, 1maj-H), 7.24–7.31 (m, 2H, Ph), 7.11–7.20 (m, 2H, Ph), 6.10 (dd, J = 2.7, 1.2 Hz, 1H, 3maj-H), 5.75 (d, J = 3.3 Hz, 1H, 3min-H), 4.00 (dd, J = 8.4, 2.1 Hz, 1H, 3bα-H), 3.62 (dd, J = 8.6, 6.2 Hz, 1H, 6aαmin-H), 3.54 (dd, J = 8.4, 6.0 Hz, 1H, 6aαmaj-H), 3.19–3.26 (m, 1H, 9aαmaj-H), 3.10–3.16 (m, 1H, 9aβmin-H), 2.76–2.87 (m, 1H, 6bα-H), 2.28 (s, 3H, 2-CH_3), 1.88–2.07 (m, 2H, cyclopent.), 1.53–1.72 (m, 3H, cyclopent.), 1.33–1.48 (m, 1H, cyclopent.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.49 (bs, 1H, 1min-H), 10.29 (bs, 1H, 1maj-H), 7.24–7.39 (m, 4H, Ph), 5.76 (dd, J = 2.1, 0.6 Hz, 1H, 3maj-H), 5.57 (app. d, J = 1.8 Hz, 1H, 3min-H), 4.17 (app. d, J = 9.3 Hz, 1H, 3bαmin-H), 4.04 (dd, J = 8.1, 1.8 Hz, 1H, 3bαmaj-H), 3.48 (dd, J = 8.4, 5.7 Hz, 1H, 6aαmaj-H), 3.44 (dd, J = 7.1, 5.0 Hz, 1H, 6aαmin-H), 3.07–3.14 (m, 1H, 9aαmaj-H), 2.98–3.04 (m, 1H, 9aβmin-H), 2.53–2.65 (m, 1H, 6bα-H), 2.02–2.18 (m, 1H, cyclopent.), 2.15 (s, 3H, 2-CH_3), 1.76–1.89 (m, 1H, cyclopent.), 1.34–1.61 (m, 3H, cyclopent.), 1.13–1.29 (m, 1H, cyclopent.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.3, 176.9, 163.9, 160.1, 128.4, 128.3, 128.1, 127.8, 116.5, 116.4, 116.1, 108.5, 105.6, 41.9, 41.6, 38.5, 36.5, 31.0, 30.5, 24.9, 22.3, 13.3; IR (thin film, cm^{-1}) 3387(bs), 2876(m), 1775(w), 1706(s), 1510(s), 1510(m), 1387(m), 1229(m), 1189(m), 1159(m); HRMS m/z (M + Na^+) calcd 361.1324, found 361.1323. Anal. Calcd for $C_{20}H_{19}FN_2O_2$: C, 70.99; H, 5.66; N, 8.28. Found: C, 71.03; H, 5.71; N, 8.21.

5-Dimethylamino-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (15). Method A gave **15** (407 mg, 45%) as a light-orange solid, a mixture of two isomers (maj:min = 19.2:1.0): mp 234–236°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.25 (bs, 1H, 1min-H), 7.65 (bs, 1H, 1maj-H), 6.15 (dd, J = 2.4, 1.2 Hz, 1H, 3maj-H), 5.74 (dd, J = 2.6, 0.7 Hz, 1H, 3min-H), 3.68 (dd, J = 2.0 Hz, 1H, 3bα-H), 3.23 (dd, J = 8.9, 5.6 Hz, 1H, 6aαmin-H), 3.17 (dd, J = 8.6, 5.6 Hz, 1H, 6aαmaj-H), 3.04–3.09 (m, 1H, 10aαmaj-H), 2.92 (s, 6H, $N(CH_3)_2$), 2.30 (dd, J = 1.1, 1.1 Hz, 3H, 2-CH_3), 2.43–2.53 (m, 1H, 6b-H), 2.08–2.16 (m, 1H, cyclohex.), 1.45–1.79 (m, 3H, cyclohex.), 1.05–1.32 (m, 4H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.5, 176.6, 127.4, 127.0, 108.9, 105.6, 44.2, 44.1, 38.2, 37.0, 32.7, 27.9, 25.5, 22.8, 21.0, 13.3; IR (thin film, cm^{-1}) 3426(bs), 2930(m), 2859(m), 2124(bw), 1770(bw), 1705(s), 1648(bs), 1446(m), 1362(m), 1193(m), 1146(m); HRMS m/z (M + Na^+) calcd 324.1683, found 324.1707. Anal. Calcd for $C_{17}H_{23}N_3O_2$: C, 67.75; H, 7.69; N, 13.94. Found: C, 67.92; H, 7.69; N, 13.76.

5-Dimethylamino-8-ethyl-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (16). Method A gave **16** (484 mg, 49%) as a light-orange solid, a mixture of three isomers (maj:min:min = 8.4:1.0:0.2): mp 230–231°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.24 (bs, 1H, 1min-H), 7.65 (bs, 1H, 1maj-H), 6.15 (dd, J = 2.4 Hz, 0.9 Hz, 1H, 3maj-H), 5.75 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H), 5.71 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H), 3.68 (dd, J = 8.4, 1.8 Hz,

1H, 3bα-H), 3.22 (dd, J = 9.0 Hz, 5.4 Hz, 1H, 6aαmin-H), 3.21 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.17 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.99–3.04 (m, 1H, 10aαmaj-H), 2.93 (s, 6H, $N(CH_3)_2$), 2.59–2.70 (m, 1H, 6bαmaj-H), 2.48–2.56 (m, 1H, 6bmin-H), 2.30 (dd, J = 0.9, 0.9 Hz, 1H, 2-CH_3), 1.70–1.99 (m, 2H, cyclohex.), 1.00–1.60 (m, 7H, cyclohex., CH_2CH_3), 0.86 (t, J = 7.2 Hz, 3H, CH_2CH_3 maj), 0.76 (t, J = 7.2 Hz, 3H, CH_2CH_3 min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.5, 177.4, 176.6, 127.5, 127.4, 127.0, 126.8, 109.1, 105.6, 103.7, 44.1, 43.9, 39.0, 38.3, 37.0, 36.0, 34.4, 34.0, 32.8, 32.7, 32.6, 29.6, 29.3, 27.8, 27.5, 26.1, 24.3, 23.6, 22.6, 13.3, 12.2, 11.4; IR (thin film, cm^{-1}) 3455(bs), 2957(m), 1704(m), 2125(bw), 1770(w), 1704(s), 1651(bs), 1558(m), 1446(m), 1194(m), 1142(m); HRMS m/z ($M + Na^+$) calcd 352.1996, found 352.2002. Anal. Calcd for $C_{19}H_{27}N_3O_2$: C, 69.27; H, 8.26; N, 12.76. Found: C, 69.50; H, 8.09; N, 12.67.

5-Dimethylamino-8-isopropyl-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (17). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL) and then a diethyl ether wash (10 mL) gave **17** (690 mg, 42%) as light-orange crystals, a single isomer: mp 237–238°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 7.65 (bs, 1H, 1-H), 6.15 (dd, J = 2.4, 0.9 Hz, 1H, 3-H), 3.69 (dd, J = 8.4, 1.8 Hz, 1H, 3bα-H), 3.16 (dd, J = 8.4, 5.4 Hz, 1H, 6aα-H), 3.00–3.05 (m, 1H, 10aα-H), 2.93 (s, 6H, $N(CH_3)_2$), 2.57–2.72 (m, 1H, 6bα-H), 2.31 (s, 3H, 2-CH_3), 1.77–1.96 (m, 3H, cyclohex.), 1.52–1.63 (m, 1H, cyclohex.), 1.10–1.44(m, 4H, $CH(CH_3)_2$, cyclohex.), 0.88 (d, J = 6.6 Hz, 6H, $CH(CH_3)_2$), 0.87 (d, J = 6.6 Hz, 6H, $CH(CH_3)_2$); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.5, 176.6, 127.4, 127.1, 109.2, 105.6, 44.0, 43.8, 39.7, 37.0, 33.0, 32.7, 25.6, 25.0, 23.0, 21.3, 20.8, 13.3; IR (thin film, cm^{-1}) 3369 (bs), 2952(s), 2868(s), 2363(w), 1769(m), 1706(s), 1602(w), 1522(w), 1449(m), 1365(m), 1312(w), 1244(w), 1192(m), 1144(m), 1046(m); HRMS m/z ($M + Na^+$) calcd 366.2153, found 366.2160. Anal. Calcd for $C_{20}H_{29}N_3O_2$: C, 69.94; H, 8.51; N, 12.23. Found: C, 69.87; H, 8.41; N, 12.08.

8-tert-Butyl-5-dimethylamino-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (18). Method A gave **18** (558 mg, 52%) as an orange solid, a mixture of three isomers (maj:min:min = 2.4:1.0:0.1): mp 175–176°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 7.65 (bs, 1H, 1min-H), 7.61 (bs, 1H, 1maj-H), 6.12 (dd, J = 2.7, 1.2 Hz, 1H, 3min-H), 5.99 (dd, J = 2.6, 1.1 Hz, 1H, 3maj-H), 5.75 (dd, J = 2.7, 1.2 Hz, 1H, 3min-H), 3.77 (dd, J = 7.8, 1.5 Hz, 1H, 3bαmaj-H), 3.68 (dd, J = 8.3, 2.0 Hz, 1H, 3bαmin-H), 3.21 (dd, J = 8.6, 5.6 Hz, 1H, 6aαmin-H), 3.09 (dd, J = 7.8, 6.0 Hz, 1H, 6aαmaj-H), 3.00–3.05 (m, 1H, 10aβmin-H), 2.94 (s, 6H, $N(CH_3)_2$ min), 2.86 (s, 6H, $N(CH_3)_2$ maj), 2.45–2.75 (m, 2H, 6b-H, 10aαmaj-H), 2.29 (dd, J = 0.9, 0.9 Hz, 3H, 2-CH_3 min), 2.24 (d, J = 0.9 Hz, 3H, 2-CH_3 maj), 0.98–2.20 (m, 7H, cyclohex.), 0.90 (s, 9H, t-Bu), 0.70 (s, 9H, t-Bu); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.6, 177.2, 176.65, 176.61, 130.0, 127.6, 127.4, 126.9, 109.1, 108.7, 105.5, 104.6, 58.3, 47.8, 44.2, 44.1, 43.8, 43.3, 40.8, 39.3, 38.9, 37.1, 34.2, 34.0, 32.9, 32.42, 32.40, 30.2, 28.3, 27.6, 27.5, 25.0, 24.2, 22.2, 18.4, 13.3, 13.2; IR (thin film, cm^{-1}) 3411(bs), 2953(m), 2866(m), 2114(bw), 1774(w), 1711(s), 1646(bm), 1365(m), 1200(m), 1148(m); HRMS m/z ($M + Na^+$) calcd 380.2309, found 380.2335. Anal. Calcd for $C_{21}H_{31}N_3O_2$: C, 70.55; H, 8.74; N, 11.75. Found: C, 69.84; H, 8.82; N, 11.09.

5-Dimethylamino-2-methyl-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (19). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **19** (870 mg, 48%) as light-orange crystals, a single isomer: mp 220–222°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 7.67 (bs, 1H, 1-H), 7.18–7.34 (m, 5H, Ph), 6.14–6.16 (m, 1H, 3-H), 3.75 (dd, J = 8.4, 1.8 Hz, 1H, 3bα-H), 3.19 (dd, J = 8.3, 5.6 Hz, 1H, 6aα-H), 2.96–3.00 (m, 1H, 10aα-H), 2.96 (s, 6H, $N(CH_3)_2$), 2.72–2.80 (m, 1H, 6bα-H), 2.32 (s, 3H, 2-CH_3), 1.70–2.05 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.4, 176.5, 128.6, 127.6, 127.3, 127.1, 125.8, 109.4, 105.5, 44.1, 43.7, 32.9–33.5 (overlapped peaks), 13.3; IR (thin film, cm^{-1}) 3380(bs), 3085(w), 3058(w), 3026(w), 2933(s), 2867(m), 2800(w), 1772(w), 1709(s), 1601(w), 1495(w), 1448(m), 1361(m), 1243(w), 1195(m), 1150(w), 1106(w), 1028(m); HRMS m/z ($M + Na^+$) calcd for $C_{23}H_{27}N_3O_2$: 400.1996, found 400.2008.

2-Methyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (20). Method A gave **20** (602 mg, 60%) as a white solid, a mixture of two isomers (maj:min = 12.5:1.0): mp 268–269°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.30 (bs, 1H, 1min-H), 7.68 (bs, 1H, 1maj-H), 7.43–7.52 (m, 3H, Ph), 7.27–7.33 (m, 2H, Ph), 6.19 (dd, J = 2.6, 1.1 Hz, 1H, 3maj-H), 5.78 (dd, J = 3.0, 0.9 Hz, 1H, 3min-H), 3.96 (dd, J = 8.6, 2.0 Hz, 1H, 3bα-H), 3.47 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.40 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.12–3.18 (m, 1H, 10aαmaj-H), 3.01–3.07 (m, 1H, 10aβmin-H), 2.51–2.60 (m, 1H, 6bα-H), 2.32 (dd, 3H, J = 0.9, 0.9 Hz, 2-CH_3), 2.11–2.20 (m, 1H, cyclohex.), 1.18–1.83 (m, 7H, cyclohex.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.52 (bs, 1H, 1maj-H), 10.26 (bs, 1H, 1min-H), 7.35–7.54 (m, 3H, Ph), 7.19–7.26 (m, 2H, Ph), 5.84 (dd, J = 2.1 Hz, 0.6 Hz, 1H, 3maj-H), 5.60 (app. d, J = 2.4 Hz, 1H, 3min-H), 4.16 (app. d, J = 7.5 Hz, 1H, 3bαmin-H), 4.02 (dd, J = 8.7, 1.8 Hz, 1H, 3bαmaj-H), 3.39 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.34 (dd, J = 8.6, 5.3 Hz, 1H, 6aαmaj-H), 2.99–3.06 (m, 1H, 10aαmaj-H), 2.90–2.96 (m, 1H, 10aβ-H), 2.04–2.40 (m, 1H, 6bα-H), 2.18 (s, 3H, 2-CH_3), 1.50–1.64 (m, 2H, cyclohex.), 1.32–1.46 (m, 1H, cyclohex.), 0.98–1.28 (m, 5H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.1, 176.8, 154.8, 132.1, 129.3, 129.2, 128.8, 128.5, 127.4, 127.1, 126.5, 109.4, 105.9, 103.7, 46.0, 38.9, 38.7, 38.4, 37.8, 33.1, 32.9, 29.1, 28.1, 26.1, 25.6, 23.1, 22.7, 21.1, 20.6, 13.3; IR (thin film, cm^{-1}) 3392(bs), 2943(m), 2855(m), 2181 (bw), 1775 (w), 1697(s), 1645(bs), 1387(m), 1186 (m), 1162(m); HRMS m/z ($M + Na^+$) calcd 357.1574, found 357.1584. Anal. Calcd for $C_{21}H_{22}N_2O_2$: C, 75.42; H, 6.63; N, 8.38. Found: C, 75.53; H, 6.80; N, 8.38.

2,8-Dimethyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (21). Method B with **3d** (785 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **21** (800 mg, 48%) as a colorless solid, a mixture of three isomers (maj:min:min = 1.8:1.0:0.3): mp 270–272°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.52 (bs, 1H, 1min-H), 10.27 (bs, 1H, 1maj-H), 7.38–7.55 (m, 3H, Ph), 7.20–7.25 (m, 2H, Ph), 5.83 (d, J = 1.2 Hz, 1H, 3maj-H), 5.61 (d, J = 2.1 Hz, 1H, 3min-H), 5.59 (d, J = 2.4 Hz, 1H, 3min-H), 4.16 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.02 (dd, J = 8.6, 1.7 Hz, 1H, 3bαmin-H), 4.01 (dd, J = 9.9, 1.5 Hz, 1H, 3bαmaj-H), 3.40

(dd, J = 8.1, 4.8 Hz, 1H, 6aαmin-H), 3.36 (dd, J = 8.1, 5.4 Hz, 1H, 6aαmaj-H), 2.93–3.02 (m, 1H, 10aα-H), 2.85–2.92 (m, 1H, 10aβ-H), 2.48–2.58 (m, 1H, 6bαmaj-H), 2.30–2.42 (m, 1H, 6bmin-H), 2.18 (s, 3H, 2-CH_3), 1.74–2.16 (m, 2H, cyclohex.), 0.89–1.65 (m, 5H, cyclohex.), 0.95 (d, J = 7.2 Hz, 3H, 8-CH_3 maj), 0.73 (d, J = 6.3 Hz, 3H, 8-CH_3 min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.0, 176.8, 132.1, 129.3, 129.2, 128.7, 128.5, 127.4, 126.5, 117.0, 109.5, 105.8, 103.8, 45.7, 38.9, 37.8, 33.1, 33.0, 32.7, 32.6, 27.1, 26.7, 22.5, 22.4, 17.7, 13.3; IR (thin film, cm^{-1}) 3384(bs), 3063(m), 2950(s), 2866(s), 2361(m), 1778(m), 1712(s), 1598(m), 1501(m), 1457(m), 1384(m), 1182(m); HRMS m/z (M + Na^+) calcd 371.1731, found 371.1737. Anal. Calcd for $C_{22}H_{24}N_2O_2$: C, 75.83; H, 6.94; N, 8.04. Found: C, 75.70; H, 7.08; N, 7.88.

8-Ethyl-2-methyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (22). Method A gave **22** (402 mg, 37%) as a cream-colored solid, a mixture of three isomers (maj:min:min = 5.6:1.0:0.1): mp 257–258°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.30 (bs, 1H, 1min-H), 7.69 (bs, 1H, 1maj-H), 7.36–7.51 (m, Ph, 3H), 7.27–7.32 (m, Ph, 2H), 6.19 (dd, J = 2.4, 1.2 Hz, 1H, 3maj-H), 5.79 (dd, J = 2.9, 1.1, 1H, 3min-H), 5.76 (dd, J = 2.7, 1.2 Hz, 1H, 3min-H), 3.96 (dd, J = 8.4, 1.8 Hz, 1H, 3bα-H), 3.46 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.43 (dd, J = 8.4, 5.1 Hz, 1H, 6aαmin-H), 3.39 (dd, J = 8.4, 5.7 Hz, 1H, 6aαmaj-H), 3.06–3.12 (m, 1H, 10aαmaj-H), 2.96–2.02 (m, 1H, 10aβmin-H), 2.65–2.75 (m, 1H, 6bαmaj-H), 2.52–2.63 (m, 1H, 6bαmin-H), 2.32 (dd, J = 0.9, 0.9 Hz, 3H, 2-CH_3), 1.72–2.02 (m, 2H, cyclohex.), 1.26–1.64 (m, 5H, cyclohex.), 1.42 (app. q, J = 7.5 Hz, 2H, CH_2CH$_3$), 0.86 (t, J = 7.2 Hz, 3H, CH$_2$CH_3); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.52 (bs, 1H, 1min-H), 10.28 (bs, 1H, 1maj-H), 10.27 (bs, 1H, 1min-H), 7.39–7.54 (m, 3H, Ph), 7.20–7.25 (m, 2H, Ph), 5.83 (d, J = 1.5 Hz, 1H, 3maj-H), 5.61 (d, J = 2.4 Hz, 1H, 3min-H), 5.59 (d, J = 2.4 Hz, 1H, 3min-H), 4.15 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.02 (dd, J = 8.4, 2.1 Hz, 1H, 3bαmin-H), 4.01 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.42 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmin-H), 3.39 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.35 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmaj-H), 2.94–3.00 (m, 1H, 10aαmaj-H), 2.85–2.91 (m, 1H, 10aβmin-H), 2.41–2.52 (m, 1H, 6bαmaj-H), 2.27–2.39 (m, 1H, 6bmin-H), 2.18 (s, 3H, 2-CH_3), 1.70–2.18 (m, 2H, cyclohex.), 0.98–1.84 (m, 5H, cyclohex.), 1.38 (app. q, J = 7.5 Hz, 2H, CH_2CH$_3$), 0.80 (t, J = 7.2 Hz, 3H, CH$_2$CH_3 maj), 0.79 (t, J = 7.2 Hz, 3H, CH$_2$CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.2, 177.4, 176.3, 133.0, 132.9, 129.6, 129.5, 128.8, 128.7, 128.2, 127.4, 127.3, 126.5, 117.0, 114.8, 108.9, 105.4, 103.0, 46.5, 45.7, 45.5, 38.1, 34.2, 34.0, 32.6–33.2 (overlapped peaks), 23.9, 23.8, 23.7, 13.5, 13.4, 12.6; IR (thin film, cm^{-1}) 3420(bs), 2955(m), 2930(m), 2866(m), 2100 (bw), 1771 (w), 1695(s), 1644(bs), 1389(m), 1193(m), 1178(m), 1164(m); HRMS m/z (M + Na^+) calcd 385.1887, found 385.1881. Anal. Calcd for $C_{23}H_{26}N_2O_2$: C, 76.21; H, 7.23; N, 7.73. Found: C, 76.40; H, 7.38; N, 7.84.

8-Isopropyl-2-methyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (23). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **23** (700 mg, 39%) as a colorless solid, a mixture of two isomers (maj:min = 5.0:1.0): mp 278–281°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.53 (bs, 1H, 1min-H), 10.28 (bs, 1H, 1maj-H), 7.38–7.55 (m, 3H, Ph), 7.17–7.24 (m, 2H, Ph), 7.82 (app. d, J = 1.5 Hz, 1H, 3maj-H), 5.62 (dd, J = 2.4, 0.6 Hz, 1H, 3min-H), 4.15 (app. d, J = 7.2 Hz, 1H, 3bαmin-H), 4.01 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.39 (dd, J = 8.7, 5.4 Hz, 1H, 6aαmin-H), 3.35 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.94–3.01 (m, 1H, 10aαmaj-H), 2.86–2.92 (m, 1H, 10aβmin-H), 2.41–2.50 (m, 1H, 6bα-H), 2.18 (s, 3H, 2-CH_3), 1.64–2.12 (m, 2H, cyclohex.), 1.08–1.58 (m, 6H, cyclohex, CH(CH$_3$)$_2$), 0.86 (d, J = 6.3 Hz, 6H, CH(CH_3)$_2$ maj), 0.79 (d, J = 6.6 Hz, 6H, CH(CH$_3$)$_2$ min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 177.4, 144.7, 133.0, 129.6, 128.7, 127.3, 126.5, 108.9, 105.4, 45.5, 32.9–33.2 (overlapped peaks), 21.7, 21.0, 13.5; IR (thin film, cm^{-1}) 3467(m), 3393(bs), 3061(w), 2951(m), 2868(m), 1773(w), 1705(s), 1599(w), 1502(m), 1454(m), 1384(s), 1193(m), 1177(m), 1161(m); HRMS m/z (M + Na^+) calcd 399.2044, found 399.2047. Anal. Calcd for $C_{24}H_{28}N_2O_2$: C, 76.56; H, 7.50; N, 7.44. Found: C, 76.72; H, 7.63; N, 7.33.

8-tert-Butyl-2-methyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (24). Method A gave **24** (445 mg, 38%) as a light-orange solid, a mixture of three isomers (maj:min:min = 8.3:1.0:0.2): mp 221–222°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.08 (bs, 1H, 1min-H), 7.65 (bs, 1H, 1min-H), 7.61 (bs, 1H, 1maj-H), 7.21–7.56 (m, 5H, Ph), 6.17 (dd, J = 2.7, 1.2 Hz, 1H, 3min-H), 6.03 (dd, J = 2.6, 1.1 Hz, 1H, 3maj-H), 5.74 (dd, J = 2.7, 1.1 Hz, 1H, 3min-H), 4.04 (dd, J = 7.8, 1.5 Hz, 1H, 3bαmaj-H), 3.96 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.43 (dd, J = 8.6, 5.6 Hz, 1H, 6aαmin-H), 3.34 (dd, J = 7.7, 5.6 Hz, 1H, 6aαmaj-H), 3.10–3.15 (m, 1H, 10aβmin-H), 2.69–2.78 (m, 1H, 6bαmaj-H), 2.61–2.68 (m, 1H, 10aαmaj-H), 2.53–2.62 (m, 1H 6bmin-H), 2.26 (d, J = 0.9 Hz, 3H, 2-CH_3), 1.77–2.07 (m, 3H, cyclohex.), 1.62 (ddd, J = 13.9, 10.1, 7.1 Hz, 1H, cyclohex.), 0.83–1.43 (m, 3H, cyclohex.), 0.91 (s, 9H, t-Bu), 0.74 (s, 9H, t-Bu); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.37 (bs, 1H, 1min-H), 10.34 (bs, 1H, 1maj-H), 10.29 (bs, 1H, 1min-H), 7.36–7.50 (m, 3H, Ph), 7.10–7.13 (m, 2H, Ph), 5.67 (dd, J = 2.3, 0.8 Hz, 1H, 3maj-H), 5.55 (dd, J = 2.1, 0.6 Hz, 1H, 3min-H), 4.12 (app. d, J = 8.7 Hz, 1H, 3bαmin-H), 4.03 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmin-H), 3.90 (dd, J = 7.7, 1.4 Hz, 1H, 3bαmaj-H), 3.50 (dd, J = 8.4, 6.6 Hz, 1H, 6aαmin-H), 3.47 (dd, J = 7.7, 5.6 Hz, 1H, 6aαmaj-H), 3.38 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmin-H), 2.98–3.03 (m, 1H, 10aβmin-H), 2.56–2.65 (m, 1H, 10aαmaj-H), 2.42–2.53 (m, 1H, 6b-H), 0.90–2.20 (m, 7H, cyclohex.), 2.12 (s, 3H, 2-CH_3), 0.86 (s, 9H, t-Bu maj), 0.68 (s, 9H, t-Bu min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.9, 177.1, 176.3, 173.8, 146.2, 134.3, 132.3, 130.4, 129.7, 129.4, 129.3, 129.29, 129.26, 129.20, 129.1, 128.6, 128.5, 127.0, 126.8, 126.5, 126.4, 126.2, 119.7, 114.0, 109.7, 104.7, 53.1, 47.8, 46.2, 45.7, 43.9, 43.7, 41.7, 41.2, 40.7, 39.2, 39.0, 38.3, 34.3, 34.2, 32.9, 32.6, 32.5, 32.4, 31.4, 30.5, 28.7, 28.5, 27.7, 27.5, 27.4, 25.5, 24.8, 24.3, 22.2, 13.2; IR (thin film, cm^{-1}) 3390(bs), 2951(m), 2866(w), 2357 (w), 2088(bw), 1772(w), 1708(s), 1647(bs), 1500(m), 1386(m), 1372(m), 1199(m), 1176(m); HRMS m/z (M + Na^+) calcd 413.2200, found 413.2181. Anal. Calcd for $C_{25}H_{30}N_2O_2$: C, 76.89; H, 7.74; N, 7.17. Found: C, 76.65; H, 7.43; N, 7.39.

2-Methyl-5,8-diphenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (25). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **25** (850 mg, 43%) as a colorless solid, a mixture of two isomers (maj:min = 3.8:1.0): mp 282–285°C; ^{1}H NMR (300 MHz,

DMSO-d_6, δ) 10.56 (bs, 1H, 1min-H), 10.38 (bs, 1H, 1maj-H), 7.15–7.56 (m, 10H, Ph), 5.78–5.87 (m, 1H, 3maj-H), 5.68–5.67 (m, 1H, 3min-H), 4.19 (d, J = 8.1 Hz, 1H, 3bαmin-H), 4.02 (d, J = 7.5 Hz, 1H, 3bαmaj-H), 3.36–3.54 (m, 1H, 6aα-H), 2.82–2.98 (m, 1H, 10a-H), 2.48–2.60 (m, 1H, 6bα-H), 2.19 (s, 3H, 2-CH_3), 1.34–2.10 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.0, 176.8, 129.3, 128.6, 127.7, 127.3, 126.6, 125.8, 109.8, 105.6, 105.5, 105.4, 45.6, 33.2–33.6 (overlapped peaks), 13.3; IR (thin film, cm^{-1}) 3462(m), 3431(m), 3394(bs), 3060(w), 3024(w), 2934(s), 2868(m), 1776(w), 1706(s), 1599(w), 1499(m), 1383(m), 1189(m), 1168(m); HRMS m/z (M + Na^+) calcd 433.1887, found 433.1908. Anal. Calcd for $C_{27}H_{26}N_2O_2$: C, 79.00; H, 6.38; N, 6.82. Found: C, 78.88; H, 6.58; N, 6.68.

