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ORIGINAL PAPER

Enhanced rate of intramolecular nitrile oxide cycloaddition and rapid synthesis of isoxazoles and isoxazolines

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Abstract An enhanced rate of intramolecular nitrile oxide cycloaddition and hence a rapid synthesis of isoxazoles and isoxazolines is described. Formation of nitrile oxides from oximes using only 1 or 2 Eq (equivalents) of aqueous sodium hypochlorite solution is also described.

Keywords Carbonyl compounds · Heterocycles · Amines · 1,3-dipolar cycloaddition · Nitrile oxide

Introduction

Isoxazoles and isoxazolines are important molecules as they exhibit a wide range of biological activities. Recent reports have described isoxazoles as lysophosphatidic acid (LPA) antagonists [1], inhibitors of human rhinovirus 2 replication [2], and they also exhibit antitubulin [3] and insect antifeedant [4] activities. Isoxazolines have been described as antimicrobial [5, 6], protein tyrosine phosphatase 1B (PTPIB) inhibitors [7], and anti-inflammatory agents [8]. Some isoxazolines and isoxazoles have applications as dyes, electric insulating oils, and high temperature lubricants [9]. Synthetic manipulation of isoxazolines and isoxazoles provides easy access to β-hydroxy ketones, β-hydroxy acids, esters, β-hydroxy nitriles, α,β-unsaturated ketones, γ-amino alcohols, and so on [10]. So easy and rapid syntheses of these compounds are very much essential.

1,3-Dipolar cycloaddition reaction (inter- or intramolecular) of nitrile oxides and olefins/alkynes is one of the versatile methods for the preparation of synthetically useful isoxazolines/isoxazoles [11, 12]. There are several methods for the preparation of nitrile oxides [10, 11, 13–19]. One of those methods is the reaction of aldoximes with alkali hypochlorite. Existing reports describe that an excess, up to 40 Eq (equivalents), of aqueous sodium hypochlorite [20–22] is required to give nitrile oxides from oximes, and an organic base [21, 22] is generally used for the concomitant cycloaddition reaction with olefin/alkyne. It has been shown that benzonitrile oxides can be formed in water from benzohydroximoyl chloride [23] and these undergo cyclization in aqueous or organic biphasic medium in the presence of a catalytic amount of organic base, e.g., triethylamine [24]. A recent study showed the rate enhancement of cyclization reaction of nitrile oxides and electron-rich dipolarophiles in water and protic solvents [25]. No additional base was required when the cycloaddition was performed in biphasic medium. Water in biphasic medium enhances the rate, whereas the reaction in water is slow [26].

Results and discussion

Existing reports describe the isoxazoline- and isoxazole-forming reactions (inter- or intramolecular nitrile oxide cycloaddition) as slow [20, 27]. Herein, we describe the nitrile oxide formation and its concomitant cycloaddition as a fast reaction. A rapid synthesis of benzopyrano- and naphthopyrano-fused isoxazolines **2** and isoxazoles **5** is also reported in a one-pot reaction from oximes **1** and **4** (Scheme 1) in dichloromethane–water or chloroform–water biphasic medium at room temperature (rt) in good to excellent yields with 4% aqueous sodium hypochlorite solution within 10 min.

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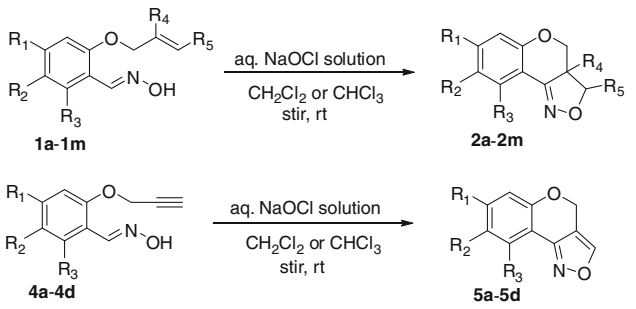


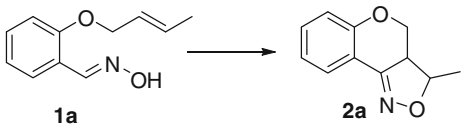
Scheme 1

For our study we prepared different *o*-allylated arylaldehyde oximes (**1a**–**1m**, Table 2). Firstly, an aqueous suspension of oxime **1a** was treated with 1 Eq of 4% aqueous sodium hypochlorite at room temperature and the desired isoxazoline **2a** was obtained. The reaction was monitored by TLC and the product was characterized from its spectral data. The characteristic peak of –OH stretching of oxime **1a** at 3,250 cm^{-1} was missing in the IR spectrum of **2a**. In the ^1H NMR spectrum, the peaks at $\delta = 7.79$ and 8.54 for –OH and –N=CH, respectively, disappeared and a new peak at $\delta = 4.57$ appeared for $-OCH_2$ of compound **2a**. The HRMS mass peak changed from 192.0945 of oxime **1a** to 190.0867 for the product **2a**.

