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Expanding the Synthetic Method and Structural Diversity Potential for the Intramolecular Aza Diels−Alder Cyclization

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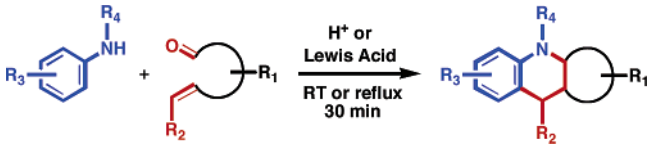

New experimental facets have been examined to expand upon the known methods for an aromatic variant of the intramolecular Aza Diels−Alder cyclization. The specific transformation under study is one that uses functionalized anilines and an aldehyde−olefin tether to provide tetrahydroquinoline cycloadducts under mild acidic conditions. Variations investigated encompass the use of N-alkylated anilines, including one with ring-constrained nitrogen, in the context of glycine, phenylalanine, and glyoxyl ester bridging elements; bridge components with structural perturbations; modified dienophile segments; and different acid catalysts. Substituted tetrahydroquinolines obtained from many of the preceding experiments were obtained in good chemical yield, generally in excess of 80%. Designed as a platform for combinatorial chemical synthesis, this reaction manifold accommodates a range of starting materials with structurally and electronically distinct characteristics. The results of this report, in combination with the discoveries from previous work in this area, enhance the ability of the intramolecular Aza Diels−Alder transformation to generate a diverse array of quinolinic structures with multiple stereogenic centers, many of which resemble lignan and arylnaphthalene-type natural products.

Introduction

The design and execution of combinatorial synthetic routes that lead to functionally, structurally, and stereochemically diverse compound libraries are to many current practitioners of organic synthesis what natural product and total synthesis represented to an earlier generation of chemists. The two fields share a common operational agenda: to devise streamlined, high-yielding syntheses with minimal reaction and purification steps, under defined stereochemical control, using readily attainable starting materials, and (if possible without compromising any of the preceding) with an innovative flair. Yet the principles of combinatorial synthetic design, which are at the core of the emerging discipline of *diversity-oriented synthesis*,[1] give rise to challenges distinct from those of *target-oriented synthesis*, originating in large measure from the inherent *forward* design nature of the former.[2]

One particular organic reaction class that lends itself to combinatorial thinking is the hetero Diels−Alder cyclization,[3] an expansive synthetic genre that includes the extensively studied *aza Diels−Alder* (or *imino Diels−Alder*) category of reactions.[4,5] Among these, of particular note is the subclass that involves the formation of an aromatic imine from an aniline and aldehyde, which subsequently reacts as a 2-azadiene with a suitable dienophile to form a tetrahydroquinoline (Figure 1A). Since its initial disclosure,[6] this type of three-component assembly, in which the imine is either preformed or generated in situ, has been employed to generate a range of heterocyclic products with stereochemical and functional group variability. As the iminium diene component is relatively electron-deficient, this reflects an *inverse electron demand* Diels−Alder transformation, which has influenced the type of components used in the reaction. Aside from studies that have explored the use of different diene and dienophile components, other distinct advances from a methodology perspective have centered on catalysis,[7] especially in the realm of asymmetric induction,[8] and

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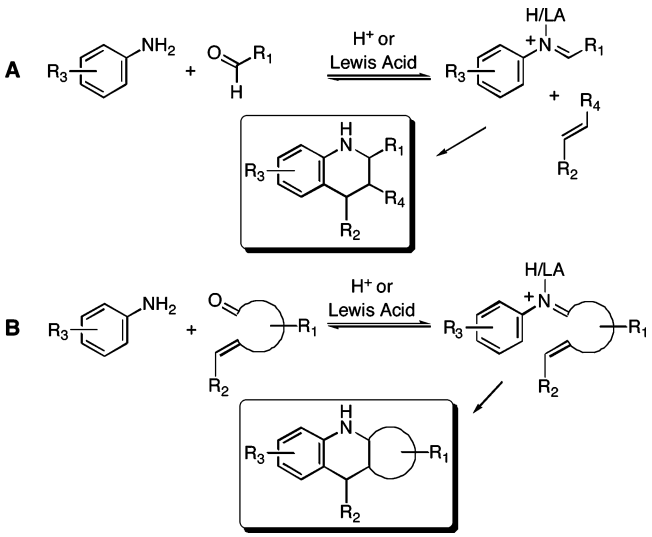
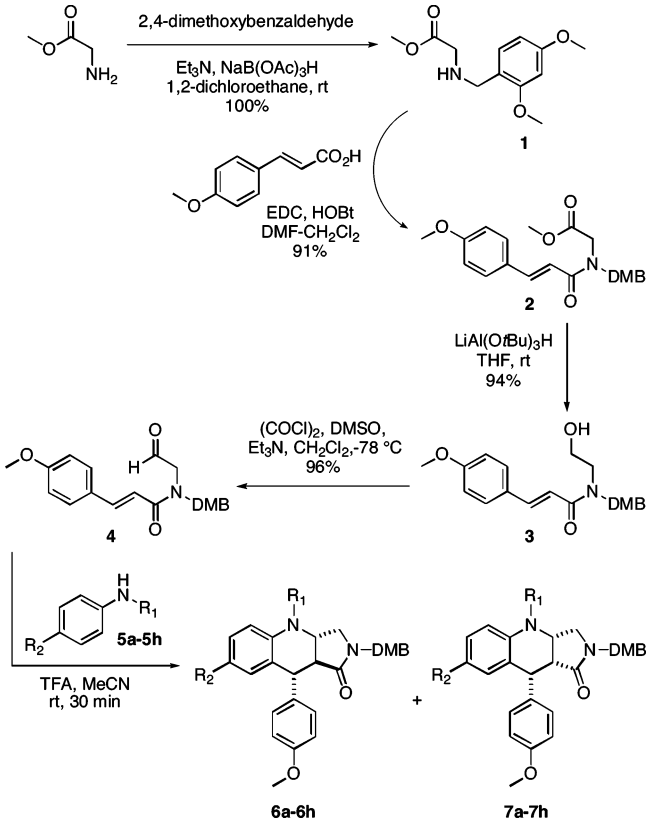


FIGURE 1. (A) Intermolecular and (B) intramolecular versions of the aza Diels−Alder cyclization.

the use of aqueous and nontraditional reaction media, such as ionic liquids[9−11] and supercritical carbon dioxide.[12,13]

The *intramolecular* implementation of the aza Diels−Alder using 2-azadienes has also been examined; here the added advantages include better control over the formation of stereoisomers and the creation of an additional ring.[14−16] This, in conjunction with the observation that the quinolinic substructure is present in a number of biologically active compounds,[17] has served as justification for developing combinatorial-based syntheses around this reaction framework. An early effort that specifically focused on such a translation developed an appropriately modular route and systematically investigated the suitability of various diene, dienophile, and intramolecular bridging units, both in solution and on a solid-phase matrix (Figure 1B).[18,19] The chemical diversity was derived from the anilines used to assemble the iminium diene, the cinnamic acids used as the dienophile, and the way the two were tethered through varied aldehyde−olefin components, which were based on salicylaldehyde, glyoxyl ester, glyoxyl amide, and amino acid aldehydes. The configurational diversity of the resulting products is reminiscent of the structures of biologically active arylnaphthalene lignan[20] and heterolignan[21] natural products. Other studies exploring the combinatorial avenues possible with the intermolecular aza Diels−Alder transformation have also been reported.[22,23]

SCHEME 1. Synthetic Route to Aza Diels−Alder Cyclization with N-Alkylated Anilines and 4-Methoxycinnamate-Based Dienophile with Glycine Bridge



In this report, we revisit the intramolecular aza Diels−Alder previously implemented on a combinatorial scaffold,[18] with an eye toward expanding the methodology associated with its use while adhering to the guiding precepts of combinatorial synthetic design.[24] The modifications we chose to examine were (1) use of secondary (N-alkylated) anilines, (2) use of various Brønsted and Lewis acid catalysts, (3) variation of the aldehyde−alkene bridge, and (4) a noncinnamic acid dienophile unit.

Results and Discussion

Although primary anilines were previously shown to be efficiently incorporated into the aza Diels−Alder reaction manifold, less investigated was the use of secondary anilines. The application of these as input would allow for the additional display of chemical diversity, and a convenient one at that, considering the availability (and ease of preparation) of N-alkylated anilines. For this investigation, the model system used to test the ensuing chemistry was a glycine-derived aldehyde tethered to a suitable dienophile, in this case, 4-methoxycinnamic acid (Scheme 1). This was prepared by first carrying out a reductive amination[25] using 2,4-dimethoxybenzaldehyde and glycine methyl ester, and the resulting secondary amine **1** was then coupled to the 4-methoxycinnamic acid under carbodiimide

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TABLE 1. Tetrahydroquinoline Products from the Aza Diels−Alder Using Variably Substituted N-Alkylated Anilines

entry	aniline	R1	R2	ratio **6**:**7**	combined yield (%)
1	**5a**	$-CH_3$	−H	7.0:1	96
2	**5b**	$-CH_3$	−F	4.3:1	95
3	**5c**	$-CH_3$	$-OCH_3$	4.6:1	95
4	**5d**	$-CH_2CH_3$	−H	8.1:1	91
5	**5e**	$-CH_2CH{=}CH_2$	−H	4.2:1	98
6	**5f**	$-CH(CH_3)_2$	−H	2.4:1	89
7	**5g**	−cyclohexyl	−H	2.0:1	42
8	**5h**	−Ph	−H	5.5:1	91

conditions to yield ester **2**. The 2,4-dimethoxybenzyl (DMB) group was used based on the prior finding that it serves both as a directing element in the cyclization reaction and as an isosteric and isoelectronic model for the corresponding resin linker when prepared on a solid-phase matrix.[18]

Selective reduction of the ester moiety in **2** using lithium tri-*tert*-butoxyaluminohydride furnished alcohol **3** without perturbing the cinnamyl amide functionality. This was followed by a Swern oxidation to yield the cyclization precursor aldehyde **4**. The aza Diels−Alder reaction was then performed using each of eight different secondary anilines, representing a modest assortment of electronic and structural diversity (Table 1). The conditions used were quite mild, using 3 equiv of trifluoroacetic acid (TFA) at room temperature for 30 min. In all cases, two diastereomers were formed, although the ring fusion trans, trans isomer **6** predominated. Whether *N*-methylaniline was substituted with electron-acceptor fluorine (**5b**) or an electron-donor methoxy (**5c**), the yields of the formed tetrahydroquinoline were quite high and comparable to unsubstituted *N*-methylaniline (**5a**), although the stereoisomer ratios were somewhat diminished. The remaining four entries were N-alkylated anilines possessing larger substituents, with ethyl (**5d**), *n*-propenyl (**5e**), isopropyl (**5f**), cyclohexyl (**5g**), and phenyl (**5h**). Yields of cycloadduct with these anilines were also on the order of 90% or greater, with the notable exception of the cyclohexyl aniline, which was less than half that of the others (Table 1). The reaction with **5a** was also performed with several other solvents in addition to acetonitrile; combined yields and the ratio of **6**:**7** were generally reduced in all cases (EtOAc: 86%, 6.8:1; CH_2Cl_2: 71%, 4.0:1; $CHCl_3$: 79%, 3.0:1; CCl_4: 80%, 3.5:1).

When visualizing the transition states leading to the cycloadducts, the configuration obtained by way of an *endo* transition state (defined based on position of the phenyl substituent of the olefin) leads to the observed product stereochemistry (Figure 2). While the major isomers **6a**−**g** reflect the expected pathway, the presence of the cis, cis diastereomers **7a**−**g** suggests that the reaction is in fact not concerted and may constitute a stepwise process that represents a formal aza Diels−Alder transformation.[26]

The relative stereochemical disposition of the favored endo product was confirmed by NMR analysis, in accord with the same stereoisomer form reported previously for other products of the intramolecular aza Diels−Alder cyclization.[18] The stereochemistry for the cis, cis isomer **7a** was confirmed by NMR spectroscopy; on irradiation of H_2, nOe values of 5.9 and 8.4% were observed with H_1 and H_3, respectively (Figure 3). In addition, a similar enhancement of 8.1% was detected between H_2 and H_3 upon irradiation of H_3.

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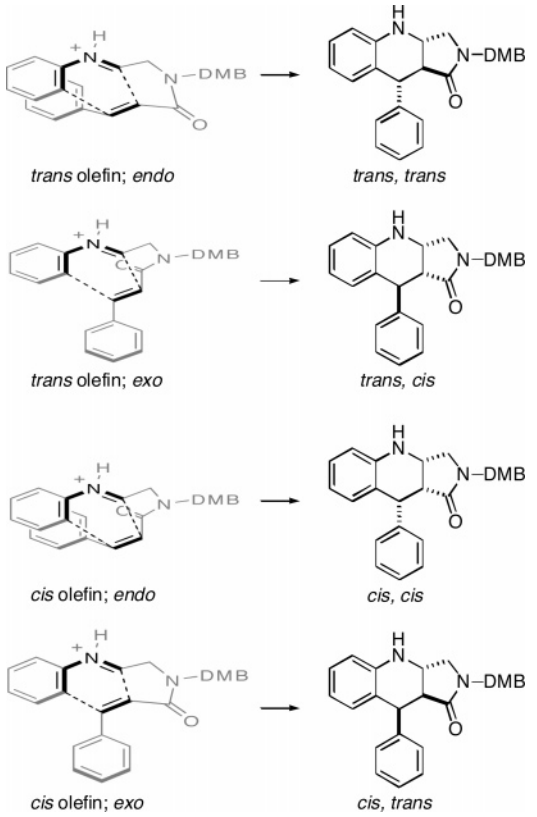

FIGURE 2. Transition states for a hypothetical concerted aza Diels−Alder reaction leading to corresponding stereoisomeric products.

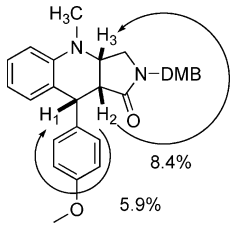

FIGURE 3. Measured nOe values for cis-fused cycloadduct **7a**.

Invoking a mechanism that involves two discrete steps (intramolecular attack of the iminium and aromatic closure onto the benzylic cation) that are separated by a possible bond rotation can lead to the formation of the observed tetrahydroquinolines (Figure 4). In forming both **6** and **7**, the reaction could be rationalized as a consequence of maximizing secondary orbital overlap through π-stacking of the aromatic groups, leading to the respective endo products.