2-Methyl-5-(4-methylphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (26). Method B with **3c** (687 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **26** (700 mg, 42%) as a colorless solid, a mixture of two isomers (maj:min = 1.6:1.0): mp 276–278°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.51 (bs, 1H, 1maj-H), 10.25 (bs, 1H, 1min-H), 7.30 (d, J = 8.1 Hz, 2H, Ph maj), 7.28 (d, J = 7.8 Hz, 2H, Ph min), 7.12 (d, J = 8.1 Hz, 2H, Ph maj), 7.09 (d, J = 8.4 Hz, 2H, Ph min), 5.83 (dd, J = 2.4, 1.2 Hz, 1H, 3min-H), 5.59 (dd, J = 2.4, 0.6 Hz, 1H, 3maj-H), 4.14 (app. d, J = 7.8 Hz, 1H, 3bαmaj-H), 3.99 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.37 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.32 (dd, J = 8.6, 5.3 Hz, 1H, 6aαmin-H), 2.99–3.04 (m, 1H, 10aαmin-H), 2.92 (m, 1H, 10aβmaj-H), 2.06–2.40 (m, 2H, cyclohex., 6bα-H), 2.35 (s, 3H, 4′-CH_3 maj), 2.34 (s, 3H, 4′-CH_3 min), 2.18 (s, 3H, 2-CH_3), 0.98–1.64 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 178.3, 177.6, 176.4, 138.4, 138.3, 130.4, 130.2, 130.1, 130.0, 128.2, 127.2, 126.9, 126.5, 119.0, 116.9, 108.8, 105.5, 102.8, 46.2, 45.9, 38.7, 38.5, 38.4, 38.2, 33.1, 33.0, 29.3, 27.6, 26.1, 25.7, 23.2, 22.9, 21.4, 21.3, 20.8, 13.52, 13.45; IR (thin film, cm^{-1}) 3400(bs), 2927(m), 2857(m), 1776(w), 1702(s), 1516(m), 1387(m), 1182(m), 1161(m); HRMS m/z (M + Na^+) calcd 371.1731, found 371.1743. Anal. Calcd for $C_{22}H_{24}N_2O_2$: C, 75.83; H, 6.94; N, 8.04. Found: C, 75.98; H, 6.92; N, 7.90.

2,8-Dimethyl-2-(4-methylphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (27). Method B with **3d** (785 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **27** (650 mg, 37%) as a colorless solid, a mixture of three isomers (maj:min:min = 1.1:1.0:0.1): mp 255–257°C; ^{1}H NMR (200 MHz, DMSO-d_6, δ) 10.50 (bs, 1H, 1min-H), 10.25 (bs, 1H, 1maj-H), 7.29 (d, J = 8.4 Hz, 2H, Ph min), 7.27 (d, J = 8.2 Hz, 2H, Ph maj), 7.09 (d, J = 8.2 Hz, 1H, Ph min), 7.07 (d, J = 8.2 Hz, 1H, Ph maj), 5.80 (app. d, J = 1.8 Hz, 1H, 3maj-H), 5.58 (dd, J = 2.2, 0.8 Hz, 1H, 3min-H), 5.56–5.58 (m, overlapped, 1H, 3min-H), 4.12 (app. d, J = 7.8 Hz, 1H, 3bαmin-H), 3.97 (dd, J = 8.3, 1.7 Hz, 1H, 3bαmaj-H), 3.38 (dd, J = 7.8, 5.0 Hz, 1H, 6aαmin-H), 3.37 (dd, J = 8.6, 5.4 Hz, 1H, 6aαmin-H), 3.32 (dd, J = 8.6, 5.4 Hz, 1H, 6aαmaj-H), 2.91–2.98 (m, 1H, 10amaj-H), 2.83–2.89 (m, 1H, 10amin-H), 1.64–2.60 (m, 3H, cyclohex., 6bα-H), 2.32 (s, 3H, 4′-CH_3), 2.16 (s, 3H, 2-CH_3), 0.98 (m, 5H, cyclohex.), 0.94 (d, J = 7.0 Hz, 3H, 8-CH_3 maj), 0.70 (d, J = 6.2 Hz, 3H, 8-CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.2, 177.5, 176.4, 138.2, 130.4, 130.1, 130.0, 127.2, 126.5, 109.0, 105.4, 45.6, 38.8, 38.0, 33.1, 32.3–32.7 (overlapped peaks), 26.8, 26.7, 21.3, 13.4; IR (thin film, cm^{-1}) 3460(m), 3396(bs), 3075(w), 3040(w), 2927(s), 2892(m), 2867(m), 2362(w), 2336(w), 1776(m), 1708(s), 1516(s), 1387(s), 1180(s); HRMS m/z (M + Na^+) calcd 385.1887, found 385.1900. Anal. Calcd for $C_{23}H_{26}N_2O_2$: C, 76.21; H, 7.23; N, 7.73. Found: C, 76.01; H, 7.03; N, 7.58.

8-Ethyl-2-methyl-5-(4-methylphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (28). Method B with **3e** (883 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **28** (680 mg, 38%) as a colorless solid, a mixture of two isomers (maj:min = 2.1:1.0): mp 269–271°C; ^{1}H NMR (200 MHz, DMSO-d_6, δ) 10.50 (bs, 1H, 1min-H), 10.25 (bs, 1H, 1maj-H), 7.29 (d, J = 8.0 Hz, 2H, Ph min), 7.27 (d, J = 8.2 Hz, 2H, Ph maj), 7.08 (d, J = 8.2 Hz, 2H, Ph min), 7.06 (d, J = 8.4 Hz, 2H, Ph maj), 5.80 (dd, J = 2.0, 0.8 Hz, 1H, 3maj-H), 5.59 (app. d, J = 2.2 Hz, 1H, 3min-H), 4.11 (app. d, J = 8.2 Hz, 1H, 3bαmin-H), 3.96 (dd, J = 8.6, 1.4 Hz, 1H, 3bαmaj-H), 3.35 (dd, J = 8.7, 5.3 Hz, 1H, 6aαmin-H), 3.31 (dd, J = 8.5, 5.3 Hz, 1H, 6aαmaj-H), 2.92–2.99 (m, 1H, 10aαmaj-H), 2.83–2.88 (m,1H, 10aβmin-H), 1.64–2.50 (m, 2H, cyclohex., 6bα-H), 2.38 (s, 3H, 4′CH_3), 2.16 (s, 3H, 2-CH_3), 1.00–1.88 (m, 6H, cyclohex.), 1.35 (app. q, J = 7.4 Hz, 2H, CH_2CH_3), 0.78 (t, J = 7.2 Hz, 3H, CH_2CH_3 maj), 0.77 (t, J = 7.2 Hz, 1H, CH_2CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.3, 177.5, 176.4, 138.4, 138.2, 130.4, 130.4, 130.1, 130.0, 128.2, 127.2, 126.8, 126.5, 117.0, 108.9, 105.4, 104.5, 103.0, 45.6, 33.8–34.2 (overlapped peaks), 33.1, 32.6–32.8 (overlapped peaks), 23.8, 23.7, 21.3, 13.5, 12.6; IR (thin film, cm^{-1}) 3468(m), 3394(bs), 3038(w), 2958(m), 2932(s), 2867(m), 1776(m), 1706(s), 1516(s), 1386(s), 1181(m), 1165(m); HRMS m/z (M + Na^+) calcd 399.2044, found 399.2051. Anal. Calcd for $C_{24}H_{28}N_2O_2$: C, 76.56; H, 7.50; N, 7.44. Found: C, 76.70; H, 7.49; N, 7.43.

8-Isopropyl-2-methyl-5-(4-methylphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (29). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **29** (760 mg, 41%) as a colorless solid, a mixture of two isomers (maj:min = 3.2:1.0): mp 296–298°C; ^{1}H NMR (200 MHz, DMSO-d_6, δ) 10.50 (bs, 1H, 1min-H), 10.26 (bs, 1H, 1maj-H), 7.30 (d, J = 8.2 Hz, 2H, Ph min), 7.28 (d, J = 8.0 Hz, 2H, Ph maj), 7.07 (d, J = 8.2 Hz, 2H, Ph min), 7.05 (d, J = 8.2 Hz, 2H, Ph maj), 5.80 (app. d, J = 1.2 Hz, 1H, 3maj-H), 5.59 (dd, J = 2.8, 0.6 Hz, 1H, 3min-H), 4.11 (app. d, J = 9.0 Hz, 1H, 3bαmin-H), 3.96 (dd, J = 8.3, 1.7 Hz, 1H, 3bαmaj-H), 3.35 (dd, J = 8.0, 5.2 Hz, 1H, 6aαmin-H), 3.31 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.91–2.99 (m, 1H, 10aαmaj-H), 2.84–2.89 (m, 1H, 10aβmin-H), 2.29–2.49 (m, 1H, 6bα-H), 2.32 (s, 3H, 4′-CH_3), 0.73–2.20 (m, 8H, cyclohex., $CH(CH_3)_2$), 2.16 (s, 3H, 2-CH_3), 0.84 (d, J = 6.6 Hz, 6H, $CH(CH_3)_2$), 0.77 (d, J = 6.4 Hz, 6H, $CH(CH_3)_2$); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.6, 177.5, 138.2, 130.5, 130.0, 128.4, 127.1, 126.5, 108.9, 105.4, 45.5, 32.7–33.2 (overlapped peaks), 21.7, 21.3, 21.0, 13.5; IR (thin film, cm^{-1}) 3393(bs), 2948(m), 2925(m), 2867(m), 1773(w), 1696(s), 1516(m), 1387(m), 1192(m), 1180(m), 1162(m); HRMS m/z (M + Na^+) calcd 413.2200, found 413.2201. Anal. Calcd for $C_{25}H_{30}N_2O_2$: C, 76.89; H, 7.74; N, 7.17. Found: C, 76.58; H, 7.82; N, 6.93.

8-tert-Butyl-2-methyl-5-(4-methylphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (30). Method B with **3g** (1079 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **30** (530 mg, 27%) as a pink solid, a mixture of three isomers (maj:min:min = 12.4:1.0:0.6): mp 220–222°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.32 (bs, 1H, 1maj-H), 10.28 (bs, 1H, 1min-H), 10.22 (bs, 1H, 1min-H), 7.25 (d, J = 7.8 Hz, 2H, Ph), 6.99 (d, J = 7.8 Hz, 2H, Ph), 5.79–5.82 (m, 1H, 3min-H), 5.75–5.77 (m, 1H, 3min-H), 5.65–5.68 (m, 1H, 3maj-H), 4.06 (d, J = 6.9 Hz, 1H, 3bαmin-H), 3.89 (d, J = 7.5 Hz, 1H, 3bαmaj-H), 3.44 (dd, J = 7.4, 5.9 Hz, 1H, 6aαmaj-H), 1.46–2.68 (m, 7H, cyclohex., 10a-H, 6b-H), 2.32 (s, 3H, 4′-CH_3), 2.12 (s, 3H, 2-CH_3), 0.90–1.20 (m, 2H, cyclohex.), 0.85 (s, 9H, t-Bu maj), 0.68 (s, 9H, t-Bu min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.0, 177.6, 141.5, 137.7, 133.3, 133.1, 131.3, 129.8, 129.7, 126.9, 126.5, 114.2, 109.9, 104.9, 51.9, 47.0, 45.6, 44.2, 41.6, 40.6, 34.2, 32.8, 32.5, 31.9, 27.7, 27.5, 26.8, 24.7, 23.9, 23.8, 23.5, 23.4, 21.3, 18.6, 13.2; IR (thin film, cm^{-1}) 3390(bs), 3038(w), 2953(s), 2869(m), 2360(w), 2340(w), 1767(m), 1708(s), 1516(m), 1384(s), 1367(m), 1175(m), 1169(m); HRMS m/z (M + Na$^+$) calcd for $C_{26}H_{32}N_2O_2$: 427.2357, found 427.2356.

2-Methyl-5-(4-methylphenyl)-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (31). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **31** (830 mg, 41%) as a colorless solid, a mixture of two isomers (maj:min = 11.9:1.0): mp 298–300°C; ^{1}H NMR (200 MHz, DMSO-d_6, δ) 10.53 (bs, 1H, 1min-H), 10.35 (bs, 1H, 1maj-H), 6.96–7.30 (m, 9H, Ph), 5.78–5.83 (m, 1H, 3maj-H), 5.63 (dd, J = 2.1, 1.5 Hz, 1H, 3min-H), 4.15 (d, J = 9.2 Hz, 1H, 3bαmin-H), 3.98 (d, J = 7.4 Hz, 1H, 3bαmaj-H), 3.36–3.45 (m, 1H, 6aα-H), 2.80–2.96 (m, 1H, 10a-H), 1.70–2.60 (m, 8H, cyclohex., 6bα-H), 2.33 (s, 3H, 4′-CH_3), 2.17 (s, 3H, 2-CH_3); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.7, 177.5, 138.3, 130.5, 130.0, 128.9, 127.6, 127.3, 126.8, 126.1, 109.2, 45.4, 33.1–33.7 (overlapped peaks), 21.3, 13.5; IR (thin film, cm^{-1}) 3431(bs), 3025(w), 2941(m), 2872(m), 1772(m), 1688(m), 1516(m), 1452(m), 1379(m), 1192(m); HRMS m/z (M + Na$^+$) calcd 447.2044, found 447.2065. Anal. Calcd for $C_{28}H_{28}N_2O_2$: C, 79.22; H, 6.65; N, 6.60. Found: C, 78.98; H, 6.70; N, 6.49.

5-(4-Methoxyphenyl)-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (32). Method A gave **32** (372 mg, 34%) as a cream-colored solid, a mixture of two isomers (maj:min = 3.5:1.0): mp 239–240°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.31 (bs, 1H, 1min-H), 7.70 (bs, 1H, 1maj-H), 7.21 (d, J = 9.0 Hz, 2H, Ph), 6.98 (d, J = 9.0 Hz, 2H, Ph), 6.19 (dd, J = 2.4, 0.9 Hz, 1H, 3maj-H), 5.77 (dd, J = 3.3, 1.2 Hz, 1H, 3min-H), 3.94 (dd, J = 8.6, 2.0 Hz, 1H, 3bα-H), 3.83 (s, 3H, OCH_3), 3.45 (dd, J = 8.6, 5.6 Hz, 1H, 6aαmin-H), 3.38 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.11–3.17 (m, 1H, 10aαmaj-H), 3.00–3.07 (m, 1H, 10aβmin-H), 2.50–2.58 (m, 1H, 6b-H), 2.31–2.32 (dd, J = 0.9, 0.9 Hz, 3H, 2-CH_3), 2.10–2.19 (m, 1H, cyclohex.) 1.17–1.80 (m, 7H, cyclohex.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.50 (d, J = 1.5 Hz, 1H, 1min-H), 10.24 (app. bs, 1H, 1maj-H), 7.13 (d, J = 9.0 Hz, 2H, Ph), 7.02 (d, J = 9.0 Hz, 2H, Ph), 5.83 (d, J = 0.9 Hz, 1H, 3maj-H), 5.59 (d, J = 1.5 Hz, 1H, 3min-H), 4.12 (d, J = 8.1 Hz, 1H, 3bαmin-H), 3.98 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.78 (s, 3H, OCH_3), 3.36 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.31 (dd, J = 8.4, 5.1 Hz, 1H, 6aαmaj-H), 2.98–3.05 (m, 1H, 10aαmaj-H), 2.89–2.95 (m, 1H, 10aβmin-H), 2.04–2.40 (m, 2H, cyclohex., 6bα-H), 2.18 (s, 3H, 2-CH_3), 0.99–1.64 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.4, 178.3, 177.1, 176.5, 159.4, 127.7, 127.4, 127.1, 114.6, 114.5, 109.4, 103.7, 55.6, 46.0, 38.8, 38.6, 38.4, 37.8, 33.0, 32.9, 29.1, 28.1, 26.1, 25.6, 23.0, 22.7, 21.1, 20.6, 13.3; IR (thin film, cm^{-1}) 3447(bs), 2935(m), 2858(m), 2150(bw), 1772(w), 1697(s), 1651(bs), 1518(m), 1392(m), 1252(m), 1183(m), 1162(m); HRMS m/z (M + Na$^+$) calcd 387.1680, found 387.1701. Anal. Calcd for $C_{22}H_{24}N_2O_3$: C, 72.50; H, 6.64; N, 7.69. Found: C, 72.61; H, 6.84; N, 7.64.

5-(4-Methoxyphenyl)-2,8-dimethyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (33). Method B with **3d** (785 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **33** (1100 mg, 61%) as a colorless solid, a mixture of three isomers (maj:min:min = 2.3:1.0:0.3): mp 265–268°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.50 (bs, 1H, 1min-H), 10.25 (bs, 1H, 1maj-H), 10.23 (bs, 1H, 1min-H), 7.14 (d, J = 9.0 Hz, 2H, Ph min), 7.12 (d, J = 9.0 Hz, 2H, Ph maj), 7.04 (d, J = 8.4 Hz, 2H, Ph min), 7.02 (d, J = 8.7 Hz, 2H, Ph maj), 5.81 (dd, J = 1.5, 0.6 Hz, 1H, 3maj-H), 5.60 (app. d, J = 2.4 Hz, 1H, 3min-H), 5.58 (app. d, J = 2.4 Hz, 1H, 3min-H), 4.12 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 3.98 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmin-H), 3.97 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.79 (s, 3H, OCH_3 min), 3.78 (s, 3H, OCH_3 maj), 3.37 (dd, J = 8.3, 5.0 Hz, 1H, 6aαmin-H), 3.34 (dd, J = 8.3, 5.6 Hz, 1H, 6aαmin-H), 3.33 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.93–3.00 (m, 1H, 10aαmaj-H), 2.85–2.90 (m, 1H, 10aβmin-H), 1.74–2.56 (m, 4H, cyclohex., 6b-H), 2.17 (s, 3H, 2-CH_3), 0.90–1.66 (m, 4H, cyclohex.), 0.95 (d, J = 6.9 Hz, 3H, 8-CH_3 maj), 0.72 (d, J = 6.6 Hz, 3H, 8-CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 178.0, 159.3, 128.6, 126.5, 125.7, 114.7, 109.0, 108.9, 105.0, 104.9, 55.9, 45.5, 33.1, 32.2–32.7 (overlapped peaks), 13.4; IR (thin film, cm^{-1}) 3462(m), 3390(bs), 3068(w), 3012(w), 2957(m), 2924(m), 2863(m), 1776(m), 1705(s), 1516(s), 1391(m), 1303(m), 1253(m), 1177(s); HRMS m/z (M + Na$^+$) calcd 401.1836, found 401.1841. Anal. Calcd for $C_{23}H_{26}N_2O_3$: C, 72.99; H, 6.92; N, 7.40. Found: C, 72.74; H, 6.66; N, 7.38.

8-Ethyl-5-(4-methoxyphenyl)-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (34). Method A gave **34** (424 mg, 36%) as a white solid, a mixture of three isomers (maj:min:min = 4.3:1.0:0.1): mp 249–251°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.30 (bs, 1H, 1min-H), 7.71 (bs, 1H, 1maj-H), 7.16–7.24 (m, 2H, Ph), 6.96–7.02 (m, 2H, Ph), 6.18 (dd, J = 2.4, 0.9 Hz, 1H, 3maj-H), 5.79 (d, J = 2.4 Hz, 1H, 3min-H), 5.75 (d, J = 3.3 Hz, 1H, 3min-H), 3.94 (dd, J = 8.4, 1.8 Hz, 1H, 3bα-H), 3.83 (s, 3H, OCH_3), 3.44 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.41 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.37 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.05–3.11 (m, 1H, 10aαmaj-H), 2.95–3.01 (m, 1H, 10aβmin-H), 2.64–2.74 (m, 1H, 6bαmaj-H), 2.53–2.62 (m, 1H, 6bβmin-H), 2.30–2.32 (m, 3H, 2-CH_3), 1.81–1.97 (m, 2H, cyclohex.), 1.10–1.60 (m, 7H, cyclohex., CH_2CH$_3$), 0.86 (t, J = 7.4 Hz, 3H, CH$_2$CH_3); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.51 (bs, 1H, 1min-H), 10.26 (bs, 1H, 1maj-H), 10.23 (bs, 1H, 1min-H), 7.02–7.16 (m, 4H, Ph), 5.82 (dd, J = 2.4, 1.5 Hz, 1H, 3maj-H), 5.60 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H),

5.58 (dd, J = 2.7, 1.2 Hz, 1H, 3min-H), 4.11 (app. d, J = 8.7 Hz, 1H, 3bαmin-H), 3.98 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.97 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.791 (s, 3H, OCH_3 min), 3.787 (s, 3H, OCH_3 min), 3.78 (s, 3H, OCH_3 maj), 3.36 (dd, J = 8.9, 5.6 Hz, 1H, 6aαmin-H), 3.34 (dd, J = 8.1, 5.4 Hz, 1H, 6aαmin-H), 3.32 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.98–3.02 (m, 1H, 10amin-H), 2.93–2.99 (m, 1H, 10aαmaj-H), 2.85–2.90 (m, 1H, 10aβmin-H), 2.37–2.50 (m, 1H, 6bαmaj-H), 2.26–2.38 (m, 1H, 6bmin-H), 2.17 (s, 3H, 2-CH3), 1.74–2.17 (m, 1H, cyclohex.), 1.68–1.84 (m, 1H, cyclohex.), 0.98–1.66 (m, 5H, cyclohex.), 1.40 (app. q, J = 7.5 Hz, 2H, CH_2CH_3), 0.80 (t, J = 7.2 Hz, 3H, CH_2CH_3 maj), 0.78 (t, J = 7.8 Hz, 3H, CH_2CH_3 min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.3, 178.2, 177.1, 159.4, 127.7, 127.4, 126.8, 124.8, 114.6, 114.5, 109.6, 105.8, 103.8, 55.5, 45.8, 45.6, 39.1, 38.8, 38.4, 34.5, 34.0, 32.7–33.0 (overlapped peaks), 29.7, 27.9, 27.3, 26.5, 24.4, 23.7, 22.9, 13.3, 12.3; IR (thin film, cm^{-1}) 3393(bs), 2916(m), 2862(m), 2400(w), 2150(bw), 1774(w), 1694(s), 1644(bs), 1518(m), 1388(m), 1256(m), 1178(m), 1160(m); HRMS m/z (M + Na^+) calcd 415.1993, found 415.1986. Anal. Calcd for $C_{24}H_{28}N_2O_3$: C, 73.44; H, 7.19; N, 7.14. Found: C, 73.31; H, 7.06; N, 7.03.

8-Isopropyl-5-(4-methoxyphenyl)-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (35). Method B with **3f** (981 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **35** (1450 mg, 74%) as a colorless solid, a mixture of four isomers (maj:min:min:min = 2.9:1.0:0.3:0.3): mp 300–303°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.51 (d, J = 2.1 Hz, 1H, 1min-H), 10.47–10.50 (m, overlapped, 1H, 1min-H), 10.27 (d, J = 2.1 Hz, 1H, 1maj-H), 10.22 (d, J = 2.7 Hz, 1H, 1min-H), 7.10 (d, J = 9.0 Hz, 2H, Ph), 7.04 (d, J = 9.3 Hz, 2H, Ph), 5.82 (dd, J = 2.4, 0.6 Hz, 1H, 3maj-H), 5.79–5.81 (m, overlapped, 1H, 3min-H), 5.61 (dd, J = 2.1, 0.6 Hz, 1H, 3min-H), 5.58 (dd, J = 2.7, 1.5 Hz, 1H, 3min-H), 4.13 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.11 (dd, J = 9.6, 1.2 Hz, 1H, 3bαmin-H), 3.99 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.96 (dd, J = 9.6, 1.8 Hz, 1H, 3bαmaj-H), 3.78 (s, 3H, OCH_3), 3.40 (dd, J = 8.7, 5.3 Hz, 1H, 6aαmin-H), 3.36 (dd, J = 8.7, 5.4 Hz, 1H, 6aαmin-H), 3.35 (dd, J = 8.4, 5.1 Hz, 1H, 6aαmin-H), 3.32 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.93–2.99 (m, 1H, 10aαmaj-H), 2.86–2.91 (m, 1H, 10aβmin-H), 2.25–2.50 (m, 1H, 6b-H), 2.17 (s, 3H, 2-CH_3), 0.94–2.17 (m, 8H, cyclohex., $CH(CH_3)_2$), 0.854 (d, J = 6.3 Hz, 6H, $CH(CH_3)_2$ maj), 0.845 (d, J = 6.6 Hz, 6H, $CH(CH_3)_2$ min), 0.79 (d, J = 6.3 Hz, 6H, $CH(CH_3)_2$ min), 0.75 (d, J = 6.6 Hz, 6H, $CH(CH_3)_2$ min), 0.70 (d, J = 6.6 Hz, 6H, $CH(CH_3)_2$ min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.8, 177.6, 159.4, 128.9, 128.7, 127.6, 126.8, 126.7, 126.0, 125.6, 114.9, 114.8, 109.2, 104.5, 45.4, 33.1–33.9 (overlapped peaks), 13.5; IR (thin film, cm^{-1}) 3397(bs), 3063(w), 2948(s), 2867(s), 1774(m), 1706(s), 1516(s), 1454(m), 1389(s), 1304(m), 1252(s), 1175(s); HRMS m/z (M + Na^+) calcd 429.2149, found 429.2138. Anal. Calcd for $C_{25}H_{30}N_2O_3$: C, 73.86; H, 7.44; N, 6.89. Found: C, 74.01; H, 7.61; N, 6.98.

8-tert-Butyl-5-(4-methoxyphenyl)-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (36). Method A gave **36** (442 mg, 35%) as a light-orange solid, a mixture of three isomers (maj:min:min = 4.4:1.0:0.3): mp 239–240°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.07 (bs, 1H, 1min-H), 7.62 (bs, 1H, 1min-H), 7.58 (bs, 1H, 1maj-H), 7.12–7.24 (m, 2H, Ph), 6.92–7.02 (m, 2H, Ph), 6.03 (dd, J = 2.6, 1.1 Hz, 1H, 3maj-H), 5.75 (dd, J = 2.6, 1.1 Hz, 1H, 3min-H), 5.73 (dd, J = 1.2, 2.7 Hz, 1H, 3min-H), 4.02 (dd, J = 7.8, 1.5 Hz, 1H, 3bαmaj-H), 3.94 (dd, J = 7.8, 1.5 Hz, 1H, 3bαmin-H), 3.84 (s, 3H, OCH_3 min), 3.81 (s, 3H, OCH_3 maj), 3.41 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.40 (dd, J = 7.8, 5.4 Hz, 1H, 6aαmin-H), 3.32 (dd, J = 7.7, 5.6 Hz, 1H, 6aαmaj-H), 3.08–3.12 (m, 1H, 10aβmin-H), 2.70–2.77 (m, 1H, 6bαmaj-H), 2.59–2.67 (m, 1H, 10aαmaj-H), 2.53–2.60 (m, 1H, 6bαmin-H), 2.26 (d, J = 0.9 Hz, 3H, 2-CH_3), 1.77–2.06 (m, 3H, cyclohex.), 1.55–1.66 (m, 1H, cyclohex.), 0.83–1.42 (m, 3H, cyclohex.), 0.90 (s, 9H, *t*-Bu maj), 0.74 (s, 9H, *t*-Bu min); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.36 (d, J = 2.1 Hz, 1H, 1min-H), 10.32 (d, J = 2.4 Hz, 1H, 1maj-H), 10.21 (d, J = 1.8 Hz, 1H, 1min-H), 6.97–7.10 (m, 4H, Ph), 5.67 (dd, J = 2.4, 0.9 Hz, 1H, 3maj-H), 5.58 (app. d, J = 2.1 Hz, 1H, 3min-H), 5.54 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H), 4.08 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 3.99 (dd, J = 7.8, 2.1 Hz, 1H, 3bαmin-H), 3.88 (dd, J = 7.7, 1.4 Hz, 1H, 3bαmaj-H), 3.79 (s, 3H, OCH_3 min), 3.78 (s, 3H, OCH_3 min), 3.77 (s, 3H, OCH_3 maj), 3.46 (dd, J = 8.3, 6.5 Hz, 1H, 6aαmin-H), 3.43 (dd, J = 7.5, 5.7 Hz, 1H, 6aαmaj-H), 3.38 (dd, J = 7.5, 5.1 Hz, 1H, 6aαmaj-H), 2.88–2.91 (m, 1H, 10amin-H), 2.30–2.65 (m, 3H, 6b-H, 10aαmaj-H, 10amin-H), 0.78–2.20 (m, 7H, cyclohex.), 2.18 (s, 3H, 2-CH_3 min), 2.13 (s, 3H, 2-CH_3 min), 2.12 (s, 3H, 2-CH_3 maj), 0.85 (s, 9H, *t*-Bu maj), 0.84 (s, 9H, *t*-Bu min), 0.68 (s, 9H, *t*-Bu min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.8, 178.5, 178.2, 177.3, 159.4, 130.3, 128.0, 127.68, 127.67, 127.2, 127.0, 125.0, 124.9, 114.6, 114.5, 109.7, 109.3, 105.9, 104.7, 55.6, 47.9, 46.1, 45.6, 45.3, 41.6, 40.7, 39.1, 38.9, 34.6, 34.3, 34.2, 32.9, 32.6, 32.5, 30.5, 28.6, 28.4, 27.6, 25.5, 24.3, 22.2, 13.2; ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.7, 177.5, 176.3, 159.4, 159.3, 130.2, 128.7, 128.4, 126.9, 125.7, 125.6, 117.0, 114.8, 114.6, 109.4, 104.0, 55.9, 45.2, 44.8, 41.6, 34.2–34.6 (overlapped peaks), 33.9, 33.2, 33.0, 30.7, 28.2, 28.0, 25.7, 13.4; IR (thin film, cm^{-1}) 3386(bs), 2952(m), 2865(m), 2050(bw), 1774(w), 1702(s), 1654(bs), 1513(s), 1390(m), 1251(s), 1168(m); HRMS m/z (M + Na^+) calcd 443.2306, found 443.2292. Anal. Calcd for $C_{26}H_{32}N_2O_3$: C, 74.26; H, 7.67; N, 6.66. Found: C, 74.39; H, 7.82; N, 6.49.

