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Full Paper

A Green and Convenient Route for the Regioselective Synthesis of New Substituted 3-Aryl-5*H*-indeno[1,2-*c*] pyridazines as Potential Monoamine Oxidase Type A Inhibitors

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Several indeno[1,2-*c*]pyridazines were efficiently synthesised using the one-pot, three-component reaction of substituted indanones, arylglyoxalmonohydrates, and hydrazine in the presence of 1,5-diazabicyclo[4,3,0]non-5-ene (DBN) in water at room temperature. These substituted 3-aryl indeno[1,2-*c*]pyridazines can be considered as potential monoamine oxidase type A (MAO_A) inhibitors. The advantages of this new strategy are the novelty of the indenopyridazine derivatives, high regioselectivity, use of water as the solvent, no requirement for toxic metal catalysts, and good to excellent yields.

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Introduction

Monoamino oxidases (MAOs) are flavin adenine dinucleotide (FAD)-containing enzymes localised in the outer mitochondrial membrane.[1] MAOs are involved in the oxidative deamination of important endogenous amines, including the monoamine neurotransmitters serotonin (5-HT), norepinephrine (NE), and dopamine (DA), as well as exogenous amines, including the hypertensive dietary amine tyramine.[2] Two different isoenzymatic forms have been identified, namely MAO_A and MAO_B,[3] which differ in their amino acid sequences, three-dimensional structures,[4–6] substrate specificity, and sensitivity to inhibitors.[7] Due to their role in the metabolism of monoamine neurotransmitters, MAO_A and MAO_B present a considerable pharmacological interest. The lack of selective inhibition, irreversible mechanism of action, severe side effects, e.g. hepatotoxicity and life-threatening hypertensive crisis, associated with the first-generation of antidepressant MAO_A inhibitors, have stimulated further research aimed to discover novel, less toxic drugs.[8,9] Several selective MAO_A inhibitors[10] acting as antidepressants (i.e. moclobemide **1**,[11] brofaromine **2**, clorgyline **3**, and toloxatone **4**; Fig. 1), and selective MAO_B inhibitors acting as anti-Parkinson agents (i.e. lazabemide **5**, selegiline **6**, safinamide **7**, and rasagiline **8**; Fig. 2)[12–14] have been discovered.

The pyridazine heterocycle and hetero-fused analogues continue to attract attention due to their wide variety of interesting biological activities.[15,16] The synthesis and utility of many pyridazine derivatives with analgesic, insecticide, fungicide, cardiotonic, and bacteriocide properties have been reported.[17–23] Substituted pyrimidopyridazines **9**–**13** were recognised as potential MAO_B inhibitors[24–26] (Fig. 2). Recently, indenopyridazine derivatives have been studied for their valuable properties.[27–29] Carotti and co-workers have reported that 3-phenyl-5*H*-indeno [l,2-*c*]pyridazin-5-one **14** shows MAO_B inhibitory activity, however the reduction of the carbonyl group in these systems led to a decrease in their MAO_B inhibitory property along with an increase in their MAO_A inhibitory effect.[30,31] Hence, 3-phenyl-5*H*-indeno[l,2-*c*]pyridazine **15a** showed a measurable MAO_A inhibitory activity.[32]

As shown in Scheme 1, the reported synthetic strategy for the preparation of the 3-phenyl-5*H*-indenopyridazine **15** involved a multistep procedure, with the initial preparation of 3-phenyl-5*H*-indenopyridazin-5-one **14** through the reaction of acetophenone **16** with ninhydrin **17** in acetic acid at reflux, and the condensation of the resulting intermediate **18** with hydrazine and subsequent reduction of the compound **14** using $TsNHNH_2$/$NaBH_3CN$.[31]

Following our recent investigations on the one-pot synthesis of pyridazine derivatives,[33,34] and due to the probable pharmaceutical value of the 3-aryl-5*H*-indeno[1,2-*c*]pyridazines as potential MAO_A inhibitors,[32] herein we report a convenient approach to the regioselective synthesis of new substituted 3-arylindenopyridazines using the one-step reaction of substituted indanones with arylglyoxalmonohydrates and hydrazine hydrate in the presence of catalytic 1,4-diazabicyclo[2.2.2]octane (DABCO), 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), or 1,5-diazabicyclo

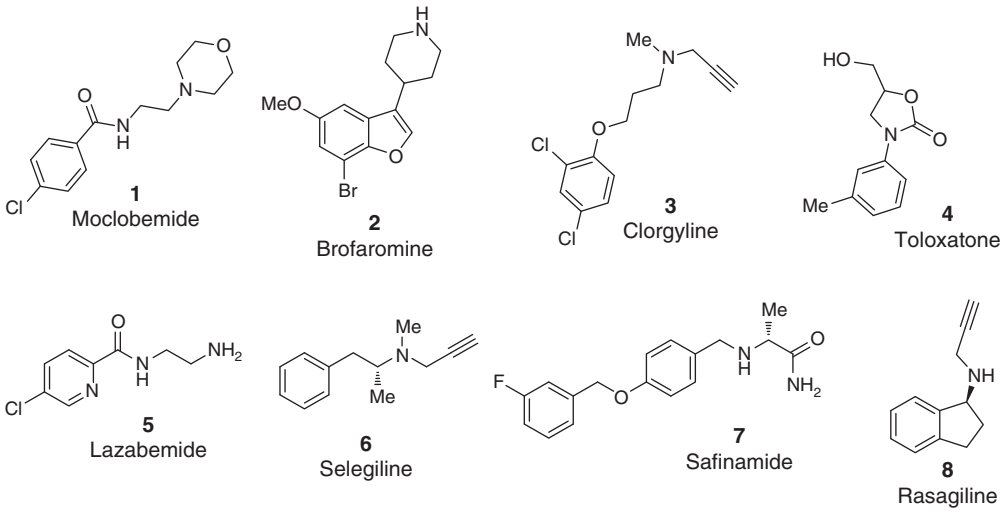


Fig. 1. Representative MAO_A and MAO_B inhibitors.