After the success with this N-alkylated aniline series, a representative entry (the *N*-methylaniline) was chosen to explore the effect of other acid catalysts aside from TFA. Specifically, we sought to determine whether the nature of the catalyst (i.e., Brønsted versus Lewis acid) would have a discernible impact upon the stereoselectivity of the intramolecular cyclization with substituted anilines. Strong Brønsted acids such as TFA have long since been known to catalyze the intermolecular imino Diels−Alder cyclization,[27] although a variety of other acidic catalysts have also been used, including Lewis acids such as the lanthanide triflates[28] and bismuth(III) chloride.[29,30]

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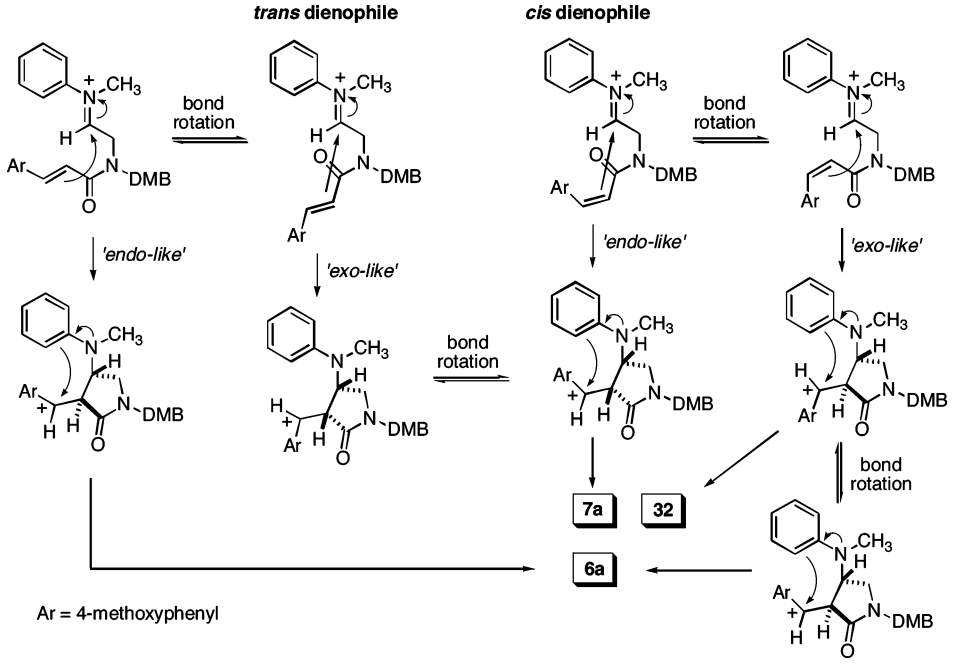
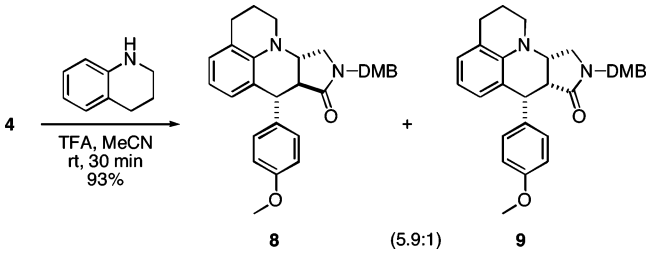


FIGURE 4. Possible two-step pathways for the *trans*- and *cis*-4-methoxyphenyl dienophile cyclizations leading to observed stereoisomeric products.

TABLE 2. Effect of Different Acid Catalysts upon Product Formation for the Aza Diels−Alder Cyclization with *N*-Methylaniline

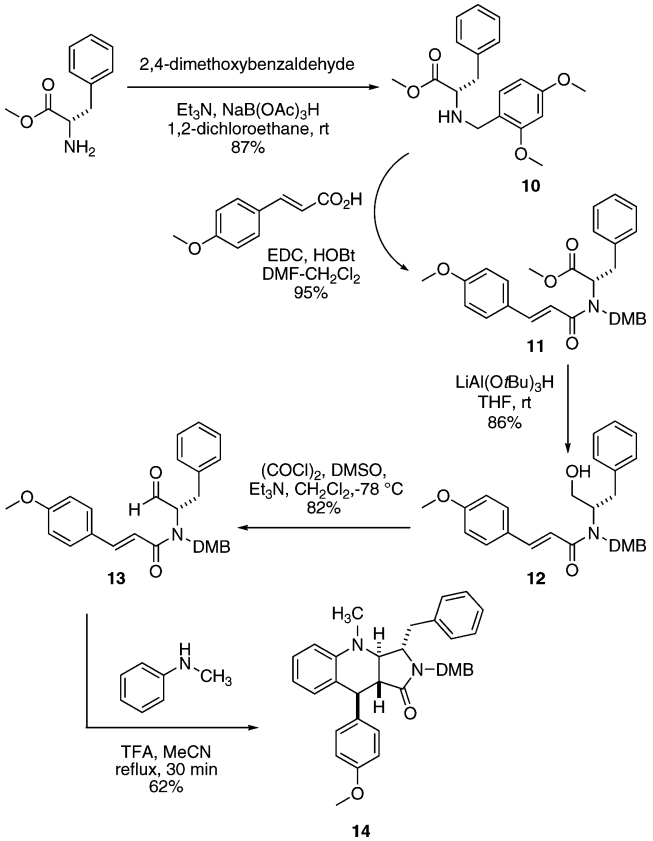
entry	catalyst	ratio **6**:**7**	combined yield (%)
1	TFA	7.0:1	96
2	TFMSA	6.6:1	84
3	acetic acid	6.0:1	87
4	*p*-TsOH	5.5:1	52
5	(L)-tartaric acid	5.8:1	89
6	$Yb(OTf)_3$	6.5:1	98
7	$BiCl_3$	6.3:1	86

SCHEME 2. Aza Diels−Alder Cyclization of 1,2,3,4-Tetrahydroquinoline and 4-Methoxycinnamate-Based Dienophile with Glycine Bridge



In addition to $Yb(OTf)_3$ and $BiCl_3$, trifluoromethanesulfonic acid (TFMSA), acetic acid, and tartaric acid were tested for the reaction with **5a**. The results showed that the identity of the acid had a negligible effect on either selectivity or overall yields (Table 2). Since the transition state would not be expected to accommodate the acid (at least not on the nitrogen, which is coordinately saturated after imine formation), the influence of the catalyst is likely felt at the stage of aldehyde activation, immediately prior to generation of the imine. One cannot rule out partial coordination of the catalyst to the amide carbonyl, although it seems unlikely that an energetically favorable transition state would hold two positive charges. Despite the lack of improvement in product isomer distribution, it was

SCHEME 3. Aza Diels−Alder Cyclization with *N*-Methylaniline and 4-Methoxycinnamate-Based Dienophile with L-Phenylalanine Bridge



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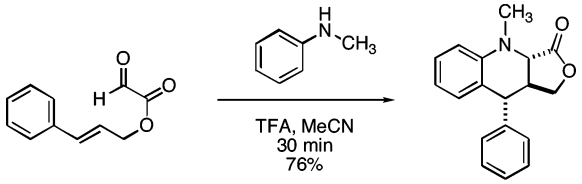
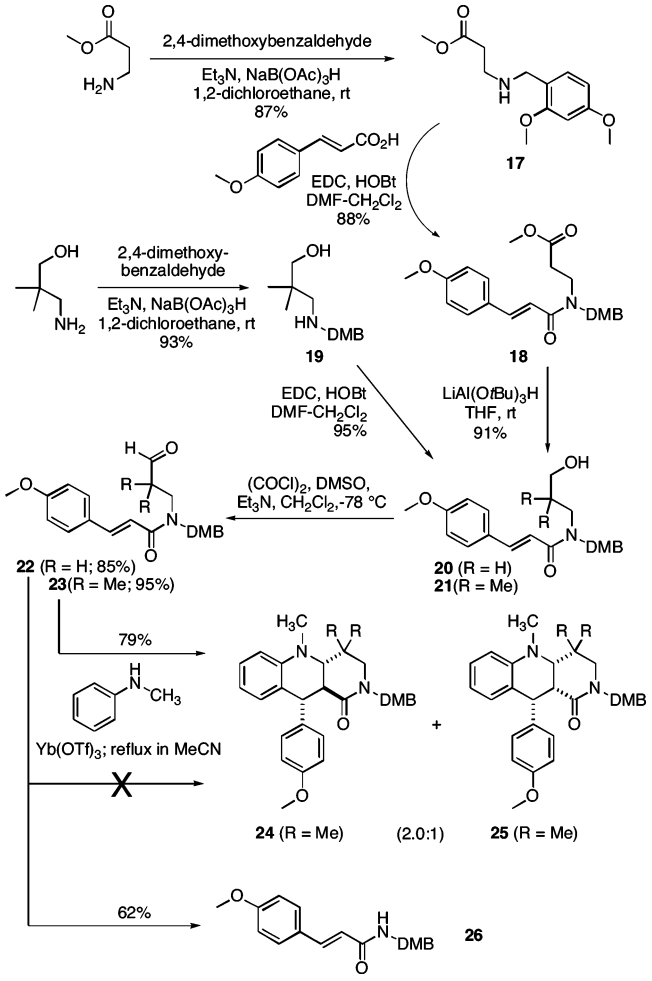
SCHEME 4. Aza Diels−Alder Cyclization with *N*-Methylaniline and Cinnamyl-Based Dienophile with Glyoxyl Ester Bridge



15 **16**

SCHEME 5. Synthetic Route to Aza Diels−Alder Cyclization for N-Alkylated Anilines and 4-Methoxycinnamate-Based Dienophile with 3-Aminopropionate-Derived Bridges

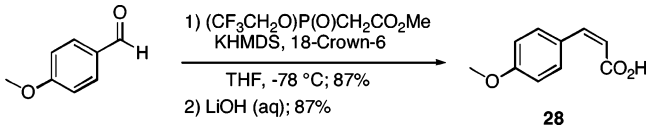


agreeable to see that less harsh agents, such as the acetate and tartrate acids, could replace TFA with comparable results.

Before moving beyond the system presented in Scheme 1, one additional aniline variation was examined, in which the *N*-alkyl substitution was rigidified through inclusion in a saturated ring. Using 1,2,3,4-tetrahydroquinoline as the aniline moiety, we obtained cycloadducts **8** and **9** in an approximate 6:1 ratio with an overall yield of 93% (Scheme 2), a satisfying result considering the increased complexity and constraint that the polycyclic product possessed. The stereochemistry was confirmed for both isomers with NMR by evaluating the coupling constants and nOe values.

Next, a series of modifications at the bridging unit (the segment tethering the aldehyde and olefin) were explored. First

SCHEME 6. Synthesis of *cis*-4-Methoxycinnamic Acid



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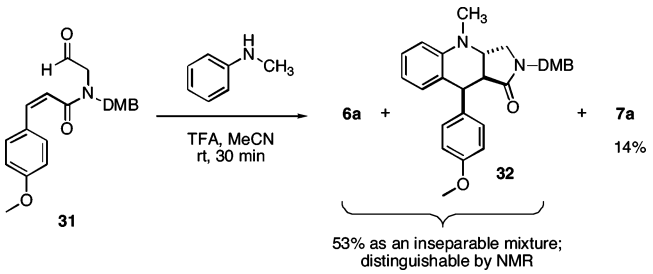
SCHEME 7. Aza Diels−Alder Cyclization with *N*-Methylaniline and *cis*-Cinnamyl-Based Dienophile with Glycine Bridge

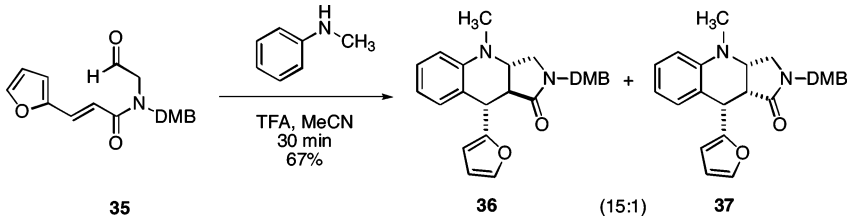


we examined the consequence of moving from an unsubstituted glycine unit to a chiral amino acid, a modification previously shown to accommodate the aza Diels−Alder cyclization when using primary anilines.[18] An aza Diels−Alder precursor was prepared from L-phenylalanine methyl ester in the same manner in which precursor **4** had been prepared from glycine; namely, via reductive amination of the amino acid ester with DMB to yield **10**, which was then coupled to 4-methoxycinnamic acid to form ester **11**, followed by reduction to alcohol **12** that was then oxidized to aldehyde **13** using the Swern protocol. Under essentially the same conditions as employed previously, albeit under refluxing conditions, the Phe-based **13** did form exclusively the expected trans, trans adduct **14** in 62% yield (Scheme 3), the benzyl moiety effectively controlling the facial selectivity of the cyclization. When performed at room temperature, the reaction delivered only 29% of **14**, the remainder of the mass balance being recovered starting material.

The cyclization was then tested with a non-amino acid bridge, the glyoxyl ester of cinnamate, **15**. The resulting tetrahydroquinoline **16**, obtained in a 76% yield, bears a ring fusion to a lactone instead of a lactam (and with a reversed carbonyl orientation) as seen in the previous products (Scheme 4).

Investigating the possibility of elongating the bridging unit was next on the agenda, specifically, modifying the amino acid linker by adding an additional methylene within the "backbone". In this manner, the larger ring in the polycyclic product would represent another degree of structural diversity, with a conformational nature distinct from that of the earlier five-membered rings. Further modifications could then be based on adding various functional groups on such an expanded bridge. To begin, the glycine unit was replaced with a 3-aminopropionate ("β-alanine") unit, and the same protocol was followed to prepare the requisite aldehyde−alkene intermediate **22**, accessed by way of formation of ester **17**, amide **18**, and alcohol **20** (Scheme 5). When subjected to the aza Diels−Alder conditions, however, the anticipated six-membered lactam cycloadduct was not obtained. Rather, cinnamyl amide byproduct **26** was isolated. We rationalized that this may be due to a "retro-Michael"-type of side reaction, in which deprotonation at the α-carbon of aldehyde **22** results in the formation of a double bond while expelling the amide nitrogen, thereby forming **26** and acrylaldehyde (the latter, though, was never isolated). The results were the same irrespective of the choice of catalyst; trials included using TFA, $Yb(OTf)_3$, and $BiCl_3$.

SCHEME 8. Aza Diels−Alder Cyclization with *N*-Methylaniline and Furanyl-Based Dienophile with Glycine Bridge



If such an elimination reaction were to blame, we reasoned that removal of the acidic protons by alkyl substitution should circumvent the problem. A dimethylated analogue, **23**, was prepared in a manner commensurate with the earlier elaboration of bridge units, except here the preparation began with the reductive amination of 3-amino-2-dimethylpropanol with 2,4-dimethoxyenzaldehyde to yield **19**. This was followed by EDC coupling of **19** with 4-methoxycinnamic acid to generate the 4-methoxycinnamyl amide alcohol **21**, which was then oxidized to cyclization precursor **23**.