5-(4-Methoxyphenyl)-2-methyl-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (37). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **37** (1200 mg, 57%) as a colorless solid, a mixture of three isomers (maj:min:min = 4.7:1.0:0.8): mp 306–309°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.54 (app. bs, 1H, 1min-H), 10.36 (d, J = 0.9 Hz, 1H, 1maj-H), 10.33 (app. bs, 1H, 1min-H), 6.98–7.37 (m, 9H, Ph), 5.84–5.87 (m, 1H, 3min-H), 5.77–5.85 (m, 1H, 3maj-H), 5.63–5.66 (m, 1H, 3min-H), 4.15 (d, J = 8.1 Hz, 1H, 3bαmin-H), 4.03 (dd, J = 8.1, 2.4 Hz, 1H, 3bαmin-H), 3.98 (d, J = 8.1 Hz, 1H, 3bαmaj-H), 3.79 (s, 3H, OCH_3 maj), 3.74 (s, 3H, OCH_3 min), 3.48 (dd, J = 8.1, 5.4 Hz, 1H, 6aαmin-H), 3.43 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.07–3.13 (m, 1H, 10amin-H), 2.88–2.96 (m, 1H, 10amaj-H), 2.82–2.88 (m, 1H, 10amin-H), 1.55–2.60 (m, 8H, cyclohex., 6b-H), 2.18(s, 3H, 2-CH_3); IR (thin film, cm^{-1}) 3446(m), 3393(bs), 3056(w), 3023(w), 2935(m), 2868(m), 1773(w), 1705(s), 1514(s), 1389(m), 1302(m), 1252(m), 1189(m), 1172(m); HRMS m/z (M + Na^+) calcd 463.1993, found 463.2013. Anal. Calcd for $C_{28}H_{28}N_2O_3$: C, 76.34; H, 6.41; N, 6.36. Found: C, 76.11; H, 6.41; N, 6.16.

2-Methyl-5-(4-phenoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (38). Method B with **3c** (687 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **38** (910 mg, 44%) as a colorless solid, a mixture of two isomers (maj:min = 5.0:1.0): mp 282–284°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.52 (bs, 1H, 1maj-H), 10.25 (bs, 1H, 1min-H), 7.41–7.47 (m, 2H, Ph), 7.17–7.31 (m, 3H, Ph), 7.07–7.11 (m, 4H, Ph), 5.84 (d, J = 1.8 Hz, 1H, 3min-H), 5.60 (d, J = 1.8 Hz, 1H, 3maj-H), 4.15 (app. d, J = 7.8 Hz, 1H, 3bαmaj-H), 4.01 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmin-H), 3.38 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmaj-H), 3.34 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.00–3.05 (m, 1H, 10aβmin-H), 2.90–2.95 (m, 1H, 10aαmaj-H), 2.04–2.40 (m, 2H, cyclohex., 6bα-H), 2.18 (s, 3H, 2-CH_3), 1.02–1.64 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.1, 176.3, 157.7, 130.0, 128.9, 127.9, 124.1, 123.9, 120.4, 119.7, 119.6, 118.9, 103.7, 46.0, 38.8, 38.7, 38.4, 37.8, 33.0, 32.9, 29.1, 28.1, 26.1, 23.0, 22.7, 21.1, 20.6, 13.4; IR (thin film, cm^{-1}) 3390(bs), 2925(m), 2855(m), 1777(w), 1701(s), 1590(m), 1508(s), 1489(s), 1392(m), 1244(s), 1180(m), 1165(m); HRMS m/z (M + Na$^+$) calcd for $C_{27}H_{26}N_2O_3$: 449.1836, found 449.1837.

2,8-Dimethyl-5-(4-phenoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (39). Method B with **3d** (785 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **39** (1100 mg, 52%) as a colorless solid, a mixture of three isomers (maj:min:min = 1.2:1.0:0.1): mp 278–280°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.67 (bs, 1H, 1min-H), 10.52 (bs, 1H, 1min-H), 10.27 (bs, 1H, 1maj-H), 7.38–7.46 (m, 2H, Ph), 7.15–7.26 (m, 3H, Ph), 7.06–7.14 (m, 4H, Ph), 5.82–5.84 (m, 1H, 3maj-H), 5.60 (d, J = 1.8 Hz, 1H, 3min-H), 5.58 (d, J = 2.1 Hz, 1H, 3min-H), 4.14 (app. d, J = 8.1 Hz, 1H, 3bαmin-H), 4.01 (dd, J = 8.3, 2.3 Hz, 1H, 3bαmin-H), 3.99 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.41 (dd, J = 8.4, 6.0 Hz, 1H, 6aαmin-H), 3.40 (dd, J = 8.0, 5.6 Hz, 1H, 6aαmin-H), 3.35 (dd, J = 8.4, 5.7 Hz, 1H, 6aαmaj-H), 2.93–3.00 (m, 1H, 10amaj-H), 2.85–2.92 (m, 1H, 10amin-H), 2.70–2.74 (m, 1H, 10amin-H), 2.44–2.54 (m, 1H, 6bmaj-H), 2.30–2.40 (m, 1H, 6bmin-H), 2.18 (s, 3H, 2-CH_3), 0.98–2.16 (m, 7H, cyclohex.), 0.95 (d, J = 6.9 Hz, 3H, 8-CH_3 maj), 0.71 (d, J = 6.0 Hz, 3H, 8-CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 177.5, 157.0, 156.6, 130.7, 129.3, 129.1, 127.9, 126.6, 124.5, 119.7, 119.0, 108.9, 105.0, 45.6, 33.0–33.3 (overlapped peaks), 32.2–32.6 (overlapped peaks), 26.7, 13.5; IR (thin film, cm^{-1}) 3461(m), 3394(bs), 3077(w), 2954(m), 2923(m), 2864(m), 1777(w), 1711(s), 1591(m), 1508(s), 1489(s), 1391(m), 1245(s), 1192(m), 1165(m); HRMS m/z (M + Na$^+$) calcd 463.1993, found 463.1993. Anal. Calcd for $C_{28}H_{28}N_2O_3$: C, 76.34; H, 6.41; N, 6.36. Found: C, 76.19; H, 6.21; N, 6.23.

8-Ethyl-2-methyl-5-(4-phenoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (40). Method B with **3e** (883 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **40** (1000 mg, 46%) as a colorless solid, a mixture of three isomers (maj:min:min = 1.7:1.0:0.3): mp 272–274°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.52 (bs, 1H, 1min-H), 10.27 (bs, 1H, 1maj-H), 10.24 (bs, 1H, 1min-H), 7.40–7.47 (m, 2H, Ph), 7.17–7.25 (m, 3H, Ph), 7.06–7.13 (m, 4H, Ph), 5.83 (dd, J = 2.1, 0.6 Hz, 1H, 3maj-H), 5.61 (dd, J = 2.4, 0.6 Hz, 1H, 3min-H), 5.58 (dd, J = 2.7, 0.9 Hz, 1H, 3min-H), 4.15 (app. d, J = 8.7 Hz, 1H, 3bαmin-H), 4.14 (app. d, J = 8.1 Hz, 1H, 3bαmin-H), 3.99 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.41 (dd. J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.38 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmin-H), 3.34 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmaj-H), 2.99–3.04 (m, 1H, 10amin-H), 2.93–2.99 (m, 1H, 10aαmaj-H), 2.85–2.91 (m, 1H, 10aβmin-H), 2.24–2.50 (m, 1H, 6b-H), 2.27 (s, 3H, 2-CH_3), 0.84–2.16 (m, 7H, cyclohex.), 1.37 (app. q, J = 7.8 Hz, 2H, CH_2CH$_3$), 0.80 (t, J = 7.2 Hz, 3H, CH$_2$CH_3 maj), 0.78 (t, J = 7.2 Hz, 3H, CH$_2$CH_3 min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.1, 178.0, 176.9, 159.8, 157.9, 130.0, 127.9, 127.4, 127.0, 126.8, 123.9, 119.7, 118.9, 109.5, 105.8, 45.7, 38.9, 34.5, 34.0, 32.8–33.1 (overlapped peaks), 13.3, 12.3; IR (thin film, cm^{-1}) 3462(m), 3393(bs), 3073(w), 2958(m), 2922(s), 2868(m), 1776(w), 1702(s), 1590(m), 1508(s), 1489(s), 1390(m), 1243(s), 1190(m), 1164(m); HRMS m/z (M + Na$^+$) calcd for $C_{29}H_{30}N_2O_3$: 477.2149, found 477.2153.

8-Isopropyl-2-methyl-5-(4-phenoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (41). Method B with **3f** (981 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **41** (950 mg, 42%) as a colorless solid, a mixture of three isomers (maj:min:min = 2.1:1.0:0.3): mp 158–160°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.52 (bs, 1H, 1min-H), 10.28 (bs, 1H, 1maj-H), 10.24 (bs, 1H, 1min-H), 7.40–7.48 (m, 2H, Ph), 7.16–7.26 (m, 3H, Ph), 7.04–7.16 (m, 4H, Ph), 5.80–5.84 (m, 1H, 3maj-H), 5.60–5.63 (m, 1H, 3min-H), 5.57–5.59 (m, 1H, 3min-H), 4.15 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.14 (d, J = 8.1 Hz, 1H, 3bαmin-H), 3.99 (dd, J = 8.7, 1.5 Hz, 1H, 3bαmaj-H), 3.42 (dd, J = 8.1, 4.5 Hz, 1H, 6aαmin-H), 3.38 (dd, J = 8.7, 5.4 Hz, 1H, 6aαmin-H), 3.34 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmaj-H), 2.93–3.10 (m, 1H, 10aαmaj-H), 2.86–2.92 (m, 1H, 10aβmin-H), 2.22–2.50 (m, 1H, 6b-H), 2.18 (s, 3H, 2-CH_3), 0.90–2.12 (m, 8H, cyclohex., CH(CH$_3$)$_2$), 0.74–0.88 (m, 6H, CH(CH_3)$_2$); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.3, 176.9, 157.5, 156.5, 130.1, 130.0, 127.9, 127.4, 126.8, 124.1, 124.0, 119.8, 119.7, 118.8, 109.6, 109.5, 105.8, 105.0, 103.8, 45.6, 39.7, 38.9, 33.1, 33.07, 32.8–33.2 (overlapped peaks), 23.3, 21.4, 21.3, 29.1, 25.6, 23.1–23.7 (overlapped peaks), 21.3, 20.9, 20.1, 13.3; IR (thin film, cm^{-1}) 3394(bs), 3064(w), 2930(m), 2866(m), 1776(w), 1705(s), 1591(m), 1508(s), 1490(s), 1389(m), 1243(s), 1189(m), 1165(m); HRMS m/z (M + Na$^+$) calcd 491.2306, found 491.2325. Anal. Calcd for $C_{30}H_{32}N_2O_3$: C, 76.90; H, 6.88; N, 5.98. Found: C, 76.76; H, 6.79; N, 5.78.

8-tert-Butyl-2-methyl-5-(4-phenoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (42). Method B with **3g** (1080 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **42** (700 mg, 30%) as a colorless solid, a mixture of three isomers (maj:min:min = 4.8:1.0:0.9): mp 243–245°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.50 (d, J = 1.8 Hz, 1H, 1min-H), 10.36 (d, J = 1.5 Hz, 1H, 1maj-H), 10.33 (d, J = 2.1 Hz, 1H, 1min-H), 7.38–7.48 (m, 2H, Ph), 7.04–7.27 (m, 7H, Ph), 5.67 (dd, J = 2.7, 1.5 Hz, 1H, 3min-H), 5.58 (dd, J = 2.4, 0.6 Hz, 1H, 3min-H), 5.54 (dd, J = 2.4, 0.9 Hz, 1H, 3maj-H), 4.16 (app. d, J = 7.5 Hz, 1H, 3bαmin-H), 4.10 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 3.85 (dd, J = 7.5, 1.2 Hz, 1H, 3bαmaj-H), 3.49 (dd, J = 8.4, 6.3 Hz, 1H, 6aαmin-H), 3.46 (dd, J = 7.2, 8.7 Hz, 1H, 6aαmin-H), 3.42 (dd, J = 8.1, 5.1 Hz, 1H, 6aαmaj-H), 2.86–2.92 (m, 1H, 10amin-H), 2.50–

2.66 (m, 1H, 10aαmaj-H), 2.24–2.50 (m, 1H, 6b-H), 2.17 (s, 3H, 2-CH_3 min), 2.13 (s, 3H, 2-CH_3 maj), 2.12 (s, 3H, 2-CH_3 min), 1.38–2.00 (m, 4H, cyclohex.), 0.95–1.24 (m, 3H, cyclohex.), 0.84 (s, 9H, *t*-Bu maj), 0.67 (s, 9H, *t*-Bu min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.7, 176.1, 157.2, 156.5, 130.8, 129.2, 128.8, 128.5, 127.9, 124.6, 120.1, 119.8, 119.6, 119.0, 116.9, 104.2, 102.7, 44.9, 34.3–34.6 (overlapped peaks), 33.2, 32.6, 28.2, 28.0, 27.8, 13.4; IR (thin film, cm^{-1}) 3388(bs), 3070(w), 2952(s), 2866(m), 1777(w), 1705(s), 1591(m), 1507(s), 1489(s), 1392(m), 1244(s), 1180(m), 1165(m); HRMS *m*/*z* (M + Na^+) calcd 505.2462, found 505.2467. Anal. Calcd for $C_{31}H_{34}N_2O_3$: C, 77.15; H, 7.10; N, 5.80. Found: C, 76.92; H, 6.98; N, 5.66.

2-Methyl-5-(4-phenoxyphenyl)-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (43). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **43** (1150 mg, 48%) as a colorless solid, a mixture of four isomers (maj:min:min = 5.4:1.0:1.0:0.7): mp 297–299°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.57 (bs, 1H, 1min-H), 10.55 (bs, 1H, 1min-H), 10.38 (bs, 1H, 1maj-H), 10.34 (bs, 1H, 1min-H), 7.37–7.46 (m, 2H, Ph), 6.97–7.35 (m, 12H, Ph), 5.86–5.88 (m, 1H, 3min-H), 5.78–5.86 (m, 1H, 3maj-H), 5.63–5.66 (m, 1H, 3min-H), 4.17 (app. d, *J* = 8.4 Hz, 1H, 3bαmin-H), 4.06 (dd, *J* = 8.6, 1.7 Hz, 1H, 3bαmin-H), 4.01 (d, *J* = 7.8 Hz, 1H, 3bαmaj-H), 3.50 (dd, *J* = 8.4, 5.1 Hz, 1H, 6aαmin-H), 3.45 (dd, *J* = 8.1, 5.1 Hz, 1H, 6aαmaj-H), 3.07–3.13 (m, 1H, 10amin-H), 2.96–3.04 (m, 1H, 10amin-H), 2.86–2.97 (m, 1H, 10aαmaj-H), 2.80–2.89 (m, 1H, 10amin-H), 2.46–2.58 (m, 1H, 6b-H), 1.20–2.30 (m, 7H, cyclohex.), 2.19 (s, 3H, 2-CH_3); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 178.3, 177.0, 176.3, 156.5, 130.7, 129.2, 128.9, 127.6, 124.6, 124.5, 119.9, 119.7, 119.1, 45.4, 33.2–33.5 (overlapping peaks), 13.5; IR (thin film, cm^{-1}) 3390(bs), 3060(w), 2932(m), 2866(m), 1774(w), 1702(s), 1590(m), 1507(s), 1490(s), 1391(m), 1243(s), 1191(m), 1165(m); HRMS *m*/*z* (M + Na^+) calcd for $C_{33}H_{30}N_2O_3$: 525.2149, found 525.2140.

2-Methyl-5-(3-nitrophenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (44). Method B with **3c** (600 mg, 6.12 mmol), 1-h reflux, reprecipitation from diethyl ether (20 mL), and then a diethyl ether wash (10 mL) gave **44** (700 mg, 40%) as a yellow solid, a mixture of two isomers (maj:min = 2.8:1.0): mp 223–225°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.24–8.31 (m, 3H, Ph, Ph, 1min-H), 7.62–7.77 (m, 3H, Ph, 1maj-H), 6.19 (dd, *J* = 2.4, 1.2 Hz, 1H, 3maj-H), 5.79 (dd, *J* = 2.4, 0.6 Hz, 1H, 3min-H), 4.04 (app. d, *J* = 8.7 Hz, 1H, 3bαmin-H), 4.01 (dd, *J* = 8.6, 2.0 Hz, 1H, 3bαmaj-H), 3.52 (dd, *J* = 8.9, 5.6 Hz, 1H, 6aαmin-H), 3.45 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.14–3.20 (m, 1H, 10aαmaj-H), 3.03–3.08 (m, 1H, 10aβmin-H), 2.53–2.61 (m, 1H, 6bα-H), 2.33 (s, 3H, 2-CH_3), 2.12–2.28 (m, 1H, cyclohex.), 1.43–1.85 (m, 3H, cyclohex.), 1.17–1.38 (m, 4H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.4, 176.1, 148.5, 133.5, 133.1, 132.3, 130.1, 130.0, 127.7, 127.1, 123.3, 123.1, 121.6, 120.6, 108.8, 105.8, 103.8, 46.0, 38.9, 38.7, 38.4, 37.9, 33.0, 32.8, 29.1, 28.0, 26.0, 25.6, 23.1, 22.7, 21.0, 20.5, 13.3; IR (thin film, cm^{-1}) 3414(bs), 3081(m), 2928(m), 2858(m), 1779(w), 1707(s), 1532(s), 1384(w), 1350(m), 1164(m); HRMS *m*/*z* (M + Na^+) calcd for $C_{21}H_{21}N_3O_4$: 402.1425, found 402.1434.

2,8-Dimethyl-5-(3-nitrophenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (45). Method B with **3d** (750 mg, 6.70 mmol), 1-h reflux, reprecipitation from diethyl ether (20 mL), and then a diethyl ether wash (10 mL) gave **45** (820 mg, 44%) as a colorless solid, a mixture of two isomers (maj:min = 3.7:1.0): mp 236–238°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.24–8.30 (m, 3H, Ph, Ph, 1min-H), 7.64–7.76 (m, 3H, Ph, 1maj-H), 6.18 (dd, *J* = 2.6, 1.1 Hz, 1H, 3maj-H), 5.80 (dd, *J* = 2.6, 1.1 Hz, 1H, 3min-H), 4.04 (app. d, *J* = 8.4 Hz, 1H, 3bαmin-H), 4.01 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.53 (dd, *J* = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.46 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmaj-H), 3.08–3.14 (m, 1H, 10aαmaj-H), 2.98–3.04 (m, 1H, 10aβmin-H), 2.72–2.82 (m, 1H, 6bαmaj-H), 2.55–2.67 (m, 1H, 6bαmin-H), 2.32 (s, 3H, 2-CH_3), 1.81–2.12 (m, 3H, cyclohex.), 1.45–1.68 (m, 2H, cyclohex.), 1.18–1.34 (m, 1H, cyclohex.), 0.95–1.14 (m, 1H, cyclohex.), 1.03 (s, 3H, 8-CH_3 maj), 1.01 (s, 3H, 8-CH_3 min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.3, 176.1, 148.5, 132.2, 129.9, 127.7, 123.1, 121.7, 109.0, 105.7, 45.7, 39.0, 32.9, 32.6, 26.7, 13.3; IR (thin film, cm^{-1}) 3430(bs), 2924(m), 2850(m), 1773(w), 1705(s), 1534(m), 1385(m), 1350(m), 1168(m); HRMS *m*/*z* (M + Na^+) calcd 416.1582, found 416.1567. Anal. Calcd for $C_{22}H_{23}N_3O_4$: C, 67.16; H, 5.89; N, 10.68. Found: C, 67.12; H, 5.64; N, 10.53.

8-Ethyl-2-methyl-5-(3-nitrophenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (46). Method B with **3e** (820 mg, 6.50 mmol), 1-h reflux, reprecipitation from diethyl ether (20 mL), and then a diethyl ether wash (10 mL) gave **46** (800 mg, 41%) as a yellow solid, a mixture of two isomers (maj:min = 3.3:1.0): mp 213–215°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.24–8.31 (m, 3H, Ph, Ph, 1min-H), 7.64–7.75 (m, 3H, Ph, 1maj-H), 6.18 (dd, *J* = 2.1, 0.9 Hz, 1H, 3maj-H), 5.81 (app. d, *J* = 2.7 Hz, 1H, 3min-H), 4.01 (dd, *J* = 8.4 Hz, 1H, 3bα-H), 3.52 (dd, *J* = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.45 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmaj-H), 3.08–3.14 (m, 1H, 10aαmaj-H), 2.97–3.04 (m, 1H, 10aβmin-H), 2.67–2.76 (m, 1H, 6bα-H), 2.33 (s, 3H, 2-CH_3), 1.80–2.04 (m, 2H, cyclohex.), 1.18–1.66 (m, 6H, cyclohex., 8-CH_2CH_3), 1.10–1.26 (m, 1H, cyclohex.), 0.86 (t, *J* = 7.2 Hz, 3H, 8-CH_2CH_3), 0.85 (t, *J* = 7.2 Hz, 3H, 8-CH_2CH_3); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.3, 176.2, 148.5, 133.1, 132.2, 130.7, 130.0, 127.7, 123.7, 123.1, 121.6, 109.0, 105.7, 45.7, 39.0, 33.9, 32.9, 23.8, 13.3, 12.3; IR (thin film, cm^{-1}) 3401(bs), 2928(m), 2868(m), 1778(w), 1714(s), 1532(s), 1353(m), 1160(m); HRMS *m*/*z* (M + Na^+) calcd 430.1738, found 430.1732. Anal. Calcd for $C_{23}H_{25}N_3O_4$: C, 67.80; H, 6.18; N, 10.31. Found: C, 68.29; H, 6.20; N, 10.51.

8-Isopropyl-2-methyl-5-(3-nitrophenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (47). Method B with **3f** (910 mg, 6.50 mmol), 1-h reflux, reprecipitation from diethyl ether (20 mL), and then a diethyl ether wash (10 mL) gave **47** (600 mg, 30%) as a yellow solid, a mixture of three isomers (maj:min:min = 2.8:1.0:0.2): mp 205–207°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.24–8.31 (m, 3H, Ph, Ph, 1min-H), 7.63–7.74 (m, 3H, Ph, 1maj-H), 6.18 (dd, *J* = 2.7, 1.5 Hz, 1H, 3maj-H), 5.81 (dd, *J* = 2.7, 0.6 Hz, 1H, 3min-H), 5.77 (dd, *J* = 2.3, 1.1 Hz, 1H, 3min-H), 4.01 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bα-H), 3.55 (dd, *J* = 8.4, 5.7 Hz, 1H, 6aαmin-H), 3.51 (dd, *J* = 8.9, 5.9 Hz, 1H, 6aαmin-H), 3.44 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.07–3.15 (m, 1H, 10aαmaj-H), 2.98–3.04 (m, 1H, 10aβmin-H), 2.66–2.75 (m,

1H, 6bαmaj-H), 2.53–2.62 (m, 1H, 6bmin-H), 2.33 (s, 3H, 2-CH_3), 1.78–2.02 (m, 3H, cyclohex.), 1.20–1.65 (m, 5H, cyclohex., C*H*(CH_3)$_2$), 0.90 (d, J = 6.3 Hz, 6H, CH(CH_3)$_2$), 0.86 (d, J = 6.6 Hz, 6H, CH(CH_3)$_2$); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.4, 176.1, 148.5, 133.1, 132.2, 130.0, 127.7, 123.1, 121.6, 108.9, 105.7, 45.7, 40.1, 39.6, 39.0, 33.1, 32.9, 26.1, 23.3, 21.3, 20.8, 13.3; IR (thin film, cm^{-1}) 3408(bs), 2937(m), 2850(m), 1778(w), 1708(s), 1531(m), 1381(m), 1353(m), 1195(m), 1164(m); HRMS m/z (M + Na^+) calcd 444.1895, found 444.1889. Anal. Calcd for $C_{24}H_{27}N_3O_4$: C, 68.39; H, 6.46; N, 9.97. Found: C, 68.38; H, 6.26; N, 9.75.

8-tert-Butyl-2-methyl-5-(3-nitrophenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (48). Method B with **3g** (1000 mg, 6.490 mmol), 3-h reflux, removal of solvent under vacuum, elution through a 5-cm silica gel plug with CH_2Cl_2, and then reprecipitation twice from diethyl ether/hexanes (2:1, 20 mL) gave **48** (650 mg, 31%) as a yellow solid, a mixture of two isomers (maj:min = 2.1:1.0): mp 203–205°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.18–8.26 (m, 3H, Ph, 1maj-H), 7.57–7.71 (m, 3H, Ph, Ph, 1min-H), 7.01 (dd, J = 2.9, 1.1 Hz, 1H, 3maj-H), 7.00 (m, overlapped, 1H, 3min-H), 4.14 (dd, J = 7.5, 2.1 Hz, 1H, 3bαmin-H), 4.09 (dd, J = 7.5, 1.5 Hz, 1H, 3bαmaj-H), 3.39 (dd, J = 7.4, 5.4 Hz, 1H, 6aαmaj-H), 3.35 (dd, J = 7.5, 3.9 Hz, 1H, 6aαmin-H), 2.70–2.78 (m, 1H, 6b-H), 2.62–2.70 (m, 1H, 10aβmaj-H), 2.44–2.54 (m, 1H, 10aαmin-H), 1.75–2.32 (m, 3H, cyclohex.), 2.27 (d, J = 0.6 Hz, 3H, 2-CH_3), 1.63 (ddd, J = 13.9, 11.5, 7.1 Hz, 1H, cyclohex.), 1.07–1.40 (m, 3H, cyclohex.), 0.94 (s, 9H, *t*-Bu min), 0.92 (s, 9H, *t*-Bu maj); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.38 (d, J = 1.8 Hz, 1H, 1maj-H), 10.32 (d, J = 2.1 Hz, 1H, 1min-H), 8.21–8.28 (m, 1H, Ph), 8.06–8.08 (m, 1H, Ph), 7.62–7.80 (m, 2H, Ph), 5.67–5.70 (m, 1H, 3-H), 3.98 (dd, J = 7.2, 1.8 Hz, 1H, 3bαmin-H), 3.96 (dd, J = 7.2, 1.2 Hz, 1H, 3bαmaj-H), 3.50–3.55 (m, overlapped, 1H, 6aαmin-H), 3.53 (dd, J = 7.5, 5.7 Hz, 1H, 6aαmaj-H), 2.55–2.70 (m, 1H, 10aβmaj-H), 2.50 (s, 3H, 2-CH_3), 1.49–2.33 (m, 5H, cyclohex., 6bα-H), 1.00–1.40 (m, 3H, cyclohex.), 0.89 (s, 9H, *t*-Bu min), 0.86 (s, 9H, *t*-Bu maj); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.2, 176.1, 148.5, 133.4, 132.7, 132.3, 130.4, 129.9, 129.7, 128.0, 127.7, 123.0, 122.8, 122.0, 121.7, 109.3, 105.0, 104.1, 49.0, 46.0, 45.9, 42.2, 41.8, 41.1, 40.8, 34.3, 34.2, 34.1, 32.8, 32.7, 30.5, 28.9, 27.7, 27.6, 26.3, 25.6, 13.2; IR (thin film, cm^{-1}) 3393(bs), 3097(m), 2958(s), 2868(s), 2361(m), 2255(m), 1778(m), 1716 (s), 1532(s), 1478(m), 1356(s), 1171(m); HRMS m/z (M + Na^+) calcd for $C_{25}H_{29}N_3O_4$: 458.2051, found 458.2036.

2-Methyl-5-(3-nitrophenyl)-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (49). Method B with **3h** (1140 mg, 6.555 mmol), 1-h reflux, reprecipitation twice from ethanol/diethyl ether (4:1, 20 mL), and then a diethyl ether wash (10 mL) gave **49** (900 mg, 40%) as a yellow solid, a mixture of three isomers (maj:min:min = 4.2:1.0:0.6): mp 236–238°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.59 (bs, 1H, 1min-H), 10.41 (bs, 1H, 1maj-H), 8.28–8.33 (m, 2H, 5-Ph), 7.74–7.86 (m, 2H, 5-Ph), 7.27–7.35 (m, 4H, 8-Ph), 7.15–7.21 (m, 1H, 8-Ph), 5.86–5.89 (m, 1H, 3min-H), 5.80–5.86 (m, 1H, 3maj-H), 5.65–5.67 (m, 1H, 3min-H), 4.22 (d, J = 9.0 Hz, 1H, 3bαmin-H), 4.13 (d, J = 9.0 Hz, 1H, 3bαmin-H), 4.06 (d, J = 8.1 Hz, 1H, 3bαmaj-H), 3.28–3.58 (m, obscured by H_2O, 1H, 6aα-H), 3.08–3.17 (m, 1H, 10amin-H), 2.83–3.00 (m, 1H, 10aαmaj-H), 2.51–2.62 (m, 1H, 6b-H), 2.19 (s, 3H, 2-CH_3), 1.34–2.09 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.3, 148.4, 134.0, 131.0, 128.8, 127.6, 126.1, 123.6, 122.2, 45.5, 40.9, 39.2, 33.4, 13.5; IR (thin film, cm^{-1}) 3417(bs), 3071(m), 2928(m), 2865(m), 1776(w), 1707(s), 1533(m), 1382(m), 1350(m), 1160(m); HRMS m/z (M + Na^+) calcd for $C_{27}H_{25}N_3O_4$: 478.1738, found 478.1757.