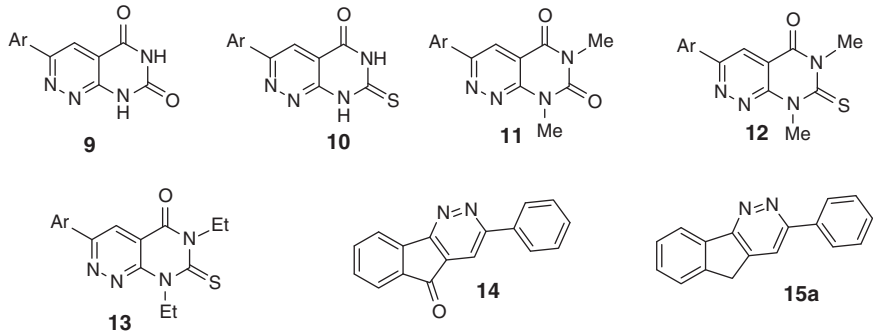
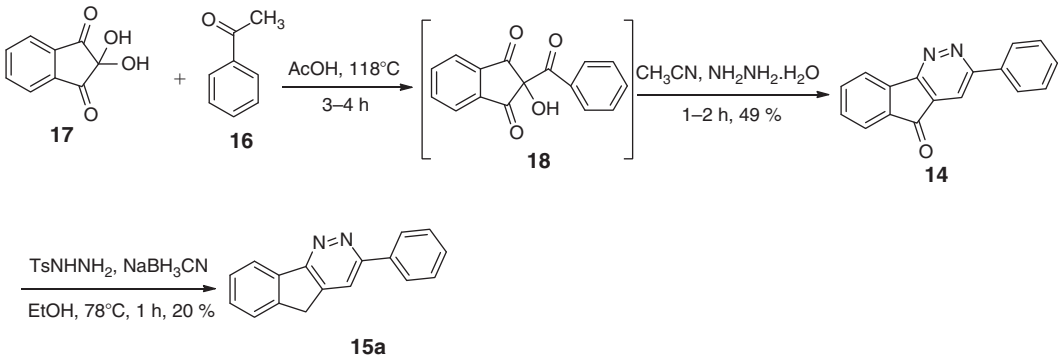


Fig. 2. Selected pyridazine derivatives as potential MAO_B and MAO_A inhibitors.



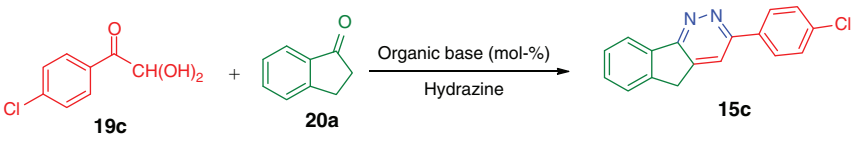
Scheme 1. Multi-step synthesis of 3-phenyl-5*H*-indeno[1,2-*c*]pyridazine.[31]

[4,3,0]non-5-ene (DBN). This procedure is an extension of that we have used in the synthesis of new heterocyclic compounds using multicomponent reaction (MCR) techniques.[33–37]

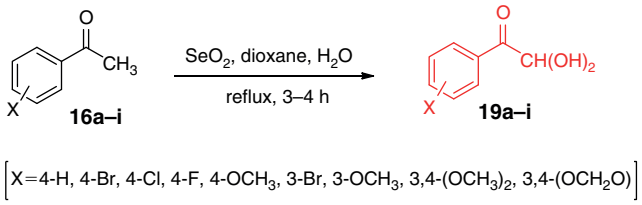
Results and Discussion

As a trial reaction, the one-pot reaction of 4-chlorophenylglyoxalmonohydrate **19c** and indanone **20a**, in the presence of excess hydrazine hydrate in water at different temperatures (25, 50, and 100°C) was tested, but all of these reactions failed to deliver the desired product (Table 1, entries 1–3), which may be due to the low acidity of the indanone's α-hydrogens under these conditions. As we have recently focussed on using green organic bases to catalyse the combinatorial reactions of arylglyoxalmonohydrates with various active methylene-carrying compounds,[33–36] the reactions were repeated in the presence of 10 mol-% DABCO, DBU, and DBN at room temperature (Table 1, entries 4–6), and now led to traces of the desired product **15c**. Monitoring the reaction progress by thin-layer chromatography showed that upon increasing the reaction temperature to 50°C, formation of the undesired hydrazone by-products was facilitated (Table 1, entries 7–9). However,

Table 1. Optimisation of the reaction conditions



Entry	Catalyst [mol-%]	Temp. [°C]	Time [h]	Yield [%]
1	–	25	12	0
2	–	50	12	0
3	–	100	12	0
4	DABCO (10)	25	6	10
5	DBU (10)	25	6	8
6	DBN (10)	25	6	20
7	DABCO (10)	50	6	0
8	DBU (10)	50	6	0
9	DBN (10)	50	6	0
10	DABCO (15)	25	6	18
11	DBU (15)	25	6	15
12	DBN (15)	25	6	30
13	DABCO (20)	50	3	32
14	DBU (20)	50	3	25
15	DBN (20)	50	3	58
16	DABCO (25)	50	3	38
17	DBU (25)	50	3	30
18	DBN (25)	25	3	85
19	DABCO (30)	50	3	30
20	DBU (30)	50	3	23
21	DBN (30)	50	3	72



Scheme 2. Synthesis of arylglyoxalmonohydrates.

increasing the catalyst loading from 15 to 30 mol-% (Table 1, entries 10–24) showed that 25 mol-% DBN (Table 1, entry 18) was the best catalyst.

Various arylglyoxalmonohydrates **19a**–**i** were prepared from the corresponding acetophenones **16a**–**i** via the literature procedure as shown in Scheme 2.[38]

Under the optimised reaction conditions, the arylglyoxalmonohydrates **19a**–**i** were then reacted with substituted indanones **20a**–**c** and hydrazine hydrate in the presence of DBN (25 mol-%) as shown in Scheme 3.

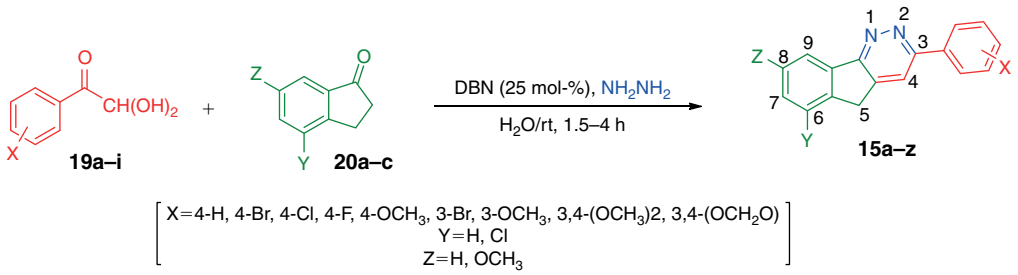
It was found that the obtained yields using 4-chloroindanone were higher than that with indanone or 6-methoxyindanone due to the increased acidity of the α-hydrogens of the 4-chloroindanone. It is worth noting that all the obtained substituted indeno[1,2-*c*]pyridazines are the 3-aryl regioisomers as listed in Table 2.