The subsequent aza Diels−Alder transformation with *N*-methylaniline proved relatively sluggish and required refluxing with $Yb(OTf)_3$, leading to a combined yield of 77% for the product diastereomers **24** and **25**. In this case, under refluxing conditions the TFA-mediated reaction led to crude isolates that were much less clean than those obtained when using $Yb(OTf)_3$. The observed isomer ratio of 2:1 may be attributable to ring products possessing comparable energetic stability, coupled to a prolonged opportunity for single-bond rotation to occur before final ring closure (this assuming that a two-step mechanism is responsible as raised earlier). The presence of the *gem*-dimethyl groups might also contribute, both in terms of sites of additional steric contact or in conformational biasing of what might otherwise be a very flexible alkyl bridge.

Moving on to considerations of the dienophile, we thought to examine the consequence of using an olefin with *Z* stereochemistry. The commercial availability of the *trans*-cinnamic-type acids is far larger than that of their cis counterparts, and therefore the former represents the more useful pool for starting materials in a combinatorial synthesis. Nonetheless, if the cis-based dienophiles yielded distinctly different product stereoisomers (or stereoisomeric distributions), then that could justify their inclusion into a combinatorial synthesis. To make the most direct comparison with a previously performed experiment with a trans olefin, we prepared the *cis*-4-methoxycinnamic acid (Scheme 6).

Using a variation of the Still−Gennari method,[31] we hydrolyzed the methyl ester of the olefin, **27**, to yield acid **28**. A sequence analogous to the preparation of aldehyde−olefin **4** was followed: acid **28** was coupled to glycine−DMB amine **1** to form ester **29** (88%), which was reduced to alcohol **30** (94%), followed by oxidation to the cyclization precursor **31** (91%). After application of the standard aza-Diels−Alder conditions, **31** provided a mixture of three identifiable stereoisomers (Scheme 7). The mixture of the chromatographically inseparable **6a** and **32**, found in a 1:1 ratio, both maintain a trans orientation at the ring fusion with the lactam ring. But only **32** retains the original stereochemistry of the cis alkene, and **6a** reflects, as described previously, a possible isomerization event through a two-step mechanism (Figure 4). The final result is also influenced by the possibility that the cis double bond loses its integrity when exposed to acid. This additional effect was bolstered by the results from a test reaction in which morpholine was added to **31** under the cyclization reaction conditions; upon quenching and workup, the NMR spectrum indicated the presence of two forms of the alkene.

A final adjustment to the dienophile was made by replacing the phenyl unit with a smaller, and electronically distinct, aromatic ring. A furan group was selected, and a sequence was initiated that again mirrored the assembly of cyclization precursor **4**: glycine methyl ester was coupled to *trans*-3-(2-furyl)acrolein to form ester **33** (97%), reduced to alcohol **34** (96%), followed by Swern oxidation to generate **35**. The aza Diels−Alder reaction provided the endo-derived tetrahydroquinoline **36** (92%) as the predominant product with a combined yield of 67% (Scheme 8). While the efficiency is lower than that of the 4-methoxycinnamate dienophile, this recovery nevertheless represents a usable outcome and demonstrates the feasibility of including nonphenyl moieties in the aza Diels−Alder combinatorial synthesis.

In summary, we have demonstrated that there are chemical aspects of the intramolecular Aza Diels−Alder that can be modified or tuned, which enrich the combinatorial chemical possibilities and will supplement what had earlier been demonstrated with this system.[18] Previously, a variety of amino acids and electron-rich and -poor cinnamyl dienophiles and primary anilines contributed to structural diversity; to this we can now introduce selected variations on the dienophile and linking bridge, as well as a range of secondary anilines that form the diene component. Further, it is now possible to employ other catalysts in addition to those originally investigated. Taken together, these accumulated results will permit the use of this transformation for the generation of chemical libraries comprising a variety of structurally diverse compounds built around a quinoline core.

Experimental Section

General procedures for preparation of the reported compounds may be found in the Supporting Information.

(2,4-Dimethoxybenzylamino)-acetic Acid Methyl Ester (1). 2,4-Dimethoxybenzaldehyde (1.50 g, 9.0 mmol) was dissolved in dichloroethane (100 mL); to this was added triethylamine (1.88 mL, 13.5 mmol) and glycine methyl ester (2.55 g, 13.5 mmol) and stirred. To this solution was added, with vigorous stirring, sodium triacetoxyborohydride (3.83 g, 18.1 mmol). The solution was stirred under argon for 24 h, after which it was quenched with saturated $NaHCO_3$ (60 mL), extracted with CH_2Cl_2 (3 × 20 mL), and then concentrated. Flash chromatography was performed using CH_2Cl_2/EtOAc (1:4) to give a clear thick liquid in quantitative yield. ^1H NMR (500 MHz, $CDCl_3$) δ 7.12 (d, J = 8.0 Hz, 1H), 6.45 (d, J = 2.0 Hz, 1H), 6.42 (dd, J = 8.0, 2.5 Hz, 1H), 4.15 (q, J = 7.3 Hz, 2H), 3.81 (s, 3H), 3.79 (s, 3H), 3.74 (s, 3H), 3.37 (s, 1H), 1.25 (t, J = 7.3 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.7, 160.5, 158.9, 130.8, 120.34, 103.9, 98.7, 60.8, 55.6, 55.5, 50.3, 48.3, 14.4;

(31) Still, W. C.; Gennari, C. *Tetrahedron Lett.* **1983**, *24*, 4405−4408.

IR 3340, 2996, 2949, 2833, 1736, 1613, 1578, 1497 cm^{-1}. HRMS calcd for $C_{13}H_{18}O_4N$ 238.10793 (M^+), found 238.10803.

{(2,4-Dimethoxybenzyl)-[3-(4-methoxyphenyl)-acroyl]amino}-acetic Acid Methyl Ester (2). To a solution of **1** (2.00 g, 8.4 mmol) in DMF/CH_2Cl_2 (21 mL, 1:3) was dissolved 4-methoxycinnamic acid (2.23 g, 12.3 mmol). To this was added HOBt (0.162 g, 1.2 mmol) and EDCI (2.36 g, 12.3 mmol). The reaction mixture was allowed to stir for 12 h. The organic layer was washed with 50 mL of 0.1 M HCl solution and extracted with CH_2Cl_2 (3 × 20 mL). The organic layers were combined and successively washed with distilled water (1 × 100 mL) and saturated $NaHCO_3$ (1 × 60 mL), dried over $MgSO_4$, and then concentrated. The residue was purified by flash chromatography using CH_2Cl_2/EtOAc (8:1) as eluant to afford 3.04 g (91%) as a white flaky solid. mp 94.2−95.5 °C; white flaky solid. ^1H NMR (500 MHz, $CDCl_3$) δ 7.73 (d, J = 15.0 Hz, 1H), 7.46 (d, J = 8.5 Hz, 2H), 7.09 (d, J = 8.0 Hz, 1H), 6.91 (d, J = 14.5 Hz, 1H), 6.87 (d, J = 8.5 Hz, 2H), 6.48 (d, J = 2.0 Hz, 1H), 6.45 (dd, J = 7.5 Hz, 1H), 4.66 (s, 2H), 4.15 (s, 2H), 3.82 (s, 3H), 3.80 (s, 6H), 3.73 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 170.0, 167.8, 160.8, 160.7, 158.3, 143.0, 129.4, 129.0, 128.0, 116.7, 114.7, 114.1, 103.9, 98.6, 55.4, 55.3, 55.2, 52.0, 47.6, 47.5, 44.8; IR 3072, 3008, 2943, 2844, 1754, 1643, 1590, 1508, 1438 cm^{-1}; HRMS calcd for $C_{22}H_{25}O_6N$ 399.16819 (M^+), found 399.16823.

***N*-(2,4-Dimethoxybenzyl)-*N*-(2-hydroxyethyl)-3-(4-methoxyphenyl)-acryl Amide (3).** To a solution of **2** (3.04 g, 7.6 mmol) in 50 mL of THF was added via a syringe 1 M lithium tri-*tert*-butoxyaluminohydride (21.8 mL, 21.8 mmol). The mixture was stirred for 6 h, and 60 mL of diethyl ether was added, followed by dropwise addition of 5 mL of 10% $KHSO_4$. The mixture was left to stir until two separate layers formed. Vacuum filtration was used to remove the white salt layer, which was washed with 60 mL of EtOAc. The combined organic layers were washed with brine (40 mL), dried ($MgSO_4$), and concentrated to give a thick light-green slurry. The slurry was chromatographed using silica gel to afford 2.79 g (98%) as clear glass. ^1H NMR (500 MHz, $CDCl_3$) δ 7.72 (d, J = 15.5 Hz, 1H), 7.42 (d, J = 9.0 Hz, 2H), 7.03 (d, J = 8.5 Hz, 1H), 6.86 (d, J = 9.0 Hz, 2H), 6.77 (d, J = 15.5 Hz, 1H), 6.48 (d, J = 2.0 Hz, 1H), 6.47 (dd, J = 9.0, 2.0 Hz, 1H), 4.62 (s, 2H), 4.12 (t, J = 4.5 Hz, 1H), 3.81 (s, 3H), 3.80 (s, 3H), 3.797 (s, 3H), 3.77 (m, 1H), 3.63 (t, J = 4.8 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) 169.7, 160.9, 160.6, 156.0, 143.1, 129.4, 128.2, 127.8, 116.8, 114.9, 114.1, 113.8, 104.0, 98.6, 62.4, 55.3, 55.3, 55.2, 50.4, 47.8; IR 3382, 3072, 2996, 2932, 1643, 1584, 1520, 1444 cm^{-1}; HRMS calcd for $C_{21}H_{25}O_5N$ 371.17327 (M^+), found 371.17336.

***N*-(2,4-Dimethoxybenzyl)-3-(4-methoxyphenyl)-*N*-(2-oxoethyl)-acryl Amide (4).** Oxalyl chloride (179 μL, 1.4 mmol) was dissolved in 6 mL of CH_2Cl_2 and cooled to −78 °C. DMSO (200 μL, 2.8 mmol) dissolved in 2 mL of CH_2Cl_2 was added cautiously, and the mixture was allowed to stir for 15 min, after which the alcohol (210 mg, 0.6 mmol) was added, and the reaction was stirred for 1 h at −78 °C. Triethylamine (437 μL, 3.1 mmol) was added, and the reaction was stirred for another 30 min and allowed to warm to ambient temperature. The mixture was quenched with 30 mL of saturated NH_4Cl and extracted using CH_2Cl_2 (3 × 15 mL), washed with brine, and dried using $MgSO_4$. Flash chromatography was performed using hexane/CH_2Cl_2/EtOAc (1:1:1) system to yield a yellow-green thick liquid weighing 199 mg. Yield 95%. ^1H NMR (500 MHz, $CDCl_3$) δ 9.44 (s, 1H), 7.74 (d, J = 15.5 Hz, 1H), 7.50 (d, J = 8.5 Hz, 2H), 7.08 (d, J = 8.0 Hz, 1H), 6.98 (d, J = 15.5 Hz, 1H), 6.91 (d, J = 8.5 Hz, 2H), 6.47 (m, 2H), 4.66 (s, 2H), 4.10 (s, 2H), 3.82 (s, 3H), 3.82 (s, 3H), 3.80 (s, 3H), 3.78 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 198.5, 167.9, 160.95, 160.92, 158.3, 143.1, 129.5, 129.4, 127.8, 116.5, 114.22, 114.16, 104.1, 98.7, 56.4, 55.4, 55.33, 55.27, 55.1, 48.2; IR 3061, 2996, 2938, 2844, 1736, 1643, 1590, 1514, 1444 cm^{-1}; HRMS calcd for $C_{21}H_{23}O_5N$ was 369.15678 (M^+), found 369.15716.

2-(2,4-Dimethoxybenzyl)-9-(4-methoxyphenyl)-4-methyl-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (6a). To a solution of aldehyde **4** (155 mg, 0.4 mmol) in 5 mL of MeCN were added *N*-methylaniline (65 μL, 0.6 mmol) and then TFA (94 μL, 1.2 mmol). The reaction mixture was stirred for 30 min, quenched with $NaHCO_3$ (30 mL), and extracted using CH_2Cl_2 (3 × 10 mL). The combined organic layers were dried ($MgSO_4$) and concentrated to give a light brown film. Flash chromatographed with silica gel using hexane/CH_2Cl_2/EtOAc (3:1:1) eluant to afford 159 mg (83%) of 6**a** as a white solid and 22 mg (12%) of **7a** as a film. mp 173.8−174.8 °C; white crystalline solid. ^1H NMR (500 MHz, $CDCl_3$) δ 7.18 (dd, J = 8.5 Hz, 2H), 7.12 (d, J = 8.0 Hz, 1H), 7.11 (t, J = 6.0 Hz, 1H), 6.86 (d, J = 8.5 Hz, 2H), 6.81 (d, J = 8.5 Hz, 1H), 6.76 (d, J = 8.0 Hz, 1H), 6.68 (t, J = 7.5 Hz, 1H), 6.45 (d, J = 2.0 Hz, 1H), 6.42 (dd, J = 8.0, 2.5 Hz, 1H), 4.49 (d, J = 14.5 Hz, 1H), 4.36 (d, J = 15.0 Hz, 1H), 4.32 (d, J = 12.0 Hz, 1H), 3.80 (s, 3H), 3.79 (s, 6H), 3.48 (dd, J = 8.8, 6.5 Hz, 1H), 3.37 (ddd, J = 11.5, 9.8, 6.5 Hz, 1H), 3.22 (t, J = 8.0 Hz, 1H), 2.92 (t, J = 12.0 Hz, 1H), 2.76 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.5, 160.5, 158.6, 158.1, 148.3, 136.0, 131.03, 131.0, 130.3, 128.8, 127.3, 119.1, 117.0, 113.7, 113.4, 104.2, 98.4, 59.5, 55.3, 55.1, 50.6, 49.8, 45.1, 40.4, 36.2; IR 3061, 2996, 2932, 2874, 2827, 1695, 1613, 1503, 1462, 1362 cm^{-1}; HRMS calcd for $C_{28}H_{30}O_4N_2$ was 458.22056 (M^+), found 458.22002.

2-(2,4-Dimethoxybenzyl)-9-(4-methoxyphenyl)-4-methyl-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (7a). Light yellow film. ^1H NMR (500 MHz, CD_2Cl_2) δ 7.19 (dt, J = 7.8, 1.5 Hz, 1H), 7.13 (dt, J = 11.0, 2.5 Hz, 2H), 6.99 (dd, J = 7.3, 2.0 Hz, 1H), 6.81 (dt, J = 9.1, 2.5 Hz, 2H), 6.71 (dt, J = 7.5, 1.5 Hz, 1H), 6.61 (d, J = 8.0 Hz, 1H), 6.52 (d, J = 8.0 Hz, 1H), 6.40 (d, J = 2.5 Hz, 1H), 6.29 (dd, J = 8.5, 2.5 Hz, 1H), 4.42 (d, J = 15.0 Hz, 1H), 4.40 (d, J = 2.5 Hz, 1H), 4.16 (d, J = 15.5 Hz, 1H), 3.95 (ddd, J = 7.9, 5.3, 2.5 Hz, 1H), 3.76 (s, 3H), 3.74 (s, 3H), 3.71 (s, 3H), 3.29 (m, 2H), 3.25 (dd, J = 8.3, 3.0 Hz, 1H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 173.1, 160.7, 158.8, 158.5, 145.9, 135.7, 129.6, 129.1, 128.3, 126.4, 118.1, 116.8, 114.0, 112.2, 104.6, 98.4, 55.7, 55.6, 55.5, 50.6, 49.9, 42.7, 40.7, 36.0; IR 3066, 2996, 2938, 2833, 1678, 1596, 1509, 1456 cm^{-1}; HRMS calcd for $C_{28}H_{30}O_4N_2$ was 458.22056 (M^+), found 458.22056.