4-(2-Methyl-4,6-dioxo-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indol-5-yl)benzoic acid (50). Method B with **3c** (600 mg, 6.12 mmol), 1.5-h reflux, reprecipitation from diethyl ether (10 mL), and then a diethyl ether wash (5 mL) gave **50** (550 mg, 31%) as a colorless solid, a mixture of two isomers (maj:min = 1.5:1.0): mp 257–259°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 13.16 (s, 1H, CO_2H), 10.55 (d, J = 2.4 Hz, 1H, 1min-H), 10.27 (d, J = 2.1 Hz, 1H, 1maj-H), 8.07 (d, J = 8.7 Hz, 2H, Ph min), 8.05 (d, J = 8.4 Hz, 2H, Ph maj), 7.41 (d, J = 8.7 Hz, 2H, Ph min), 7.40 (d, J = 8.7 Hz, 2H, Ph maj), 5.84 (dd, J = 2.1, 0.6 Hz, 1H, 3maj-H), 5.60 (dd, J = 2.1, 0.6 Hz, 1H, 3min-H), 4.19 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.05 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.37–3.44 (m, 1H, 6aα-H), 3.00–3.06 (m, 1H, 10aαmaj-H), 2.90–2.96 (m, 1H, 10aβmin-H), 2.27–2.41 (m, 1H, 6bα-H), 2.18 (s, 3H, 2-CH_3), 0.95–1.62 (m, 8H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.1, 177.9, 177.2, 167.2, 136.7, 130.9, 130.5, 127.4, 126.9, 126.6, 119.0, 108.6, 105.4, 105.0, 65.5, 46.3, 46.5, 46.0, 38.4, 33.0, 25.7, 21.4, 13.5; IR (thin film, cm^{-1}) 3472(bs), 3409(bs), 2950(m), 2847(m), 2294(w), 1770(w), 1686(s), 1514(w), 1422(m), 1378(m), 1279(m), 1170(m); HRMS m/z (M + Na^+) calcd 401.1473, found 401.1490. Anal. Calcd for $C_{22}H_{22}N_2O_4$: C, 69.83; H, 5.86; N, 7.40. Found: C, 69.59; H, 6.20; N, 7.45.

4-(2,8-Dimethyl-4,6-dioxo-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]indol-5-yl)benzoic acid (51). Method B with **3d** (750 mg, 6.70 mmol), 1.5-h reflux, reprecipitation from ethanol/diethyl ether (1:3, 20 mL), and then a diethyl ether wash (5 mL) gave **51** (550 mg, 31%) as a colorless solid, a mixture of two isomers (maj:min = 1.6:1.0): mp 258–260°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 13.15 (s, 1H, CO_2H), 10.54 (d, J = 2.2 Hz, 1H, 1min-H), 10.28 (d, J = 1.6 Hz, 1H, 1maj-H), 8.07 (d, J = 8.4 Hz, 2H, Ph min), 8.05 (d, J = 8.7 Hz, 2H, Ph maj), 7.41 (d, J = 8.4 Hz, 2H, Ph min), 7.39 (d, J = 8.4 Hz, 2H, Ph maj), 5.82 (d, J = 1.2 Hz, 1H, 3maj-H), 5.61 (d, J = 2.1 Hz, 1H, 3min-H), 4.19 (app. d, J = 8.7 Hz, 1H, 3bαmin-H), 4.04 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.43 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.39 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmaj-H), 2.94–3.00 (m, 1H, 10aαmaj-H), 2.86–2.92 (m, 1H, 10aβmin-H), 2.35–2.60 (m, overlapped by DMSO, 1H, 6bα-H), 2.17 (s, 3H, 2-CH_3), 1.74–1.94 (m, 2H, cyclohex.), 1.30–1.46 (m, 2H, cyclohex.), 1.08–1.22 (m, 1H, cyclohex.), 0.88–1.04 (m, 2H, cyclohex.), 0.96 (d, J = 7.2 Hz, 3H, 8-CH_3); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.0, 177.2, 167.2, 136.7, 130.8, 130.5, 127.4, 126.6, 108.8, 105.3, 45.7, 38.3, 34.7, 33.1, 32.6, 27.0, 26.7, 13.5; IR (thin film, cm^{-1}) 3458(bs), 3381(bs), 2919(m), 2285(w), 1780(w), 1700(s), 1515(w), 1425(m), 1382(m), 1285(m), 1161(m); HRMS m/z (M + Na^+) calcd for $C_{23}H_{24}N_2O_4$: 415.1629, found 415.1628.

4-(8-Ethyl-2-methyl-4,6-dioxo-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]-5-indolyl)benzoic acid (52). Method B with **3e** (820 mg, 6.50 mmol), 1-h reflux, reprecipitation from diethyl ether (20 mL), and then a diethyl ether

wash (10 mL) gave **52** (600 mg, 31%) as a colorless solid, a mixture of three isomers (maj:min:min = 3.7:1.0:0.1): mp 233–235°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 13.14 (s, 1H, CO_2H), 10.54 (d, J = 2.4 Hz, 1H, 1min-H), 10.29 (d, J = 2.1 Hz, 1H, 1maj-H), 10.24–10.28 (app. bs, 1H, 1min-H), 8.06 (d, J = 8.7 Hz, 2H, Ph), 7.38 (d, J = 8.4 Hz, 2H, Ph), 5.83 (app. s, 1H 3maj-H), 5.62 (d, J = 1.8 Hz, 1H, 3min-H), 5.59 (app. s, 1H, 3min-H), 4.18 (d, J = 8.1 Hz, 1H, 3bαmin-H), 4.04 (dd, J = 8.6, 1.7, 1H, 3bαmaj-H), 3.42 (dd, J = 8.4, 5.7 Hz, 1H, 6aαmin-H), 3.38 (dd, J = 8.7, 5.1 Hz, 1H, 6aαmaj-H), 2.93–3.00 (m, 1H, 10aαmaj-H), 2.86–2.92 (m, 1H, 10aβ-H), 2.40–2.54 (m, 1H, 6b-H), 1.86–2.40 (m, 2H, cyclohex.), 2.18 (s, 3H, 2-CH_3), 0.92–1.84 (m, 7H, cyclohex., CH_2CH$_3$), 0.87 (t, J = 7.5 Hz, 3H, CH$_2$CH_3 min), 0.80 (t, J = 7.2 Hz, 3H, CH$_2$CH_3 maj); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.1, 177.2, 167.2, 136.7, 130.8, 130.6, 127.3, 126.6, 108.7, 105.3, 45.6, 39.2, 35.0, 33.9, 33.3, 33.1, 32.8, 13.5, 12.6; IR (thin film, cm^{-1}) 3396(bs), 2922(m), 2860(m), 2293(w), 1693(s), 1513(w), 1426(m), 1387(m), 1284(m), 1166(m); HRMS m/z (M + Na$^+$) calcd 429.1786, found 429.1797. Anal. Calcd for $C_{24}H_{26}N_2O_4$: C, 70.92; H, 6.45; N, 6.89. Found: C, 70.92; H, 6.37; N, 6.75.

4-(8-Isopropyl-2-methyl-4,6-dioxo-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]-5-indolyl)benzoic acid (53). Method B with **3f** (910 mg, 6.50 mmol), 1-h reflux, reprecipitation from diethyl ether (20 mL), and then a diethyl ether wash (10 mL) gave **53** (600 mg, 30%) as a colorless solid, a mixture of three isomers (maj:min:min = 5.3:1.0:0.2): mp 260–262°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 13.16 (s, 1H, CO_2H), 10.55 (d, J = 1.8 Hz, 1H, 1min-H), 10.30 (d, J = 2.1 Hz, 1H, 1maj-H), 10.24–10.27 (app. bs, 1H, 1min-H), 8.07 (d, J = 8.4 Hz, 2H, Ph), 7.37 (d, J = 8.7 Hz, 2H, Ph), 5.83 (app. s, 1H, 3maj-H), 5.62 (d, J = 1.2 Hz, 1H, 3min-H), 5.59 (app. s, 1H, 3min-H), 4.18 (app. d, 1H, 3bαmin-H), 4.03 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.34–3.48 (m, 1H, 6aα-H), 2.93–3.03 (m, 1H, 10aαmaj-H), 2.86–2.92 (m, 1H, 10aβmin-H), 2.39–2.50 (m, 1H, 6b-H), 2.18 (s, 3H, 2-CH$_{-3}$), 1.70–2.12 (m, 2H, cyclohex.), 1.10–1.55 (m, 6H, cyclohex., CH(CH$_3$)$_2$), 0.85 (d, J = 6.6 Hz, 6H, CH(CH_3)$_2$ maj), 0.79 (d, J = 6.6 Hz, CH(CH$_3$)$_2$ min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.2, 177.1, 167.2, 136.8, 130.9, 130.6, 127.2, 126.6, 108.7, 105.3, 65.5, 45.6, 39.4, 33.0, 21.7, 21.0, 15.7, 13.5; IR (thin film, cm^{-1}) 3398(bs), 2950(m), 2865(m), 1770(w), 1696(s), 1514(w), 1430(m), 1388(m), 1285(m), 1184(m); HRMS m/z (M + Na$^+$) calcd 443.1942, found 443.1938. Anal. Calcd for $C_{25}H_{28}N_2O_4$: C, 71.41; H, 6.71; N, 6.66. Found: C, 71.16; H, 6.46; N, 6.49.

4-(2-Methyl-4,6-dioxo-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-cyclopenta[g]pyrrolo[3,4-e]-5-indolyl)benzoic acid (54). Method B with **3h** (1140 mg, 6.550 mmol), 1-h reflux, reprecipitation from ethanol/diethyl ether (1:2, 20 mL), and then a diethyl ether wash (10 mL) gave **54** (1000 mg, 46%) as a colorless solid, a mixture of two isomers (maj:min = 4.3:1.0): mp 255–257°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 13.16 (s, 1H, CO_2H), 10.57 (bs, 1H, 1min-H), 10.39 (bs, 1H, 1maj-H), 8.06 (d, J = 8.4 Hz, 2H, 5-Ph), 7.41 (d, J = 8.7 Hz, 2H, 5-Ph), 7.27–7.35 (m, 4H, 8-Ph), 7.15–7.23 (m, 1H, 8-Ph), 5.78–5.86 (m, 1H, 3maj-H), 5.64–5.67 (m, 1H, 3min-H), 4.22 (d, J = 8.7 Hz, 1H, 3bαmin-H), 4.04 (d, J = 7.5 Hz, 1H, 3bαmaj-H), 3.25–3.58 (m, obscured by H_2O, 1H, 6aα-H), 2.80–3.00 (m, 1H, 10aαmaj-H), 2.40–60 (m, overlapped by DMSO, 1H, 6bα-H), 2.18 (s, 3H, 2-CH_3), 1.45–2.10 (m, 7H, 1H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.2, 177.1, 167.2, 136.8, 130.7, 130.6, 128.8, 127.2, 127.1, 126.6, 108.7, 99.7, 65.5, 45.6, 42.5, 42.1, 39.2, 33.0, 21.8, 15.7, 13.5; IR (thin film, cm^{-1}) 3475(bs), 3399(bs), 2933(m), 2861(m), 1770(w), 1703(s), 1510(w), 1427(m), 1386(m), 1286(m), 1188(m); HRMS m/z (M + Na$^+$) calcd 477.1786, found 477.1804. Anal. Calcd for $C_{28}H_{26}N_2O_4$: C, 73.99; H, 5.77; N, 6.16. Found: C, 73.66; H, 5.42; N, 6.00.

5-(4-Bromophenyl)-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (55). Method B with **3c** (687 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **55** (820 mg, 41%) as a colorless solid, a mixture of two isomers (maj:min = 1.8:1.0): mp 284–286°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.53 (bs, 1H, 1min-H), 10.26 (bs, 1H, 1maj-H), 7.72 (d, J = 8.4 Hz, 2H, Ph min), 7.70 (d, J = 8.7 Hz, 2H, Ph maj), 7.23 (d, J = 8.7 Hz, 2H, Ph min), 7.22 (d, J = 8.7 Hz, 2H, Ph maj), 5.83 (dd, J = 1.2, 0.6 Hz, 1H, 3maj-H), 5.59 (dd, J = 2.1, 0.6 Hz, 1H, 3min-H), 4.16 (dd, J = 8.4, 0.9 Hz, 1H, 3bαmin-H), 4.02 (dd, J = 8.6, 1.7 Hz, 1H, 3bαmaj-H), 3.40 (dd, J = 8.1, 5.1 Hz, 1H, 6aαmin-H), 3.35 (dd, J = 8.6, 5.3 Hz, 1H, 6aαmaj-H), 2.99–3.05 (m, 1H, 10aα-H), 2.90–2.95 (m, 1H, 10aβ-H), 2.26–2.40 (m, 2H, cyclohex., 6b-H), 2.25–2.40 (m, 2H, cyclohex.), 2.18 (s, 3H, 2-CH_3), 1.32–1.44 (m, 1H, cyclohex.), 0.96–1.24 (m, 4H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.1, 178.0, 177.2, 176.1, 132.6, 132.55, 132.2, 132.0, 129.5, 128.3, 126.9, 126.6, 121.9, 121.7, 119.0, 116.7, 114.0, 108.6, 105.4, 102.9, 94.5, 46.3, 46.0, 38.8, 38.5, 38.4, 38.2, 33.0, 29.3, 27.6, 26.1, 25.7, 23.2, 22.9, 21.4, 20.8, 13.5, 13.4; IR (thin film, cm^{-1}) 3400(bs), 2923(m), 2855(m), 1776(w), 1701(s), 1492(m), 1386(m), 1179(m), 1159(m); HRMS m/z (M + Na$^+$) calcd 435.0679, found 435.0696. Anal. Calcd for $C_{21}H_{21}BrN_2O_2$: C, 61.03; H, 5.12; N, 6.78. Found: C, 61.11; H, 5.03; N, 6.67.

5-(4-Bromophenyl)-2,8-dimethyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (56). Method B with **3d** (785 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **56** (1000 mg, 49%) as a colorless solid, a mixture of three isomers (maj:min:min = 3.6:1.0:0.2): mp 274–276°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.53 (bs, 1H, 1min-H), 10.27 (bs, 1H, 1maj-H), 10.26 (bs, 1H, 1min-H), 7.72 (d, J = 9.0 Hz, 2H, Ph min), 7.71 (d, J = 9.6 Hz, 2H, Ph min), 7.70 (d, J = 8.7 Hz, 2H, Ph maj), 7.23 (d, J = 8.4 Hz, 2H, Ph min), 7.21 (d, J = 8.7 Hz, 2H, Ph min), 7.20 (d, J = 8.7 Hz, 2H, Ph maj), 5.82 (dd, J = 2.1, 0.6 Hz, 1H, 3maj-H), 5.60 (dd, J = 2.1, 0.6 Hz, 1H, 3min-H), 5.59 (d, J = 2.1, 0.6 Hz, 1H, 3min-H), 4.15 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.02 (dd, J = 8.6, 1.7 Hz, 1H, 3bαmin-H), 4.01 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.41 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.36 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.93–3.02 (m, 1H, 10aαmaj-H), 2.85–2.92 (m, 1H, 10aβmin-H), 2.30–2.52 (m, 1H, 6b-H), 2.17 (s, 3H, 2-CH_3), 1.74–2.16 (m, 2H, cyclohex.), 0.86–1.68 (m, 5H, cyclohex.), 0.95 (d, J = 7.2 Hz, 3H, 8-CH_3 maj), 0.72 (d, J = 6.6 Hz, 3H, 8-CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.1, 177.2, 176.0, 132.6, 132.5, 132.2, 132.1, 129.5, 129.4, 128.4, 128.3, 126.6, 121.8, 121.7, 118.5, 116.8, 108.8, 108.6, 105.3, 105.0, 45.6, 39.0, 38.9, 39.8, 33.1, 32.3–32.9 (overlapped peaks), 26.7, 13.4; IR (thin film, cm^{-1}) 3460(m), 3393(bs), 3095(w), 3066(w), 2959(m),

2922(s), 2889(m), 2866(m), 2854(m), 1777(w), 1705(s), 1492(s), 1383(s), 1188(m), 1177(m), 1159(m); HRMS *m/z* (M + Na^+) calcd 449.0836, found 449.0840. Anal. Calcd for $C_{22}H_{23}BrN_2O_2$: C, 61.83; H, 5.42; N, 6.56. Found: C, 62.02; H, 5.21; N, 6.59.

5-(4-Bromophenyl)-8-ethyl-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (57). Method B with **3e** (883 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **57** (1000 mg, 47%) as a colorless solid, a mixture of four isomers (maj:min:min:min = 3.0:1.0:0.3:0.3): mp 277–279°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.53 (bs, 1H, 1min-H), 10.28 (bs, 1H, 1maj-H), 10.25 (bs, 1H, 1min-H), 7.73 (d, *J* = 8.7 Hz, 2H, Ph min), 7.71 (d, *J* = 9.0 Hz, 2H, Ph maj), 7.22 (d, *J* = 8.4 Hz, 2H, Ph min), 7.20 (d, *J* = 8.7 Hz, 2H, Ph maj), 5.81–5.83 (m, 1H, 3maj-H), 5.80–5.82 (m, 1H, 3min-H), 5.61 (d, *J* = 2.4 Hz, 1H, 3min-H), 5.58 (app. d, *J* = 2.1 Hz, 1H, 3min-H), 4.16 (app. d, *J* = 8.1 Hz, 1H, 3bαmin-H), 4.15 (d, *J* = 8.4 Hz, 1H, 3bαmin-H), 4.02 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmin-H), 4.00 (dd, *J* = 8.6, 2.0 Hz, 1H, 3bαmaj-H), 3.43 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.39 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.38 (dd, *J* = 7.5, 5.4 Hz, 1H, 6aαmin-H), 3.35 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.99–3.04 (m, 1H, 10amin-H), 2.93–2.99 (m, 1H, 10aαmaj-H), 2.86–2.90 (m, 1H, 10amin-H), 2.10–2.50 (m, 1H, 6b-H), 2.17 (s, 3H, 2-CH_3), 0.96–2.17 (m, 9H, cyclohex., CH_2CH$_3$), 0.79 (t, *J* = 7.5 Hz, 3H, CH$_2$CH_3 maj), 0.77 (t, *J* = 7.2 Hz, 3H, CH$_2$CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.1, 177.9, 177.2, 176.0, 132.7, 132.6, 132.2, 129.4, 129.3, 128.3, 126.9, 126.6, 121.7, 116.8, 108.7, 105.3, 103.0, 45.6, 33.9, 33.0, 32.6–32.9 (overlapped peaks), 13.5, 12.6, 11.8; IR (thin film, cm^{-1}) 3468(m), 3388(bs), 3093(w), 3065(w), 2957(m), 2927(m), 2869(m), 1777(w), 1705(s), 1492(s), 1383(m), 1188(m), 1176(m), 1157(m), ; HRMS *m/z* (M + Na^+) calcd 463.0992, found 463.0980. Anal. Calcd for $C_{23}H_{25}BrN_2O_2$: C, 62.59; H, 5.71; N, 6.35. Found: C, 62.62; H, 5.63; N, 6.55.

5-(4-Bromophenyl)-8-isopropyl-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (58). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **58** (900 mg, 41%) as a colorless solid, a mixture of three isomers (maj:min:min = 1.8:1.0:0.3): mp 291–293°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.53 (bs, 1H, 1min-H), 10.28 (bs, 1H, 1maj-H), 10.24 (bs, 1H, 1min-H), 7.74 (d, *J* = 8.4 Hz, 2H, Ph min), 7.71 (d, *J* = 8.1 Hz, 2H, Ph maj), 7.20 (d, *J* = 8.4 Hz, 2H, Ph min), 7.18 (d, *J* = 8.7 Hz, 2H, Ph maj), 5.79–5.84 (m, 1H, 3maj-H), 5.60–5.63 (m, 1H, 3min-H), 5.58–5.60 (m, 1H, 3min-H), 4.12–4.18 (m, overlapped, 3bαmin-H), 4.14 (d, *J* = 7.8 Hz, 1H, 3bαmin-H), 4.00 (d, *J* = 8.4 Hz, 1H, 3bαmaj-H), 3.31–3.46 (m, 1H, 6a-H), 2.93–3.02 (m, 1H, 10aαmaj-H), 2.86–2.92 (m, 1H, 10aβmin-H), 2.26–2.50 (m, 1H, 6b-H), 2.17 (s, 3H, 2-CH_3), 0.90–2.08 (m, 8H, cyclohex., CH(CH$_3$)$_2$), 0.67–0.86 (m, 6H, CH(CH_3)$_2$); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.1, 176.0, 165.3, 132.7, 132.6, 129.3, 128.2, 126.6, 121.9, 119.0, 116.7, 105.2, 103.0, 45.9, 35.0, 32.7–33.2 (overlapped peaks), 28.9, 25.5, 21.1, 21.0, 20.9, 13.5; IR (thin film, cm^{-1}) 3467(m), 3398(s), 3094(w), 3067(w), 2946(m), 2888(m), 2867(m), 1777(w), 1705(s), 1492(s), 1386(m), 1176(m), 1162(m); HRMS *m/z* (M + Na^+) calcd 477.1149, found 477.1152. Anal. Calcd for $C_{24}H_{27}BrN_2O_2$: C, 63.30; H, 5.98; N, 6.15. Found: C, 63.07; H, 5.67; N, 6.16.

5-(4-Bromophenyl)-8-tert-butyl-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (59). Method B with **3g** (1080 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **59** (700 mg, 31%) as a colorless solid, a mixture of three isomers (maj:min:min = 3.3:1.0:0.5): mp 263–265°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.51 (d, *J* = 1.8 Hz, 1H, 1min-H), 10.38 (d, *J* = 1.5 Hz, 1H, 1maj-H), 10.34 (d, *J* = 1.8 Hz, 1H, 1min-H), 7.77 (d, *J* = 8.7 Hz, 2H, Ph min), 7.71 (d, *J* = 9.0 Hz, 2H, Ph maj), 7.66 (d, *J* = 9.0 Hz, 2H, Ph min), 7.17 (d, *J* = 9.0 Hz, 2H, Ph min), 7.15 (d, *J* = 8.7 Hz, 2H, Ph maj), 7.11 (d, *J* = 8.7 Hz, 2H, Ph min), 5.66 (dd, *J* = 2.4, 0.9 Hz, 1H, 3min-H), 5.59 (dd, *J* = 2.4, 0.6 Hz, 1H, 3min-H), 5.54 (dd, *J* = 2.4, 0.9 Hz, 1H, 3maj-H), 4.16 (app. d, *J* = 8.4 Hz, 1H, 3bαmin-H), 4.11 (app. d, *J* = 8.1 Hz, 1H, 3bαmaj-H), 3.90 (dd, *J* = 7.5, 1.2 Hz, 1H, 3bαmin-H), 3.50 (dd, *J* = 8.1, 6.3 Hz, 1H, 6aαmaj-H), 3.47 (dd, *J* = 7.5, 5.4 Hz, 1H, 6aαmin-H), 3.44 (dd, *J* = 8.4, 5.1 Hz, 1H, 6aαmin-H), 2.22–2.68 (m, 2H, 6b-H, 10a-H), 2.18 (s, 3H, 2-CH_3 min), 2.12 (s, 3H, 2-CH_3 maj), 2.11 (s, 3H, 2-CH_3 min), 0.92–2.04 (m, 7H, cyclohex.), 0.85 (s, 9H, *t*-Bu min), 0.84 (s, 9H, *t*-Bu maj), 0.67 (s, 9H, *t*-Bu min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.3, 178.2, 177.0, 175.8, 175.7, 175.1, 132.6, 132.4, 132.3, 129.5, 129.4, 128.6, 121.8, 116.8, 104.5, 104.2, 44.9, 34.3–34.5 (overlapped peaks), 33.2, 28.2, 28.0, 27.9, 13.4; IR (thin film, cm^{-1}) 3403(bs), 2923(m), 2353(w), 1770(w), 1713(s), 1492(m), 1390(m), 1163(m); HRMS *m/z* (M + Na^+) calcd 491.1305, found 491.1328. Anal. Calcd for $C_{25}H_{29}BrN_2O_2$: C, 63.97; H, 6.23; N, 5.97. Found: C, 63.94; H, 6.00; N, 5.73.

5-(4-Bromophenyl)-2-methyl-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (60). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **60** (1000 mg, 43%) as a colorless solid, a mixture of four isomers (maj:min:min:min = 2.2:1.0:0.6:0.1): mp 294–296°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.59 (bs, 1H, 1min-H), 10.57 (bs, 1H, 1min-H), 10.38 (bs, 1H, 1maj-H), 10.35 (bs, 1H, 1min-H), 7.64–7.75 (m, 2H, Ph), 7.08–7.35 (m, 7H, Ph), 5.82–5.92 (m, 1H, 3min-H), 5.76–5.88 (m, 1H, 3maj-H), 5.63–5.70 (m, 1H, 3min-H), 5.50–5.55 (d, *J* = 7.2 Hz, 1H, 3min-H), 4.30 (d, *J* = 7.2 Hz, 1H, 3bαmin-H), 4.18 (d, *J* = 8.1 Hz, 1H, 3bαmin-H), 4.01 (d, *J* = 8.1 Hz, 1H, 3bαmaj-H), 3.38–3.54 (m, 1H, 6aα-H), 2.78–2.96 (m, 1H, 10a-H), 2.46–2.58 (m, 1H, 6b-H), 2.18 (s, 3H, 2-CH_3), 1.48–1.98 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 177.1, 132.7, 132.6, 132.34, 132.3, 132.3, 132.2, 129.6, 128.9, 127.6, 126.8, 126.7, 126.0, 121.7, 116.9, 105.0, 45.5, 33.1–33.6 (overlapped peaks), 13.5; IR (thin film, cm^{-1}) 3464(m), 3397(s), 3087(m), 3061(m), 3025(m), 2939(s), 2871(m), 1777(m), 1712(s), 1601(m), 1491(s), 1454(m), 1387(s), 1333(m), 1162(s), 1072(m); HRMS *m/z* (M + Na^+) calcd 511.0992, found 511.1012. Anal. Calcd for $C_{27}H_{25}BrN_2O_2$: C, 66.26; H, 5.15; N, 5.72. Found: C, 66.25; H, 5.17; N, 5.63.

5-(4-Fluorophenyl)-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (61). Method B with **3c** (687 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **61** (760 mg, 45%) as a light-brown solid, a mixture of two isomers (maj:min = 1.8:1.0): mp 266–268°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.53 (d, *J* = 1.8 Hz, 1H, 1maj-H), 10.26 (d,

J = 1.2 Hz, 1H, 1min-H), 7.25–7.42 (m, 4H, Ph), 5.83 (dd, J = 2.1, 0.6 Hz, 1H, 3min-H), 5.60 (dd, J = 2.4, 0.6 Hz, 1H, 3maj-H), 4.15 (dd, J = 8.1, 0.9 Hz, 1H, 3bαmaj-H), 4.01 (dd, J = 8.4 Hz, 1H, 3bαmin-H), 3.40 (dd, J = 8.4, 5.1 Hz, 1H, 6aαmaj-H), 3.35 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 2.99–3.05 (m, 1H, 10aαmin-H), 2.90–2.95 (m, 1H, 10aβmaj-H), 2.06–2.40 (m, 2H, cyclohex., 6b-H), 2.18 (s, 3H, 2-CH_3), 0.98–1.64 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.2, 177.4, 176.3, 163.8, 160.0, 129.7, 129.5, 129.3, 129.0, 128.3, 126.9, 126.6, 119.0, 116.8, 116.7, 116.6, 116.4, 116.3, 108.6, 105.4, 102.8, 46.2, 45.9, 38.7, 38.5, 38.4, 38.2, 33.0, 29.3, 27.6, 26.1, 25.7, 23.2, 22.9, 21.4, 20.8, 13.5, 13.4; IR (thin film, cm^{-1}) 3460(m), 3390(s), 3072(w), 2928(m), 2856(m), 1777(w), 1701(s), 1604(w), 1512(s), 1391(m), 1230(m), 1180(m), 1161(m); HRMS m/z (M + Na^+) calcd 375.1480, found 375.1488. Anal. Calcd for $C_{21}H_{21}FN_2O_2$: C, 71.57; H, 6.01; N, 7.95. Found: C, 71.66; H, 6.28; N, 7.73.

5-(4-Fluorophenyl)-2,8-dimethyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (62). Method B with **3d** (785 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **62** (670 mg, 38%) as a colorless solid, a mixture of three isomers (maj:min:min = 1.6:1.0:0.2): mp 265–267°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.52 (d, J = 1.5 Hz, 1H, 1min-H), 10.27 (d, J = 2.1 Hz, 1H, 1maj-H), 10.24–10.27 (app. bs, 1H, 1min-H), 7.24–7.40 (m, 4H, Ph), 5.82 (dd, J = 2.1, 0.9 Hz, 1H, 3maj-H), 5.60 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H), 5.59 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H), 4.15 (dd, J = 8.1, 1.8 Hz, 1H, 3bαmin-H), 4.01 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 4.00 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.41 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmin-H), 3.37 (dd, J = 8.7 Hz, 1H, 6aαmin-H), 3.36 (dd, J = 8.1, 5.1 Hz, 1H, 6aαmaj-H), 2.93–3.00 (m, 1H, 10aαmaj-H), 2.87–2.92 (m, 1H, 10aβmin-H), 2.30–2.58 (m, 1H, 6b-H), 2.17 (s, 3H, 2-CH_3), 0.85–2.10 (m, 7H, cyclohex.), 0.951 (d, J = 7.2 Hz, 3H, 8-CH_3 maj), 0.949 (d, J = 7.2 Hz, 3H, 8-CH_3 min), 0.72 (d, J = 6.6 Hz, 1H, 8-CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.3, 178.1, 177.3, 176.2, 163.5, 160.3, 129.7, 129.5, 129.4, 129.2, 129.1, 129.0, 128.2, 126.6, 116.9, 116.7, 116.6, 116.4, 116.3, 108.8, 105.3, 103.0, 45.9, 45.6, 38.5, 38.1, 33.1, 32.6, 32.5, 27.0, 26.7, 18.2, 13.5, 13.4; IR (thin film, cm^{-1}) 3462(m), 3390(bs), 3071(w), 2956(m), 2920(m), 2889(m), 2856(m), 1777(w), 1701(s), 1604(m), 1512(s), 1391(m), 1231(m), 1189(m), 1174(m), 1161(m); HRMS m/z (M + Na^+) calcd 389.1637, found 389.1651. Anal. Calcd for $C_{22}H_{23}FN_2O_2$: C, 72.11; H, 6.33; N, 7.64. Found: C, 72.11; H, 6.28; N, 7.48.