The reaction was slow (Table 1, entries 1–3) in aqueous medium. Then the reaction was carried out with 1 Eq of 4% aqueous sodium hypochlorite solution in the presence of a base (Et_3N) in dichloromethane as solvent, which gave better yield in shorter reaction time (Table 1, entries 4–6). In all cases we isolated some starting oxime and corresponding deoximation compound (amount depending on the reaction time) along with the desired isoxazoline. When the same reaction was performed in the absence of base at room temperature, a dramatic rate enhancement occurred with excellent yield (Table 1, entry 7). For this case a very small amount of deoximation product (less than 5%) [19] and no starting oxime was isolated. We screened several other solvents for the reaction; it was observed that in tetrahydrofuran (THF) the reaction was slow (Table 1, entries 9, 10), and in acetonitrile and ethyl acetate rates were somewhat faster (Table 1, entries 11, 12). When chloroform was used as solvent the reaction rate and yield were comparable with that in dichloromethane (entry 8). TLC indicated the completion of the reaction within 10 min, when the reaction was performed in dichloromethane or chloroform.

With optimized reaction conditions in hand, we carried out the reaction with other *o*-allylated arylaldehyde oximes **1b**–**1m** in chloroform or dichloromethane with 1 Eq of 4% aqueous sodium hypochlorite (Table 2) for 10 min to provide isoxazolines **2b**–**2m** in 77–95% yields and for some cases we recovered small amounts of starting material (Table 2, entries 2, 5, 9, 10, 13). Then we performed the reactions with 2 Eq of sodium hypochlorite solution. This time all reactions were complete within 10 min with better yields (86–95%) and no starting materials were recovered.

Table 1 Reaction of oxime **1a** with 1 Eq of 4% aq. NaOCl solution

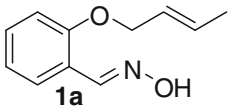
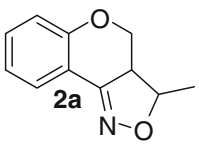
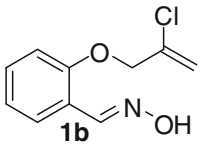
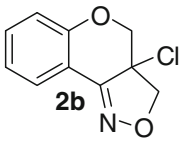
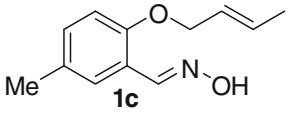
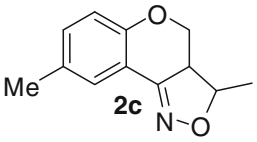
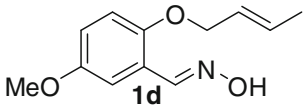
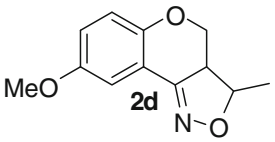
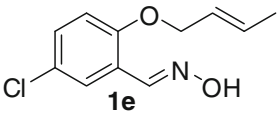
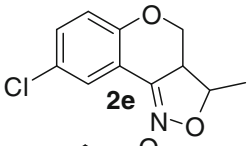
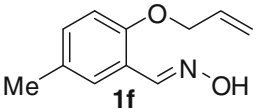
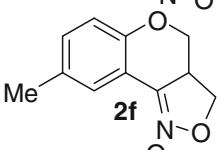
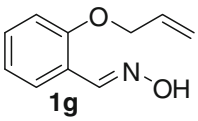
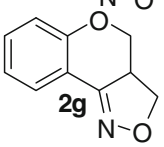


Entry	Medium	Base	Temperature (°C)	Time (h)	Yield (%)
1	H_2O	–	rt	0.17	15
2	H_2O	–	rt	3.0	30
3	H_2O	–	rt	14.0	45
4	CH_2Cl_2–H_2O (84:16)	Et_3N	0	0.17	50
5	CH_2Cl_2–H_2O (84:16)	Et_3N	0	1.0	60
6	CH_2Cl_2–H_2O (84:16)	Et_3N	rt	2.0	65
7	CH_2Cl_2–H_2O (84:16)	–	rt	0.17	92
8	$CHCl_3$–H_2O (84:16)	–	rt	0.17	90
9	THF–H_2O (65:35)	–	rt	22.0	70
10	THF–H_2O (84:16)	–	rt	45.0	72
11	CH_3CN–H_2O (84:16)	–	rt	2.0	50
12	EtOAc–H_2O (84:16)	–	rt	2.0	65