The structures of the substituted 3-aryl-5*H*-indeno[1,2-*c*]pyridazines **15a**–**z** were characterised using IR, ^{1}H NMR, and ^{13}C NMR spectroscopy, and microanalysis. The characteristic singlet around ~7.69–7.97 ppm in the ^{1}H NMR spectra was ascribed to the C_4–H of the pyridazine ring, and all new substituted indeno[1,2-*c*]pyridazines **15a**–**z** are believed to be the only regioisomers present, with no evidence for the formation of alternative isomers. Furthermore, a distinct singlet near ~3.84–4.05 ppm was ascribed to the methylene group of the indene moiety and was present in all new products. The spectroscopic data obtained for compound **15a** was also in accordance with the data reported by Carotti and co-workers.[32] As shown in Scheme 4, this regioselectivity may be due to the high tendency of the carbanion **21** to attack the formyl group of arylglyoxal **22** (path a) leading to regioselective formation of the Knoevenagel adduct **23**. In the ^{13}C NMR spectra of the products **15a**–**z**, two signals located around ~155–162 ppm were attributed to the carbimide groups and neither was attached to a H atom, thereby ruling out the 4-aryl substituted isomeric structure **24,** which would result from the less likely attack of carbanion **21** on the keto carbonyl of **22** (path b), as shown in Scheme 4. Similar to the previous reports on the synthesis of 3-aryl substituted pyridazine derivatives,[39,40] H-3 in structure **25** would also have a lower chemical shift than that of H-4 in **15**. In addition, the carbon signal for the CH_2 group appeared at 33.6–34.6 ppm in all derivatives **15a**–**z**. In the IR spectra, the characteristic absorption bands at ~1580 and ~1450 cm^{-1} could be assigned to the stretching and bending vibrations of C=N and CH_2 bonds of the indenopyridazine ring, respectively.

Experimental

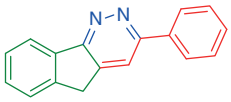
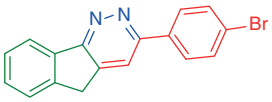
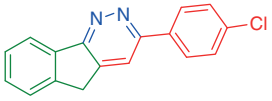
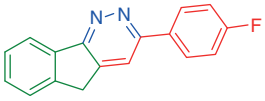
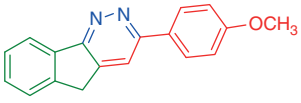
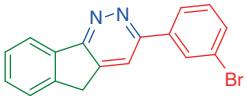
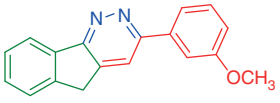
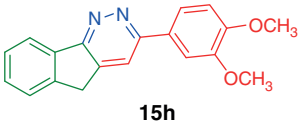
General Procedures

Melting points were determined on an Electrothermal 9200 apparatus. ^{1}H (300 MHz) and ^{13}C (75 MHz) NMR spectra were recorded on a BRUKER DRX-300 AVANCE spectrometer in $CDCl_3$ with tetramethylsilane as internal standard. Infrared spectra were recorded on a Perkin Elmer Spectrum Two FT-IR spectrophotometer, measured as KBr disks. Microanalyses were performed on a Leco Analyzer 932.



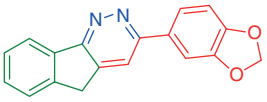
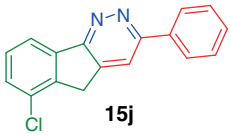
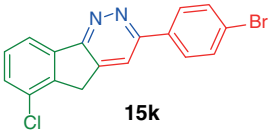
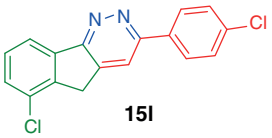
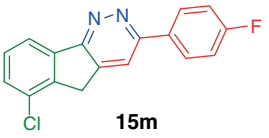
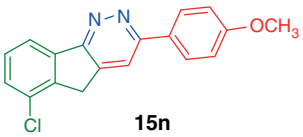
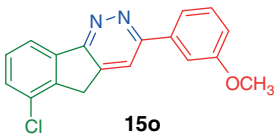
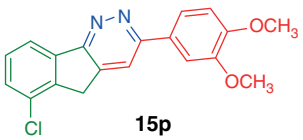
Scheme 3. One pot synthesis of substituted 3-arylindeno[1,2-*c*]pyridazines.

Table 2. Substituted 3-arylindeno[1,2-*c*]pyridazines

Entry	Indeno[1,2-*c*]pyridazine	Reaction time [h]	Yield [%]
1	 **15a**	3	78
2	 **15b**	3	80
3	 **15c**	3	85
4	 **15d**	3	79
5	 **15e**	3	69
6	 **15f**	3	80
7	 **15g**	3	71
8	 **15h**	3	75

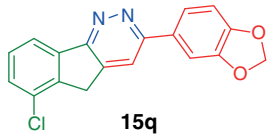
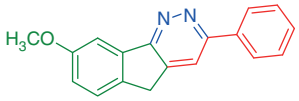
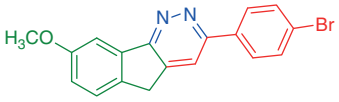
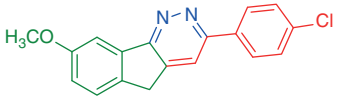
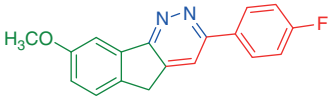
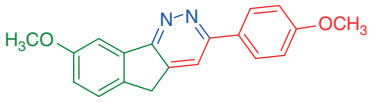
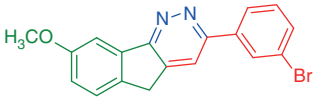
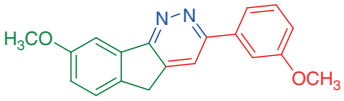
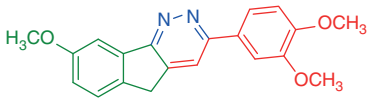
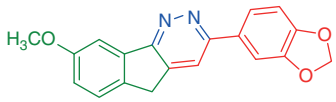
(*continued*)

Table 2. (Continued)

Entry	Indeno[1,2-*c*]pyridazine	Reaction time [h]	Yield [%]
9	 **15i**	3	72
10	 **15j**	1.5	89
11	 **15k**	1.5	90
12	 **15l**	1.5	92
13	 **15m**	1.5	77
14	 **15n**	1.5	74
15	 **15o**	1.5	79
16	 **15p**	1.5	78

(*continued*)

Table 2. (Continued)

Entry	Indeno[1,2-*c*]pyridazine	Reaction time [h]	Yield [%]
17	 **15q**	1.5	75
18	 **15r**	4	76
19	 **15s**	4	79
20	 **15t**	4	80
21	 **15u**	4	68
22	 **15v**	4	69
23	 **15w**	4	72
24	 **15x**	4	70
25	 **15y**	4	71
26	 **15z**	4	69

*General Procedures for Regioselective DBN Catalysed One-Pot Synthesis of Substituted 3-Aryl-5*H*-indeno[1,2-*c*]pyridazines **15a–z***

To a stirred mixture of indanone (1 mmol), arylglyoxalmonohydrate (1 mmol), and DBN (25 mol-%) in water (5 mL) was added hydrazine hydrate (3 mmol) at room temperature. The suspension was then stirred for 2–4 h, as shown in Table 2. After the appropriate time, the heterogeneous mixture was filtered and the product recrystallised from ethanol.