2-(2,4-Dimethoxybenzyl)-7-fluoro-9-(4-methoxyphenyl)-4-methyl-2,3,3a,4, 9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (6b). Prepared according to the general procedure for the cyclization. Yield: 77%; mp 199.0−200.1 °C; white solid. ^1H NMR (500 MHz, $CDCl_3$) δ 7.16 (d, J = 8.5 Hz, 2H), 7.12 (d, J = 8.0 Hz, 1H), 6.87 (d, J = 8.5 Hz, 2H), 6.83 (dt, J = 8.3, 3.0 Hz, 1H), 6.73 (dd, J = 8.0, 5.0 Hz, 1H), 6.48 (ddd, J = 9.8, 3.0, 1.0 Hz, 1H), 6.44 (d, J = 2.0 Hz, 1H), 6.42 (dd, J = 8.3, 2.0 Hz, 1H), 4.48 (d, J = 14.5 Hz, 1H), 4.36 (d, J = 15.0 Hz, 1H), 4.27 (d, J = 12.5 Hz, 1H), 3.80 (s, 6H), 3.79 (s, 3H), 3.48 (dd, J = 8.5, 6.0 Hz, 1H), 3.32 (ddd, J = 9.5, 9.3, 6.5 Hz, 1H), 3.21 (t, J = 9.3 Hz, 1H), 2.91 (t, J = 11.8 Hz, 1H), 2.73 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.3, 160.5, 158.5, 158.3, 157.3, 155.4, 144.7, 135.2, 131.0, 130.6, 130.6, 130.2, 117.3, 117.1, 116.9, 114.4, 114.3, 114.0, 113.9, 104.2, 98.4, 59.8, 55.3, 55.1, 50.4, 49.7, 45.3, 40.5, 36.7; IR 2996, 2938, 2874, 2833, 1701, 1614, 1584, 1503, 1503, 1409, 1368 cm^{-1}; HRMS calcd for $C_{28}H_{29}O_4N_2F$ 476.21114 (M^+), found 476.21135.

2-(2,4-Dimethoxybenzyl)-7-fluoro-9-(4-methoxyphenyl)-4-methyl-2,3,3a,4, 9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (7b). Prepared according to the general procedure for the cyclization. Yield: 18%; clear film. ^1H NMR (500 MHz, CD_2Cl_2) δ 7.13 (d, J = 9.0 Hz, 2H), 6.88 (dt, J = 8.9, 3.0 Hz, 1H), 6.82 (d, J = 9.5 Hz, 2H), 6.78 (dd, J = 8.5, 3.0 Hz, 1H), 6.56 (d, J = 8.0 Hz, 1H), 6.53 (dd, J = 9.3, 5.0 Hz, 1H), 6.41 (d, J = 2.0 Hz, 1H), 6.31 (dd, J = 8.3, 3.0 Hz, 1H), 4.44 (d, J = 15.0 Hz, 1H), 4.37 (d, J = 3.5 Hz, 1H), 4.15 (d, J = 14.5 Hz, 1H), 3.93 (m, 1H), 3.77 (s, 3H), 3.76 (s, 3H), 3.72 (s, 3H), 3.32 (dd, J = 10.5, 5.5 Hz, 1H), 3.26 (m, 2H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 129.7, 129.1, 116.0, 114.1, 104.5, 98.43, 55.5, 50.4, 49.8, 43.0, 40.7, 36.2, 24.4; HRMS calcd for $C_{28}H_{29}O_4N_2F$ 476.21114 (M^+), found 476.2106.

2-(2,4-Dimethoxybenzyl)-7-methoxy-9-(4-methoxyphenyl)-4-methyl-2,3,3 a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (6c). Prepared according to the general procedure for the cyclization.

Yield: 78%; mp 178.5−179.7 °C; white solid. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.17 (d, J = 8.5 Hz, 2H), 7.11 (d, J = 8.0 Hz, 1H), 6.85 (d, J = 8.5 Hz, 2H), 6.77 (d, J = 9.0 Hz, 1H), 6.71 (dd, J = 9.0, 3.0 Hz, 1H), 6.44 (d, J = 2.0 Hz, 1H), 6.43 (dd, J = 8.3, 2.0 Hz, 1H), 6.34 (d, J = 2.5 Hz, 1H), 4.48 (d, J = 14.5 Hz, 1H), 4.35 (d, J = 14.5 Hz, 1H), 4.29 (d, J = 12.0 Hz, 1H), 3.79 (s, 3H), 3.78 (s, 6H), 3.57 (s, 3H), 3.46 (dd, J = 8.8, 6.0 Hz, 1H), 3.27 (m, 2H), 2.90 (t, J = 11.3 Hz, 1H), 2.71 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.5, 160.4, 158.5, 158.1, 152.7, 142.6, 135.7, 130.9, 130.2, 117.0, 116.5, 114.5, 113.7, 112.8, 104.2, 98.4, 60.1, 55.4, 55.3, 55.1, 50.7, 49.8, 45.3, 40.4, 37.0; IR 3066, 3031, 3002, 2938, 2874, 2836, 1689, 1607, 1579, 1491, 1462, 1421, 1369 cm^{-1}; HRMS calcd for $C_{29}H_{32}O_5N_2$ 488.23112 (M^+), found 488.23100.

2-(2,4-Dimethoxybenzyl)-7-methoxy-9-(4-methoxyphenyl)-4-methyl-2,3,3 a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (7c). Prepared according to the general procedure for the cyclization. Yield: 17%; light yellow film. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.16 (dd, J = 9.5, 2.0 Hz, 2H), 6.81 (dt, J = 9.0, 2.5 Hz, 2H), 6.76 (dd, J = 8.8, 3.0 Hz, 1H), 6.66 (d, J = 3.0 Hz, 1H), 6.56 (d, J = 9.0 Hz, 1H), 6.45 (d, J = 8.5 Hz, 1H), 6.40 (d, J = 2.5 Hz, 1H), 6.28 (dd, J = 8.3, 2.5 Hz, 1H), 4.42 (d, J = 15.5 Hz, 1H), 4.38 (d, J = 3.5 Hz, 1H), 4.14 (d, J = 15.0 Hz, 1H), 3.95 (ddd, J = 8.6, 5.8, 2.0 Hz, 1H), 3.76 (s, 3 H), 3.75 (s, 3H), 3.72 (s, 3H), 3.70 (s, 3H), 3.31 (dd, J = 5.8, 3.5 Hz, 1H), 3.27 (t, J = 5.3 Hz, 1H), 3.24 (dd, J = 10.8, 2.5 Hz, 1H), 2.71 (s, 3H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 173.3, 160.6, 158.7, 158.5, 152.8, 139.9, 135.3, 129.4, 129.1, 128.7, 116.7, 115.4, 114.5, 113.9, 113.3, 113.2, 104.5, 98.4, 55.8, 55.7, 55.7, 55.6, 55.5, 50.8, 49.6, 43.5, 40.7, 36.2; MS calcd for $C_{29}H_{32}O_5N_2$ 527.3 (MK^+), found 527.2.

2-(2,4-Dimethoxybenzyl)-4-ethyl-9-(4-methoxyphenyl)-2,3,3a,4,9,9a-hexahydropyrrol[3,4-*b*]quinolin-1-one (6d). Prepared according to the general procedure for the cyclization. Yield 81%; mp 166.6−167.6 °C; creamy white solid. ^{13}H NMR (500 MHz, $CDCl_3$) δ 7.17 (d, J = 8.0 Hz, 2H), 7.12 (d, J = 8.0 Hz, 1H), 7.08 (t, J = 7.8 Hz, 1H), 6.85 (d, J = 8.5 Hz, 2H), 6.79 (d, J = 7.5 Hz, 1H), 6.73 (d, J = 7.5 Hz, 1H), 6.62 (t, J = 7.3 Hz, 1H), 6.45 (d, J = 2.0 Hz, 1H), 6.44 (dd, J = 8.3,2.0 Hz, 1H), 4.51 (d, J = 15.0 Hz, 1H), 4.34 (d, J = 14.5 Hz, 1H), 4.30 (d, J = 12.0 Hz, 1H), 3.80 (s, 3H), 3.79 (s, 6H), 3.61 (m, 2H), 3.47 (dd, J = 8.8, 6.5 Hz, 1H), 3.22 (t, J = 9.3 Hz, 1H), 3.09 (s, J = 7.5 Hz, 1H), 2.89 (t, J = 11.5 Hz, 1H), 1.09 (t, J = 7.3 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.5, 160.4, 158.5, 158.1, 146.0, 135.9, 131.3, 131.0, 130.3, 128.8, 127.3, 118.3, 117.1, 113.7, 113.1, 104.2, 98.4, 56.8, 55.3, 55.1, 50.4, 49.6, 45.2, 41.9, 40.5, 9.9; IR 3066, 2967, 2926, 2868, 2838, 1695, 1602, 1509, 1462, 1369 cm^{-1}; HRMS calcd for $C_{29}H_{32}O_4N_2$ was 472.23621 (M^+), found 472.23669.

2-(2,4-Dimethoxybenzyl)-4-ethyl-9-(4-methoxyphenyl)-2,3,3a,4,9,9a-hexahydropyrrol[3,4-*b*]quinolin-1-one (7d). Prepared according to the general procedure for the cyclization. Yield: 10%; creamy white foam. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.16 (dt, J = 7.8, 2.0 Hz, 1H), 7.12 (dt, J = 10.0, 2.5 Hz, 2H), 6.98 (dd, J = 7.5, 1.5 Hz, 1H), 6.81 (dt, J = 9.5, 2.5 Hz, 2H), 6.68 (d, J = 7.5 Hz, 2H), 6.55 (d, J = 8.5 Hz, 1H), 6.40 (d, J = 2.0 Hz, 1H), 6.30 (dd, J = 8.3, 3.0 Hz, 1H), 4.42 (d, J = 15.5 Hz, 1H), 4.35 (d, J = 3.5 Hz, 1H), 4.17 (d, J = 15.0 Hz, 1H), 3.99 (ddd, J = 8.0, 5.5, 2.0 Hz, 1H), 3.76 (s, 3H), 3.75 (s, 3H), 3.71 (s, 3H), 3.45 (dq, J = 13.7, 7.2 Hz, 1H), 3.33 (dd, J = 10.0, 5.5 Hz, 1H), 3.21 (dd, J = 10.8, 2.5 Hz, 1H), 3.18 (dd, J = 8.0, 3.5 Hz, 1H), 3.03 (dq, J = 14.1, 7.0 Hz, 1H), 1.11 (t, J = 7.3 Hz, 3H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 173.2, 160.6, 158.8, 158.5, 144.7, 135.7, 129.8, 129.6, 129.3, 128.2, 126.6, 117.8, 116.9, 113.9, 112.8, 104.6, 98.4, 55.7, 55.6, 55.5, 53.2, 51.2, 50.4, 42.5, 42.5, 40.7, 12.0; IR 3060, 2961, 2926, 2839, 1695, 1602, 1509, 1456 cm^{-1}; HRMS calcd for $C_{29}H_{32}O_4N_2$ was 472.23621 (M^+), found 472.23603.

4-Allyl-2-(2,4-dimethoxybenzyl)-9-(4-methoxyphenyl)-2,3,3a,4,9,9a,-hexahydropyrrolo[3,4-*b*]quinolin-1-one (6e). Prepared according to the general procedure for the cyclization. Yield: 79%; mp 132.2−133.0 °C; white powder. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.17 (d, J = 8.5 Hz, 2H), 7.12 (d, J = 8.0 Hz, 1H), 7.09 (t, J = 7.3 Hz, 1H), 6.87 (d, J = 8.0 Hz, 2H), 6.78 (d, J = 8.5 Hz, 1H), 6.73 (d, J = 8.5 Hz, 1H), 6.64 (t, J = 7.3 Hz, 1H), 6.44 (d, J = 3.0 Hz, 1H), 6.43 (dd, J = 8.3, 2.5 Hz, 1H), 5.91 (dddd, J = 16.8, 11.8, 6.0, 4.5 Hz, 1H), 5.28 (dd, J = 17.0, 1.5 Hz, 1H), 5.21 (d, J = 10.0, 1.5 Hz, 1H), 4.49 (d, J = 15.0 Hz, 1H), 4.33 (d, J = 15.0 Hz, 1H), 4.31 (d, J = 12.5 Hz, 1H), 4.08 (t, J = 2.3 Hz, 1H), 4.04 (dt, J = 16.9, 2.3 Hz, 1H), 3.80 (s, 9H), 3.61 (m, 2H), 3.47 (dd, J = 8.8, 6.5 Hz, 1H), 3.22 (t, J = 9.0 Hz, 1H), 2.93 (t, J = 11.8 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 172.6, 160.7, 158.4, 146.9, 136.0, 133.6, 131.4, 131.2, 130.5, 128.8, 127.5, 118.9, 117.5, 117.3, 114.0, 113.8, 104.5, 98.7, 58.2, 55.6, 55.6, 55.4, 51.5, 50.7, 49.9, 45.4, 40.8; IR 3061, 3008, 2938, 2827, 1701, 1608, 1509, 1462, 1415, 1357 cm^{-1}; HRMS calcd for $C_{30}H_{32}O_4N_2$ was 484.23621 (M^+), found 484.23627.

4-Allyl-2-(2,4-dimethoxybenzyl)-9-(4-methoxyphenyl)-2,3,3a,4,9,9a,-hexahydropyrrolo[3,4-*b*]quinolin-1-one (7e). Prepared according to the general procedure for the cyclization. Yield: 19%; light yellow film. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.11 (d, J = 8.5 Hz, 2H), 6.98 (d, J = 7.5 Hz, 1H), 6.81 (d, J = 8.5 Hz, 2H), 6.69 (t, J = 7.5 Hz, 1H), 6.63 (d, J = 8.0 Hz, 1H), 6.59 (d, J = 8.0 Hz, 1H), 6.40 (d, J = 2.5 Hz, 1H), 6.31 (dd, J = 8.8, 2.5 Hz, 1H), 5.79 (dtt, J = 11.1, 10.5, 5.5 Hz, 1H), 5.13 (d, J = 9.0, 1H), 5.12 (d, J = 10.0 Hz, 1H), 4.38 (d, J = 3.5 Hz, 1H), 4.37 (d, J = 15.0 Hz, 1H), 4.22 (d, J = 15.5 Hz, 1H), 3.99 (ddd, J = 7.3, 5.3, 2.5 Hz, 1H), 3.92 (dd, J = 16.3, 5.0 Hz, 1H), 3.76 (s, 3H), 3.74 (s, 3H), 3.71(s, 3H), 3.68 (dd, J = 16.5, 5.5 Hz, 1H), 3.29 (dd, J = 10.8, 5.0 Hz, 1H), 3.26 (dd, J = 10.0, 3.5 Hz, 1H), 3.18 (dd, J = 8.0, 3.5 Hz, 1H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 173.1, 160.7, 158.8, 158.5, 145.0, 135.8, 134.8, 129.9, 129.7, 129.2, 128.1, 126.2, 118.2, 117.2, 116.9, 114.0, 113.3, 104.6, 98.4, 55.6, 55.5, 53.7, 51.9, 51.0, 50.3, 42.2, 41.0, 40.8; IR 3061, 3002, 2932, 2827, 1684, 1608, 1509, 1444, 1340 cm^{-1}; HRMS calcd for $C_{30}H_{32}O_4N_2$ was 484.23621 (M^+), found 484.23605.