The rate enhancements for some *o*-propargyloxy arylaldehyde oximes (**4a**–**4d**, Table 3) were also similarly tested. Oxime **4a** was treated with only 1 Eq of 4% aqueous sodium hypochlorite at room temperature for 10 min (Scheme 1). The product (**5a**, Table 3) was obtained in good yield (79%) and characterized from its spectral data. The characteristic peak at 3,291 cm^{-1} for –OH stretching of oxime **4a** disappeared in the IR spectrum of **5a**. In the ^1H NMR spectrum the characteristic peaks at $\delta = 2.51$ and 7.83 ppm for propargylic hydrogen and N–O–H disappeared and a new peak at $\delta = 8.19$ appeared for O–CH= in the product **5a**. Similarly oximes **4b**–**4d** were treated and corresponding isoxazoles were obtained in 65–77% yield within 10 min. Even better yields (82–90%) were obtained by using 2 Eq of sodium hypochlorite solution.

Encouraged by these results, we also performed intermolecular 1,3-dipolar cycloaddition reactions to test whether the reaction might show a similar rate enhancement. Benzaldoxime **6** in dichloromethane was reacted with dienophiles **7a** and **7b** (Scheme 2) in the presence of 1 Eq aqueous sodium hypochlorite solution. With dienophile **7a**, the reaction was complete in 45 min while with dienophile **7b**, the reaction required more than 1 h for completion. However for both cases the reaction was

Table 2 Reaction of various oximes with aqueous NaOCl solution

Entry	Substrates	Products	Yields[a,b](%)
1			92 (92)
2			78 (86)
3			95 (95)
4			94 (94)
5			77 (88)
6			91 (93)
7			90 (93)

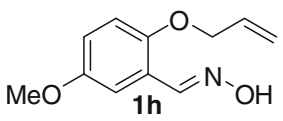
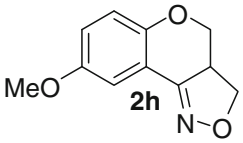
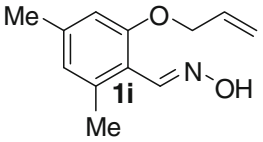
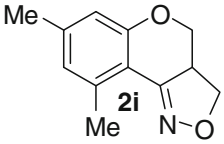
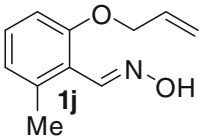
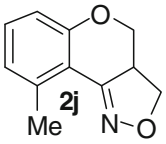
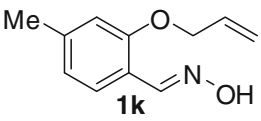
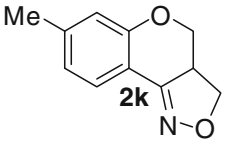
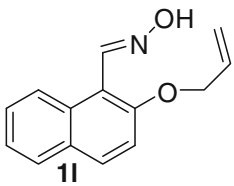
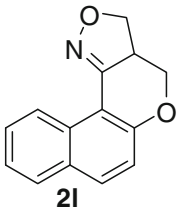
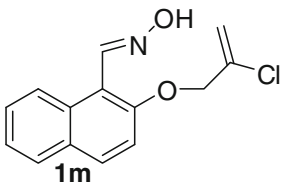
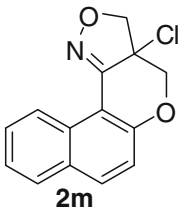
complete in 10 min, if 2 Eq of aqueous sodium hypochlorite solution was used and the yields were 82 and 85%, respectively (Table 4). More deoximation product was obtained when 1 Eq of aqueous sodium hypochlorite solution was used (Table 4).

We studied the rate enhancement in large-scale intramolecular nitrile oxide cycloaddition reactions also. Compound **1g** (5.0 g) in dichloromethane was treated with 2 Eq of 4% aqueous sodium hypochlorite solution and 1.5 Eq of triethylamine. The reaction was not complete even after 24 h. We performed the same reaction with 1 Eq of 4% aqueous sodium hypochlorite solution in the absence of base and the reaction remained incomplete even after 2 h. But when the reaction was repeated with 2 Eq of 4% aqueous sodium hypochlorite solution, it was complete within 20 min with satisfactory yield (88%).