*3-Phenyl-5*H*-indeno[1,2-*c*]pyridazine (**15a**)*

Compound **15a** was obtained as white needles, mp 161–162°C. δ_H (300 MHz, $CDCl_3$) 3.99 (s, 2H, CH_2), 7.63–7.44 (m, 6H, Ar), 7.95 (s, 1H, Ar), 8.12 (dd, J_1 6.6, J_2 1.5, 2H, Ar), 8.42–8.33 (m, 2H, Ar). δ_C (75 MHz, $CDCl_3$) 34.6, 120.1, 121.9, 121.5, 125.3, 127.1, 127.9, 128.9, 129.6, 130.2, 137.0, 137.7, 141.3, 142.8, 156.5, 161.6. ν_{max} (KBr)/cm^{-1} 3060, 3030, 2895, 1463, 1384, 1074, 761, 692. Found C 83.60, H 4.96, N 11.65. $C_{17}H_{12}N_2$ requires C 83.58, H 4.95, N 11.4 7 %.[32]

*3-(4-Bromophenyl)-5*H*-indeno[1,2-*c*]pyridazine (**15b**)*

Compound **15b** was obtained as yellow needles, mp 218–220°C. δ_H (300 MHz, $CDCl_3$) 3.96 (s, 2H, CH_2), 7.44–7.67 (m, 5H, Ar), 7.88 (s, 1H, Ar), 7.98 (d, *J* 8.4, 2H, Ar), 8.31 (d, *J* 6, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.6, 119.8, 121.9, 124.2, 125.4, 127.9, 128.5, 130.3, 132.0, 135.7, 137.5, 141.5, 142.8, 155.3, 161.8. ν_{max} (KBr)/cm^{-1} 3057, 1615, 1590, 1493, 1351, 1069, 832. Found C 63.21, H 3.45, N 8.84. $C_{17}H_{11}BrN_2$ requires C 63.18, H 3.43, N 8.67 %.

*3-(4-Chlorophenyl)-5*H*-indeno[1,2-*c*]pyridazine (**15c**)*

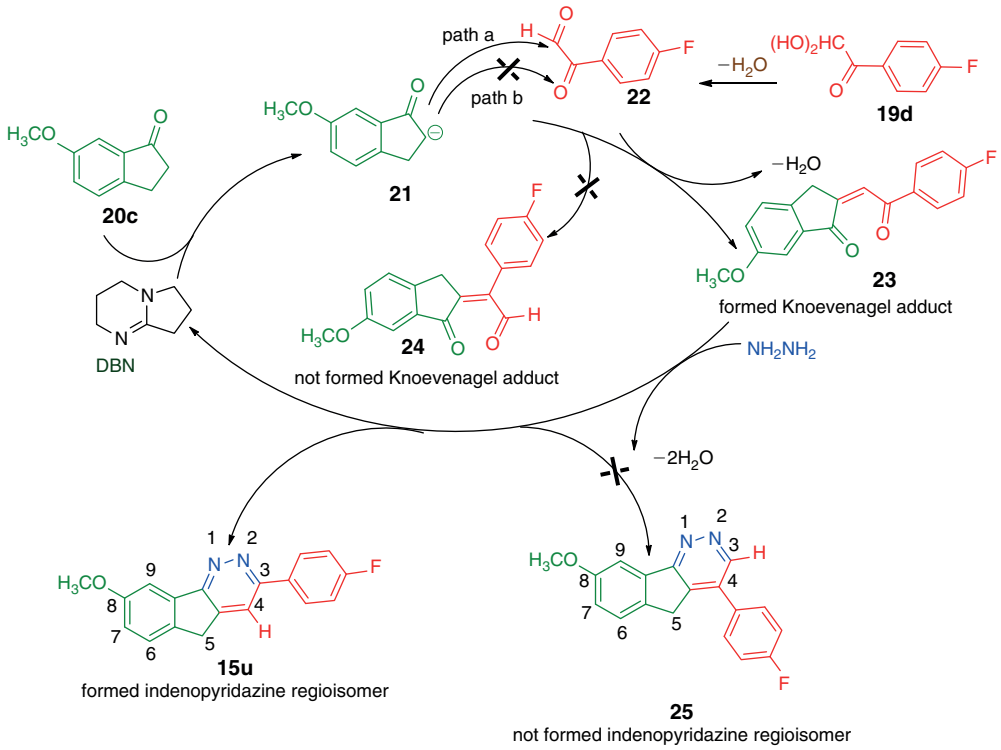
Compound **15c** was obtained as white needles, mp 244–245°C. δ_H (300 MHz, $CDCl_3$) 4.03 (s, 2H, CH_2), 7.41 (t, *J* 7.8, 1H, Ar), 7.48–7.67 (m, 4H, Ar), 7.96 (s, 1H, Ar), 8.10 (d, *J* 7.8, 2H, Ar), 8.28–8.42 (m, 2H, Ar). δ_C (75 MHz, $CDCl_3$) 34.6, 120.1, 122.1, 123.1, 125.4, 125.6, 128.0, 130.1, 130.4, 132.5, 137.9, 141.8, 142.9, 156.1, 162.3. ν_{max} (KBr)/cm^{-1} 3057, 2939, 2906, 1611, 1588, 1462, 1407, 1386, 1004, 780. Found C 73.20, H 3.94, N 10.17. $C_{17}H_{11}ClN_2$ requires C 73.25, H 3.98, N 10.05 %.