2-(2,4-Dimethoxybenzyl)-4-isopropyl-9-(4-methoxyphenyl)-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (6f). Prepared according to the general procedure for the cyclization, but was refluxed for 12 h and $Yb(OTf)_3$ was used as the catalyst. Yield: 63%. ^{1}H NMR (400 MHz, CD_2Cl_2) δ 7.13 (dd, J = 8.8, 4.0 Hz, 3H), 7.05 (t, J = 7.4 Hz, 1H), 6.94 (d, J = 8.0 Hz, 1H), 6.86 (d, J = 8.8 Hz, 2H), 6.66 (d, J = 7.6 Hz, 1H), 6.60 (t, J = 7.4 Hz, 1 H), 6.45 (d, J = 1.6 Hz, 1H), 6.44 (dd, J = 11.8, 2.4 Hz, 1H), 4.48 (d, J = 14.8 1H), 4.33 (d, J = 14.8 Hz, 1H), 4.23 (d, J = 12.4 Hz, 1H), 3.93 (p, J = 6.9 Hz, 1H), 3.80 (s, 3H), 3.79 (s, 6H), 3.56 (m, 2H), 3.29 (dt, J = 9.9, 2.4 Hz, 1H), 2.92 (t, J = 11.6 Hz, 1H), 1.30 (d, J = 6.4 Hz, 3H), 1.26 (d, J = 7.6 Hz, 3H); ^{13}C NMR (100 MHz, CD_2Cl_2) δ 171.9, 160.4, 158.5, 158.1, 146.8, 135.5, 131.0, 130.9, 130.2, 129.9, 126.9, 118.3, 117.1, 115.3, 113.7, 104.2, 98.4, 56.8, 55.3, 55.3, 55.1, 51.5, 51.5, 50.3, 45.3, 40.5, 20.6, 19.8; HRMS calcd for $C_{30}H_{34}O_4N_2$ was 486.2519 (M^+), found 486.2517.

2-(2,4-Dimethoxybenzyl)-4-isopropyl-9-(4-methoxyphenyl)-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (7f). Prepared according to the general procedure for the cyclization, but was refluxed for 12 h and $Yb(OTf)_3$ was used as the catalyst. Yield: 26%. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.17 (dt, J = 8.4, 2.0 Hz, 1H), 7.11 (dt, J = 8.5, 2.5 Hz, 2H), 7.04 (dd, J = 12.5, 1.5 Hz, 1H), 6.79 (dt, J = 9.0, 2.8 Hz, 3H), 6.73 (dt, J = 7.5, 1.0 Hz, 1H), 6.42 (d, J = 9.0 Hz, 1H), 6.38 (d, J = 2.5 Hz, 1H), 6.26 (dd, J = 8.3, 2.0 Hz, 1H), 4.45 (d, J = 15.0 Hz, 1H), 4.30 (d, J = 3.0 Hz, 1H), 4.19 (tt, J = 6.5, 3.5 Hz, 1H), 4.11 (d, J = 15.5 Hz, 1H), 3.75 (s, 3H), 3.74 (s, 3H), 3.70 (s, 3H), 3.39 (dd, J = 10.5, 7.0 Hz, 1H), 3.36 (dd, J = 9.8, 3.5 Hz, 1H), 3.12 (dd, J = 10.5, 3.5 Hz, 1H), 1.15 (d, J = 7.0 Hz, 3H), 1.08 (d, J = 6.5 Hz, 3H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 173.8, 160.5, 158.6, 158.4, 145.9, 135.7, 131.0, 130.5, 129.8, 129.5, 129.3, 129.21, 129.16, 128.1, 118.5, 116.7, 115.1, 114.5, 113.7, 104.6, 98.4, 55.6, 55.5, 51.3, 49.4, 48.6, 43.3, 40.7, 21.5, 20.8; MS calcd for $C_{30}H_{34}O_4N_2$ was 487.3 (MH^+), found 487.1.

4-Cyclohexyl-2-(2,4-dimethoxybenzyl)-9-(4-methoxyphenyl)-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (6g). Prepared according to the general procedure for the cyclization. Yield: 28%. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.12 (m, 3H), 7.06 (ddd, J = 7.8, 1.5, 1.0 Hz, 1H), 6.93 (d, J = 8.5 Hz, 1H), 6.86 (d, J = 8.5 Hz, 1H), 6.63 (d, J = 7.5 Hz, 1H), 6.58 (dt, J = 8.5, 1.0 Hz, 1H), 6.45 (d, J = 2.5 Hz, 1H), 6.44 (dd, J = 8.0, 2.5 Hz, 1H), 4.49 (d, J = 14.5 Hz, 1H), 4.30 (d, J = 15.0 Hz, 1H), 4.21 (d, J = 12.0 Hz, 1H), 3.81 (s, 3H), 3.79 (s, 6H), 3.67 (m, 2H), 3.51 (t, J = 12.3 Hz, 1H), 3.28 (t, J = 8.0 Hz, 1H), 2.91 (t, J = 11.5 Hz, 1H), 1.86 (d, J = 12.0 Hz, 4H), 1.71 (d, J = 13.0 Hz, 1H), 1.64 (d, J = 9.0 Hz, 1H), 1.54 (t, J = 11.5 Hz, 1H), 1.38 (q, J = 13.0 Hz, 1H), 1.28 (q, J = 13.0 Hz, 1H), 1.12 (q, J = 13.0 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.7, 160.3, 158.4, 158.1, 147.3, 135.3, 130.7, 130.7, 130.2, 130.0, 127.0, 118.1, 117.2, 115.4, 113.7, 104.2, 98.4, 61.9, 56.8, 55.3, 55.2, 55.1, 52.1, 50.2, 45.3, 40.5, 32.4, 26.8, 26.7, 25.9; IR 3057, 2993, 2932, 2850, 1697, 1610, 1502, 1480, 1450; HRMS calcd for $C_{33}H_{38}O_4N_2$ was 526.28316 (M^+), found 526.28285.

4-Cyclohexyl-2-(2,4-dimethoxybenzyl)-9-4(4-methoxyphenyl)-2,3,3a,4,9,9 a,-hexahydropyrrolo[3,4-*b*]quinolin-1-one (7g). Prepared according to the general procedure for the cyclization. Yield: 14%. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.17 (ddd, J = 7.6, 7.3, 2.0 Hz, 1H), 7.13 (dd, J = 9.0, 3.0 Hz, 2H), 7.05 (dd, J = 7.6, 1.0 Hz, 1H), 6.79 (dt, J = 9.3, 2.5 Hz, 2H), 6.76 (d, J = 7.5 Hz, 1H), 6.74 (dt, J = 7.5, 1.0 Hz, 1H), 6.38 (d, J = 2.5 Hz, 1H), 6.35 (d, J = 9.0 Hz, 1H), 6.24 (dd, J = 8.3, 2.5 Hz, 1H), 4.45 (d, J = 15.0 Hz, 1H), 4.31 (d, J = 3.0 Hz, 1H), 4.22 (ddd, J = 10.5, 6.3, 3.5 Hz, 1H), 4.09 (d, J = 15.0 Hz, 1H), 3.76 (s, 3H), 3.75 (s, 3H), 3.71 (s, 3H), 3.40 (m, 3H), 3.11 (dd, J = 10.0, 3.5 Hz, 1H), 1.89 (d, J = 12.0 Hz, 1H), 1.79 (m, 1H), 1.72 (m, 1H), 1.64 (m, 1H), 1.32 (m, 5H), 1.15 (m, 1H), 0.90 (t, J = 6.8 Hz, 1H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 174.0, 160.5, 158.6, 158.3, 145.9, 135.6, 129.8, 129.6, 129.2, 129.1, 128.1, 118.6, 116.7, 115.2, 113.6, 104.6, 98.4, 58.2, 55.6, 55.6, 55.5, 51.3, 48.8, 43.4, 40.6, 32.6, 31.3, 26.5, 26.4, 26.3; IR 3001, 2937, 1688, 1610, 1584, 1510, 1489, 1450; HRMS calcd for $C_{33}H_{38}O_4N_2$ was 526.28316 (M^+), found 526.2823.

2-(2,4-Dimethoxybenzyl)-9-(4-methoxyphenyl)-4-phenyl-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (6h). Prepared according to the general procedure for the cyclization. Yield: 77%; mp 175.6−177.8 °C; creamy white solid. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.45 (t, J = 7.75 Hz, 2H), 7.32 (dt, J = 7.25, 1.5 Hz, 1H), 7.26 (d, J = 7.5 Hz, 2H), 7.23 (dd, J = 8.3, 1.5 Hz, 2H), 7.06 (d, J = 7.5 Hz, 1H), 6.91 (m, 3H), 6.81 (d, J = 8.0 Hz, 1H), 6.67 (dt, J = 7.5, 1.0 Hz, 1H), 6.40 (d, J = 2.0 Hz, 1H), 6.38 (s, 1H), 6.37 (dd, J = 7.5, 1.0 Hz, 1H), 4.47 (d, J = 12.5 Hz, 1H), 4.44 (d, J = 15.0 Hz, 1H), 4.24 (d, J = 15.0 Hz, 1H), 3.93 (dt, J = 11.0, 8.0 Hz, 1H), 3.82 (s, 3H), 3.77 (s, 3H), 3.68 (s, 3H), 3.07 (m, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.2, 160.3, 158.4, 158.2, 147.4, 145.1, 135.7, 131.0, 130.6, 130.3, 129.9, 128.3, 128.2, 126.9, 119.4, 117.1, 115.6, 113.7, 104.1, 98.4, 59.5, 55.3, 55.1, 50.7, 50.0, 45.2, 40.4; IR 3062, 3028, 2997, 2928, 2833, 1709, 1610, 1593, 1510, 1485; HRMS calcd for $C_{33}H_{32}O_4N_2$ was 520.23621 (M^+), found 520.23612.

2-(2,4-Dimethoxybenzyl)-9-(4-methoxyphenyl)-4-phenyl-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (7h). Prepared according to the general procedure for the cyclization. Yield: 14%; light green-yellow film. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.24 (dd, J = 7.5, 1.5 Hz, 1H), 7.21 (dd, J = 7.0, 2.0 Hz, 1H), 7.20 (d, J = 7.5 Hz, 1H), 7.17 (dd, J = 6.5, 2.0 Hz, 2H), 7.16 (dd, J = 9.0, 1.5 Hz, 1H), 7.02 (d, J = 7.5 Hz, 1H), 6.98 (dt, J = 7.5, 1.5 Hz, 1H), 6.92 (d, J = 7.0 Hz, 1H), 6.89 (d, J = 7.5 Hz, 2H), 6.76 (dd, J = 6.5, 2.0 Hz, 2H), 6.37 (d, J = 3.0 Hz, 1H), 6.26 (d, J = 8.5 Hz, 2H), 6.21 (dd, J = 8.5, 2.0 Hz, 1H), 4.57 (d, J = 2.5 Hz, 1H), 4.48 (d, J = 15.5 Hz, 1H), 4.38 (ddd, J = 8.8, 6.8, 1.5 Hz, 1H), 4.12 (d, J = 15.5 Hz, 1H), 3.74 (s, 3H), 3.71 (s, 3H), 3.69 (s, 3H), 3.68 (dd, J = 10.0, 6.5 Hz, 1H), 3.62 (dd, J = 9.0, 2.5 Hz, 1H), 3.31 (dd, J = 10.5, 1.5 Hz, 1H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 173.1, 160.5, 158.6, 158.5, 145.8, 142.9, 134.7, 131.3, 130.5, 129.5, 129.3, 128.8, 128.0, 122.3, 121.7, 120.5, 119.0, 116.4, 114.0, 104.7, 98.3, 55.7, 55.6, 55.4, 53.6, 52.8, 52.5, 43.3, 40.6; IR 3066, 3036, 3006, 2924, 2837, 1687, 1614, 1593, 1515, 1493, 1455, 1385; HRMS calcd for $C_{33}H_{32}O_4N_2$ was 520.23621 (M^+), found 520.23665.

9-(2,4-Dimethoxybenzyl)-7-(4-methoxyphenyl)-2,3,7a,9,10,10a-hexahydro-1*H*,7*H*-9,10*b*-diazacyclopenta[*a*]phenalen-8-one (8). Prepared according to the general procedure for the cyclization. Yield: 80%; mp 159.6−160.8 °C; creamy white solid. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.19 (d, J = 7.5 Hz, 2H), 7.12 (d, J = 8.0 Hz, 1H), 6.85 (m, 3H), 6.58 (d, J = 4.5 Hz, 1H), 6.44 (d, J = 2.0 Hz, 1H), 6.43 (dd, J = 8.0, 2.5 Hz, 1H), 4.50 (d, J = 14.5 Hz, 1H), 4.33 (d, J = 14.5 Hz, 1H), 4.30 (d, J = 12.5 Hz, 1H), 3.79 (s, 3H), 3.79 (s, 6H), 3.45 (dd, J = 8.5, 6.0 Hz, 1H), 3.32 (ddd, J = 11.0, 9.8, 6.0 Hz, 1H), 3.17 (t, J = 9.8 Hz, 1H), 3.09 (dt, J = 10.0, 3.5 Hz, 1H), 2.91 (t, J = 11.5 Hz, 1H), 2.90 (q, J = 11.5 Hz, 1H), 2.81 (d, J = 5.5 Hz, 1H), 2.77 (dt, J = 11.0, 3.0 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.7, 160.4, 158.6, 158.1, 144.5, 136.3, 131.0, 130.3, 128.9, 128.2, 127.8, 123.6, 118.9, 117.1, 113.6, 104.2, 98.4, 58.9, 55.3, 55.1, 50.5, 49.4, 46.4, 45.2, 40.4, 27.0, 22.0; IR 2990, 2926, 2845, 1707, 1619, 1584, 1509, 1450, 1351 cm^{-1}; HRMS calcd for $C_{30}H_{32}O_4N_2$ was 484.23621 (M^+), found 484.23564.