8-Ethyl-5-(4-fluorophenyl)-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (63). Method B with **3e** (883 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **63** (800 mg, 44%) as a colorless solid, a mixture of three isomers (maj:min:min = 2.3:1.0:0.6): mp 283–285°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.53 (d, J = 2.1 Hz, 1H, 1maj-H), 10.28 (d, J = 1.5 Hz, 1H, 1min-H), 7.23–7.41 (m, 4H, Ph), 5.82 (dd, J = 1.8, 0.9 Hz, 1H, 3min-H), 5.61 (dd, J = 2.4, 1.2 Hz, 1H, 3maj-H), 5.58 (dd, J = 2.1, 0.9 Hz, 1H, 3min-H), 4.15 (dd, J = 7.8, 1.2 Hz, 1H, 3bαmaj-H), 4.14 (dd, J = 8.1, 0.6 Hz, 1H, 3bαmin-H), 4.00 (dd, J = 8.6, 1.7 Hz, 1H, 3bαmin-H), 3.43 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.39 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmaj-H), 3.35 (dd, J = 8.1, 5.41 Hz, 1H, 6aαmin-H), 2.94–2.99 (m, 1H, 10aαmin-H), 2.90–2.93 (m, 1H, 10amin-H), 2.85–2.91 (m, 1H, 10aβmaj-H), 2.40–2.50 (m, 1H, 6bαmaj-H), 2.25–2.40 (m, 1H, 6bmin-H), 2.18 (s, 3H, 2-CH_3), 0.82–2.18 (m, 9H, cyclohex., CH_2CH_3), 0.80 (t, 3H, CH_2CH_3 min), 0.78 (t, 3H, CH_2CH_3 maj); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.2, 177.4, 176.2, 129.6, 129.5, 129.4, 129.0, 128.2, 126.6, 119.0, 118.7, 116.9, 116.8, 116.7, 116.5, 116.3, 108.8, 103.0, 45.9, 38.2, 38.1, 34.3, 33.9, 33.0, 32.9, 32.8, 32.7, 30.0, 23.6–24.0 (overlapped peaks); IR (thin film, cm^{-1}) 3461(m), 3393(bs), 3071(w), 2959(m), 2925(s), 2866(m), 1779(w), 1702(s), 1512(s), 1391(m), 1231(m), 1186(m), 1161(m); HRMS m/z (M + Na^+) calcd for $C_{23}H_{25}FN_2O_2$: 403.1793, found 403.1809.

5-(4-Fluorophenyl)-8-isopropyl-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (64). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **64** (770 mg, 41%) as a colorless solid, a mixture of three isomers (maj:min:min = 1.9:1.0:0.2): mp 286–288°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.53 (d, J = 2.4 Hz, 1H, 1min-H), 10.28 (d, J = 2.4 Hz, 1H, 1maj-H), 10.24 (d, J = 2.4 Hz, 1H, 1min-H), 7.21–7.43 (m, 4H, Ph), 5.82 (dd, J = 1.8, 0.6 Hz, 1H, 3maj-H), 5.61 (dd, J = 2.4, 0.6 Hz, 1H, 3min-H), 5.58 (dd, J = 1.8, 0.9 Hz, 1H, 3min-H), 4.16 (dd, J = 9.3, 1.2 Hz, 1H, 3bαmin-H), 4.14 (dd, J = 7.8, 0.9 Hz, 1H, 3bαmin-H), 3.99 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.43 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmin-H), 3.39 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.35 (dd, J = 8.4, 5.7 Hz, 1H, 6aαmaj-H), 2.93–3.01 (m, 1H, 10aαmaj-H), 2.86–2.92 (m, 1H, 10aβmin-H), 2.20–2.50 (m, 1H, 6b-H), 2.18 (s, 3H, 2-CH_3), 0.95–2.16 (m, 8H, cyclohex., $CH(CH_3)_2$), 0.85 (d, J = 6.3 Hz, 6H, $CH(CH_3)_2$ maj), 0.84 (d, J = 6.3 Hz, 6H, $CH(CH_3)_2$ min), 0.79 (d, J = 6.3 Hz, 6H, $CH(CH_3)_2$ min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 177.4, 176.2, 129.5, 129.4, 129.2, 129.1, 129.02, 129.0, 128.5, 128.2, 126.5, 118.9, 116.8, 116.7, 116.5, 116.4, 108.8, 105.3, 105.0, 104.5, 102.9, 45.5, 32.7–33.1, 21.8, 21.7, 21.0, 13.6, 13.5; IR (thin film, cm^{-1}) 3402(bs), 2922(m), 1770(w), 1730(s), 1453(m), 1231(m), 1157(m), 1110(m); HRMS m/z (M + Na^+) calcd 417.1950, found 417.1964. Anal. Calcd for $C_{24}H_{27}FN_2O_2$: C, 73.07; H, 6.90; N, 7.10. Found: C, 72.91; H, 6.76; N, 6.90.

8-tert-Butyl-5-(4-fluorophenyl)-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (65). Method B with **3g** (1080 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **65** (670 mg, 34%) as a light-yellow solid, a mixture of four isomers (maj:min:min:min = 1.6:1.0:0.3:0.2): mp 223–225°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.51 (d, J = 1.8 Hz, 1H, 1min-H), 10.37 (d, J = 2.4 Hz, 1H, 1min-H), 10.34 (d, J = 2.4 Hz, 1H, 1maj-H), 10.23 (d, J = 2.7 Hz, 1H, 1min-H), 7.13–7.46 (m, 4H, Ph), 5.81 (dd, J = 2.1, 0.6 Hz, 1H, 3min-H), 5.67 (dd, J = 2.4, 0.9 Hz, 1H, 3maj-H), 5.59 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H), 5.54 (dd, J = 2.4, 0.9 Hz, 1H, 3min-H), 4.16 (dd, J = 1.8 Hz, 9.6 Hz, 1H, 3bαmin-H), 4.11 (app. d, J = 8.7 Hz, 1H, 3bαmin-H), 4.02 (dd, J = 8.4. 1.8 Hz, 1H, 3bαmin-H), 3.90 (dd, J = 7.7, 1.4 Hz, 1H, 3bαmaj-H), 3.50 (dd, J = 8.3, 6.2 Hz, 1H, 6aαmin-H), 3.46 (dd, J = 7.8, 5.7 Hz, 1H, 6aαmaj-H), 3.44 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.38 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 2.96–3.02 (m, 1H, 10amin-H), 2.86–2.92 (m, 1H, 10amin-H), 2.24–2.64 (m, 2H, 6b-H, 10aαmaj-H), 0.94–2.22 (m, 7H, cyclohex.),

2.18 (s, 3H, 2-CH_3 min), 2.13 (2, 3H, 2-CH_3 min), 2.11 (s, 3H, 2-CH_3 maj), 0.86 (s, 9H, *t*-Bu maj), 0.84 (s, 9H, *t*-Bu min), 0.68 (s, 9H, *t*-Bu min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.6, 178.5, 177.2, 176.0, 130.3, 129.7, 129.6, 129.3, 129.25, 129.2, 129.1, 128.5, 126.9, 126.8, 126.6, 122.3, 119.0, 116.8, 116.7, 116.5, 116.4, 116.2, 109.3, 104.2, 104.0, 46.26, 45.3, 44.9, 41.8, 34.4, 33.9, 33.2, 33.0, 32.6, 30.6–30.9 (overlapped peaks), 28.9–29.3 (overlapped peaks), 28.2, 28.0, 27.8, 27.7, 25.9, 13.4 ; IR (thin film, cm^{-1}) 3388(bs), 2921(m), 2864(m), 1774(w), 1713(s), 1512(s), 1391(m), 1231(m), 1160(m); HRMS *m*/*z* (M + Na^+) calcd for $C_{25}H_{29}FN_2O_2$: 431.2106, found 431.2109.

5-(4-Fluorophenyl)-2-methyl-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (66). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **66** (1000 mg, 49%) as a colorless solid, a mixture of four isomers (maj:min:min:min = 8.0:1.0:0.5:0.4): mp 310–312°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.58 (app. bs, 1H, 1min-H), 10.56 (app. bs, 1H, 1min-H), 10.38 (d, *J* = 1.2 Hz, 1H, 1maj-H), 10.35 (app. bs, 1H, 1min-H), 7.15–7.39 (m, 9H, Ph), 5.87 (dd, *J* = 2.7, 0.6 Hz, 1H, 3min-H), 5.77–5.85 (app. m, 1H, 3maj-H), 5.65 (dd, *J* = 1.5, 0.6 Hz, 1H, 3min-H), 5.50–5.52 (app. m, 1H, 3min-H), 4.29 (dd, *J* = 6.9, 0.9 Hz, 1H, 3bαmin-H), 4.18 (dd, *J* = 8.7, 0.6 Hz, 1H, 3bαmin-H), 4.06 (dd, *J* = 8.6, 1.7 Hz, 1H, 3bαmin-H), 4.01 (app. d, *J* = 8.1 Hz, 1H, 3bαmaj-H), 3.34–3.52 (m, 1H, 6aα-H), 3.06–3.12 (m, 1H, 10amin-H), 2.86–2.96 (m, 1H, 10aαmaj-H), 2.80–2.90 (m, 1H, 10aβmin-H), 2.46–2.58 (m, 1H, 6b-H), 2.18 (s, 3H, 2-CH_3), 1.40–2.18 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.6, 177.4, 163.5, 160.5, 129.8, 129.7, 129.3, 129.2, 128.9, 127.6, 126.8, 126.1, 116.6, 116.4, 116.3, 75.0, 45.4, 33.1–33.8 (overlapped peaks), 13.5; IR (thin film, cm^{-1}) 3461(m), 3391(bs), 2935(m), 2871(m), 1775(w), 1701(s), 1603(w), 1512(s), 1391(m), 1228(m), 1191(m), 1165(m); HRMS *m*/*z* (M + Na^+) calcd 451.1793, found 451.1797. Anal. Calcd for $C_{27}H_{25}FN_2O_2$: C, 75.68; H, 5.88; N, 6.54. Found: C, 75.53; H, 5.76; N, 6.40.

2-Methyl-5-phenyl-3b,6a,6b,7,8,9,10,11,11a-nonahydro-1H,5H-cyclohepta[g]pyrrolo[3,4-e]indole-4,6-dione (67). Method B with **3b** (1100 mg, 9.820 mmol), 4-h reflux, and then a diethyl ether wash (10 mL) gave **67** (350 mg, 21%) as a colorless solid, a mixture of two isomers (maj:min = 16.7:1.0): mp 232–233°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.20 (bs, 1H, 1min-H), 7.64 (bs, 1H, 1maj-H), 7.31–7.52 (m, 4H, Ph), 7.24–7.31 (m, 1H, Ph), 6.05 (d, *J* = 1.5 Hz, 1H, 3maj-H), 5.77 (d, *J* = 2.1 Hz, 1H, 3min-H), 3.97 (dd, *J* = 7.8, 1.8 Hz, 1H, 3bα-H), 3.45 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.34 (dd, *J* = 8.0, 5.0 Hz, 1H, 6aαmaj-H), 2.98–3.10 (m, 1H, 11a-H), 2.52–2.63 (m, 1H, 6bα-H), 2.05–2.30 (m, 1H, cyclohept.), 2.27 (s, 3H, 2-CH_3), 1.30–1.92 (m, 9H, cyclohept.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.44 (d, *J* = 1.8 Hz, 1H, 1min-H), 10.34 (d, *J* = 2.4 Hz, 1H, 1maj-H), 7.35–7.51 (m, 4H, Ph), 7.11–7.16 (m, 1H, Ph), 5.67 (dd, *J* = 2.1, 0.9 Hz, 1H, 3maj-H), 5.57 (dd, *J* = 2.4, 0.9 Hz, 1H, 3min-H), 4.09 (dd, *J* = 9.0, 1.8 Hz, 3bαmin-H), 3.87 (dd, *J* = 7.7, 1.7 Hz, 1H, 3bαmaj-H), 3.44 (dd, *J* = 5.1, 4.2 Hz, 1H, 6aαmin-H), 3.42 (dd, *J* = 7.5, 4.8 Hz, 1H, 6aαmaj-H), 2.88–2.96 (m, 1H, 11a-H), 2.34–2.43 (m, 1H, 6bα-H), 2.10–2.23 (m, 1H, cyclohept.), 2.13 (s, 3H, 2-CH_3), 1.17–1.91 (m, 9H, cyclohept.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.4, 176.7, 132.1, 130.2, 129.1, 128.3, 127.9, 126.5, 109.9, 104.7, 45.3, 40.7, 39.5, 36.8, 31.0, 30.4, 28.0, 26.6, 13.3; IR (thin film, cm^{-1}) 3393(bs), 3059(m), 2937(m), 2907(m), 2854(m), 1775(w), 1704(s), 1505(m), 1498(m), 1455(m), 1383(m), 1190(m), 1166(m), 1112(m); HRMS *m*/*z* (M + Na^+) calcd 371.1731, found 371.1734. Anal. Calcd for $C_{22}H_{24}N_2O_2$: C, 75.83; H, 6.94; N, 8.04. Found: C, 75.66; H, 6.93; N, 7.78.

5-(4-Isopropylphenyl)-2-methyl-3b,6a,6b,7,8,9,10,11,11a-nonahydro-1H,5H-cyclohepta[g]pyrrolo[3,4-e]indole-4,6-dione (68). Method B with **3b** (1100 mg, 9.820 mmol), 2-h reflux, and then reprecipitation from diethyl ether (5 mL) gave **68** (50 mg, 3%) as a colorless solid, a mixture of three isomers (maj:min:min = 1.2:1.0:0.05): mp 188–192°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.82 (bs, 1H, 1min-H), 8.20 (bs, 1H, 1min-H), 7.62 (bs, 1H, 1min-H), 7.32 (d, *J* = 8.4 Hz, 2H, Ph min), 7.29 (d, *J* = 8.7 Hz, 2H, Ph maj), 7.18 (d, *J* = 8.4 Hz, 2H, Ph min), 7.17 (d, *J* = 8.7 Hz, 1H, Ph maj), 6.05 (dd, *J* = 2.1, 0.6 Hz, 1H, 3maj-H), 5.92 (dd, *J* = 2.4, 1.2 Hz, 1H, 3min-H), 5.77 (dd, *J* = 2.7, 0.9 Hz, 1H, 3min-H), 4.07 (app. d, *J* = 8.1 Hz, 1H, 3bαmin-H), 4.00 (d, *J* = 8.4 Hz, 1H, 3bαmin-H), 3.96 (dd, *J* = 7.8, 1.8 Hz, 1H, 3bαmaj-H), 3.45 (dd, *J* = 8.6, 5.6 Hz, 1H, 6aαmin-H), 3.33 (dd, *J* = 7.8, 5.1 Hz, 1H, 6aαmaj-H), 2.97–3.09 (m, 1H, 11a-H), 2.93 (septet, *J* = 6.8 Hz, 1H, C*H*$(CH_3)_2$), 2.52–2.63 (m, 1H, 6b-H), 2.35 (s, 3H, 2-CH_3 min), 2.29 (s, 3H, 2-CH_3 min), 2.27 (s, 3H, 2-CH_3 maj), 2.10–2.26 (m, 1H, cyclohept.), 1.65–1.90 (m, 6H, cyclohept.), 1.32–1.63 (m, 3H, cyclohept.), 1.27 (d, *J* = 6.9 Hz, 6H, CH(C*H*$_3)_2$ min), 1.25 (d, *J* = 6.9 Hz, 6H, CH(C*H*$_3)_2$ maj); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.4, 149.1, 127.3, 127.2, 126.2, 104.8, 45.3, 40.6, 36.8, 34.0, 31.0, 30.4, 26.7, 24.0, 13.3; IR (thin film, cm^{-1}) 3395(bs), 3047(m), 2957(m), 2919(m), 2858(m), 2361(w), 1774(w), 1698(s), 1516(m), 1389(m), 1181(m), 1171(m); HRMS *m*/*z* (M + Na^+) calcd for $C_{25}H_{30}N_2O_2$: 413.2200, found 413.2203.

5-(4-Methoxyphenyl)-2-methyl-3b,6a,6b,7,8,9,10,11,11a-nonahydro-1H,5H-cyclohepta[g]pyrrolo[3,4-e]indole-4,6-dione (69). Method B with **3b** (1100 mg, 9.820 mmol), 4-h reflux, removal of solvent under reduced pressure, column chromatography eluting with CH_2Cl_2, and then reprecipitation from diethyl ether (20 mL) gave **69** (400 mg, 11%) as a colorless solid, a mixture of three isomers (maj:min:min = 5.8:1.0:0.3): mp 191–193°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.16 (bs, 1H, 1min-H), 7.61 (bs, 1H, 1maj-H), 7.17 (d, *J* = 9.0 Hz, 2H, Ph), 6.95 (d, *J* = 9.0 Hz, 2H, Ph), 6.05 (d, *J* = 2.4 Hz, 1H, 3maj-H), 5.99 (d, *J* = 2.4 Hz, 1H, 3min-H), 5.77 (d, *J* = 2.1 Hz, 1H, 3min-H), 4.06 (dd, *J* = 7.4, 2.0 Hz, 1H, 3bαmin-H), 3.93 (dd, *J* = 7.8, 1.8 Hz, 1H, 3bαmaj-H), 3.84 (s, 3H, OCH_3 min), 3.82 (s, 3H, OCH_3 maj), 3.81 (s, 3H, OCH_3 min), 3.44 (dd, *J* = 8.7, 5.7, 1H, 6aαmin-H), 3.35 (dd, *J* = 7.7, 3.5 Hz, 1H, 6aαmin-H), 3.33 (dd, *J* = 7.8, 4.8 Hz, 1H, 6aαmaj-H), 3.01–3.09 (m, 1H, 11a-H), 2.53–2.62 (m, 1H, 6b-H), 2.10–2.30 (m, 1H, cyclohept.), 2.29 (s, 3H, 2-CH_3 min), 2.27 (s, 3H, 2-CH_3 maj), 2.24 (s, 3H, 2-CH_3 min), 1.70–1.98 (m, 6H, cyclohept.), 1.32–1.55 (m, 3H, cyclohept.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.5, 177.0, 159.3, 130.2, 127.8, 127.7, 124.8, 114.4, 114.2, 109.9, 105.2, 104.7, 55.6, 48.2, 45.3, 43.2, 42.5, 40.6, 39.6, 37.6, 36.8, 31.9, 31.8, 31.0, 30.9, 30.4, 27.9, 27.5, 26.6, 26.4, 24.8, 13.3; IR (thin film, cm^{-1}) 3379(bs), 2925(m), 2858(m), 1773(w), 1705(s), 1513(s), 1387(m), 1252(m), 1169(m); HRMS *m*/*z* (M + Na^+) calcd 401.1836, found 401.1837. Anal. Calcd for $C_{23}H_{26}N_2O_3$: C, 72.99; H, 6.92; N, 7.40. Found: C, 72.80; H, 6.92; N, 7.41.

2-Methyl-5-(3-nitrophenyl)-3b,6a,6b,7,8,9,10,11,11a-nonahydro-1H,5H-cyclohepta[g]pyrrolo[3,4-e]indole-4,6-dione (70). Method B with **3b** (1100 mg, 9.820 mmol), 1.5 h-reflux, removal of solvent under reduced pressure, column chromatography eluting with CH_2Cl_2, and then reprecipitation from diethyl ether (20 mL) gave **70** (450 mg, 24%) as a colorless solid, a mixture of two isomers (maj:min = 9.0:1.0): mp 169–170°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.21–8.29 (m, 2H, Ph), 8.13 (bs, 1H, 1min-H), 7.60–7.72 (m, 3H, Ph, Ph, 1maj-H), 6.02 (dd, J = 2.4, 0.9 Hz, 1H, 3maj-H), 5.78 (dd, J = 3.0, 0.9 Hz, 1H, 3min-H), 4.07 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 4.02 (dd, J = 7.8, 1.8 Hz, 1H, 3bαmaj-H), 3.49 (dd, J = 8.6, 5.7 Hz, 1H, 6aαmin-H), 3.38 (dd, J = 7.7, 5.0 Hz, 1H, 6aαmaj-H), 3.02–3.10 (m, 1H, 11a-H), 2.54–2.63 (m, 1H, 6bα-H), 2.20–2.38 (m, 1H, cyclohex.), 2.28 (s, 3H, 2-CH_3), 1.57–1.95 (m, 6H, cyclohept.), 1.35–1.58 (m, 3H, cyclohept.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.5, 176.0, 148.4, 133.2, 132.3, 130.4, 129.8, 128.3, 122.9, 121.6, 109.5, 104.5, 45.3, 41.0, 39.2, 36.8, 36.7, 31.1, 31.0, 30.6, 28.5, 28.3, 26.3, 13.3; IR (thin film, cm^{-1}) 3394(bs), 2926(m), 2859(m), 1777(w), 1713(s), 1533(s), 1351(m), 1195(w), 1165(m); HRMS m/z (M + Na^+) calcd 416.1582, found 416.1589. Anal. Calcd for $C_{22}H_{23}N_3O_4$: C, 67.16; H, 5.89; N, 10.68. Found: C, 67.33; H, 5.86; N, 10.80.

5-(4-Chlorophenyl)-2-methyl-3b,6a,6b,7,8,9,10,11,11a-nonahydro-1H,5H-cyclohepta[g]pyrrolo[3,4-e]indole-4,6-dione (71). Method B with **3b** (1100 mg, 9.820 mmol), 4-h reflux, removal of solvent under reduced pressure, column chromatography eluting with CH_2Cl_2, and then reprecipitation from diethyl ether (20 mL) gave **71** (300 mg, 17%) as a colorless solid, a mixture of two isomers (maj:min = 28.0:1.0): mp 219–220°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.15 (bs, 1H, 1min-H), 7.63 (bs, 1H, 1maj-H), 7.41 (d, J = 8.7 Hz, 2H, Ph), 7.23 (d, J = 9.0 Hz, 2H, Ph), 6.03 (dd, J = 2.4, 0.9 Hz, 1H, 3maj-H), 5.77 (dd, J = 2.7, 0.9 Hz, 1H, 3min-H), 4.08 (dd, J = 7.2, 2.1, 1H, 3bαmin-H), 3.96 (dd, J = 7.8, 2.1 Hz, 1H, 3bαmaj-H), 3.45 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.33 (J = 7.7, 5.0 Hz, 1H, 6aαmaj-H), 3.00–3.08 (m, 1H, 11a-H), 2.52–2.61 (m, 1H, 6bα-H), 2.15–2.25 (m, 1H, cyclohept.), 2.27 (s, 3H, 2-CH_3), 1.68–1.92 (m, 6H, cyclohept.), 1.32–1.55 (m, 3H, cyclohept.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.0, 176.4, 134.0, 130.6, 130.2, 129.2, 128.0, 127.7, 109.7, 104.6, 45.3, 40.7, 39.4, 36.8, 31.0, 30.4, 28.1, 27.8, 26.5, 13.3; IR (thin film, cm^{-1}) 3394(bs), 2925(m), 2858(m), 1775(w), 1709(s), 1494(m), 1381(m), 1195(w), 1166(w), 1092(w); HRMS m/z (M + Na^+) calcd 405.1341, found 405.1340. Anal. Calcd for $C_{22}H_{23}ClN_2O_3$: C, 69.01; H, 6.05; N, 7.32. Found: C, 69.21; H, 6.33; N, 7.40.

5-Dimethylamino-2-methyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (72). Method A gave **72** (265 mg, 28%) as a light-brown solid, a mixture of two isomers (maj:min = 8.5:1.0): mp 218–219°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.29 (bs, 1H, 1min-H), 7.68 (bs, 1H, 1maj-H), 6.18 (d, J = 2.7 Hz, 1H, 3maj-H), 5.76 (d, J = 2.4 Hz, 1H, 3min-H), 3.70 (dd, J = 8.6, 1.6 Hz, 1H, 3bα-H), 3.23 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.17 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.04–3.10 (m, 1H, 10aαmaj-H), 2.80–2.92 (m, 1H, 10aβmin-H), 2.92 (s, 6H, $N(CH_3)_2$), 2.43–2.71 (m, 3H, 6bα-H, CH_2CH$_3$), 0.87–2.23 (m, 11H, cyclohex., CH_2CH_3); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 177.6, 176.7, 133.4, 126.8, 108.4, 103.6, 44.0, 43.8, 38.2, 36.8, 32.9, 27.6, 25.7, 23.0, 21.4, 21.0, 14.7; IR (thin film, cm^{-1}) 3371(bs), 2932(m), 2857(m), 2380(w), 1770(w), 1704(s), 1445(m), 1369(m), 1194(m); HRMS m/z (M + Na^+) calcd 338.1840, found 338.1844. Anal. Calcd for $C_{18}H_{25}N_3O_2$: C, 68.54; H, 7.99; N, 13.32. Found: C, 68.36; H, 8.06; N, 13.12.

5-Dimethylamino-2,8-diethyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (73). Method A gave **73** (319 mg, 31%) as a white solid, a mixture of two isomers (maj:min = 4.7:1.0): mp 221–222°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.29 (bs, 1H, 1min-H), 7.68 (bs, 1H, 1maj-H), 6.18 (d, J = 2.4 Hz, 1H, 3maj-H), 5.78 (d, J = 3.0 Hz, 1H, 3min-H), 3.73 (dd, J = 8.6, 2.0 Hz, 1H, 3bαmin-H), 3.70 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.22 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.17 (d, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.01–3.06 (m, 1H, 10aαmaj-H), 2.92–2.96 (m, 1H, 10aβmin-H), 2.93 (s, 6H, $N(CH_3)_2$), 2.56–2.75 (m, 3H, 6bα-H, 2-CH_2CH$_3$), 1.80–1.99 (m, 2H, cyclohex.), 1.07–1.52 (m, 10H, cyclohex., 2-CH_2CH_3, 8-CH_2CH$_3$), 0.85 (t, J = 7.4 Hz, 3H, 8-CH_2CH_3); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 177.6, 177.4, 176.7, 175.6, 135.0, 133.3, 126.7, 116.9, 112.5, 108.5, 105.0, 103.6, 101.2, 43.8, 43.7, 36.8, 33.9, 33.0, 32.6, 21.1, 14.7, 12.5; IR (thin film, cm^{-1}) 3378(bs), 2931(m), 2857(m), 2342(m), 1770(w), 1703(s), 1447(m), 1362(m), 1194(m); HRMS m/z (M + Na^+) calcd 366.2153, found 366.2161. Anal. Calcd for $C_{20}H_{29}N_3O_2$: C, 69.94; H, 8.51; N, 12.23. Found: C, 69.78; H, 8.35; N, 12.08.

8-tert-Butyl-5-dimethylamino-2-ethyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (74). Method A gave **74** (256 mg, 23%) as a light-orange solid, a mixture of two isomers (maj:min = 14.0:1.0): mp 190–191°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 7.61 (bs, 1H, 1maj-H), 6.13 (d, J = 2.7 Hz, 1H, 3min-H), 6.02 (d, J = 2.4 Hz, 1H, 3maj-H), 3.79 (dd, J = 7.8, 1.5 Hz, 1H, 3bα-H), 3.19 (dd, J = 12.3, 8.1 Hz, 1H, 6aαmin-H), 3.10 (dd, J = 5.9, 8.0 Hz, 1H, 6aαmaj-H), 2.87 (s, 6H, $N(CH_3)_2$), 2.52–2.75 (m, 4H, 6bα-H, 10a-H, CH_2CH$_3$), 1.72–2.04 (m, 4H, cyclohex.), 1.51 (ddd, J = 13.5, 10.2, 6.6 Hz, 1H, cyclohex.), 1.11–1.33 (m, 2H, cyclohex.), 1.25 (t, J = 7.5 Hz, 3H, CH_2CH_3), 0.90 (s, 9H, t-Bu); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 177.7, 176.4, 133.4, 129.9, 108.8, 102.4, 43.5, 43.1, 34.3, 34.0, 33.0, 30.6, 28.4, 28.0, 25.6, 21.0, 14.3; IR (thin film, cm^{-1}) 3386(bs), 2961(m), 2359(w), 1774(w), 1712(s), 1448(m), 1365(m), 1203(m), 1148(m); HRMS m/z (M + Na^+) calcd 394.2466, found 394.2473. Anal. Calcd for $C_{22}H_{33}N_3O_2$: C, 71.12; H, 8.95; N, 11.31. Found: C, 71.32; H, 8.75; N, 11.31.