*3-(4-Fluorophenyl)-5*H*-indeno[1,2-*c*]pyridazine (**15d**)*

Compound **15d** was obtained as white needles, mp 208–209°C. δ_H (300 MHz, $CDCl_3$) 3.98 (s, 2H, CH_2), 7.20 (t, *J* 8.4, 2H, Ar), 7.48–7.67 (m, 3H, Ar), 7.89 (s, 1H, Ar), 8.08–8.18 (m, 2H, Ar), 8.32 (d, *J* 6, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.6, 115.8, 116.0, 119.8, 121.9, 125.3, 127.9, 128.8, 129.0, 130.3, 133.1, 137.6, 141.5, 142.8, 155.5, 161.6, 162.2, 165.5. ν_{max} (KBr)/cm^{-1} 3045, 2895, 1599, 1419, 1230, 839. Found C 77.87, H 4.21, N 10.78. $C_{17}H_{11}FN_2$ requires C 77.85, H 4.23, N 10.68 %.

*3-(4-Methoxyphenyl)-5*H*-indeno[1,2-*c*]pyridazine (**15e**)*

Compound **15e** was obtained as white needles, mp 174–176°C. δ_H (300 MHz, $CDCl_3$) 3.85 (s, 3H, OCH_3), 3.91 (s, 2H, CH_2), 6.99 (d, *J* 8.7, 2H, Ar), 7.4–7.6 (m, 3H, Ar), 7.82 (s, 1H, Ar), 8.05 (d, *J* 8.7, 2H, Ar), 8.29 (d, *J* 6.6, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.6, 55.3, 114.3, 119.4, 121.7, 125.3, 127.8, 128.3, 129.4, 130.0, 137.9, 141.3, 142.7, 156.0, 161.1, 162.3. ν_{max} (KBr)/cm^{-1} 3042, 2968, 2918, 2839, 1606, 1512, 1412, 1253, 1183, 1030, 836. Found C 78.84, H 5.15, N 10.32. $C_{18}H_{14}N_2O$ requires C 78.81, H 5.14, N 10.21 %.



Scheme 4. Proposed mechanism for the DBN catalysed regioselective synthesis of substituted 3-arylindeno[1,2-*c*]pyridazines.

*3-(3-Bromophenyl)-5*H*-indeno[1,2-c]pyridazine (**15f**)*

Compound **15f** was obtained as yellow needles, mp 194–195°C. δ_H (300 MHz, $CDCl_3$) 3.96 (s, 2H, CH_2), 7.41–7.54 (m, 4H, Ar), 7.57 (t, *J* 6.9, 1H, Ar), 7.88 (s, 1H, Ar), 8.03 (d, *J* 8.4, 2H, Ar), 8.31 (d, *J* 6.3, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.6, 118.2, 119.8, 122.0, 125.4, 128.0, 128.3, 129.1, 130.3, 131.5, 135.4, 135.9, 137.6, 141.5, 142.8, 155.4, 161.8. ν_{max} (KBr)/cm^{-1} 3057, 2928, 2893, 1613, 1595, 1417, 1096, 834, 718. Found C 63.20, H 3.46, N 8.80. $C_{17}H_{11}BrN_2$ requires C 63.18, H 3.43, N 8.67 %.

*3-(3-Mehoxyphenyl)-5*H*-indeno[1,2-c]pyridazine (**15g**)*

Compound **15g** was obtained as white needles, mp 150–151°C. δ_H (300 MHz, $CDCl_3$) 3.92 (s, 3H, OCH_3), 3.99 (s, 2H, CH_2), 7.03 (d, *J* 8.4, 1H, Ar), 7.42 (t, *J* 8.1, 1H, Ar), 7.48–7.68 (m, 4H, Ar), 7.82 (s, 1H, Ar), 7.94 (s, 1H, Ar), 8.34 (d, *J* 6.3, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.6, 55.4, 112.0, 116.0, 119.2, 120.2, 121.9, 125.3, 127.9, 129.8, 130.2, 137.8, 138.4, 141.4, 142.9, 156.3, 160.2, 161.8. ν_{max} (KBr)/cm^{-1} 2963, 2888, 2839, 1606, 1584, 1449, 1249, 1045, 782. Found C 78.78, H 5.12, N 10.35. $C_{18}H_{14}N_2O$ requires C 78.81, H 5.14, N 10.21 %.

*3-(3,4-Dimethoxyphenyl)-5*H*-indeno[1,2-c] pyridazine (**15h**)*

Compound **15h** was obtained as white needles, mp 208–210°C. δ_H (300 MHz, $CDCl_3$) 3.93 (s, 3H, OCH_3), 3.95 (s, 2H, CH_2), 4.01 (s, 3H, OCH_3), 6.94 (d, *J* 8.4, 1H, Ar), 7.43–7.6 (m, 4H, Ar), 7.88 (s, 1H, Ar), 7.95 (s, 1H, Ar), 8.30 (d, *J* 7.5, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.6, 55.9, 56.0, 110.0, 110.9, 119.3, 119.4, 121.8, 125.3, 127.9, 129.7, 130.1, 137.8, 141.3, 142.7, 149.4, 150.5, 155.8, 161.3. ν_{max} (KBr)/cm^{-1} 3003, 2943, 2839, 1596, 1518, 1422, 1273, 1136, 1019, 848. Found C 74.95, H 5.27, N 9.30. $C_{19}H_{16}N_2O_2$ requires C 74.98, H 5.30, N 9.20 %.

*3-(3,4-Methylenedioxyphenyl)-5*H*-indeno[1,2-c] pyridazine (**15i**)*

Compound **15i** was obtained as white needles, mp 210–212°C. δ_H (300 MHz, $CDCl_3$) 3.96 (s, 2H, CH_2), 6.03 (s, 2H, OCH_2O), 6.92 (d, *J* 6.9, 1H, Ar), 7.45–7.64 (m, 4H, Ar), 7.71 (s, 1H, Ar), 7.83 (s, 1H, Ar), 8.31 (d, *J* 6.3, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.5, 101.4, 107.4, 108.5, 119.6, 121.1, 121.8, 125.3, 127.8, 130.0, 131.2, 137.7, 141.3, 142.8, 148.4, 149.0, 155.9, 161.3. ν_{max} (KBr)/cm^{-1} 3002, 2910, 2792, 1607, 1499, 1414, 1390, 1241, 834, 747. Found C 75.05, H 4.25, N 9.84. $C_{18}H_{12}N_2O_2$ requires C 74.99, H 4.20, N 9.72 %.