9-(2,4-Dimethoxybenzyl)-7-(4-methoxyphenyl)-2,3,7a,9,10,10a-hexahydro-1*H*,7*H*-9,10*b*-diazacyclopenta[*a*]phenalen-8-one (9). Prepared according to the general procedure for the cyclization. Yield: 13%; creamy white foam. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.11 (d, J = 8.5 Hz, 2H), 6.84 (d, J = 7.0 Hz, 1H), 6.82 (dd, J = 7.0, 2.0 Hz, 2H), 6.76 (d, J = 9.0 Hz, 1H), 6.71 (d, J = 7.5 Hz, 1H), 6.50 (t, J = 7.3 Hz, 1H), 6.42 (d, J = 2.5 Hz, 1H), 6.34 (dd, J = 8.8, 2.5 Hz, 1H), 4.49 (d, J = 15.0 Hz, 1H), 4.26 (d, J = 5.0 Hz, 1H), 4.19 (d, J = 15.5 Hz, 1H), 3.82 (ddd, J = 7.6, 4.5, 3.0 Hz, 1H), 3.77 (s, 3 H), 3.76 (s, 3H), 3.70 (s, 3H), 3.37 (dd, J = 11.0, 3.0 Hz, 1H), 3.35 (dd, J = 10.3, 4.5 Hz, 1H), 3.16 (ddd, J = 12.4, 7.3, 4.5 Hz, 1H), 3.08 (dd, J = 7.5, 4.5 Hz, 1H), 2.77 (p, J = 5.5 Hz, 1H), 2.66 (p, J = 5.5 Hz, 1H), 1.85 (m, 2H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 173.0, 160.8, 158.9, 158.5, 141.6, 136.2, 130.0, 129.3, 127.9, 127.6, 124.2, 122.8, 117.0, 116.7, 114.1, 104.6, 98.4, 55.7, 55.6, 55.5, 54.6, 50.6, 49.9, 47.5, 42.0, 40.7, 28.2, 22.5; IR 3002, 2933, 2842, 1688, 1619, 1593, 1506, 1459.

2-(2,4-Dimethoxybenzylamino)-3-phenyl-propionic Acid Methyl Ester (10). Prepared according to the general procedure for the synthesis of amines. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.27 (ddt, J = 7.0, 6.5, 1.5 Hz, 2H), 7.22 (dt, J = 7.5, 1.3 Hz, 1H), 7.14 (d, J = 7.0 Hz, 2H), 7.03 (d, J = 8.0 Hz, 1H), 6.38 (d, J = 2.5 Hz, 1H), 6.37 (dd, J = 8.3, 2.5 Hz, 1H), 3.77 (s, 3H), 3.74 (d, J = 13.5 Hz, 1 H), 3.63 (s, 3H), 3.61 (d, J = 13.0 Hz, 1H), 3.61 (s, 3H), 3.52 (dd, J = 8.0, 7.0 Hz, 1H), 2.99 (dd, J = 13.5, 6.3 Hz, 1H), 2.93 (dd, J = 13.5, 7.8 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 174.8, 160.0, 158.5, 137.3, 130.3, 129.0, 128.3, 126.5, 119.9, 103.4, 98.2, 61.8, 55.2, 55.0, 51.5, 47.2, 39.5; IR 3329, 3066, 3026, 2996, 2944, 2827, 1736, 1619, 1584, 1503, 1450 cm^{-1}; HRMS calcd for $C_{19}H_{22}O_4N$ 328.15488 (M^+), found 328.15465.

2-{(2,4-Dimethoxybenzyl)-[3-(4-methoxyphenyl)-acryoyl]-amino}-3-phenyl-propionic Acid Methyl Ester (11). Prepared according to the general procedure for the synthesis of amides. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.74 (d, J = 15.6 Hz, 1H), 7.42 (d, J = 9.2 Hz, 1H), 7.25 (m, 3H), 7.13 (d, J = 6.4 Hz, 2H), 7.03 (d, J = 8.0 Hz, 1H), 6.86 (d, J = 8.8 Hz, 2H), 6.78 (d, J = 15.2 Hz, 1H), 6.34 (d, J = 2.4 Hz, 1H), 6.37 (dd, J = 8.4, 2.8 Hz, 1H), 4.52 (d, J = 16.8 Hz, 1H), 4.46 (dd, J = 7.6, 2.4 Hz, 1H), 4.11 (d, J = 17.2 Hz, 1H), 3.80 (s, 3H), 3.78 (s, 3H), 3.74 (s, 3H), 3.64 (s, 3H), 3.45 (dd, J = 13.6, 6.4 Hz, 1H), 3.25 (dd, J = 13.6, 8.0 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 171.1, 167.5, 160.7, 160.4, 157.9, 142.7, 138.2, 129.5, 129.3, 129.2, 128.2, 127.9, 126.2, 116.8, 115.3, 114.0, 103.5, 98.2, 60.9, 55.2, 55.2, 55.0, 52.0, 47.2, 35.2; IR 3066, 3002, 2944, 2839, 1742, 1654, 1596, 1503, 1444, 1345 cm^{-1}; HRMS calcd for $C_{29}H_{31}O_6N$ 489.21514 (M^+), found 489.21539.

***N*-(1-Benzyl-2-hydroxyethyl)-*N*-(2,4-dimethoxybenzyl)-3-(4-methoxyphenyl)-acryl Amide (12).** Prepared according to the general procedure for the synthesis of esters. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.73 (d, J = 15.0 Hz, 1H), 7.43 (d, J = 8.0 Hz, 2H), 7.27 (t, J = 7.5 Hz, 3H), 7.21 (m, 3H), 7.00 (d, J = 8.5 Hz, 1H), 6.89 (d, J = 9.0 Hz, 2H), 6.77 (d, J = 16.0 Hz, 1H), 6.43 (d, J = 2.0 Hz, 1H), 6.43 (dd, J = 8.5, 2.0 Hz, 1H), 4.52 (d, J = 17.0 Hz, 1H), 4.14 (d, J = 17.0 Hz, 1H), 3.82 (d, J = 5.0 Hz, 1H), 3.79 (s, 3H), 3.77 (s, 3H), 3.76 (s, 3H), 3.71 (m, 2H), 3.19 (dd, J = 13.5, 7.5 Hz, 1H), 3.13 (dd, J = 13.3, 7.5 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 169.3, 160.9, 160.6, 158.1, 142.6, 139.0, 129.4, 129.2, 128.4, 127.9, 126.2, 117.0, 116.3, 114.2, 104.0, 98.6, 64.3, 64.1, 55.4, 55.3, 55.2, 48.5, 34.3; IR 3376, 3066, 2996, 2938, 2833, 1643, 1608, 1584, 1520, 1456, 1357 cm^{-1}; HRMS calcd for $C_{28}H_{31}O_5N$ 461.22022 (M^+), found 461.22076.

***N*-(1-Benzyl-2-oxoethyl)-*N*-(2,4-dimethoxybenzyl)-3-(4-methoxyphenyl)-acryl Amide (13).** Prepared according to the general procedure for the oxidation of alcohols. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.37 (s, 1H), 7.76 (d, J = 15.5 Hz, 1H), 7.49 (d, J = 9.5 Hz, 2H), 7.32 (t, J = 7.3 Hz, 2H), 7.24 (d, J = 7.5 Hz, 1H), 7.19 (d, J = 7.5 Hz, 2H), 6.91 (d, J = 9.0 Hz, 2H), 6.88 (d, J = 2.5 Hz, 2H), 6.41 (s, 1H), 6.39 (d, J = 2.5 Hz, 1H), 4.62 (d, J = 16.0 Hz, 1H), 3.84 (s, 3H), 3.78 (s, 3H), 3.72 (s, 3H), 3.69 (m, 1H), 3.57 (d, J = 16.0 Hz, 1H), 3.47 (dd, J = 14.0, 4.5 Hz, 1H), 3.22 (dd, J = 13.5, 9.5 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 197.5, 167.3, 161.1, 158.5, 143.2, 138.6, 130.4, 129.5, 129.3, 128.6, 127.8, 126.5, 116.6, 114.3, 113.9, 104.1, 98.7, 68.3, 55.4, 55.3, 55.1, 48.9, 33.1; IR 3061, 3002, 2938, 2827, 1718, 1637, 1596, 1514, 1444, 1287 cm^{-1}; HRMS calcd for $C_{28}H_{29}O_5N$ 459.20457 (M^+), found 459.20499.

3-Benzyl-2-(2,4-dimethoxybenzyl)-9-(4-methoxyphenyl)-4-methyl-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (14). Prepared according to the general procedure for the cyclization but was refluxed for 12 h. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.24 (m, 4H), 7.13 (d, J = 12.5, 3.0 Hz, 2H), 7.10 (d, J = 7.0 Hz, 2H), 7.07 (d, J = 1.0 Hz, 1H), 7.01 (d, J = 8.5 Hz, 1H), 6.85 (dd, J = 11.5, 2.5 Hz, 1H), 6.78 (d, J = 8.0 Hz, 1H), 6.74 (d, J = 8.5 Hz, 1H), 6.67 (t, J = 7.3 Hz, 1H), 6.42 (d, J = 2.0 Hz, 1H), 6.40 (dd, J = 9.8 Hz, 1H), 5.08 (d, J = 15.5 Hz, 1H), 4.22 (d, J = 11.0 Hz, 1H), 4.20 (d, J = 15.5 Hz, 1H), 3.82 (m, 1H), 3.80 (s, 3H), 3.79 (s, 3H), 3.69 (s, 3H), 3.40 (dd, J = 15.0, 3.5 Hz, 1H), 3.22 (dd, J = 11.0, 8.5 Hz, 1H), 3.08 (dd, J = 15.0, 4.5 Hz, 1H), 2.93 (t, J = 11.0 Hz, 1H), 2.81 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 173.0, 160.3, 158.3, 158.1, 148.7, 136.4, 136.2, 131.1, 130.7, 130.1, 129.8, 129.4, 128.6, 127.2, 126.7, 119.3, 116.4, 114.5, 113.6, 104.2, 98.3, 94.7, 61.0, 58.2, 55.3,55.2, 55.1, 49.5, 45.8, 38.4, 36.5, 36.2; IR 3061, 3031, 2996, 2932, 2874, 1707, 1608, 1509, 1462, 1398 cm^{-1}; HRMS calcd for $C_{35}H_{36}O_4N_2$ was 548.26751 (M^+), found 548.26693.

4-Methyl-9-phenyl-3a,4,9,9a-tetrahydrofuro[3,4-*b*]quinolin-3(1*H*)-one (16). Prepared according to the general procedure for the cyclization. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.36 (m, 3H), 7.19 (m, 3H), 6.95 (d, J = 8.5 Hz, 1H), 6.69 (t, J = 7.5 Hz, 1H), 6.64 (d, J = 7.5 Hz, 1H), 4.20 (d, J = 12.0 Hz, 1H), 4.15 (dd, J = 8.5, 7.0 Hz, 1H), 3.98 (dd, J = 11.0, 8.5 Hz, 1H), 3.64 (d, J = 12.5 Hz, 1H), 3.26 (s, 3H), 3.12 (o, J = 7.0 Hz, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.8, 148.5, 129.7, 129.0, 128.4, 128.1, 127.9, 127.5, 126,3, 119.2, 114.7, 68.8, 60.1, 48.3, 46.4, 35.9; HRMS calcd for $C_{18}H_{17}O_2N$ was 279.12593 (M^+), found 279.12593.

3-(2,4-Dimethoxybenzylamino)-propionic Acid Methyl Ester (17). This compound was prepared according to the general procedure for the synthesis of amines. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.13 (d, J = 8.0 Hz, 1H), 6.45 (d, J = 2.5 Hz, 1H), 6.43 (dd, J = 8.3 Hz, 1H), 3.81 (s, 3H), 3.79 (s, 3H), 3.73 (s, 2H), 3.67 (s, 3H), 2.86 (t, J = 6.5 Hz, 2H), 2.54 (t, J = 6.8 Hz, 2H), 1.99 (s, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 173.2, 160.0, 158.5, 130.3, 120.6, 103.6, 98.5, 55.3, 55.2, 51.5, 48.6, 44.2, 34.6; IR 3335, 3002, 2944, 2827, 1730, 1614, 1584, 1497, 1450 cm^{-1}; HRMS calcd for $C_{13}H_{18}O_4N$ was 252.12358 (M^+), found 252.12359.

3-{(2,4-Dimethoxybenzyl)-[3-(4-methoxyphenyl)-acryoyl]-amino}-3-propionic Acid Methyl Ester (18). Prepared according to the general procedure for the synthesis of amides. Major rotamer ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.69 (d, J = 16.0 Hz, 1H), 7.42 (d, J = 8.5 Hz, 2H), 7.03 (d, J = 8.0 Hz, 1H), 6.86 (d, J = 8.5 Hz, 2H), 6.77 (d, J = 15.5 Hz, 1H), 6.47 (d, J = 2.5 Hz, 1H), 6.45 (dd, J = 8.5, 3.0 Hz, 1H), 4.62 (s, 2H), 3.81 (s, 3H), 3.81 (s, 3H), 3.79 (s, 3H), 3.71 (t, J = 6.8 Hz, 2H), 3.65 (s, 3H), 2.66 (t, J = 7.5 Hz, 2H); minor rotamer ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.73 (d, J = 15.5 Hz, 1H), 7.50 (d, J = 8.0 Hz, 2H), 7.25 (d, J = 8.5 Hz, 1H), 6.91 (d, J = 8.0 Hz, 2H), 6.84 (d, J = 16.5 Hz, 1H), 4.66 (s, 2H), 3.75 (t, J = 7.3 Hz, 2H), 2.61 (t, J = 7.5 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) for both rotamers δ 172.5, 167.4, 160.7, 160.5, 158.0, 142.9, 142.2, 130.8, 129.4, 129.3, 128.3, 128.1, 117.5, 115.4, 114.7, 114.2, 114.1, 104.3, 103.9, 98.6, 98.3, 55.3, 55.3, 55.2, 51.8, 51.6, 47.2, 43.0, 34.0, 32.6; IR 3002, 2944, 2839, 1736, 1649, 1503, 1439, 1369 cm^{-1}; HRMS calcd for $C_{23}H_{27}O_6N$ was 413.18384 (M^+), found 413.18391.

3-(2,4-Dimethoxybenzylamino)-2,2-dimethylpropan-1-ol (19). This compound was prepared according to the general procedure for the synthesis of amines. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.08 (d, J = 8.5 Hz, 1H), 6.45 (d, J = 3.0 Hz, 1H), 6.43 (dd, J = 8.0, 2.0 Hz, 1H), 3.79 (s, 3H), 3.78 (s, 3H), 3.68 (s, 2H), 3.47 (s, 2H), 2.54 (s, 2H), 0.88 (s, 6H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 160.1, 158.6, 130.4, 119.9, 103.5, 98.4, 74.5, 60.8, 55.2, 55.1, 49.6, 34.8, 23.0; IR 3328, 3002, 2950, 2836, 1614, 1590, 1509, 1462 cm^{-1}; HRMS calcd for $C_{14}H_{23}O_3N$ 253.16779 (M^+) found 253.16777.