2-Ethyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (75). Method A gave **75** (502 mg, 48%) as a white solid, a mixture of two isomers (maj:min = 1.4:1.0): mp 219–220°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.20 (bs, 1H, 1min-H), 7.83 (bs, 1H, 1maj-H), 7.44–7.54 (m, 3H, Ph), 7.27–7.31 (m, 2H, Ph), 6.14 (d, J = 2.7 Hz, 1H, 3maj-H), 5.82 (d, J = 2.4 Hz, 1H, 3min-H), 4.02 (dd, J = 8.9, 2.0 Hz, 1H, 3bαmin-H), 3.97 (dd, J = 8.6, 2.0 Hz, 1H, 3bαmaj-H), 3.78 (dd, J = 8.6, 5.6 Hz, 1H, 6aαmin-H), 3.40 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.13–3.19 (m, 1H, 10aαmaj-H), 3.03–3.13 (m, 1H, 10aβmin-H), 2.67 (q, J = 7.5 Hz, 2H, 2-CH_2CH$_3$), 2.47–2.57 (m, 1H, 6b-H), 2.17–2.30 (m, 1H, cyclohex.), 1.10–1.83 (m, 10H, cyclohex., 2-CH_2CH_3); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.2, 177.5, 176.3, 135.2, 133.5, 133.0, 132.8, 129.6, 129.5, 128.9, 128.8, 127.5, 127.4, 126.9, 118.7, 116.8, 108.5, 103.8,

101.1, 46.3, 46.0, 33.1, 33.07, 29.3, 27.6, 26.1, 25.7, 23.3, 22.9, 21.5, 21.0, 20.9, 14.8, 14.7; IR (thin film, cm^{-1}) 3394(bs), 2938(m), 2857(m), 2310(w), 1774(w), 1698(s), 1499(m), 1387(m), 1190(m), 1160(m); HRMS *m*/*z* ($M + Na^+$) calcd 371.1731, found 371.1738. Anal. Calcd for $C_{22}H_{24}N_2O_2$: C, 75.83; H, 6.94; N, 8.04. Found: C, 75.92; H, 7.03; N, 8.11.

2,8-Diethyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (76). Method A gave **76** (395 mg, 35%) as a cream-colored solid, a mixture of four isomers (maj:min:min:min = 1.2:1.0:0.2:0.1): mp 243–244°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.38 (bs, 1H, 1maj-H), 7.82 (bs, 1H, 1min-H), 7.42–7.55 (m, 3H, Ph), 7.27–7.30 (m, 2H, Ph), 6.13 (d, *J* = 2.4 Hz, 1H, 3min-H), 5.82 (d, *J* = 2.7 Hz, 1H, 3maj-H), 5.79 (d, *J* = 2.7 Hz, 1H, 3min-H), 4.02 (dd, *J* = 8.7 Hz, 2.1 Hz, 1H, 3bαmaj-H), 3.97 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.51 (dd, *J* = 5.6 Hz, 8.6 Hz, 1H, 6aαmin-H), 3.47 (dd, *J* = 8.7, 5.7 Hz, 1H 6aαmaj-H), 3.43 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.39 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.09–3.16 (m, 1H, 10aαmin-H), 2.98–3.06 (m, 1H, 10aβmaj-H), 2.50–2.72 (m, 1H, 6b-H), 2.67 (q, *J* = 7.5 Hz, 2H, 2-C*H*$_2$CH$_3$), 1.89–2.30 (m, 2H, cyclohex.), 1.19–1.57 (m, 10H, cyclohex., 2-CH$_2$C*H*$_3$, 8-C*H*$_2$CH$_3$), 0.87 (t, *J* = 7.2 Hz, 3H, 8-CH$_2$C*H*$_3$ min), 0.86 (t, *J* = 7.5 Hz, 1H, 8-CH$_2$CH$_3$ maj); IR (thin film, cm^{-1}) 3384(bs), 2953(m), 2923(m), 1773(w), 1694(s), 1497(w), 1456(w), 1447(w), 1389(m), 1192(m); HRMS *m*/*z* ($M + Na^+$) calcd 399.2044, found 399.2059. Anal. Calcd for $C_{24}H_{28}N_2O_2$: C, 76.56; H, 7.50; N, 7.44. Found: C, 76.41; H, 7.73; N, 7.24.

8-tert-Butyl-2-ethyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (77). Method A gave **77** (328 mg, 27%) as a cream-colored solid, a mixture of three isomers (maj:min:min = 4.4:1.0:0.3): mp 209–210°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.20 (bs, 1H, 1min-H), 7.76 (bs, 2H, 1maj-H, 1min-H), 7.37–7.56 (m, 3H, Ph), 7.19–7.29 (m, 2H, Ph), 6.12 (d, *J* = 2.4 Hz, 1H, 3min-H), 5.98 (d, *J* = 2.7 Hz, 1H, 3maj-H), 5.76 (d, *J* = 2.4 Hz, 1H, 3min-H), 4.03 (dd, *J* = 7.8, 1.5 Hz, 1H, 3bαmaj-H), 3.98 (dd, *J* = 8.6, 2.0 Hz, 1H, 3bαmin-H), 3.442 (dd, *J* = 8.3, 5.9 Hz, 1H, 6aαmin-H), 3.437 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.36 (dd, *J* = 7.8, 5.7 Hz, 1H, 6aαmaj-H), 3.10–3.13 (m, 1H, 10aβmin-H), 2.74–2.81 (m, 1H, 6bαmaj-H), 2.52–2.71 (m, 3H, 10aαmaj-H, C*H*$_2$CH$_3$), 1.76–2.28 (m, 3H, cyclohex.), 1.62 (ddd, *J* = 13.7, 11.0, 6.9 Hz, 1H, cyclohex.), 0.84–1.46 (m, 6H, cyclohex., CH$_2$C*H*$_3$), 0.92 (s, 9H, *t*-Bu), 0.75 (s, 9H, *t*-Bu); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 178.46, 177.5, 177.3, 133.7, 133.5, 133.1, 133.0, 130.3, 129.6, 129.4, 128.8, 128.6, 127.5, 127.2, 126.9, 115.5, 109.1, 108.4, 103.8, 102.3, 47.5, 46.0, 45.9, 45.3, 41.8, 34.5, 33.9, 33.0, 32.7, 32.6, 30.6–31.0 (multiple peaks), 28.8–29.3 (multiple peaks), 27.6–28.3 (multiple peaks), 25.8–26.0 (multiple peaks), 22.4, 21.1, 21.0, 14.8, 14.3; IR (thin film, cm^{-1}) 3386(bs), 2961(m), 2923(m), 1771(w), 1708(s), 1496(m), 1372(m), 1314(m), 1176(m), 1163(m); HRMS *m*/*z* ($M + Na^+$) calcd 427.2357, found 427.2340. Anal. Calcd for $C_{26}H_{32}N_2O_2$: C, 77.19; H, 7.97; N, 6.92. Found: C, 77.34; H, 8.23; N, 7.07.

2-Ethyl-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (78). Method A gave **78** (466 mg, 41%) as a cream-colored solid, a mixture of two isomers (maj:min = 5.6:1.0): mp 242–243°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.37 (bs, 1H, 1maj-H), 7.70 (bs, 1H, 1min-H), 7.16–7.28 (m, 2H, Ph), 6.95–7.02 (m, 2H, Ph), 6.24 (d, *J* = 2.7 Hz, 1H, 3min-H), 5.80 (d, *J* = 3.0 Hz, 1H, 3maj-H), 3.97 (dd, *J* = 8.9, 2.0 Hz, 1H, 3bαmaj-H), 3.96 (dd, *J* = 8.6, 2.0 Hz, 1H, 3bαmin-H), 3.843 (s, 3H, OCH$_3$ maj), 3.841 (s, 3H, OCH$_3$ min), 3.46 (dd, *J* = 8.7, 5.7 Hz, 1H, 6aαmaj-H), 3.39 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmin-H), 3.12–3.17 (m, 1H, 10aαmin-H), 3.02–3.07 (m, 1H, 10aβmaj-H), 2.66 (q, *J* = 7.8 Hz, 2H, C*H*$_2$CH$_3$), 2.49–2.59 (m, 1H, 6bα-H), 2.14–2.27 (m, 1H, cyclohex.), 1.20–1.77 (m, 10H, cyclohex., CH$_2$C*H*$_3$); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 176.5, 159.4, 135.1, 133.5, 128.6, 125.4, 118.7, 116.9, 114.8, 114.7, 108.5, 101.1, 55.9, 46.2, 46.0, 33.1, 29.3, 26.1, 22.9, 21.1, 20.9, 14.7; IR (thin film, cm^{-1}) 3399(bs), 2935(m), 1774(w), 1697(s), 1518(m), 1456(m), 1395(m), 1304(m), 1256(m), 1182(m); HRMS *m*/*z* ($M + Na^+$) calcd 401.1836, found 401.1851. Anal. Calcd for $C_{23}H_{26}N_2O_3$: C, 72.99; H, 6.92; N, 7.40. Found: C, 72.78; H, 6.88; N, 7.32.

2,8-Diethyl-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (79). Method A gave **79** (439 mg, 36%) as a cream-colored solid, a mixture of three isomers (maj:min:min = 6.5:1.0:0.3): mp 252–253°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.35 (bs, 1H, 1min-H), 7.73 (bs, 1H, 1maj-H), 7.16–7.23 (m, 2H, Ph), 6.97–7.01 (m, 2H, Ph), 6.21 (d, *J* = 2.7 Hz, 1H, 3maj-H), 5.81 (d, *J* = 2.4 Hz, 1H, 3min-H), 5.78 (d, *J* = 2.4 Hz, 1H, 3min-H), 3.97 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.94 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 3.84 (s, 3H, OCH$_3$), 3.45 (dd, *J* = 8.7, 6.0 Hz, 1H, 6aαmin-H), 3.41 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmin-H), 3.38 (dd, *J* = 8.6, 5.6 Hz, 1H, 6aαmaj-H), 3.05–3.16 (m, 1H, 10aαmaj-H), 2.98–3.05 (m, 1H, 10aβmin-H), 2.59–2.74 (m, 3H, 6b-H, 2-C*H*$_2$CH$_3$), 1.03–2.20 (m, 9H, cyclohex., 8-C*H*$_2$CH$_3$), 1.29 (t, *J* = 7.5 Hz, 3H, 2-CH$_2$C*H*$_3$), 0.86 (t, *J* = 7.5 Hz, 3H, 8-CH$_2$C*H*$_3$); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.6, 177.7, 159.3, 133.4, 128.6, 125.6, 114.9, 114.8, 108.7, 103.8, 103.7, 55.9, 45.5, 34.0, 33.9, 33.1, 32.9, 32.8, 32.75, 32.7, 21.0, 14.7, 12.6; IR (thin film, cm^{-1}) 3383(bs), 2932(m), 2356(w), 1772(w), 1695(s), 1518(m), 1392(m), 1258(m), 1195(m), 1176(m); HRMS *m*/*z* ($M + Na^+$) calcd 429.2149, found 429.2167. Anal. Calcd for $C_{25}H_{30}N_2O_3$: C, 73.86; H, 7.44; N, 6.89. Found: C, 70.74; H, 6.94; N, 6.62.

8-tert-Butyl-2-ethyl-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (80). Method A gave **80** (365 mg, 28%) as a cream-colored solid, a mixture of three isomers (maj:min:min = 2.1:1.0:0.1): mp 207–208°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.10 (bs, 1H, 1min-H), 7.64 (bs, 1H, 1min-H), 7.61 (bs, 1H, 1maj-H), 7.19 (d, *J* = 8.7 Hz, 2H, Ph min), 7.14 (d, *J* = 8.7 Hz, 2H, Ph maj), 7.00 (d, *J* = 9.3 Hz, 2H, Ph min), 6.95 (d, *J* = 8.7 Hz, 2H, Ph maj), 6.20 (d, *J* = 2.4 Hz, 1H, 3min-H), 6.06 (d, *J* = 2.7 Hz, 1H, 3maj-H), 5.77 (d, *J* = 2.4 Hz, 1H, 3min-H), 4.04 (dd, *J* = 7.5, 1.2 Hz, 1H, 3bmaj-H), 3.96 (dd, *J* = 8.6, 2.0 Hz, 1H, 3bmin-H), 3.84 (s, 3H, OCH$_3$ min), 3.82 (s, 3H, OCH$_3$ maj), 3.42 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmin-H), 3.33 (dd, *J* = 7.8, 5.4 Hz, 1H, 6aαmaj-H), 3.05–3.14 (m, 1H, 10aβmin-H), 2.55–2.76 (m, 4H, 6b-H, 10aαmaj-H, C*H*$_2$CH$_3$), 0.91–2.25 (m, 7H, cyclohex.), 1.27 (t, *J* = 7.8 Hz, 3H, CH$_{-2}$C*H*$_3$), 0.91 (s, 9H, *t*-Bu maj), 0.73 (s, 9H, *t*-Bu min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.7, 178.7, 177.7, 177.5, 176.2, 159.4, 159.3, 135.2, 133.6, 133.5, 130.0, 128.7, 128.6, 128.3, 128.2, 126.9, 125.7, 126.6, 126.55, 114.9, 114.8, 114.6, 109.1, 108.4, 103.8, 102.4, 55.9, 47.5, 46.0, 45.1, 44.7, 34.5, 33.9, 33.3, 33.0, 32.7, 32.6, 28.2, 28.0, 27.8, 21.1, 21.0, 14.8, 14.5, 14.3; IR (thin

film, cm^{-1}) 3390(bs), 2963(m), 2935m), 2357(w), 1513, 1770(w), 1705(s), 1640(bm), 1514(s), 1389(m), 1252(m), 1168(m); HRMS *m*/*z* ($M + Na^+$) calcd 457.2462, found 457.2471. Anal. Calcd for $C_{27}H_{34}N_2O_3$: C, 74.62; H, 7.89; N, 6.45. Found: C, 74.73; H, 7.83; N, 6.36.

2-Benzyl-5-(dimethylamino)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (81). Method A gave **81** (396 mg, 35%) as a light-brown solid, a mixture of two isomers (maj:min = 3.2:1.0): mp 238–239°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.22 (bs, 1H, 1min-H), 7.57 (bs, 1H, 1maj-H), 7.29–7.36 (m, 2H, Ph), 7.21–7.28 (m, 3H, Ph), 6.25 (d, *J* = 2.7 Hz, 1H, 3maj-H), 5.79 (d, *J* = 2.7 Hz, 1H, 3min-H), 4.04 (AA′d, *J* = 16.2 Hz, 1H, Bn maj), 4.02 (AA′d, *J* = 15.9 Hz, 1H, Bn min), 3.95 (AA′d, *J* = 16.2 Hz, 1H, Bn maj), 3.94 (AA′d, *J* = 16.2 Hz, 1H, Bn min), 3.69 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bα-H), 3.21 (dd, *J* = 8.6, 5.4 Hz, 1H, 6aαmin-H), 3.16 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmaj-H), 3.01–3.05 (m, 1H, 10a-H), 2.93 (s, 6H, $N(CH_3)_2$), 2.42–2.52 (m, 1H, 6b-H), 1.99–2.22 (m, 1H, cyclohex.), 0.99–1.74 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.6, 177.4, 176.7, 175.6, 141.53, 141.47, 132.3, 130.4, 129.1, 128.7, 127.4, 126.3, 119.0, 117.4, 108.9, 105.7, 102.9, 44.3, 44.0, 43.8, 38.2, 36.7, 36.3, 34.0, 33.0, 29.2, 27.6, 26.0, 25.7, 23.1, 22.7, 21.4, 20.8; IR (thin film, cm^{-1}) 3450(bs), 2923(m), 2100(bw), 1770(w), 1703(s), 1648(bs), 1442(m), 1366(m), 1194(m), 1148(m); HRMS *m*/*z* ($M + Na^+$) calcd for $C_{23}H_{27}N_3O_2$: 400.1996, found 400.1992.

2-Benzyl-5-dimethylamino-8-ethyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (82). Method A gave **82** (353 mg, 29%) as a cream-colored solid, a mixture of three isomers (maj:min:min = 5.3:1.0:0.3): mp 239–240°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 7.54 (bs, 1H, 1min-H), 7.35 (bs, 2H, 1maj-H, 1min-H), 7.23–7.36 (m, 5H, Ph), 6.25 (d, *J* = 2.7 Hz, 1H, 3maj-H), 5.81 (d, *J* = 2.7 Hz, 1H, 3min-H), 5.77 (d, *J* = 2.7 Hz, 1H, 3min-H), 4.04 (AA′d, *J* = 16.2 Hz, 1H, Bn maj), 4.03 (AA′d, *J* = 15.9 Hz, 1H, Bn min), 3.95 (AA′d, *J* = 15.9 Hz, 1H, Bn maj), 3.94 (AA′d, *J* = 15.9 Hz, 1H, Bn min), 3.69 (dd, *J* = 8.4, 2.1 Hz, 1H, 3bα-H), 3.24 (dd, *J* = 6.0 Hz, 1H, 6aαmin-H), 3.20 (dd, *J* = 5.9 Hz, 1H, 6aαmin-H), 3.20 (dd, *J* = 5.4 Hz, 1H, 6aαmin-H), 3.16 (dd, *J* = 5.3 Hz, 1H, 6aαmaj-H), 2.86–3.01 (m, 7H, 10a-H, $N(CH_3)_2$), 2.58–2.67 (m, 1H, 6bαmaj-H), 2.46–2.55 (m, 1H, 6bmin-H), 0.98–2.08 (m, 9H, cyclohex., CH_2CH_3), 0.85 (t, *J* = 7.2 Hz, 3H, CH_2CH_3 maj), 0.77 (t, *J* = 7.2 Hz, 3H, CH_2CH_3 min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.4, 177.3, 176.6, 139.6, 130.4, 128.8, 128.7, 128.7, 127.8, 127.6, 126.5, 120.0, 117.6, 117.5, 109.2, 44.1, 43.9, 39.3, 38.9, 38.5, 38.2, 37.0, 36.0, 34.5, 34.45, 34.37, 33.9, 32.9, 32.8, 32.7, 29.7, 29.6, 29.2, 29.0, 27.7, 27.4, 27.0, 26.1, 24.3, 23.6, 12.2, 11.4; IR (thin film, cm^{-1}) 3452(bs), 2923(m), 2122(bw), 1770(w), 1703(s), 1645(bs), 1446(m), 1367(m), 1190(m), 1151(m); HRMS *m*/*z* ($M + Na^+$) calcd for $C_{25}H_{31}N_3O_2$: 428.2309, found 428.2327.

2-Benzyl-8-tert-butyl-5-(dimethylamino)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,3bH-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (83). Method A gave **83** (325 mg, 25%) as cream-colored crystals, a single isomer: mp 195–196°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 7.46 (bs, 1H, 1-H), 7.20–7.35 (m, 5H, Ph), 6.11 (d, *J* = 2.7 Hz, 1H, 3-H), 3.98 (AA′d, *J* = 16.2 Hz, 1H, Bn), 3.90 (AA′d, *J* = 16.2 Hz, 1H, Bn), 3.78 (dd, *J* = 8.1, 1.5 Hz, 1H, 3bα-H), 3.08 (dd, *J* = 6.0, 8.1 Hz, 1H, 6aα-H), 2.87 (s, 6H, $N(CH_3)_2$), 2.63–2.70 (m, 1H, 10aα-H), 2.51–2.58 (m, 1H, 6bα-H), 1.66–2.08 (m, 4H, cyclohex.), 1.50 (ddd, *J* = 13.7, 10.4, 6.8 Hz, 1H, cyclohex.), 1.04–1.30 (m, 2H, cyclohex.), 0.89 (s, 9H, *t*-Bu); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.1, 176.5, 139.5, 130.7, 130.6, 128.8, 128.7, 126.5, 109.3, 105.5, 43.8, 43.3, 40.8, 39.3, 34.3, 34.1, 33.9, 32.9, 30.1, 27.7, 24.8; IR (thin film, cm^{-1}) 3388(bs), 2957(m), 2108(bw), 1774(w), 1709(s), 1604(bs), 1448(m), 1364(m), 1202(m), 1146(m); HRMS *m*/*z* ($M + Na^+$) calcd for $C_{27}H_{35}N_3O_2$: 456.2622, found 456.2631.

2-Benzyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (84). Method A gave **84** (690 mg, 56%) as a cream-colored solid, a mixture of two isomers (maj:min = 3.6:1.0): mp 252–253°C; ^{1}H NMR (300 MHz, DMSO-*d*6, δ) 10.70 (bs, 1H, 1min-H), 10.41 (bs, 1H, 1maj-H), 7.38–7.53 (m, 4H, Ph), 7.15–7.32 (m, 6H, Ph), 5.85 (d, *J* = 2.4 Hz, 1H, 3maj-H), 5.60 (d, *J* = 2.4 Hz, 1H, 3min-H), 4.19 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmin-H), 4.03 (dd, *J* = 8.6, 1.7 Hz, 1H, 3bαmaj-H), 3.90 (AA′d, *J* = 15.9 Hz, 1H, Bn), 3.85 (AA′d, *J* = 16.5 Hz, 1H, Bn), 3.40 (dd, *J* = 8.1, 4.8 Hz, 1H, 6aαmin-H), 3.35 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmaj-H), 3.02–3.08 (m, 1H, 10aαmaj-H), 2.91–2.96 (m, 1H, 10aβmin-H), 2.07–2.42 (m, 2H, 6b-H, cyclohex.), 1.04–1.62 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.2, 177.4, 176.3, 141.5, 133.0, 132.8, 132.4, 130.5, 129.6, 129.5, 129.1, 128.9, 128.7, 127.5, 127.48, 127.4, 126.3, 119.2, 117.4, 108.9, 105.8, 103.0, 46.2, 45.9, 38.7, 38.5, 38.3, 38.2, 34.1, 34.0, 33.1, 29.3, 27.6, 26.1, 25.7, 23.3, 22.9, 21.4, 20.9; IR (thin film, cm^{-1}) 3390(bs), 2924(m), 2853(m), 2110(bw), 1772(w), 1697(s), 1651(bs), 1496(w), 1455(w), 1444(w), 1382(m), 1187(m), 1157(m), 1004(m); HRMS *m*/*z* ($M + Na^+$) calcd 433.1887, found 433.1901. Anal. Calcd for $C_{27}H_{26}N_2O_2$: C, 79.00; H, 6.38; N, 6.82. Found: C, 79.03; H, 6.30; N, 6.87.

2-Benzyl-8-ethyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (85). Method A gave **85** (474 mg, 36%) as a light-orange solid, a mixture of four isomers (maj:min:min:min = 1.7:1.0:0.6:0.4): mp 225–226°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.60 (bs, 1H, 1maj-H), 7.61 (bs, 1H, 1min-H), 7.59 (bs, 1H, 1min-H), 7.23–7.52 (m, 10H, Ph), 6.30 (d, *J* = 2.4 Hz, 1H, 3min-H), 5.85 (d, *J* = 3.0 Hz, 1H, 3maj-H), 5.82 (d, *J* = 2.7 Hz, 1H, 3min-H), 4.05 (AA′d, *J* = 15.9 Hz, 1H, Bn min), 4.03 (AA′d, *J* = 15.9 Hz, 1H, Bn maj), 3.97 (dd, *J* = 10.5, 1.8 Hz, 1H, 3bα-H), 3.95 (AA′d, *J* = 16.2 Hz, 2H, Bn min, Bn maj), 3.48 (dd, *J* = 9.3, 5.7 Hz, 1H, 6aαmin-H), 3.45 (dd, *J* = 8.7, 5.7 Hz, 1H, 6aαmaj-H), 3.42 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmin-H), 3.39 (dd, *J* = 8.6, 5.3 Hz, 1H, 6aαmin-H), 3.08–3.13 (m, 1H, 10amin-H), 3.02–3.07 (m, 1H, 10aαmin-H), 2.98–3.03 (m, 1H, 10aβmaj-H), 2.66–2.75 (m, 1H, 6bαmaj-H), 2.53–2.62 (m, 1H, 6bmin-H), 1.00–2.30 (m, 7H, cyclohex.), 1.44 (app. q, *J* = 7.5 Hz, 2H, CH_2CH_3), 0.86 (t, *J* = 7.5 Hz, CH_2CH_3 maj), 0.80 (t, *J* = 7.5 Hz, 3H, CH_2CH_3 min); ^{13}C NMR (75 MHz, $CDCl_3$, δ); IR (thin film, cm^{-1}) 3422(bs), 2929(m), 2863(m), 2100(bw), 1777(w), 1694(s), 1651(bs), 1500(m), 1454(m), 1388(m), 1188(m), 1166(m); HRMS *m*/*z* ($M + Na^+$) calcd 461.2200, found 461.2205. Anal. Calcd for $C_{29}H_{30}N_2O_2$: C, 79.42; H, 6.89; N, 6.39. Found: C, 79.19; H, 7.02; N, 6.40.

2-Benzyl-8-isopropyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (86). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **86** (1325 mg, 61%) as a light-pink solid, a mixture of three

isomers (maj:min:min = 4.1:1.0:0.7): mp 246–247°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.72 (d, J = 2.4 Hz, 1H, 1maj-H), 10.70 (d, J = 2.4 Hz, 1H, 1min-H), 10.45 (d, J = 1.2 Hz, 1H, 1min-H), 7.40–7.57 (m, 3H, Ph), 7.14–7.33 (m, 7H, Ph), 5.83 (d, J = 2.7 Hz, 1H, 3min-H), 5.62 (d, J = 2.4 Hz, 1H, 3maj-H), 5.60 (d, J = 2.4 Hz, 1H, 3min-H), 4.20 (dd, J = 7.8, 1.2 Hz, 1H, 3bαmin-H), 4.18 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 4.02 (dd, J = 8.7, 1.2 Hz, 1H, 3bαmin-H), 3.90 (AA′d, J = 15.9 Hz, 1H, Bn), 3.85 (AA′d. J = 15.9 Hz, 1H, Bn), 3.44 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.39 (dd, J = 8.1, 5.4 Hz, 1H, 6aαmaj-H), 3.35 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 2.96–3.02 (m, 1H, 10aαmin-H), 2.86–2.93 (m, 1H, 10aβmaj-H), 2.40–2.50 (m, 1H, 6bαmaj-H), 2.28–2.38 (m, 1H, 6bmin-H), 0.94–2.18 (m, 8H, cyclohex., CH(CH$_3$)$_2$), 0.84 (d, J = 6.3 Hz, 6H, CH(CH_3)$_2$ maj), 0.77 (d, J = 6.3 Hz, 6H, CH(CH_3)$_2$ min), 0.70 (d, J = 6.6 Hz, 6H, CH(CH_3)$_2$ min); ^{13}C NMR (75 MHz, CDCl$_3$, δ) 178.0, 176.1, 139.6, 132.1, 131.9, 129.4, 129.3, 129.2, 128.8, 128.7, 128.5, 126.5, 126.44, 126.41, 117.7, 117.6, 106.7, 104.3, 104.2, 46.0, 45.6, 43.9, 40.3, 38.9, 37.9, 34.6, 34.5, 34.4, 33.3, 33.2, 33.0, 32.94, 32.88, 32.8, 29.0, 26.3, 24.0, 21.4, 21.0, 20.0, 19.9; IR (KBr, cm^{-1}) 3462(w), 3381(bs), 3061(w), 3029(w), 2928(w), 2864(m), 2359(w), 1777(w), 1699(s), 1598(w), 1498(m), 1453(m), 1387(m), 1173(m); HRMS m/z (M + Na$^+$) calcd 475.2357, found 475.2372. Anal. Calcd for $C_{30}H_{32}N_2O_2$: C, 79.61; H, 7.13; N, 6.19. Found: C, 79.80; H, 7.24; N, 6.33.

2-Benzyl-8-tert-butyl-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (87). Method A gave **87** (546 mg, 39%) as a cream-colored solid, a mixture of four isomers (maj:min:min:min = 3.0:1.0:0.5:0.3): mp 184–185°C; ^{1}H NMR (300 MHz, CDCl$_3$, δ) 8.26 (bs, 1H, 1min-H), 8.04 (bs, 1H, 1min-H), 7.55 (bs, 1H, 1maj-H), 7.22–7.51 (m, 10H, Ph), 6.28 (d, J = 2.4 Hz, 1H, 3maj-H), 6.06 (d, J = 2.4 Hz, 1H, 3min-H), 5.85 (d, J = 2.4 Hz, 1H, 3min-H), 1.59 (d, J = 2.7 Hz, 1H, 3min-H), 4.05 (dd, J = 7.8, 1.2 Hz, 1H, 3bα-H), 3.99 (AA′d, J = 15.9 Hz, 1H, Bn maj), 3.91 (AA′d, J = 15.9 Hz, 1H, Bn maj), 3.41 (dd, J = 8.1, 5.7 Hz, 1H, 6aαmin-H), 3.34 (dd, J = 7.4, 5.3 Hz, 1H, 6aαmaj-H), 2.60–2.73 (m, 2H, 6b-H, 10a-H), 2.15–2.22 (m, 1H, cyclohex.), 2.01–2.07 (m, 1H, cyclohex.), 1.73–1.88 (m, 2H, cyclohex.), 1.59 (ddd, J = 13.8, 11.3, 6.9 Hz, 1H, cyclohex.), 1.29–1.40 (m, 1H, cyclohex.), 1.06–1.19 (m, 1H, cyclohex.), 0.91 (s, 9H, t-Bu); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 177.3, 141.1, 133.2, 131.0, 130.9, 129.4, 129.1, 128.8, 128.6, 127.5, 126.4, 109.4, 104.4, 45.3, 41.7, 34.4, 34.2, 33.9, 33.0, 30.4–30.8 (multiple peaks), 28.8–29.1 (multiple peaks), 28.0, 25.6–26.0 (multiple peaks); IR (thin film, cm^{-1}) 3386(bs), 2951(m), 2866(m), 2126(bw), 1774(w), 1708(s), 1648(bs), 1500(m), 1400(m), 1371(m), 1200(m), 1176(m); HRMS m/z (M + Na$^+$) calcd 489.2513, found 489.2517. Anal. Calcd for $C_{31}H_{34}N_2O_2$: C, 79.79; H, 7.34; N, 6.00. Found: C, 79.69; H, 7.20; N, 6.01.