*3-Phenyl-6-chloro-5*H*-indeno[1,2-c]pyridazine (**15j**)*

Compound **15j** was obtained as white needles, mp 207–208°C. δ_H (300 MHz, $CDCl_3$)3.97 (s, 2H, CH_2), 7.41–7.58 (m, 5H, Ar),7.96 (s, 1H, Ar), 8.11 (d, *J* 7.5, 2H, Ar), 8.18–8.27 (m, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.3, 120.3, 123.4, 127.2, 129.0, 129.7, 129.8, 130.0, 131.4, 136.8, 138.9, 140.7, 140.8, 157.0, 161.2. ν_{max} (KBr)/cm^{-1} 3066, 2935, 2897, 1614, 1588, 1451, 1385, 1067, 778. Found C 73.28, H 4.01, N 10.16. $C_{17}H_{11}ClN_2$ requires C 73.25, H 3.98, N 10.05 %.

*3-(4-Bromophenyl)-6-chloro-5*H*-indeno[1,2-c] pyridazine (**15k**)*

Compound **15k** was obtained as white needles, mp 174–176°C. δ_H (300 MHz, $CDCl_3$) 4.00 (s, 2H, CH_2), 7.38 (t, J 7.5, 1H, Ar), 7.50 (s, 2H, Ar), 7.61 (d, *J* 8.7, 1H, Ar), 7.96 (s, 1H, Ar), 8.05 (d, *J* 7.2, 1H, Ar), 8.23 (s, 1H.Ar), 8.28 (s, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.2, 120.3, 123.2, 125.6, 129.7, 130.1, 130.2, 130.4, 131.5, 132.7, 138.6, 139.4, 140.9, 155.5, 161.5. ν_{max} (KBr)/cm^{-1} 3070, 2927, 2884, 1617, 1587, 1447, 1393, 1067, 816, 781. Found C 57.06, H 2.80, N 7.96. $C_{17}H_{10}BrClN_2$ requires C 57.09, H 2.82, N 7.83 %.

*3-(4-Chlorophenyl)-6-chloro-5*H*-indeno[1,2-c] pyridazine (**15l**)*

Compound **15l** was obtained as white needles, mp 245–246°C. δ_H (300 MHz, $CDCl_3$) 4.05 (s, 2H, CH_2), 7.52 (s, 2H, Ar), 7.68 (d, *J* 7.8, 2H, Ar), 7.97–8.08 (m, 3H, Ar), 8.27 (s, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.2, 120.1, 123.2, 125.6, 128.0, 128.1, 128.7, 129.8, 130.2, 132.2, 138.6, 139.6, 140.0, 155.4, 162.3. ν_{max} (KBr)/cm^{-1} 3074, 2910, 2889, 1598, 1518, 1441, 1068, 839, 717. Found C 65.24, H 3.23, N 9.04. $C_{17}H_{10}Cl_2N_2$ requires C 65.20, H 3.22, N 8.94 %.

*3-(4-Fluorophenyl)-6-chloro-5*H*-indeno[1,2-c] pyridazine (**15m**)*

Compound **15m** was obtained as white needles, mp 175–176°C. δ_H (300 MHz, $CDCl_3$) 4.02 (s, 2H, CH_2), 7.20 (d, *J* 7.5, 1H, Ar), 7.43–7.58 (m, 3H, Ar), 7.97 (s, 1H, Ar), 8.03–8.19 (m, 2H, Ar), 8.24 (s, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.3, 115.1, 116.9, 119.9, 120.9, 126.2, 127.4, 128.1, 129.7, 130.0, 130.3, 130.6, 130.9, 131.7, 136.0, 140.8, 156.7, 161.1. ν_{max} (KBr)/cm^{-1} 3083, 2910, 2889, 1619, 1597, 1442, 1088, 839. Found C 68.85, H 3.45, N 9.60. $C_{17}H_{10}ClFN_2$ requires C 68.81, H 3.40, N 9.44 %.

*3-(4-Methoxyphenyl)-6-chloro-5*H*-indeno[1,2-c] pyridazine (**15n**)*

Compound **15n** was obtained as white needles, mp 196–198°C. δ_H (300 MHz, $CDCl_3$)3.88 (s, 3H, OCH_3), 3.98 (s, 2H, CH_2), 7.02 (d, *J* 8.7, 2H, Ar), 7.40–7.51 (m, 2H, Ar), 7.89 (s, 1H, Ar), 8.08 (d, *J* 7.8, 2H, Ar), 8.18–8.25 (m, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.2, 55.3, 114.3, 119.4, 120.0, 128.4, 129.0, 129.6, 129.7, 131.4, 139.7, 140.6, 140.7, 156.4, 160.5, 161.1. ν_{max} (KBr)/cm^{-1} 3024, 2931, 2893, 1600, 1511, 1442, 1408, 1250, 1223, 1158, 845. Found C 70.00, H 4.20, N 9.19. $C_{18}H_{13}ClN_2O$ requires C 70.02, H 4.24, N 9.07 %.

*3-(3-Mehoxyphenyl)-6-chloro-5*H*-indeno[1,2-c] pyridazine (**15o**)*

Compound **15o** was obtained as white needles, mp 150–151°C. δ_H (300 MHz, $CDCl_3$) 3.88 (s, 3H, OCH_3), 3.97 (s, 2H, CH_2), 7.03 (dd, J_1 6.6, J_2 1.8, 1H, Ar), 7.38–7.55 (m, 3H, Ar), 7.61 (d, *J* 7.5, 1H, Ar), 7.80 (s, 1H, Ar), 7.97 (s, 1H, Ar), 8.19–8.28 (m, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.3, 55.4, 112.1, 116.2, 119.3, 120.3, 129.7, 129.9, 130.0, 131.4, 138.1, 139.7, 140.7, 140.8, 156.7, 160.2, 161.3. ν_{max} (KBr)/cm^{-1} 3074, 3007, 2973, 2939, 2838, 1609, 1582, 1457, 1393, 1273, 1050, 782. Found C 69.99, H 4.22, N 9.21. $C_{18}H_{13}ClN_2O$ requires C 70.02, H 4.24, N 9.07 %.

*3-(3,4-Dimethoxyphenyl)-6-chloro-5*H*-indeno[1,2-c] pyridazine (**15p**)*

Compound **15p** was obtained as white needles, mp 195–197°C. δ_H (300 MHz, $CDCl_3$) 3.94 (s, 6H, 2 × OCH_3), 4.01 (s, 2H, CH_2), 6.93 (d, *J* 8.4, 1H, Ar), 7.43–7.56 (m, 3H, Ar), 7.91 (d, *J* 7.2, 2H, Ar), 8.18–8.23 (m, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.2, 55.9, 56.0, 110.0, 111.0, 119.4, 120.0, 129.3, 129.6, 129.8, 131.4, 139.7, 140.6, 140.7, 149.5, 150.7, 156.3, 160.7, 162.3. ν_{max} (KBr)/cm^{-1} 3095, 3057, 3032, 2952, 2897, 1589, 1516, 1461, 1244, 1024, 793. Found C 67.38, H 4.49, N 8.40. $C_{19}H_{15}ClN_2O_2$ requires C 67.36, H 4.46, N 8.27 %.