***N*-(2,4-Dimethoxybenzyl)-*N*-(3-hydroxypropyl)-3-(4-methoxyphenyl)-acryl Amide (20).** Prepared according to the general procedure for the reduction of esters. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.74 (d, J = 15.5 Hz, 1H), 7.42 (dt, J = 9.3, 2.3 Hz, 2H), 7.03 (d, J = 9.0, 2.3 Hz, 2H), 6.77 (d, J = 15.5 Hz, 1H), 6.49 (d, J = 2.5 Hz, 1H), 6.47 (dd, J = 8.5, 2.0 Hz, 1H), 4.55 (s, 2H), 3.83 (s, 3H), 3.81 (s, 3H), 3.80 (s, 3H), 3.62 (t, J = 5.8 Hz, 2H), 3.54 (t, J = 5.3 Hz, 2H), 1.70 (p, J = 5.8 Hz, 2H); minor rotamer ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.50 (d, J = 8.5 Hz, 2H), 7.23 (d, J = 8.5 Hz, 1H), 7.11 (d, J = 8.5 Hz, 1H), 6.98 (d, J = 15.5 Hz, 1H), 6.82 (d, J = 8.5 Hz, 1H), 6.44 (s, 1H), 6.42 (dd, J = 8.3, 2.0 Hz, 1H), 3.69 (m, 2H), 3.50 (t, J = 6.3 Hz, 2H), 3.45 (t, J = 5.5 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) for both rotamers δ 168.8, 160.9, 160.6, 158.0, 143.1, 129.4, 128.0, 127.9, 116.7, 114.7, 114.1, 113.8, 104.0, 98.6, 58.0, 55.3, 55.3, 55.2, 45.9, 41.9, 30.1; IR 3393, 2996, 2932, 2839, 1649, 1584, 1514, 1456, 1357 cm^{-1}; HRMS calcd for $C_{22}H_{27}O_5N$ was 385.18892 (M^+), found 385.18904.

***N*-(2,4-Dimethoxybenzyl)-*N*-(3-hydroxy-2,2-dimethylpropyl)-3-(4-methoxyphenyl)-acryl Amide (21).** Prepared according to the general procedure for the synthesis of amides. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.72 (d, J = 15.0 Hz, 1H), 7.38 (dt, J = 8.5, 2.0 Hz, 2H), 6.91 (d, J = 8.5 Hz, 1H), 6.86 (dt, J = 8.5, 2.5 Hz, 2H), 6.66 (d, J = 15.0 Hz, 1H), 6.49 (d, J = 2.0 Hz, 1H), 6.45 (dd, J = 8.3, 2.0 Hz, 1H), 4.66 (s, 2H), 3.85 (s, 3H), 3.80 (s, 3H), 3.79 (s, 3H), 3.34 (s, 1H), 3.20 (s, 2H), 0.97 (s, 6H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 169.6, 160.9, 160.4, 157.6, 143.2, 129.5, 127.8, 127.3, 117.2, 114.6, 114.1, 103.9, 98.6, 68.0, 55.4, 55.3, 55.2, 53.7, 48.8, 38.4, 23.7; IR 3364, 3008, 2961, 2839, 1649, 1584, 1509, 1439, 1363 cm^{-1}; HRMS calcd for $C_{24}H_{31}O_5N$ 413.22022 (M^+), found 413.22047.

***N*-(2,4-Dimethoxybenzyl)-3-(4-methoxyphenyl)-*N*-(3-oxopropyl)-acryl Amide (22).** Prepared according to the general procedure for the oxidation of alcohols. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.78 (s, 1H), 7.68 (d, J = 15.0 Hz, 1H), 7.43 (d, J = 9.0 Hz, 2H), 7.03 (d, J = 8.5 Hz, 1H), 6.87 (d, J = 8.5 Hz, 2H), 6.79 (d, J = 15.0 Hz, 1H), 6.47 (s, 1H), 6.46 (dd, J = 8.3 Hz, 1H), 4.61 (s, 2H), 3.81 (s, 6H), 3.80 (s, 3H), 3.73 (t, J = 6.8 Hz, 2H), 2.74 (t, J = 6.8 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 201.0, 167.6, 160.7, 160.6, 158.0, 142.3, 129.3, 128.4, 128.0, 117.2, 115.2, 114.1, 103.9, 98.6, 55.4, 55.3, 55.2, 47.2, 43.8, 42.7, 40.9; IR 3070, 3006, 2928, 2837, 2729, 1722, 1649, 1588, 1511, 1459; HRMS calcd for $C_{22}H_{25}O_5N$ was 383.17327 (M^+), found 383.17285.

***N*-(2,4-Dimethoxybenzyl)-*N*-(2,2-dimethyl-3-oxopropyl)-3-(4-methoxyphenyl)-acryl Amide (23).** Prepared according to the general procedure for the oxidation of alcohols. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.63(s, 1H), 7.66 (d, J = 15.5 Hz, 1H), 7.38 (d, J = 9.0 Hz, 2H), 6.94 (d, J = 8.0 Hz, 1H), 6.85 (d, J = 8.5 Hz, 2H), 6.70 (d, J = 15.5 Hz, 2H), 6.48 (d, J = 2.5 Hz, 1H), 6.45 (dd, J = 8.3, 2.5 Hz, 1H), 4.59 (s, 2H), 3.83 (s, 3H), 3.80 (s, 3H), 3.79 (s, 3H), 3.53 (s, 2H), 1.11 (s, 6H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 203.4, 168.3, 160.8, 160.5, 157.8, 143.2, 142.8, 129.5, 129.4, 127.9, 127.3, 117.2, 114.7, 114.6, 114.1, 114.06, 103.9, 98.6, 68.0, 55.3, 55.25, 55.2, 53.8, 48.8, 48.5, 47.7, 38.4, 23.7, 20.2; IR 3066, 2967, 2833, 1719, 1649, 1590, 1509, 1450, 1369 cm^{-1}; HRMS calcd for $C_{23}H_{29}O_5N$ 411.20457 (M^+), found 411.20434.

2-(2,4-Dimethoxybenzyl)-10-(4-methoxyphenyl)-4,4,5-trimethyl-3,4,4a,5,10,10a-hexahydro-2*H*-benzo[*b*][1,6]naphthyridin-1-one (24). Prepared according to the general procedure for the cyclization but was refluxed for 12 h and $Yb(OTf)_3$ was used as the catalyst. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.22 (d, J = 9.0 Hz, 2H), 7.13 (d, J = 7.5 Hz, 1H), 7.07 (t, J = 7.8 Hz, 1H), 6.92 (d, J = 9.0 Hz, 2H), 6.67 (d, J = 8.0 Hz, 1H), 6.53 (t, J = 7.8 Hz, 1H), 6.43 (d, J = 2.0 Hz, 1H), 6.41 (s, 1H), 7.37 (d, J = 7.5 Hz, 1H), 4.51 (d, J = 14.5 Hz, 1H), 4.38 (d, J = 14.0 Hz, 1H), 4.09 (d, J = 9.5 Hz, 1H), 3.82 (s, 3H), 3.79 (s, 3H), 3.72 (s, 3H), 3.15 (d, J = 14.0 Hz, 1H), 3.05 (s, 3H), 2.97 (s, 2H), 2.90 (d, J = 13.5 Hz, 1H), 0.97 (s, 3H), 0.87 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.9, 160.3, 158.5, 157.8, 148.0, 135.3, 131.6, 130.2, 129.9, 127.8, 127.2, 118.2, 117.3, 114.8, 113.7, 104.2, 98.3, 67.7, 58.7, 55.3, 55.2, 55.1, 48.1, 43.2, 42.4, 42.0, 37.9, 29.2, 20.4; IR 2932, 2839, 1712, 1671, 1614, 1514, 1456 cm^{-1}; HRMS calcd for $C_{18}H_{21}O_5N$ 500.26751 (M^+), found 500.26822.

2-(2,4-Dimethoxybenzyl)-10-(4-methoxyphenyl)-4,4,5-trimethyl-3,4,4a,5,10,10a-hexahydro-2*H*-benzo[*b*][1,6]naphthyridin-1-one (25). Prepared according to the general procedure for the cyclization but was refluxed for 12 h and $Yb(OTf)_3$ was used as the catalyst. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.27 (dd, J = 8.0, 1.5 Hz, 1H), 7.25 (dd, J = 7.3, 1.5 Hz, 1H), 7.06 (dd, J = 6.8, 2.0 Hz, 1H), 6.96 (d, J = 8.0 Hz, 1H), 6.86 (d, J = 8.5 Hz, 2H), 6.83 (dd, J = 9.0, 2.0 Hz, 2H), 6.35 (d, J = 2.5 Hz, 1H), 6.15 (dd, J = 8.0, 2.5 Hz, 1H), 5.96 (d, J = 8.5 Hz, 1H), 4.90 (d, J = 15.0 Hz, 1H), 4.80 (d, J = 3.0 Hz, 1H), 4.02 (d, J = 15.0 Hz, 1H), 3.78 (s, 3H), 3.76 (s, 3H), 3.71 (s, 3H), 3.39 (dd, J = 6.5, 3.5 Hz, 1H), 3.35 (d, J = 12.0 Hz, 1H), 2.93 (s, 3H), 2.68 (dd, J = 6.8, 2.0 Hz, 1H), 2.53 (dd, J = 12.3, 2.0 Hz, 1H), 1.01 (s, 3H), 0.99 (s, 3H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 169.5, 159.5, 157.9, 149.9, 135.0, 129.5, 128.9, 128.7, 128.5, 128.1, 119.8, 119.4, 116.9, 113.7, 104.6, 97.9, 63.7, 55.3, 55.2, 54.7, 49.6, 46.5, 45.2, 43.5, 37.0, 26.5, 25.2; HRMS calcd for $C_{31}H_{36}O_4N_2$ was 500.26751 (M^+), found 500.26789.

***N*-(2,4-Dimethoxybenzyl)-3-(4-methoxyphenyl)-acryl Amide (26).** Prepared according to the general procedure for the cyclization. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.52 (d, J = 15.5 Hz, 1H), 7.46 (d, J = 8.5 Hz, 2H), 7.20 (d, J = 8.5 Hz, 1H), 6.89 (d, J = 9.0 Hz, 2H), 6.48 (d, J = 2.0 Hz, 1H), 6.45 (dd, J = 8.8 Hz, 1H), 6.29 (d, J = 16.0 Hz, 1H), 6.07 (m, 1H), 4.43 (d, J = 6.0 Hz, 2H), 3.84 (s, 3H), 3.81 (s, 3H), 3.78 (s, 3H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 160.9, 140.2, 130.6, 129.5, 128.0, 119.1, 114.5, 104.2, 98.7, 55.7, 39.2; IR 3283, 3070, 3002, 2928, 2833, 1661, 1601, 1511, 1463, 1420; HRMS calcd for $C_{19}H_{21}O_4N$ was 327.14706 (M^+), found 327.14703.

3-(4-Methoxyphenyl)-acrylic Acid Methyl Ester (27). To a solution of trifluoroethylphosphonoester (800 μL, 3.8 mmol), 18-crown-6 (2.5 g, 9.5 mmol) in 60 mL of anhydrous THF was cooled to −78 °C under argon and treated with KNTMS (0.5 M in toluene) (7.56 mL, 3.78 mmol). *p*-Anisaldehyde (400 μL, 3.8 mmol) was then added, and the resulting mixture was stirred for 30 min at −78 °C. Saturated NH_4Cl (30 mL) was added, and the product was extracted with CH_2Cl_2 (3 × 25 mL), dried ($MgSO_4$), and concentrated. Flash chromatography was performed using hexane/ethyl acetate (10:1) to afford a clear liquid weighing 630 mg. Yield 87%. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.71 (dt, J = 9.0, 2.5 Hz, 2H), 6.88 (dt, J = 8.3 Hz, 2H), 6.86 (d, J = 12.5 Hz, 1H), 5.84 (d, J = 12.5 Hz, 1H), 3.82 (s, 3H), 3.72 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 166.8, 160.4, 143.5, 132.1, 127.2, 116.6, 113.4, 55.2, 51.2; IR 3008, 2921, 2851, 1724, 1625, 1608, 1514, 1450 cm^{-1}; HRMS calcd for $C_{11}H_{12}O_3$ was 192.07864 (M^+), found 192.07829.

3-(4-Methoxyphenyl)-acrylic Acid (28). To **27** (590 mg, 3.3 mmol) dissolved in 20 mL of THF/H_2O/MeOH (1:1:1) at 0 °C, lithium hydroxide (953 mg, 39.7 mmol) was added and was stirred for 48 h. Upon TLC confirmation of reaction completion, the mixture was cooled in an ice bath and HCl (1 M) added dropwise until the pH changed to about 2−3 as indicated by universal pH paper. The mixture was extracted using CH_2Cl_2 (3 × 20 mL), dried ($MgSO_4$), and concentrated to afford a creamy solid weighing 522 mg. Yield 87%. ^{1}H NMR (500 MHz, $CDCl_3$) δ 11.60 (s, 1H), 7.72 (dt, J = 9.0, 2.5 Hz, 2H), 6.97 (dd, J = 12.5, 6.0 Hz, 1H), 6.89 (dt, J = 9.0, 2.5 Hz, 2H), 5.85 (dd, J = 13.0, 6.5 Hz, 1H), 3.83 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.8, 160.7, 145.9, 132.6, 126.9, 116.0, 113.5, 55.3; IR 3037 (broad), 2949, 2839, 2570, 1695, 1602, 1503, 1450 cm^{-1}; HRMS calcd for $C_{29}H_{32}O_4N_2$ was 178.06299 (M^+), found 178.06299.

{(2,4-Dimethoxybenzyl)-[3-(4-methoxyphenyl)-acryloyl]-amino}-acetic Acid Methyl Ester (29). Prepared according to the general procedure for the synthesis of amides. Major rotamer ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.54 (dt, J = 8.9, 2.8 Hz, 2H), 6.93 (d, J = 8.5 Hz, 1H), 6.88 (dt, J = 9.0, 2.5 Hz, 2H), 6.64 (d, J = 13.0 Hz, 1H), 6.43 (d, J = 2.5 Hz, 1H), 6.40 (d, J = 8.3, 2.5 Hz, 1H), 6.10 (d, J = 13.0 Hz, 1H), 4.57 (s, 2H), 4.11 (s, 2H), 3.82 (s, 3H), 3.78 (s, 3H), 3.74 (s, 3H), 3.73 (s, 3H); minor rotamer ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.34 (d, J = 8.0 Hz, 1H), 7.26 (d, J = 8.5 Hz, 2H), 6.71 (dt, J = 9.0, 2.5 Hz, 2H), 6.52 (d, J = 12.3 Hz, 1H), 6.49 (dd, J = 8.0, 2.5 Hz, 1H), 6.43 (d, J = 3.0 Hz, 1H), 5.88 (d, J = 12.3 Hz, 1H), 4.67 (s, 2H), 3.97 (s, 2H), 3.81 (s, 3H), 3.77 (s, 3H), 3.62 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) for both rotamers δ 170.0, 169.8, 169.7, 160.8, 159.8, 159.7, 158.6, 134.6, 133.2, 132.4, 130.5, 130.1, 130.0, 127.8, 127.6, 120.6, 120.1, 116.7, 116.0, 113.8, 113.7, 104.2, 103.9, 98.5, 98.2, 55.3, 55.2, 55.2, 55.1, 52.0, 51.9, 49.3, 47.6, 45.1, 43.2; IR 3072, 3008, 2950, 2838, 1754, 1608, 1514, 1450 cm^{-1}; HRMS calcd for $C_{22}H_{25}O_6N$ was 399.16819 (M^+), found 399.16832.