2-Benzyl-5,8-diphenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (88). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **88** (1471 mg, 63%) as a dark-red solid, a mixture of two isomers (maj:min = 2.8:1.0): mp 222–224°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.73–10.77 (app. m, 1H, 1maj-H), 10.54 (d, J = 1.5 Hz, 1H, 1min-H), 7.10–7.56 (m, 15H, Ph), 5.81–5.85 (app. m, 1H, 3min-H), 5.65 (d, J = 2.1 Hz, 1H, 3maj-H), 4.22 (dd, J = 8.4, 1.2 Hz, 1H, 3bαmaj-H), 4.03 (app. d, J = 7.5 Hz, 1H, 3bαmin-H), 3.90 (s, 2H, Bn), 3.40–3.58 (m, 1H, 6aα-H), 2.80–3.20 (m, 2H, 6bα-H, 10a-H), 1.40–2.60 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 176.2, 176.1, 141.4, 132.9, 132.6, 129.7, 129.5, 129.3, 129.0, 128.9, 128.8, 128.76, 128.7, 127.6, 127.4, 127.24, 127.20, 126.3, 126.0, 34.1, 33.3–33.6 (overlapped peaks); IR (KBr, cm^{-1}) 3379(bs), 3058(w), 3026(w), 2928(s), 2858(m), 2359(w), 2334(w), 1776(w), 1709(s), 1598(w), 1496(m), 1452(w), 1383(m), 1185(m), 1155(m); HRMS m/z (M + Na$^+$) calcd 509.2200, found 509.2210. Anal. Calcd for $C_{33}H_{30}N_2O_2$: C, 81.45; H, 6.21; N, 5.76. Found: C, 81.23; H, 5.99; N, 5.47.

2-Benzyl-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (89). Method A gave **89** (780 mg, 59%) as a white solid, a mixture of three isomers (maj:min:min = 3.0:1.0:0.3): mp 247–248°C; ^{1}H NMR (300 MHz, DMSO-d6, δ) 10.67 (bs, 1H, 1min-H), 10.40 (bs, 1H, 1maj-H), 10.30 (bs, 1H, 1min-H), 7.10–7.31 (m, 7H, Ph), 6.96–7.05 (m, 2H, Ph), 5.84 (d, J = 2.4 Hz, 1H, 3maj-H), 5.67 (d, J = 2.1 Hz, 1H, 3min-H), 5.60 (d, J = 2.4 Hz, 1H, 3min-H), 4.15 (app. d, J = 7.5 Hz, 1H, 3bαmin-H), 3.99 (d, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.87 (s, 2H, Bn), 3.78 (s, 3H, OCH$_3$), 3.42 (dd, J = 7.4, 4.1 Hz, 1H, 6aαmin-H), 3.37 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.32 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.02–3.08 (m, 1H, 10aαmaj-H), 2.93–2.98 (m, 1H, 10aβmin-H), 2.20–2.42 (m, 2H, cyclohex., 6b-H), 1.02–1.85 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.6, 178.4, 177.6, 176.5, 132.4, 130.5, 129.1, 128.9, 128.7, 128.66, 128.64, 127.5, 126.3, 125.6, 125.4, 119.1, 117.5, 114.8, 114.7, 109.0, 105.8, 103.0, 55.9, 46.2, 45.8, 34.1, 34.0, 33.1, 29.3, 27.6, 26.1, 25.7, 23.3, 22.9, 21.5, 20.9; IR (thin film, cm^{-1}) 3446(bs), 2928(m), 2861(w), 2113(bw), 1770(w), 1697(s), 1646(bs), 1515(m), 1391(m), 1256(m), 1193(m), 1170(m), 1160(m); HRMS m/z (M + Na$^+$) calcd 463.1993, found 463.2008. Anal. Calcd for $C_{28}H_{28}N_2O_3$: C, 76.34; H, 6.41; N, 6.36. Found: C, 76.26; H, 6.59; N, 6.35.

2-Benzyl-8-ethyl-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (90). Method A gave **90** (506 mg, 36%) as a light-pink solid, a mixture of four isomers (maj:min:min:min = 2.5:1.0:0.3:0.2): mp 231–232°C; ^{1}H NMR (300 MHz, CDCl$_3$, δ) 8.25 (bs, 1H, 1min-H), 7.59 (bs, 1H, 1maj-H), 7.14–7.36 (m, 7H, Ph), 6.96–7.01 (m, 2H, Ph), 6.29 (d, J = 2.7 Hz, 1H, 3maj-H), 5.85 (d, J = 2.4 Hz, 1H, 3min-H), 5.81 (d, J = 2.4 Hz, 1H, 3min-H), 4.05 (AA′d, J = 16.2 Hz, 1H, Bn maj), 4.03 (AA′d, J = 16.2 Hz, 1H, Bn min), 3.96 (AA′d, J = 15.9 Hz, 1H, Bn maj), 3.954 (AA′d, J = 15.6 Hz, 1H, Bn min), 3.951 (dd, J = 8.4, 1.8 Hz, 1H, 3bα-H), 3.84 (s, 3H, OCH$_3$), 3.46 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.43 (dd, J = 8.6, 5.9 Hz, 1H, 6aαmin-H), 3.41 (dd, J = 8.6, 5.6 Hz, 1H, 6aαmin-H), 3.37 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.07–3.12 (m, 1H, 10amin-H), 3.02–3.08 (m, 1H, 10aαmaj-H), 2.96–3.02 (m, 1H, 10aβmin-H), 2.64–2.73 (m, 1H, 6bαmaj-H), 2.52–2.61 (m, 1H, 6bmin-H), 1.04–2.28 (m, 9H, cyclohex., CH_2CH$_3$), 0.85 (t, J = 7.4 Hz, 3H, CH$_2$CH_3 maj), 0.79 (t, J = 7.2 Hz, 3H, CH$_2$CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 179.0, 178.6, 178.4, 177.64, 177.6, 176.5, 159.5, 159.4, 159.3, 141.5, 132.3, 130.5, 129.1, 128.9, 128.8, 128.7, 128.6, 127.5, 127.4, 126.3, 125.6, 115.0, 114.9, 114.8, 109.2, 105.9, 105.7, 55.9, 45.5, 34.3, 34.0, 33.9, 33.3, 33.2, 33.1, 33.0, 32.9, 32.8, 32.7, 32.6, 32.0, 28.0, 27.4, 27.36, 23.6, 12.6; IR (thin film, cm^{-1}) 3444(bs), 2930(m), 2100(bw), 1694(s), 1648(bm), 1515(m),

1389(m), 1252(m), 1172(m); HRMS *m/z* (M + Na^+) calcd 491.2306, found 491.2299. Anal. Calcd for $C_{30}H_{32}N_2O_3$: C, 76.90; H, 6.88; N, 5.98. Found: C, 77.09; H, 6.76; N, 5.79.

2-Benzyl-8-isopropyl-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (91). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **91** (1459 mg, 63%) as a light-pink solid, a mixture of three isomers (maj:min:min = 3.8:1.0:0.8): mp 254–256°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.84 (d, J = 2.4 Hz, 1H, 1maj-H), 10.68 (d, J = 3.0 Hz, 1H, 1min-H), 10.44 (d, J = 3.0 Hz, 1H, 1min-H), 7.02–7.31 (m, 9H, Ph), 5.82 (d, J = 2.1 Hz, 1H, 3min-H), 5.61 (d, J = 2.4 Hz, 1H, 3maj-H), 5.60 (d, J = 2.4 Hz, 1H, 3min-H), 4.16 (dd, J = 8.7, 1.5 Hz, 1H, 3bαmin-H), 4.14 (dd, J = 8.6, 1.7 Hz, 1H, 3bαmaj-H), 3.98 (dd, J = 8.7, 1.8 Hz, 1H, 3bαmin-H), 3.87 (s, 2H, Bn), 3.79 (s, 3H, OCH_3 maj), 3.78 (s, 3H, OCH_3 min), 3.41 (dd, J = 9.0, 6.0 Hz, 1H, 6aαmin-H), 3.36 (dd, J = 8.1, 5.1 Hz, 1H, 6aαmaj-H), 3.32 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 2.96–3.02 (m, 1H, 10aαmin-H), 2.86–2.92 (m, 1H, 10aβmaj-H), 2.38–2.49 (m, 1H, 6bαmaj-H), 2.26–2.36 (m, 1H, 6bmin-H), 1.00–2.18 (m, 8H, cyclohex., C*H*$(CH_3)_2$), 0.84 (d, J = 6.6 Hz, 6H, CH(C*H*$_3)_2$ maj), 0.77 (d, J = 6.6 Hz, 1H, CH(C*H*$_3)_2$ min), 0.70 (d, J = 6.9 Hz, 1H, CH(C*H*$_3)_2$ min); IR (KBr, cm^{-1}) 3459(w), 3372(bs), 3060(w), 3029(w), 2932(s), 2864(m), 2361(w), 1776(w), 1698(s), 1611(w), 1593(w), 1514(s), 1453(m), 1390(m), 1305(m), 1256(m), 1169(s), 1107(w), 1032(w); HRMS *m/z* (M + Na^+) calcd for $C_{31}H_{34}N_2O_3$: 505.2462, found 505.2476.

2-Benzyl-8-tert-butyl-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (92). Method A gave **92** (358 mg, 24%) as a light-orange solid, a mixture of four isomers (maj:min:min:min = 24.0:1.0:0.3:0.2): mp 179–180°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.25 (bs, 1H, 1min-H), 8.05 (bs, 1H, 1min-H), 7.55 (bs, 1H, 1min-H), 7.49 (bs, 1H, 1maj-H), 7.10–7.36 (m, 7H, Ph), 6.91–7.02 (m, 2H, Ph), 6.13 (d, J = 2.7 Hz, 1H, 3maj-H), 6.10 (d, J = 2.7 Hz, 1H, 3min-H), 5.83 (d, J = 2.7 Hz, 1H, 3min-H), 4.03 (dd, J = 8.0, 1.7 Hz, 1H, 3bα-H), 3.99 (AA′d, J = 16.2 Hz, 1H, Bn), 3.90 (AA′d, J = 16.2 Hz, 1H, Bn), 3.82 (s, 3H, OCH_3), 3.39 (dd, J = 8.1, 5.7 Hz, 1H, 6aαmin-H), 3.32 (dd, J = 7.8, 5.4 Hz, 1H, 6aαmaj-H), 3.03–3.07 (m, 1H, 10amin-H), 2.98–3.02 (m, 1H, 10amin-H), 2.59–2.72 (m, 3H, 6b-H, 10aαmaj-H, 10aβmin-H), 1.00–2.22 (m, 7H, cyclohex.), 0.89 (s, 9H, *t*-Bu maj), 0.74 (s, 9H, *t*-Bu min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.7, 177.5, 159.3, 141.0, 130.9, 129.1, 128.8, 128.7, 126.4, 125.7, 114.6, 109.4, 105.0, 104.3, 55.9, 45.1, 34.2, 33.9, 33.0, 28.0, 28.0; IR (thin film, cm^{-1}) 3387(bs), 2958(m), 2100(bw), 1776(w), 1705(s), 1645(bm), 1513(s), 1391(m), 1301(m), 1252(m), 1168(m); HRMS *m/z* (M + Na^+) calcd 519.2619, found 519.2620. Anal. Calcd for $C_{32}H_{36}N_2O_3$: C, 77.39; H, 7.31; N, 5.64. Found: C, 77.56; H, 7.46; N, 5.57.

2-Benzyl-5-(4-methoxyphenyl)-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (93). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **93** (1413 mg, 57%) as a pink solid, a mixture of three isomers (maj:min:min = 3.2:1.0:0.5): mp 235–237°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.74 (app. bs, 1H, 1maj-H), 10.53 (d, J = 2.1 Hz, 1H, 1min-H), 6.97–7.34 (m, 14 H, Ph), 5.81–5.85 (app. m, 1H, 3min-H), 5.67 (d, J = 2.7 Hz, 1H, 3min-H), 5.64 (d, J = 1.8 Hz, 1H, 3min-H), 4.21 (dd, J = 8.7, 1.5 Hz, 1H, 3bαmin-H), 4.19 (dd, J = 8.4, 1.2 Hz, 1H, 3bαmaj-H), 4.00 (app. d, J = 8.1 Hz, 1H, 3bαmin-H), 3.90 (s, 2H, Bn), 3.80 (s, 3H, OCH_3 maj), 3.79 (s, 3H, OCH_3 min), 3.75 (s, 3H, OCH_3 min), 3.48 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.37–3.46 (m, 2H, 6aαmaj-H, 6aαmin-H), 2.80–3.10 (m, 2H, 6b-H, 10a-H), 1.40–2.00 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.8, 178.6, 176.4, 159.5, 159.3, 141.4, 132.5, 129.1, 129.0, 128.8, 128.7, 127.6, 126.7, 126.3, 125.9, 125.4, 117.7, 114.9, 114.8, 55.9, 34.1, 33.2–33.6 (overlapped peaks); IR (KBr, cm^{-1}) 3452(w), 3380(bs), 3083(w), 3059(w), 3026(w), 2930(s), 2859(m), 2263(w), 1775(w), 1701(s), 1601(w), 1514(s), 1451(m), 1389(m), 1302(w), 1254(m), 1170(m), 1106(w), 1301(w); HRMS *m/z* (M + Na^+) calcd 539.2306, found 539.2310. Anal. Calcd for $C_{34}H_{32}N_2O_3$: C, 79.04; H, 6.24; N, 5.42. Found: C, 79.20; H, 6.10; N, 5.27.

2-(4-Methylbenzyl)-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (94). Method B with **3c** (687 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **94** (1304 mg, 64%) as a pink solid, a mixture of two isomers (maj:min = 1.6:1.0): mp 223–225°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.67 (d, J = 1.8 Hz, 1H, 1maj-H), 10.39 (d, J = 1.8 Hz, 1H, 1min-H), 7.38–7.54 (m, 3H, Ph), 7.19–7.27 (m, 2H, Ph), 7.07–7.16 (m, 4H, Ph), 5.81 (d, J = 2.1 Hz, 1H, 3min-H), 5.56 (d, J = 2.4 Hz, 1H, 3maj-H), 4.19 (dd, J = 8.3, 1.7 Hz, 1H, 3bαmaj-H), 4.02 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.87 (s, 2H, Bn), 3.39 (dd, J = 8.4, 5.1 Hz, 1H, 6aαmaj-H), 3.35 (dd, J = 8.4, 5.1 Hz, 1H, 6aαmin-H), 3.02–3.07 (m, 1H, 10aαmin-H), 2.90–2.95 (m, 1H, 10aβmaj-H), 2.51 (s, 3H, PhC*H*$_3$), 2.03–2.44 (m, 2H, cyclohex., 6b-H), 1.00–1.64 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.2, 177.4, 176.3, 138.4, 138.3, 135.2, 133.0, 132.8, 132.7, 130.8, 129.6, 129.5, 129.3, 129.0, 128.9, 128.8, 128.7, 127.5, 127.4, 119.1, 117.3, 108.9, 105.7, 102.8, 46.2, 45.9, 38.7, 38.5, 38.4, 38.2, 33.7, 33.6, 33.1, 29.3, 28.0, 27.6, 26.1, 25.7, 23.3, 22.9, 21.5, 21.2, 20.9; IR (KBr, cm^{-1}) 3457(w), 3374(bs), 3052(w), 2924(s), 2856(m), 1777(m), 1701(s), 1597(w), 1500(m), 1444(w), 1388(s), 1310(w), 1185(s), 1160(S); HRMS *m/z* (M + Na^+) calcd 447.2044, found 447.2040. Anal. Calcd for $C_{28}H_{28}N_2O_2$: C, 79.22; H, 6.65; N, 6.60. Found: C, 79.02; H, 6.74; N, 6.37.

8-Isopropyl-2-(4-methylbenzyl)-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (95). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **95** (1366 mg, 61%) as a pink solid, a mixture of three isomers (maj:min:min = 2.7:1.0:0.8): mp 252–254°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.67 (d, J = 2.4 Hz, 1H, 1maj-H), 10.65 (d, J = 2.1 Hz, 1H, 1min-H), 10.42 (d, J = 2.4 Hz, 1H, 1min-H), 7.40–7.56 (m, 3H, Ph), 7.05–7.24 (m, 6H, Ph), 5.79 (d, J = 2.1 Hz, 1H, 3min-H), 5.58 (d, J = 2.4 Hz, 1H, 3maj-H), 5.57 (d, J = 2.7 Hz, 1H, 3min-H), 4.19 (dd, J = 8.3, 1.4 Hz, 1H, 3bαmin-H), 4.18 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 4.01 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.82 (s, 2H, Bn), 3.43 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.39 (dd, J = 8.3, 5.3 Hz, 1H, 6aαmaj-H), 3.35 (dd, J = 8.1, 5.4 Hz, 1H, 6aαmin-H), 2.96–3.02 (m, 1H, 10aαmin-H), 2.86–2.92 (m, 1H, 10aβmaj-H), 2.39–2.49 (m, 1H, 6bαmaj-H), 2.28–2.38 (m, 1H, 6bmin-H), 2.26 (s, 3H,

PhCH_3), 1.00–1.90 (m, 8H, cyclohex., CH(CH_3)$_2$), 0.84 (d, J = 6.6 Hz, 6H, CH(CH_3)$_2$ maj), 0.77 (d, J = 6.3 Hz, 6H, CH(CH_3)$_2$ min), 0.70 (d, J = 6.6 Hz, 6H, CH(CH_3)$_2$ min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.0, 176.2, 160.2, 136.6, 132.4, 129.4, 129.2, 128.8, 128.7, 128.6, 126.5, 126.4, 117.6, 106.4, 105.0, 104.1, 104.0, 46.0, 45.7, 43.9, 40.2, 39.0, 38.9, 37.9, 34.1, 34.05, 34.0, 33.2, 33.0, 32.95, 32.9, 29.0, 24.0, 21.2, 21.1, 21.0; IR (KBr, cm^{-1}) 3458(w), 3380(bs), 3054(w), 3027(w), 2926(m), 2863(s), 1776(m), 1703(s), 1595(w), 1500(m), 1452(m), 1387(s), 1315(w), 1187(s), 1172(s), 1150(s); HRMS m/z (M + Na$^+$) calcd 489.2513, found 489.2527. Anal. Calcd for $C_{31}H_{34}N_2O_2$: C, 79.79; H, 7.34; N, 6.00. Found: C, 79.61; H, 7.15; N, 5.83.

2-(4-Methylbenzyl)-5,8-diphenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (96). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **96** (1538 mg, 64%) as a dark-red solid, a mixture of two isomers (maj:min = 5.0:1.0): mp 215–217°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.71 (app. bs, 1H, 1maj-H), 10.51 (d, J = 1.8 Hz, 1H, 1min-H), 6.98–7.56 (m, 14H, Ph), 5.65 (d, J = 2.4 Hz, 1H, 3min-H), 5.61 (d, J = 2.4 Hz, 1H, 3maj-H), 4.22 (dd, J = 8.7, 1.8 Hz, 1H, 3bαmaj-H), 4.02 (dd, J = 8.4, 1.2 Hz, 1H, 3bαmin-H), 3.84 (s, 2H, Bn), 3.51 (dd, J = 9.0, 5.7 Hz, 1H, 6aαmin-H), 3.40–3.50 (m, 2H, 6aαmaj-H, 6aαmin-H), 2.80–3.10 (m, 2H, 6b-H, 10a-H), 2.26 (s, 3H, PhCH_3), 1.10–2.26 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 177.9, 177.8, 176.8, 176.1, 136.4, 136.1, 134.3, 132.7, 131.8, 131.0, 130.2, 129.4, 129.3, 128.8, 128.7, 128.6, 127.4, 127.3, 126.9, 126.7, 126.6, 126.5, 126.1, 125.8, 125.6, 117.6, 104.2, 45.6, 34.1, 34.0, 33.2–33.6 (overlapped peaks), 21.1; IR (KBr, cm^{-1}) 3454(w), 3378(s), 3055(w), 3025(m), 2926(s), 2860(m), 1776(m), 1703(s), 1598(m), 1499(m), 1450(m), 1387(s), 1331(w), 1186(s), 1155(s); HRMS m/z (M + Na$^+$) calcd for $C_{34}H_{32}N_2O_2$: 523.2357, found 523.2382.

2-(4-Methylbenzyl)-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (97). Method B with **3c** (687 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **97** (1368 mg, 65%) as a light-pink solid, a mixture of two isomers (maj:min = 1.9:1.0): mp 218–220°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.65 (d, J = 2.1 Hz, 1H, 1maj-H), 10.38 (d, J = 2.4 Hz, 1H, 1min-H), 7.00–7.17 (m, 8H, Ph), 5.80 (d, J = 2.4 Hz, 1H, 3min-H), 5.56 (d, J = 2.4 Hz, 1H, 3maj-H), 4.15 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.99 (dd, J = 8.6, 1.7 Hz, 1H, 3bαmin-H), 3.82 (s, 2H, Bn), 3.79 (s, 3H, OCH$_3$ maj), 3.78 (s, 3H, OCH$_3$ min), 3.37 (dd, J = 8.1, 5.3 Hz, 1H, 6aαmaj-H), 3.32 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.01–3.06 (m, 1H, 10aαmin-H), 2.89–2.94 (m, 1H, 10aβmaj-H), 2.03–2.42 (m, 2H, cyclohex., 6bα-H), 2.26 (s, 3H, PhCH_3), 1.02–1.62 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.6, 178.4, 177.6, 159.4, 138.4, 138.3, 135.2, 130.8, 129.3, 129.0, 128.8, 128.6127.4, 125.5, 125.3, 119.1, 117.4, 114.7, 113.8, 108.9, 105.7, 105.0, 55.9, 45.8, 38.6, 38.4, 38.1, 33.6, 33.1, 27.6, 26.1, 25.7, 23.3, 23.0, 21.5, 21.1; IR (KBr, cm^{-1}) 3457(w), 3380(s), 3050(w), 3004(w), 2926(s), 2856(m), 1776(m), 1714(s), 1610(m), 1593(m), 1514(s), 1459(m), 1443(m), 1390(s), 1302(m), 1255(s), 1189(s), 1166(s), 1108(m), 1031(m); HRMS m/z (M + Na$^+$) calcd 477.2149, found 477.2169. Anal. Calcd for $C_{29}H_{30}N_2O_3$: C, 76.63; H, 6.65; N, 6.16. Found: C, 76.40; H, 6.61; N, 5.96.

8-Isopropyl-2-(4-methylbenzyl)-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (98). Method B with **3f** (982 mg, 7.00 mmol), 3.5 h-reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **98** (1315 mg, 57%) as a light-pink solid, a mixture of three isomers (maj:min:min = 3.4:1.0:0.9): mp 244–246°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.66 (d, J = 2.4 Hz, 1H, 1maj-H), 10.64 (d, J = 1.8 Hz, 1H, 1min-H), 10.41 (d, J = 1.5 Hz, 1H, 1min-H), 7.02–7.16 (m, 8H, Ph), 5.79 (d, J = 1.8 Hz, 1H, 3min-H), 5.57 (d, J = 2.1 Hz, 1H, 3maj-H), 5.56 (d, J = 2.0 Hz, 1H, 3min-H), 4.15 (dd, J = 8.7, 1.8 Hz, 1H, 3bαmin-H), 4.14 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.97 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmin-H), 3.82 (s, 2H, Bn), 3.79 (s, 3H, OCH$_3$ maj), 3.78 (s, 3H, OCH$_3$ min), 3.40 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.36 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.32 (dd, J = 8.1, 5.4 Hz, 1H, 6aαmin-H), 2.94–3.02 (m, 1H, 10aαmin-H), 2.85–2.91 (m, 1H, 10aβmaj-H), 2.38–2.49 (m, 1H, 6bαmaj-H), 2.30–2.36 (m, 1H, 6bmin-H), 2.26 (s, 3H, PhCH_3), 0.95–2.26 (m, 8H, cyclohex., CH(CH$_3$)$_2$), 0.84 (d, J = 6.6 Hz, 6H, CH(CH_3)$_2$ maj), 0.77 (d, J = 6.3 Hz, 6H, CH(CH_3)$_2$ min), 0.70 (d, J = 6.6 Hz, 6H, CH(CH_3)$_2$ min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.3, 177.0, 176.4, 159.6, 155.1, 136.5, 136.1, 132.4, 129.4, 128.7, 128.6, 127.6, 124.4, 120.5, 120.1, 117.9, 117.5, 114.7, 14.6, 109.5, 106.5, 104.1, 103.8, 55.6, 45.9, 45.6, 45.5, 43.9, 40.3, 38.9, 38.8, 37.8, 37.7, 34.1, 34.0, 34.9, 33.2, 33.0, 32.9, 32.8, 29.0, 26.3, 24.0, 21.4, 21.1, 21.0, 20.9, 20.0, 19.9; IR (KBr, cm^{-1}) 3463(w), 3380(bs), 3087(w), 3052(w), 3005(w), 2945(bs), 2864(s), 1776(m), 1699(s), 1612(m), 1589(w), 1514(s), 1452(m), 1391(s), 1304(m), 1256(s), 1171(s), 1109(m), 1032(m); HRMS m/z (M + Na$^+$) calcd for $C_{32}H_{36}N_2O_3$: 519.2619, found 519.2637.

2-(4-Methylbenzyl)-5-(4-methoxyphenyl)-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (99). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **99** (1532 mg, 62%) as a light-brown solid, a mixture of two isomers (maj:min = 3.0:1.0): mp 227–228°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.67–10.71 (app. bs, 1H, 1maj-H), 10.49 (d, J = 3.0 Hz, 1H, 1min-H), 6.97–7.34 (m, 13H, Ph), 5.76–5.82 (app. m, 1H, 3min-H), 5.60 (d, J = 2.4 Hz, 1H, 3maj-H), 4.18 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.99 (app. d, J = 8.4 Hz, 1H, 3bαmin-H), 3.84 (s, 2H, Bn), 3.80 (d, 3H, OCH$_3$ maj), 3.79 (d, 3H, OCH$_3$ min), 3.36–3.51 (m, 1H, 6aα-H), 2.80–3.10 (m, 2H, 10a-H, 6bα-H), 1.50–2.60 (m, 7H, cyclohex.), 2.26 (s, 3H, PhCH_3); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.3, 178.2, 177.0, 176.4, 159.7, 159.5, 136.1, 132.7, 129.4, 128.7, 128.6, 128.5, 128.4, 127.8, 127.7, 127.6, 127.4, 127.3, 126.7, 125.8, 125.6, 124.4, 117.6, 114.7, 114.6, 55.6, 45.5, 34.2, 34.1, 33.1–33.7 (overlapped peaks), 21.1; IR (KBr, cm^{-1}) 3458(w), 3389(s), 3085(w), 3057(w), 3023(w), 2933(s), 2860(m), 2368(w), 1775(w), 1698(s), 1607(w), 1514(s), 1448(m), 1390(m), 1301(m), 1253(s), 1170(s), 1108(w), 1032(m); HRMS m/z (M + Na$^+$) calcd 553.2462, found 553.2488. Anal. Calcd for $C_{35}H_{34}N_2O_3$: C, 79.22; H, 6.46; N, 5.28. Found: C, 78.91; H, 6.32; N, 5.19.

5-Dimethylamino-2-(4-methoxybenzyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (100). Method A gave **100** (293 mg, 24%) as a cream-colored solid, a mixture of two isomers (maj:min = 3.8:1.0): mp 228–229°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.20 (bs, 1H, 1min-H), 7.51 (bs, 1H, 1maj-H), 7.16 (d, J = 8.7 Hz, 2H, Ph), 6.87 (d, J =

8.4 Hz, 2H, Ph), 6.23 (d, J = 2.4 Hz, 1H, 3maj-H), 5.77 (d, J = 2.7 Hz, 1H, 3min-H), 3.97 (AA′d, J = 15.9 Hz, 1H, Bn maj), 3.96 (AA′d, J = 16.2 Hz, 1H, Bn min), 3.90 (AA′d, J = 16.2 Hz, 1H, Bn maj), 3.89 (AA′d, J = 15.9 Hz, 1H, Bn maj), 3.81 (s, 3H, OCH_3), 3.69 (dd, 1H, 3bα-H), 3.21 (dd, J = 8.4, 5.7 Hz, 1H, 6aαmin-H), 3.16 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmaj-H), 3.00–3.06 (m, 1H, 10aαmaj-H), 2.93 (s, 6H, $N(CH_3)_2$), 2.91–2.92 (m, 1H, 10aβmin-H), 2.44–2.52 (m, 1H, 6bα-H), 1.05–2.22 (m, 8H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 177.6, 177.5, 176.7, 159.1, 158.0, 133.4, 131.0, 130.0, 129.9, 127.3, 114.1, 108.8, 105.4, 55.5, 44.3, 44.0, 43.8, 38.2, 36.7, 33.2, 33.1, 32.9, 27.6, 25.6, 23.0, 21.4; IR (thin film, cm^{-1}) 3371(bs), 2924(m), 2852(m), 1770(w), 1703(s), 1515(m), 1444(m), 1360(w), 1252(m), 1193(m); HRMS m/z ($M + Na^+$) calcd for $C_{24}H_{29}N_3O_3$: 430.2102, found 430.2087.