*3-(3,4-Methylenedioxyphenyl)-6-chloro-5*H*-indeno [1,2-c]pyridazine (**15q**)*

Compound **15q** was obtained as white needles, mp 244–245°C. δ_H (300 MHz, $CDCl_3$) 4.02 (s, 2H, CH_2), 6.07 (s, 2H, OCH_2O), 6.96 (d, *J* 9.3, 1H, Ar), 7.50 (s, 2H, Ar), 7.61 (d, *J* 7.8, 1H, Ar), 7.75 (s, 1H, Ar), 7.93 (s, 1H, Ar), 8.25 (s, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 34.2, 101.5, 107.4, 108.6, 119.6, 119.8, 121.3, 125.3, 127.8, 129.7, 129.9, 137.7, 141.3, 142.8, 148.4, 149.0, 155.9, 161.4. ν_{max} (KBr)/cm^{-1} 2999, 2901, 2787, 1596, 1498, 1245, 1037, 821, 798. Found C 66.95, H 3.41, N 8.80. $C_{18}H_{11}ClN_2O_2$ requires C 66.99, H 3.44, N 8.68 %.

*3-Phenyl-8-methoxy-5*H*-indeno[1,2-c]pyridazine (**15r**)*

Compound **15r** was obtained as yellow needles, mp 198–200°C. δ_H (300 MHz, $CDCl_3$) 3.86 (s, 2H, CH_2), 3.91 (s, 3H, OCH_3), 7.04 (dd, J_1 6, J_2 2.1, 1H, Ar), 7.37–7.58 (m, 4H, Ar), 7.80 (s, 1H, Ar), 7.86 (s, 1H, Ar), 8.10 (d, *J* 6.9, 2H, Ar). δ_C (75 MHz, $CDCl_3$) 33.9, 55.6, 104.9, 118.4, 120.1, 126.0, 127.1, 128.9, 129.6, 135.0, 137.0, 139.0, 142.4, 156.5, 159.8, 161.6. ν_{max} (KBr)/cm^{-1} 3062, 2995, 2927, 2839, 1615, 1482, 1471, 1402, 1247, 1205, 1050, 791. Found C 78.79, H 5.12, N 10.32. $C_{18}H_{14}N_2O$ requires C 78.81, H 5.14, N 10.21 %.

*3-(4-Bromophenyl)-8-methoxy-5*H*-indeno[1,2-c] pyridazine (**15s**)*

Compound **15s** was obtained as white needles, mp 227–228°C. δ_H (300 MHz, $CDCl_3$) 3.88 (s, 2H, CH_2), 3.93 (s, 3H, OCH_3), 7.06 (d, *J* 8.1, 1H, Ar), 7.46 (d, *J* 8.4, 1H, Ar), 7.63 (d, *J* 8.7, 2H, Ar), 7.80 (s, 1H, Ar), 7.86 (s, 1H, Ar), 7.98 (d, *J* 8.4, 2H, Ar). δ_C (75 MHz, $CDCl_3$) 33.9, 55.6, 105.0, 118.6, 119.8, 124.2, 126.0, 128.5, 132.1, 135.0, 135.9, 138.8, 142.5, 155.4, 159.9, 161.8. ν_{max} (KBr)/cm^{-1} 2990, 2965, 2901, 2838, 1598, 1465, 1392, 1234, 1051, 829. Found C 61.23, H 3.70, N 8.07. $C_{18}H_{13}BrN_2O$ requires C 61.21, H 3.71, N 7.93 %.

*3-(4-Chlorophenyl)-8-methoxy-5*H*-indeno[1,2-c] pyridazine (**15t**)*

Compound **15t** was obtained as yellow needles, mp 211–213°C. δ_H (300 MHz, $CDCl_3$) 3.89 (s, 2H, CH_2), 3.93 (s, 3H, OCH_3), 7.07 (d, *J* 8.4, 1H, Ar), 7.47 (d, *J* 8.1, 3H, Ar), 7.81 (s, 1H, Ar), 7.86 (s, 1H, Ar), 8.04 (d, *J* 8.4, 2H, Ar). δ_C (75 MHz, $CDCl_3$) 33.6, 55.3, 105.4, 118.7, 119.3, 124.8, 126.3, 128.4, 132.0, 135.9, 136.3, 138.7, 142.0, 155.3, 159.1, 161.3. ν_{max} (KBr)/cm^{-1} 3090, 3035 1595, 1438, 1389, 1110, 824. Found C 70.00, H 4.26, N 9.20. $C_{18}H_{13}ClN_2O$ requires C 70.02, H 4.24, N 9.07 %.

*3-(4-Fluorophenyl)-8-methoxy-5*H*-indeno[1,2-c] pyridazine (**15u**)*

Compound **15u** was obtained as white needles, mp 205–207°C. δ_H (300 MHz, $CDCl_3$) 3.90 (s, 2H, CH_2), 3.93 (s, 3H, OCH_3), 7.07 (dd, J_1 6, J_2 2.4, 1H, Ar), 7.20 (t, *J* 8.7, 2H, Ar), 7.46 (d, *J* 8.4, 1H, Ar), 7.82 (s, 1H, Ar), 7.87 (s, 1H, Ar), 8.04–8.18 (m, 2H, Ar). δ_C (75 MHz, $CDCl_3$) 33.9, 55.6, 105.0, 115.7, 116.0, 118.4, 119.7, 126.0, 128.9, 129.0, 133.1, 135.0, 138.9, 142.5, 155.5, 159.9, 161.6, 162.2, 162.3, 165.5. ν_{max} (KBr)/cm^{-1} 3070, 3007, 2939, 2906, 2842, 1599, 1512, 1481, 1392, 1231, 1052, 846. Found C 73.99, H 4.50, N 9.60. $C_{18}H_{13}FN_2O$ requires C 73.96, H 4.48, N 9.58 %.