***N*-(2,4-Dimethoxybenzyl)-*N*-(2-hydroxyethyl)-3-(4-methoxyphenyl)-acryl Amide (30).** Prepared according to the general procedure for the reduction of esters. Major rotamer ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.44 (d, J = 9.5 Hz, 2H), 6.92 (d, J = 8.5 Hz, 1H), 6.87 (dd, J = 9.0, 2.0 Hz, 2H), 6.63 (d, J = 12.5 Hz, 1H), 6.43 (d, J = 2.0 Hz, 1H), 6.40 (dd, J = 8.3, 2.0 Hz, 1H), 6.07 (d, J = 12.5 Hz, 1H), 4.45 (s, 2H), 3.81 (s, 3H), 3.78 (s, 3H), 3.74 (s, 3H), 3.68 (t, J = 5.3 Hz, 2H), 3.54 (t, J = 4.8 Hz, 2H), 3.19 (s, 1H); minor rotamer ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.33 (d, J = 8.0 Hz, 1H), 7.25 (d, J = 9.0 Hz, 2H), 6.70 (d, J = 9.0 Hz, 2H), 6.51 (d, J = 12.5 Hz, 1H), 6.48 (dd, J = 8.3, 2.0 Hz, 1H), 6.46 (d, J = 2.0 Hz, 1H), 6.08 (d, J = 13.0 Hz, 1H), 4.68 (s, J = 2H), 3.81 (s, 3H), 3.78 (3, 3H), 3.75 (s, 3H), 3.61 (t, J = 5.8 Hz, 2H), 3.40 (t, J = 5.5 Hz, 2H), 3.19 (s, 1H); ^{13}C NMR (125 MHz, $CDCl_3$) for both rotamers δ 171.5, 169.9, 160.7, 160.3, 159.8, 159.4, 158.4, 134.1, 132.4, 131.6, 130.1, 129.9, 129.5, 129.3, 128.1, 127.9, 121.7, 120.7, 117.4, 116.4, 114.1, 113.8, 113.6, 104.2, 103.9, 98.5, 98.2, 62.3, 62.0, 61.9, 59.9, 55.3, 55.3, 55.2, 55.2, 55.1, 49.7, 48.4, 48.2, 47.9, 41.6; IR 3376, 2996, 2932, 2839, 1602, 1503, 1456, 1351 cm^{-1}; HRMS calcd for $C_{21}H_{25}O_5N$ was 371.17327 (M^+), found 371.17315.

***N*-(2,4-Dimethoxybenzyl)-3-(4-methoxyphenyl)-*N*-(2-oxoethyl)-acrylamide (31).** Prepared according to the general procedure for the oxidation of alcohols. Major rotamer ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.40 (s, 1H), 7.48 (d, J = 8.8 Hz, 2H), 6.91 (d, J = 8.8 Hz, 1H), 6.89 (d, J = 9.2 Hz, 2H), 6.70 (d, J = 12.8 Hz, 1H), 6.41 (s, 1H), 6.39 (dd, J = 8.0, 2.4 Hz, 1H), 6.14 (d, J = 13.3 Hz, 1H), 4.50 (s, 2H), 4.07 (s, 2H), 3.82 (s, 3H), 3.78 (s, 3H), 3.73 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 197.9, 169.9, 161.0, 159.8, 158.6, 134.7, 133.5, 132.7, 130.3, 129.9, 127.8, 120.0, 115.9, 113.8, 104.1,

98.6, 55.3, 55.2, 55.1, 54.1, 48.4; minor rotamer ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.01 (s, 1H), 7.40 (d, J = 8.0 Hz, 2H), 7.24 (d, J = 8.8 Hz, 2H), 6.71 (d, J = 8.8 Hz, 1H), 6.54 (d, J = 12.0 Hz, 1H), 6.48 (m, 2H), 5.86 (d, J = 12.8 Hz, 1H), 4.67 (s, 2H), 3.96 (s, 2H), 3.81 (s, 3H), 3.77 (s, 3H), 3.71 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 198.5, 169.9, 161.0, 159.8, 158.6, 134.7, 133.5, 132.7, 130.5, 129.3, 127.5, 120.5, 116.4, 113.8, 104.4, 98.2, 58.1, 43.7; IR 3002, 2944, 2827, 1724, 1608, 1509, 1450, 1380 cm^{-1}; HRMS calcd for $C_{21}H_{23}O_5N$ was 369.15762 (M^+), found 369.15694.

2-(2,4-Dimethoxybenzyl)-9-(4-methoxyphenyl)-4-methyl-2,3,3a,4,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (32). Prepared according to the general procedure for the cyclization. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.18 (d, J = 8.5 Hz, 3H), 7.13 (d, J = 8.3, 3.5 Hz, 4H), 6.98 (dd, J = 7.8, 2.0 Hz, 1H), 6.92 (d, J = 9.0 Hz, 1H), 6.87 (d, J = 7.5 Hz, 1H), 6.86 (d, J = 9.0 Hz, 2H), 6.78 (m, 5H), 6.67 (dt, J = 7.3, 1.0 Hz, 1H), 6.45 (dd, J = 6.0, 2.5 Hz, 1H), 6.42 (d, J = 2.0 Hz, 1H), 6.39 (d, J = 2.5 Hz, 1H), 6.36 (dd, J = 8.0, 2.5 Hz, 1H), 4.68 (d, J = 4.5 Hz, 1H), 4.49 (d, J = 14.5 Hz, 1H), 4.41 (d, J = 14.5 Hz, 1H), 4.36 (d, J = 15.0 Hz, 1H), 4.32 (d, J = 12.0 Hz, 1H), 4.29 (d, J = 15.0 Hz, 1H), 3.79 (s, 3H), 3.78 (s, 6H), 3.77 (s, 3H), 3.75 (s, 3H), 3.67 (s, 3H), 3.48 (dd, J = 8.5, 6.5 Hz, 1H), 3.45 (m, 2H), 3.22 (t, J = 9.3 Hz, 1H), 3.06 (t, J = 9.0 Hz, 1H), 2.95 (dd, J = 12.0, 4.5 Hz, 1H), 2.89 (t, J = 7.0 Hz, 1H), 2.81 (s, 3H), 2.75 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 172.5, 171.4, 160.5, 158.5, 158.1, 158.0, 148.2, 148.0, 136.0, 135.3, 131.7, 131.5, 131.4, 131.0, 131.0, 130.6, 130.2, 129.2, 129.1, 128.7, 127.9, 127.3, 125.7, 119.0, 118.8, 117.0, 116.7, 113.7, 113.5, 113.3, 112.9, 104.2, 104.0, 98.4, 98.4, 59.5, 55.3, 55.3, 55.2, 55.1, 52.4, 50.6, 50.0, 49.8, 49.3, 45.1, 42.3, 40.8, 40.4, 36.2, 36.2.

[(2,4-Dimethoxybenzyl)-(3-furan-2-yl-acryloyl)-amino]-acetic Acid Methyl Ester (33). Prepared according to the general procedure for the synthesis of amides. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.53 (d, J = 15.0 Hz, 1 H), 7.42 (s, 1H), 7.07 (d, J = 8.0 Hz, 1H), 6.98 (d, J = 15.0 Hz, 1H), 6.55 (d, J = 3.5 Hz, 1H), 6.47 (dd, J = 4.3, 2.0 Hz, 2H), 6.45 (dd, J = 8.8, 2.0 Hz, 1H), 4.66 (s, 2H), 4.14 (s, 2H), 3.80 (s, 6H), 3.72 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 169.9, 167.3, 160.7, 158.4, 151.7, 143.9, 129.9, 129.2, 116.7, 114.9, 113.82, 112.1, 103.9, 98.6, 55.3, 55.1, 52.0, 47.5, 47.3; IR 3113, 2996, 2950, 2836, 1742, 1660, 1619, 1561, 1509 cm^{-1}; HRMS calcd for $C_{19}H_{21}O_6N$ was 359.13689 (M^+), found 359.13673.

***N*-(2,4-Dimethoxybenzyl)-3-furan-2-yl-*N*-(2-hydroxyethyl)-acryl Amide (34).** The above compound was prepared according to the general procedure for the reduction of esters. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.53 (d, J = 15.5 Hz, 1H), 7.40 (s, 1H), 7.02 (d, J = 8.0 Hz, 1H), 6.84 (d, J = 15.5 Hz, 1H), 6.55 (d, J = 3.0 Hz, 1H), 6.48 (dd, J = 5.5, 2.0 Hz, 1H), 6.45 (d, J = 3.0 Hz, 1H), 6.43 (dd, J = 8.7, 2.0 Hz, 1H), 4.62 (s, 2H), 3.82 (s, 3H), 3.80 (s, 3H), 3.77 (t, J = 5.0 Hz, 2H), 3.63 (t, J = 5.0 Hz, 2H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 169.4, 160.6, 158.1, 151.6, 144.0, 130.1, 128.4, 116.7, 114.9, 114.2, 112.1, 103.9, 98.6, 62.4, 55.3, 55.2, 50.3, 47.6; IR 3370, 2996, 2938, 2833, 1649, 1584, 1503, 1415 cm^{-1}; IR 3002, 2943, 2839, 1730, 1643, 1619, 1509, 1427 cm^{-1}; HRMS calcd for $C_{18}H_{21}O_5N$ was 331.14197 (M^+), found 331.14135.

***N*-(2,4-Dimethoxybenzyl)-3-furan-2-yl-*N*-(2-oxoethyl)-acryl Amide (35).** Prepared according to the general procedure for the oxidation of alcohols. ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.41 (s, 1H), 7.54 (d, J = 15.6 Hz, 1H), 7.45(s, 1H), 7.08 (d, J = 8.0 Hz, 1H), 7.05 (d, J = 14.8 Hz, 1H), 6.58 (d, J = 3.2 Hz, 1H), 6.46 (m, 3H), 4.66 (s, 2H), 4.10 (s, 2H), 3.80 (s, 3H), 3.79 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 199.0, 167.5, 161.0, 158.4, 151.6, 144.1, 130.1, 129.7, 116.4, 114.3, 112.2, 104.0, 98.7, 57.5, 56.3, 55.4, 55.1, 48.2, 45.6; IR 3002, 2944, 2839, 1730, 1643, 1619, 1509, 1427; HRMS calcd for $C_{18}H_{19}O_5N$ 329.12632 (M^+), found 329.12644

2-(2,4-Dimethoxybenzyl)-9-furan-2-yl-4-methyl-2,3,3a,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (36). Prepared according to the general procedure for the cyclization. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.32 (t, J = 1.3 Hz, 1H), 7.14 (d, J = 8.5 Hz, 1H), 7.11 (dd, J = 7.8, 1.5 Hz, 1H), 6.87 (dt, J = 7.6, 1.3 Hz, 1H), 6.78 (dd, J = 8.0, 1.0 Hz, 1H), 6.71 (dt, J = 7.3, 1.0 Hz, 1H), 6.45 (d, J = 2.0 Hz, 1H), 6.44 (dd, J = 8.0, 2.5 Hz, 1H), 6.37 (dd, J = 4.3, 2.5 Hz, 2H), 4.49 (d, J = 14.5 Hz, 1H), 4.44 (d, J = 12.0 Hz, 1H), 4.39 (d, J = 14.5 Hz, 1H), 3.79 (s, 3H), 3,77(s, 3H), 3.49 (dd, J = 8.5, 6.5 Hz, 1H), 3.34 (ddd, J = 11.1, 9.3, 6.5 Hz, 1H), 3.24 (t, J = 9.0 Hz, 1H), 3.14 (t, J = 12.0 Hz, 1H), 2.73 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 171.9, 160.4, 158.5, 154.4, 147.6, 141.4, 130.9, 129.4, 127.7, 124.8, 119.0, 116.9, 113.6, 110.1, 108.1, 104.2, 98.3, 77.2, 59.2, 55.3, 55.2, 49.8, 46.9, 40.5, 39.0, 36.0; IR 3002, 2938, 2833, 1701, 1608, 1514, 1468, 1363, 1299; HRMS calcd for $C_{25}H_{26}O_4N_2$ was 418.18926 (M^+), found 418.18927.

2-(2,4-Dimethoxybenzyl)-9-furan-2-yl-4-methyl-2,3,3a,9,9a-hexahydropyrrolo[3,4-*b*]quinolin-1-one (37). Prepared according to the general procedure for the cyclization. ^{1}H NMR (500 MHz, CD_2Cl_2) δ 7.34 (d, J = 1.5 Hz, 1H), 7.20 (dt, J = 7.9, 1.5 Hz, 1H), 7.09 (dd, J = 7.0, 1.5 Hz, 1H), 6.75 (dt, J = 7.5, 1.0 Hz, 1H), 6.58 (d, J = 3.0 Hz, 1H), 6.47 (d, J = 8.0 Hz, 1H), 6.40 (d, J = 2.0 Hz, 1H), 6.28 (d, J = 3.0 Hz, 1H), 6.27 (dd, J = 8.0, 2.5 Hz, 1H), 5.90 (d, J = 3.0 Hz, 1H), 4.48 (d, J = 3.5 Hz, 1H), 4.42 (d, J = 15.5 Hz, 1H), 4.16 (d, J = 15.0 Hz, 1H), 4.03 (ddd, J = 8.3, 6.0, 2.5 Hz, 1H), 3.76 (s, 3H), 3.71 (s, 3H), 3.50 (dd, J = 8.5 Hz, 1H), 3.34 (dd, J = 10.5, 5.5 Hz, 1H), 3.26 (dd, J = 10.5, 1.5 Hz, 1H), 2.73 (s, 3H); ^{13}C NMR (125 MHz, CD_2Cl_2) δ 172.6, 160.7, 156.4, 145.7, 141.6, 129.5, 129.4, 128.7, 123.8, 118.1, 116.6, 112.4, 110.6, 105.8, 104.6, 98.4, 55.6, 55.5, 49.9, 47.8, 40.8, 37.7, 36.0; HRMS calcd for $C_{25}H_{26}O_4N_2$ was 418.1893 (M^+), found 418.1894.

Supporting Information Available: General synthetic procedures and NMR spectra for reported compounds. This material is available free of charge via the Internet at http://pubs.acs.org.

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