5-Dimethylamino-8-ethyl-2-(4-methoxybenzyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (101). Method A gave **101** (287 mg, 22%) as a cream-colored solid, a mixture of three isomers (maj:min = 4.0:1.0:0.2): mp 179–180°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.22 (bs 1H, 1min-H), 7.55 (bs, 1H, 1maj-H), 7.16 (d, J = 8.7 Hz, 2H, Ph), 6.86 (d, J = 8.7 Hz, 2H, Ph), 6.22 (d, J = 2.4 Hz, 1H, 3maj-H), 5.78 (d, J = 2.7 Hz, 1H, 3min-H), 5.74 (d, J = 2.1 Hz, 1H, 3min-H), 3.97 (AA′d, J = 16.2 Hz, 1H, Bn maj), 3.96 (AA′d, J = 16.2 Hz, 1H, Bn min), 3.89 (AA′d, J = 16.2 Hz, 1H, Bn maj), 3.88 (AA′d, J = 16.2 Hz, 1H, Bn min), 3.81 (s, 3H, OCH_3), 3.69 (dd, J = 8.4, 1.8 Hz, 1H, 3bα-H), 3.24 (dd, J = 5.4, 9.6 Hz, 1H, 6aαmin-H), 3.20 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.19 (dd, J = 8.7, 5.6 Hz, 1H, 6aαmin-H), 3.16 (dd, J = 8.6, 5.6 Hz, 1H, 6aαmaj-H), 2.91–2.94 (m, 7H, 10a-H, $N(CH_3)_2$), 2.58–2.67 (m, 1H, 6bαmaj-H), 2.46–2.55 (m, 1H, 6bmin-H), 1.00–2.07 (m, 9H, cyclohex., CH_2CH_3), 0.84 (t, J = 7.4 Hz, 3H, CH_2CH_3 maj), 0.76 (t, J = 7.2 Hz, 3H, CH_2CH_3 maj); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 177.6, 177.4, 176.7, 176.6, 158.0, 133.3, 131.0, 130.0, 129.9, 129.8, 127.2, 127.15, 127.1, 117.3, 114.1, 114.0, 108.9, 108.8, 105.4, 105.0, 102.8, 55.5, 43.9, 43.8, 43.6, 36.7, 33.9, 33.0–33.2 (multiple peaks), 32.6, 29.9; IR (thin film, cm^{-1}) 3378(bs), 2928(m), 2358(w), 1773(w), 1709(s), 1510(m), 1246(m); HRMS m/z ($M + Na^+$) calcd for $C_{26}H_{33}N_3O_3$: 458.2415, found 458.2422.

8-tert-Butyl-5-(dimethylamino)-2-(4-methoxybenzyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (102). Method A gave **102** (292 mg, 21%) as orange crystals, a single isomer: mp 95–96°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 7.43 (bs, 1H, 1-H), 7.13 (d, J = 8.4 Hz, 2H, Ph), 6.85 (d, J = 8.7 Hz, 2H, Ph), 6.08 (d, J = 2.7 Hz, 1H, 3-H), 3.92 (AA′d, J = 17.1 Hz, 1H, Bn), 3.83 (AA′d, J = 17.1 Hz, 1H, Bn), 3.81 (s, 3H, OCH_3), 3.78 (dd, J = 7.8 Hz, 1.7 Hz, 1H, 3bα-H), 3.09 (dd, J = 8.0, 5.9 Hz, 1H, 6aα-H), 2.87 (s, 6H, $N(CH_3)_2$), 2.61–2.70 (m, 1H, 6bα), 2.50–2.57 (m, 1H, 10aα-H), 1.65–2.06 (m, 4H, cyclohex.), 1.49 (ddd, J = 13.8, 10.4, 6.8 Hz, 1H, cyclohex.), 1.07–1.30 (m, 2H, cyclohex.), 0.89 (s, 9H, t-Bu); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 177.7, 176.4, 158.0, 133.0, 131.3, 130.5, 130.0, 129.9, 114.1, 109.1, 104.2, 55.5, 43.6, 43.55, 43.1, 33.9, 33.3, 33.0, 32.8, 30.4, 28.0, 27.9; IR (thin film, cm^{-1}) 3364(bs), 2955(m), 1774(w), 1712(s), 1511(s), 1364(m), 1246(m); HRMS m/z ($M + Na^+$) calcd for $C_{28}H_{37}N_3O_3$: 486.2728, found 486.2720.

2-(4-Methoxybenzyl)-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (103). Method B with **3c** (687 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **103** (1270 mg, 60%) as a cream-colored solid, a mixture of two isomers (maj:min = 1.8:1.0): mp 234–235°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.27 (bs, 1H, 1maj-H), 7.58 (bs, 1H, 1min-H), 7.37–7.51 (m, 3H, Ph), 7.24–7.32 (m, 2H, Ph), 7.16–7.21 (m, 2H, Ph), 6.85–6.90 (m, 2H, Ph), 6.27 (d, J = 2.4 Hz, 1H, 3min-H), 5.81 (J = 2.4 Hz, 1H, 3maj-H), 3.99 (AA′d, J = 17.1 Hz, 1H, Bn maj), 3.971 (dd, J = 8.6, 2.0 Hz, 1H, 3bα-H), 3.968 (AA′d, J = 15.9 Hz, 1H, Bn min), 3.91 (AA′d, J = 15.9 Hz, 1H, Bn maj), 3.81 (s, 3H, OCH_3), 3.46 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmaj-H), 3.39 (dd, J = 8.6, 5.3 Hz, 1H, 6aαmin-H), 3.08–3.14 (m, 1H, 10aαmin-H), 3.02–3.07 (m, 1H, 10aβmaj-H), 2.50–2.58 (m, 1H, 6bα-H), 2.04–2.25 (m, 1H, cyclohex.), 1.18–1.76 (m, 7H, cyclohex.); ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.65 (d, J = 2.1 Hz, 1H, 1maj-H), 10.38 (d, J = 1.8 Hz, 1H, 1min-H), 7.38–7.54 (m, 3H, Ph), 7.12–7.26 (m, 4H, Ph), 6.81–6.87 (m, 2H, Ph), 5.80 (d, J = 2.4 Hz, 1H, 3min-H), 5.56 (d, J = 2.4 Hz, 1H, 3maj-H), 4.18 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmaj-H), 4.02 (dd, J = 8.4, 1.8 Hz, 1H, 3bαmin-H), 3.80 (s, 2H, Bn), 3.71 (s, 3H, OCH_3), 3.37 (dd, J = 8.4, 5.1 Hz, 1H, 6aαmaj-H), 3.34 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.01–3.07 (m, 1H, 10aαmin-H), 2.90–2.95 (m, 1H, 10aβmaj-H), 2.03–2.42 (m, 2H, cyclohex., 6b-H), 1.03–1.64 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.2, 177.4, 176.3, 158.0, 133.4, 138.3, 133.0, 131.1, 130.0, 129.9, 129.6, 129.5, 128.9, 128.8, 127.4, 119.1, 117.3, 114.1, 108.9, 105.6, 102.7, 55.5, 46.2, 45.9, 38.7, 38.5, 38.4, 38.2, 33.2, 33.1, 29.3, 27.6, 26.1, 25.7, 23.5, 22.9, 21.5, 20.9; IR (thin film, cm^{-1}) 3372, 2920, 1697, 1515; HRMS m/z ($M + Na^+$) calcd 463.1993, found 463.2009. Anal. Calcd for $C_{28}H_{28}N_2O_3$: C, 76.34; H, 6.41; N, 6.36. Found: C, 76.26; H, 6.59; N, 6.35.

8-Ethyl-2-(4-methoxybenzyl)-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (104). Method A gave **104** (450 mg, 32%) as a cream-colored solid, a mixture of four isomers (maj:min:min:min = 1.1:1.0:0.3:0.3): mp 214–215°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.25 (bs, 1H, 1maj-H), 7.58 (bs, 1H, 1min-H), 7.56 (bs, 1H, 1min-H), 7.37–7.51 (m, 3H, Ph), 7.28–7.32 (m, 2H, Ph), 7.15–7.20 (m, 2H, Ph), 6.85–6.90 (m, 2H, Ph), 6.26 (d, J = 2.7 Hz, 1H, 3maj-H), 5.82 (d, J = 2.4 Hz, 1H, 3min-H), 5.79 (d, J = 3.0 Hz, 1H, 3min-H), 3.98 (AA′d, J = 15.9 Hz, 1H, Bn min), 3.97 (AA′d, J = 16.2 Hz, 1H, Bn maj), 3.96 (dd, J = 8.6, 2.0 Hz, 1H, 3bα-H), 3.91 (AA′d, J = 16.2 Hz, 1H, Bn min), 3.90 (AA′d, J = 16.2 Hz, 1H, Bn maj), 3.82 (s, 3H, OCH_3), 3.48 (dd, J = 9.02, 5.3 Hz, 1H, 6aαmin-H), 3.45 (dd, J = 8.1, 5.7 Hz, 1H, 6aαmin-H), 3.42 (dd, J = 7.8, 5.4 Hz, 1H, 6aαmin-H), 3.39 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 2.97–3.08 (m, 1H, 10a-H), 2.65–2.74 (m, 2H, 6bαmaj-H, 6bαmin-H), 2.53–2.62 (m, 2H, 6bβmin-H), 1.07–2.30 (m, 7H, cyclohex.), 1.43 (app. q, J = 7.5 Hz, 2H, CH_2CH_3), 0.85 (t, J = 7.2 Hz, 3H, CH_2CH_3 maj), 0.79 (t, J = 7.2 Hz, 3H, CH_2CH_3 min); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 178.2, 177.4, 176.3, 158.0, 133.4, 133.35, 133.3, 133.0, 132.9, 132.8, 131.1, 131.0, 130.0, 129.9, 129.8, 129.7, 129.6, 129.5, 128.9, 128.7, 127.4, 127.35, 127.3, 119.0, 118.8, 117.4, 114.1, 109.0, 105.5, 105.0, 102.8, 55.5, 45.9, 45.5, 38.9, 38.7, 38.4, 38.2, 38.1, 34.3, 33.9, 32.7–33.3 (multiple peaks), 30.0, 27.4, 23.7, 23.6, 12.6, 11.8; IR (thin film, cm^{-1}) 3389(bs), 2931(m), 1777(w), 1706(s), 1509(m), 1383(m), 1246(m), 1176(m); HRMS m/z ($M + Na^+$) calcd 491.2306, found 491.2323. Anal. Calcd for

$C_{30}H_{32}N_2O_3$: C, 76.90; H, 6.88; N, 5.98. Found: C, 76.98; H, 7.19; N, 5.19.

8-Isopropyl-2-(4-methoxybenzyl)-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (105). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **105** (1181 mg, 51%) as a light-pink solid, a mixture of three isomers (maj:min:min = 4.5:1.0:0.9): mp 235–237°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.65 (d, J = 2.4 Hz, 1H, 1maj-H), 10.64 (d, J = 1.8 Hz, 1H, 1min-H), 10.40 (d, J = 1.8 Hz, 1H, 1min-H), 7.41–7.56 (m, 3H, Ph), 7.13–7.24 (m, 4H, Ph), 6.80–6.86 (m, 2H, Ph), 5.78 (d, J = 2.4 Hz, 1H, 3min-H), 5.57 (d, J = 2.4 Hz, 1H, 3maj-H), 5.55 (d, J = 2.1 Hz, 1H, 3min-H), 4.19 (dd, J = 8.4, 1.5 Hz, 1H, 3bαmin-H), 4.17 (dd, J = 7.8, 0.9 Hz, 1H, 3bαmaj-H), 4.00 (dd, J = 8.4, 2.1 Hz, 1H, 3bαmin-H), 3.80 (s, 2H, Bn), 3.713 (s, 3H, OCH_3 maj), 3.709 (s, 3H, OCH_3 min), 3.70 (s, 3H, OCH_3 min), 3.43 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.38 (dd, J = 8.4, 5.1 Hz, 1H, 6aαmaj-H), 3.35 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmin-H), 2.95–3.01 (m, 1H, 10aαmin-H), 2.85–2.91 (m, 1H, 10aβmaj-H), 2.38–2.50 (m, 1H, 6bαmaj-H), 2.20–2.36 (m, 1H, 6bmin-H), 1.20–2.02 (m, 8H, cyclohex, C*H*$(CH_3)_2$), 0.84 (d, J = 6.3 Hz, 6H, $CH(CH_3)_2$ maj), 0.77 (d, J = 6.3 Hz, 6H, $CH(CH_3)_2$ min), 0.69 (d, J = 6.6 Hz, 6H, $CH(CH_3)_2$ min); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.0, 176.2, 132.6, 131.9, 131.6, 129.8, 129.75, 129.7, 129.4, 129.3, 129.2, 128.8, 128.7, 128.4, 126.5, 126.4, 126.2, 117.6, 114.1, 106.4, 105.0, 104.0, 103.9, 55.4, 45.7, 45.7, 43.9, 40.3, 38.9, 37.8, 32.8–33.7 (overlapped peaks), 26.3, 24.0, 22.5, 21.4, 21.0, 19.9; IR (KBr, cm^{-1}) 3463(w), 3384(bs), 3064(w), 2999(w), 2929(s), 2864(s), 2836(m), 2361(w), 2329(w), 1777(m), 1698(s), 1613(m), 1595(m), 1512(s), 1454(m), 1387(s), 1248(m), 1175(s); HRMS m/z (M + Na^+) calcd for $C_{31}H_{34}N_2O_3$: 505.2462, found 505.2483.

8-tert-Butyl-2-(4-methoxybenzyl)-5-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (106). Method A gave **106** (432 mg, 29%) as a cream-colored solid, a mixture of three isomers (maj:min:min = 5.2:1.0:0.6): mp 219–220°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.26 (bs, 1H, 1min-H), 8.06 (bs, 1H, 1maj-H), 7.36–7.53 (m, 3H, Ph), 7.12–7.28 (m, 4H, Ph), 6.84–6.89 (m, 2H, Ph), 6.10 (d, J = 2.7 Hz, 1H, 3min-H), 5.81 (d, J = 2.7 Hz, 1H, 3min-H), 5.76 (d, J = 2.7 Hz, 1H, 3maj-H), 4.06 (dd, J = 8.1, 1.8 Hz, 1H, 3bα-H), 3.90–3.97 (m, overlapped, 2H, 2XBn min), 3.92 (AA′d, J = 14.4 Hz, 1H, Bn maj), 3.84 (AA′d, J = 14.4 Hz, 1H, Bn maj), 3.81 (s, 3H, OCH_3), 3.49 (dd, J = 8.6, 5.6 Hz, 1H, 6aαmin-H), 3.41 (dd, J = 8.1, 5.7 Hz, 1H, 6aαmaj-H), 3.34 (dd, J = 7.8. 5.4 Hz, 1H, 6aαmin-H), 2.53–2.75 (m, 2H, 6bα-H, 10a-H), 1.02–2.32 (m, 7H, cyclohex.), 0.90 (s, 9H, *t*-Bu), 0.74 (s, 9H, *t*-Bu); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 176.0, 158.0, 158.75, 133.2, 133.16, 130.1, 129.8, 129.7, 129.6, 129.4, 128.8, 128.6, 127.5, 127.1, 117.3, 117.26, 114.1, 104.0, 55.5, 44.9, 34.3–34.5 (multiple peaks), 34.0, 33.8, 33.3, 33.2, 33.1, 33.0, 32.7, 28.2, 27.7; IR (thin film, cm^{-1}) 3455(bs), 2950(m), 2360(w), 1770(w), 1702(s), 1648(bm), 1511(m), 1388(m), 1247(m), 1176(m); HRMS m/z (M + Na^+) calcd 519.2619, found 519.2627. Anal. Calcd for $C_{32}H_{36}N_2O_3$: C, 77.39; H, 7.31; N, 5.64. Found: C, 77.44; H, 7.68; N, 5.67.

2-(4-Methoxybenzyl)-5,8-diphenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (107). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **107** (1513 mg, 61%) as a brown solid, a mixture of two isomers (maj:min = 5.2:1.0): mp 223–225°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.69 (bs, 1H, 1maj-H), 10.50 (bs, 1H, 1min-H), 7.15–7.56 (m, 12H, Ph), 6.83–6.88 (m, 2H, Ph), 5.79 (d, J = 2.1 Hz, 1H, 3min-H), 5.61 (d, J = 2.7 Hz, 1H, 3maj-H), 4.21 (dd, J = 8.7, 0.9 Hz, 1H, 3bαmaj-H), 4.02 (dd, J = 6.6, 2.7 Hz, 1H, 3bαmin-H), 3.82 (s, 2H, Bn), 3.72 (s, 3H, OCH_3), 3.41–3.55 (m, 1H, 6aα-H), 2.70–3.10 (m, 2H, 6b-H, 10a-H), 1.40–2.10 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.0, 176.0, 158.4, 136.1, 132.9, 131.8, 131.4, 129.8, 129.4, 129.3, 129.2, 128.8, 128.6, 128.5, 128.4, 127.4, 127.3, 126.5, 126.4, 125.6, 114.1, 55.4, 45.6, 33.4–33.8 (overlapped peaks); IR (KBr, cm^{-1}) 3458(w), 3381(bs), 3061(w), 3026(w), 3003(w), 2930(s), 2861(m), 2836(m), 2360(w), 2335(w), 1777(m), 1703(s), 1599(m), 1510(s), 1452(m), 1387(s), 1249(m), 1176(s); HRMS m/z (M + Na^+) calcd for $C_{34}H_{32}N_2O_3$: 539.2306, found 539.2308.

2-(4-Methoxybenzyl)-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (108). Method A gave **108** (593 mg, 42%) as a cream-colored solid, a mixture of two isomers (maj:min = 1.2:1.0): mp 234–235°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.26 (bs, 1H, 1min-H), 7.56 (bs, 1H, 1maj-H), 7.15–7.24 (m, 4H, Ph), 6.96–7.01 (m, 2H, Ph), 6.85–6.87 (m, 2H, Ph), 6.27 (d, J = 2.7 Hz, 1H, 3maj-H), 5.81 (d, J = 2.7 Hz, 1H, 3min-H), 3.99 (AA′d, J = 15.9 Hz, 1H, Bn maj), 3.97 (AA′d, J = 16.2 H, 1H, Bn min), 3.90–3.97 (m, overlapped, 1H, 3bα-H), 3.90 (AA′d, J = 16.5 Hz, 1H, Bn maj), 3.89 (AA′d, J = 15.6 Hz, 1H, Bn min), 3.84 (s, 3H, OCH_3), 3.82 (s, 3H, OCH_3), 3.44 (dd, J = 8.9, 5.6 Hz, 1H, 6aαmin-H), 3.38 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.08–3.12 (m, 1H, 10aαmaj-H), 3.02–3.06 (m, 1H, 10aβmin-H), 2.49–2.57 (m, 1H, 6bα-H), 2.04–2.24 (m, 1H, cyclohex.), 1.08–1.76 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.4, 176.5, 159.5, 158.0, 133.3, 132.9, 130.0, 129.9, 128.6, 125.3, 119.0, 117.4, 114.8, 114.7, 114.1, 105.6, 102.7, 74.9, 58.9, 55.9, 55.5, 46.2, 38.4, 38.1, 33.2, 33.12, 33.07, 33.04, 29.3, 26.1, 22.9, 20.9; IR (thin film, cm^{-1}) 3386(bs), 2920(m), 2360(w), 1769(w), 1697(s), 1516(m), 1392(m), 1257(m), 1178(m); HRMS m/z (M + Na^+) calcd for $C_{29}H_{30}N_2O_4$: 493.2099, found 493.2116.

8-Ethyl-2-(4-methoxybenzyl)-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (109). Method A gave **109** (434 mg, 29%) as a cream-colored solid, a mixture of three isomers (maj:min:min = 1.9:1.0:0.2): mp 228–229°C; ^{1}H NMR (300 MHz, $CDCl_3$, δ) 8.25 (bs, 1H, 1min-H), 7.57 (bs, 1H, 1maj-H), 7.14–7.23 (m, 4H, Ph), 6.96–7.02 (m, 2H, Ph), 6.85–6.90 (m, 2H, Ph), 6.26 (d, J = 2.4 Hz, 1H, 3maj-H), 5.82 (d, J = 2.7 Hz, 1H, 3min-H), 5.78 (d, J = 2.7 Hz, 1H, 3min-H), 4.99 (AA′d, J = 16.2 Hz, 1H, Bn maj), 4.97 (AA′d, J = 16.8 Hz, 1H, Bn min), 3.91–3.96 (m, overlapped, 1H, 3bα-H), 3.90 (AA′d, J = 16.2 Hz, 1H, Bn maj), 3.89 (AA′d, J = 16.5 Hz, 1H, Bn min), 3.84 (s, 3H, OCH_3), 3.82 (s, 3H, OCH_3), 3.43 (dd, J = 8.7, 5.7 Hz, 1H, 6aαmin-H), 3.37 (dd, J = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.02–3.07 (m, 1H, 10aαmaj-H), 2.96–3.01 (m, 1H, 10aβmin-H), 2.64–2.73 (m, 1H, 6bαmaj-H), 2.53–2.60 (m, 1H, 6bmin-H), 1.07–1.92 (m, 9H, cyclohex., CH_2CH$_3$), 0.85 (t, J = 7.2 Hz, 3H, CH_2CH_3); ^{13}C NMR (75 MHz, DMSO-d_6, δ) 178.5, 178.45, 178.4, 177.6, 176.5, 159.5, 159.4, 159.2, 158.0, 133.3, 132.9, 132.8, 131.0, 131.95, 130.1, 129.9, 129.8, 128.6, 128.5, 127.2, 125.6, 125.4, 118.7, 117.4, 114.9, 114.8, 114.1, 109.1,

105.5, 102.8, 55.9, 55.5, 45.8, 45.5, 34.3, 34.0, 33.0–33.3 (multiple peaks), 33.9, 32.6–32.8 (multiple peaks), 23.6, 23.5, 12.6; IR (thin film, cm^{-1}) 3441(bs), 2934(m), 2100(bw), 1777(w), 1694(s), 1651(bm), 1515(s), 1388(m), 1252(m), 1174(m); HRMS *m/z* (M + Na^+) calcd 521.2412, found 521.2416. Anal. Calcd for $C_{31}H_{34}N_2O_4$: C, 74.67; H, 6.87; N, 5.62. Found: C, 72.72; H, 6.59; N, 5.45.

8-Isopropyl-2-(4-methoxybenzyl)-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (110). Method B with **3f** (982 mg, 7.00 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **110** (1304 mg, 53%) as a colorless solid, a mixture of three isomers (maj:min:min = 3.8:1.0:0.6): mp 252–254°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.64 (d, *J* = 2.4 Hz, 1H, 1maj-H), 10.63 (d, *J* = 2.7 Hz, 1H, 1min-H), 10.39 (d, *J* = 2.4, 1H, 1min-H), 7.01–7.21 (m, 6H, Ph), 6.80–6.86 (m, 2H, Ph), 5.78 (d, *J* = 2.4 Hz, 1H, 3min-H), 5.57 (d, *J* = 2.4 Hz, 1H, 3maj-H), 5.55 (d, *J* = 2.1 Hz, 1H, 3min-H), 4.15 (dd, *J* = 8.4, 1.8 Hz, 1H, 3bαmin-H), 4.13 (dd, *J* = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.97 (dd, *J* = 8.7, 2.1 Hz, 1H, 3bαmin-H), 3.80 (s, 2H, Bn), 3.79 (s, 3H, PhOCH_3 min), 3.79 (s, 3H, PhOCH_3 maj), 3.78 (s, 3H, PhOCH_3 min), 3.712 (s, 3H, BnOCH_3 min), 3.710 (s, 3H, BnOCH_3 maj), 3.70 (s, 3H, BnOCH_3 min), 3.40 (dd, *J* = 8.7, 5.4 Hz, 1H, 6aαmin-H), 3.35 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmaj-H), 3.31 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmin-H), 2.94–3.01 (m, 1H, 10aαmin-H), 2.84–2.91 (m, 1H, 10aβmaj-H), 2.37–2.48 (m, 1H, 6bαmaj-H), 2.26–2.36 (m, 1H, 6bmin-H), 0.88–2.18 (m, 8H, cyclohex., C*H*$(CH_3)_2$), 0.84 (d, *J* = 6.0 Hz, 6H, CH(CH_3)$_2$ maj), 0.76 (d, *J* = 6.6 Hz, 6H, CH(CH_3)$_2$ min), 0.69 (d, *J* = 6.9 Hz, 1H, CH(CH_3)$_2$ min); IR (KBr, cm^{-1}) 3462(w), 3377(bs), 3064(w), 2996(w), 2931(bs), 2864(m), 2837(m), 1776(m), 1695(s), 1612(m), 1589(m), 1514(s), 1452(m), 1391(m), 1303(m), 1250(s), 1169(s), 1106(m), 1033(m); HRMS *m/z* (M + Na^+) calcd 535.2568, found 535.2589. Anal. Calcd for $C_{32}H_{36}N_2O_4$: C, 74.97; H, 7.08; N, 5.46. Found: C, 74.77; H, 6.82; N, 5.28.

8-tert-Butyl-2-(4-methoxybenzyl)-5-(4-methoxyphenyl)-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (111). Method A gave **111** (348 mg, 22%) as a cream-colored solid, a mixture of four isomers (maj:min:min:min = 2.7:1.0:0.7:0.3): mp 224–225°C; ^{1}H NMR (300 MHz, CDCl3, δ) 8.25 (bs, 1H, 1min-H), 8.05 (bs, 1H, 1maj-H), 7.50 (bs, 1H, 1min-H), 7.47 (bs, 1H, 1min-H), 7.09–7.18 (m, 4H, Ph), 6.94–7.03 (m, 2H, Ph), 6.83–6.89 (m, 2H, Ph), 6.29 (d, *J* = 2.4 Hz, 1H, 3min-H), 6.10 (d, *J* = 2.4 Hz, 1H, 3min-H), 5.80 (d, *J* = 2.7 Hz, 1H, 3min-H), 5.75 (d, *J* = 2.7 Hz, 1H, 3maj-H), 4.04 (d, *J* = 8.1 Hz, 1H, 3bα-H), 3.80–3.95 (m, 2H, Bn), 3.81 (s, 6H, 2XOCH_3), 3.47 (dd, *J* = 8.7, 5.4 Hz, 1H, 6aαmin-H), 3.42 (dd, *J* = 8.4, 5.4 Hz, 1H, 6aαmin-H), 3.38 (dd, *J* = 8.1, 5.7 Hz, 1H, 6aαmaj-H), 3.32 (dd, *J* = 7.5, 5.4 Hz, 1H, 6aαmin-H), 3.03–3.07 (m, 1H, 10amin-H), 2.98–3.02 (m, 1H, 10amin-H), 2.54–2.70 (m, 3H, 6b-H, 10aαmaj-H, 10amin-H), 1.05–2.27 (m, 7H, cyclohex.), 0.89 (s, 9H, *t*-Bu maj), 0.74 (s, 9H, *t*-Bu min); IR (thin film, cm^{-1}) 3440(bs), 2952(m), 2358(m), 1770(w), 1698(s), 1514(s), 1393(m), 1303(m), 1250(m), 1174(m); HRMS *m/z* (M + Na^+) calcd for $C_{32}H_{38}N_2O_4$: 549.2725, found 549.2694.

2-(4-Methoxybenzyl)-5-(4-methoxyphenyl)-8-phenyl-3b,6a,6b,7,8,9,10,10a-octahydro-1H,5H-benzo[g]pyrrolo[3,4-e]indole-4,6-dione (112). Method B with **3h** (1220 mg, 7.000 mmol), 3.5-h reflux, ethanol wash (4 mL), and then a diethyl ether wash (10 mL) gave **112** (1548 mg, 59%) as a light-brown solid, a mixture of two isomers (maj:min = 8.1:1.0): mp 226–227°C; ^{1}H NMR (300 MHz, DMSO-d_6, δ) 10.67 (app. bs, 1H, 1maj-H), 10.48 (d, *J* = 2.4 Hz, 1H, 1min-H), 6.97–7.39 (m, 11H, Ph), 6.82–6.90 (m, 2H, Ph), 5.62 (d, *J* = 2.7 Hz, 1H, 3min-H), 5.60 (d, *J* = 2.4 Hz, 1H, 3maj-H), 4.20 (dd, *J* = 8.7, 1.8 Hz, 1H, 3bαmin-H), 4.18 (dd, *J* = 8.4, 1.5 Hz, 1H, 3bαmaj-H), 3.82 (s, 2H, Bn), 3.79 (s, 3H, PhCH_3 maj), 3.78 (s, 3H, PhCH_3 min), 3.73 (s, 3H, BnCH_3 min), 3.72 (s, 3H, BnCH_3 maj), 3.36–3.48 (m, 1H, 6aα-H), 2.80–3.20 (m, 2H, 6b-H, 10a-H), 1.20–2.30 (m, 7H, cyclohex.); ^{13}C NMR (75 MHz, $CDCl_3$, δ) 178.2, 176.4, 159.7, 158.3, 132.9, 131.5, 130.4, 129.8, 128.5, 127.8, 127.4, 127.3, 126.8, 125.6, 124.4, 117.6, 114.7, 114.3, 114.1, 104.1, 57.4, 55.6, 55.4, 45.5, 33.6, 33.2–33.6 (overlapped peaks), 32.3; IR (KBr, cm^{-1}) 3479(w), 3458(w), 3388(bs), 3059(w), 3025(w), 3002(w), 2933(s), 2860(m), 2837(m), 2360(w), 2340(w), 1776(m), 1699(s), 1610(m), 1513(s), 1451(m), 1390(m), 1302(m), 1251(s), 1174(s), 1031(s); HRMS *m/z* (M + Na^+) calcd 569.2412, found 569.2406. Anal. Calcd for $C_{35}H_{34}N_2O_4$: C, 76.90; H, 6.27; N, 5.12. Found: C, 76.84; H, 6.27; N, 4.89.

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