*3-(4-Methoxyphenyl)-8-methoxy-5*H*-indeno[1,2-c] pyridazine (**15v**)*

Compound **15v** was obtained as white needles, mp 178–179°C. δ_H (300 MHz, $CDCl_3$) 3.86 (s, 2H, CH_2), 3.86 (s, 3H, OCH_3), 3.92 (s, 3H, OCH_3), 6.97–7.13 (m, 3H, Ar), 7.43 (s, 2H, Ar), 7.82 (s, 2H, Ar), 8.07 (d, *J* 8.7, 2H, Ar). δ_C (75 MHz, $CDCl_3$) 33.9, 55.3, 55.6, 104.9, 114.3, 118.1, 119.4, 126.0, 128.3, 129.4, 134.9, 139.1, 142.3, 156.0, 159.8, 160.9, 161.1. ν_{max} (KBr)/cm^{-1} 3079, 2961, 2940, 2835, 1601, 1479, 1408, 1252, 1177, 1031, 818. Found C 74.95, H 5.28, N 9.33. $C_{19}H_{16}N_2O_2$ requires C 74.98, H 5.30, N 9.20 %.

*3-(3-Bromophenyl)-8-methoxy-5*H*-indeno[1,2-c] pyridazine (**15w**)*

Compound **15w** was obtained as yellow needles, mp 207–208°C. δ_H (300 MHz, $CDCl_3$) 3.90 (s, 2H, CH_2), 3.93 (s, 3H, OCH_3), 7.06 (d, *J* 8.4, 1H, Ar), 7.41 (t, *J* 7.8, 1H, Ar), 7.47 (d, *J* 8.4, 1H, Ar), 7.59 (d, *J* 7.8, 1H, Ar), 7.80 (s, 1H, Ar), 7.87 (s, 1H, Ar), 8.04 (d, *J* 7.5, 1H, Ar), 8.27 (s, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 33.9, 55.6, 105.0, 112.8, 115.0, 118.4, 119.0, 120.3, 126.1, 129.8, 135.3, 138.4, 139.9, 142.6, 156.5, 159.8, 160.4, 161.8. ν_{max} (KBr)/cm^{-1} 2998, 2906, 2838, 1602, 1567, 1480, 1394, 1233, 1054, 807. Found C 61.23, H 3.75, N 8.04. $C_{18}H_{13}BrN_2O$ requires C 61.21, H 3.71, N 7.93 %.

*3-(3-Mehoxyphenyl)-8-methoxy-5*H*-indeno[1,2-c] pyridazine (**15x**)*

Compound **15x** was obtained as white needles, mp 134–135°C. δ_H (300 MHz, $CDCl_3$) 3.92 (s, 2H, CH_2), 3.94 (s, 6H, $2OCH_3$), 6.99–7.12 (m, 2H, Ar), 7.38–7.52 (m, 2H, Ar), 7.63 (d, *J* 9.3, 2H, Ar), 7.83 (d, *J* 9.3, 2H, Ar), 7.92 (s, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 33.9, 55.4, 55.6, 105.0, 112.1, 115.9, 118.5, 119.3, 120.2, 126.0, 129.8, 135.1, 138.4, 139.0, 142.4, 156.3, 159.9, 160.1, 161.8. ν_{max} (KBr)/cm^{-1} 3005, 2925, 2835, 1583, 1440, 1379, 1190, 1023, 817. Found C 75.01, H 5.33, N 9.30. $C_{19}H_{16}N_2O_2$ requires C 74.98, H 5.30, N 9.20 %.

*3-(3,4-Dimethoxyphenyl)-8-methoxy-5*H*-indeno[1,2-c] pyridazine (**15y**)*

Compound **15y** was obtained as yellow needles, mp 207–209°C. δ_H (300 MHz, $CDCl_3$) 3.84 (s, 2H, CH_2), 3.91 (s, 3H, OCH_3), 3.92 (s, 3H, OCH_3), 4.00 (s, 3H, OCH_3), 6.92 (d, *J* 8.4, 1H, Ar), 7.02 (dd, J_1 6, J_2 2.4, 1H, Ar), 7.37–7.53 (m, 2H, Ar), 7.78 (s, 1H, Ar), 7.82 (s, 1H, Ar), 7.93 (s, 1H, Ar). δ_C (75 MHz, $CDCl_3$) 33.9, 55.6, 55.9, 56.0, 104.9, 110.1, 111.0, 118.1, 119.3, 119.4, 126.0, 129.7, 134.9, 139.1, 142.3, 149.4, 150.5, 155.8, 159.8, 161.2. ν_{max} (KBr)/cm^{-1} 3091, 3003, 2944, 2886, 2835, 1606, 1517, 1483, 1462, 1290, 1240, 1028, 863. Found C 71.83, H 5.41, N 8.40. $C_{20}H_{18}N_2O_3$ requires C 71.84, H 5.43, N 8.38 %.

*3-(3,4-Methylenedioxyphenyl)-8-methoxy-5*H*-indeno [1,2-c]pyridazine (**15z**)*

Compound **15z** was obtained as white needles, mp 210–211°C. δ_H (300 MHz, $CDCl_3$) 3.87 (s, 2H, CH_2), 3.93 (s, 3H, OCH_3), 6.03 (s, 2H, OCH_2O), 6.92 (d, *J* 8.4, 1H, Ar), 7.05 (dd, J_1 6, J_2 2.4, 1H, Ar), 7.44 (d, *J* 8.4, 1H, Ar), 7.54 (d, *J* 8.1, 1H, Ar), 7.69 (s, 1H, Ar), 7.80 (s, 2H, Ar). δ_C (75 MHz, $CDCl_3$) 33.9, 55.6, 101.4, 104.9, 107.5, 108.5, 118.3, 119.6, 121.1, 126.0, 131.3, 134.9, 139.0, 142.3, 148.4, 149.0, 156.0, 159.9, 161.3. ν_{max} (KBr)/cm^{-1} 3007, 2935, 2838, 2804, 1592, 1508, 1475, 1276, 1041, 812. Found C 71.71, H 4.46, N 8.94. $C_{19}H_{14}N_2O_3$ requires C 71.69, H 4.43, N 8.80 %.

Conclusions

In conclusion, we report a novel approach to a new series of 3-aryl substituted indeno[1,2-*c*]pyridazines which are potential MAO_A inhibitors, based on the DBN catalysed one-pot, three-component reaction of substituted indanones and arylglyoxalmonohydrates with hydrazine hydrate. The valuable features of this strategy are the formation of organic products in an aqueous medium, good yields, use of DBN as an environmentally friendly catalyst, high regioselectivity, and a convenient workup procedure. The MAO inhibitory activities of these new substituted indenopyridazines are currently under assessment in our research group.

Supplementary Material

Supplementary material (FT-IR, ^{1}H NMR, and ^{13}C NMR spectra of all new compounds) associated with this paper is available on the Journal's website.

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