From the above observations, it is apparent that in biphasic reaction conditions the absence of base speeds up the 1,3-dipolar cycloaddition reactions. This might be due to a difference in hypochlorous acid concentration in the reaction medium as shown schematically in Scheme 3. In the presence of a base, the concentration of hypochlorous acid decreases in the organic phase due to the formation of triethylamine hydrochloride in aqueous medium. Thus, the formation of hydroximoyl chloride and the corresponding nitrile oxide becomes slow, hence a longer time is necessary for the completion of the reaction and more deoximation product is obtained.

In conclusion, we have shown the rate enhancement of nitrile oxide cyclization and hence a rapid synthesis of isoxazolines and isoxazoles at room temperature in biphasic medium. Only 1–2 Eq of sodium hypochlorite solution is sufficient for the completion of the reaction and no additional base is required to facilitate the cycloaddition. Again dichloromethane–water or chloroform–water is the better solvent where the reaction is rapid.

Table 2 continued

Entry	Substrates	Products	Yields[a,b](%)
8	 **1h**	 **2h**	93 (94)
9	 **1i**	 **2i**	80 (89)
10	 **1j**	 **2j**	82 (87)
11	 **1k**	 **2k**	85 (91)
12	 **1l**	 **2l**	88 (88)
13	 **1m**	 **2m**	80 (86)

[a] Deoximation product (less than 5%) was isolated in each case

[b] Improved yields by using 2 Eq of NaOCl are given in parenthesis

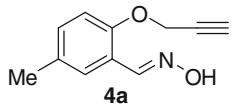
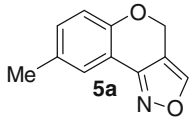
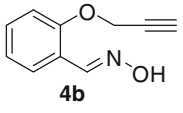
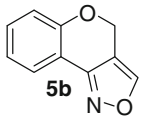
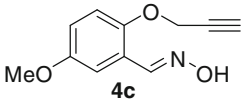
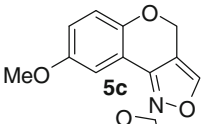
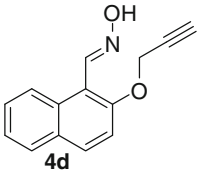
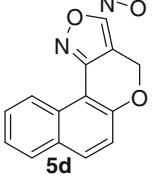
Experimental

Melting points were determined in open capillaries. IR spectra were recorded by using samples as neat liquid or solid samples on KBr disks on a PerkinElmer L 120-000A spectrophotometer. ^{1}H NMR spectra (300, 400, and 500 MHz) were recorded on Bruker DPX-300, Bruker DPX-400, Bruker DPX-500 spectrometer in $CDCl_3$ (chemical shifts in ppm) with TMS as internal standard. Elemental analyses were recorded on PerkinElmer 2400 series II CHN analyzer from the University of Kalyani, India, and results agreed with calculated values. Mass spectra were recorded on a Jeol JMS600 instrument from IICS, Kolkata. HRMS spectra were recorded on a Qtof Micro YA263 instrument. Silica gel (60–120, 230–400 mesh, Spectrochem, India) was used for chromatographic separation. Silica gel G (Spectrochem, India) was used for TLC. Petroleum (pet) ether refers to the fraction boiling at 60–80 °C.

*Representative procedure for preparation of aldoxime **1a***

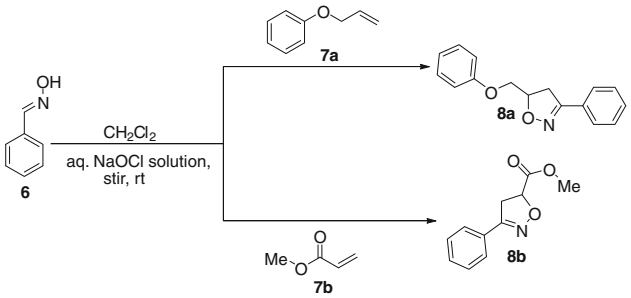
A mixture of 0.424 g (*E*)-2-(but-2-enyloxy)benzaldehyde (2.40 mmol), 0.284 g pyridine (3.6 mmol), 0.2 g hydroxylamine hydrochloride (2.88 mmol), and 5 cm^3 ethanol was refluxed for 3 h. The solvent was evaporated and the residue was acidified with 6 N HCl. It was extracted with $CHCl_3$ (3 × 20 cm^3), washed with brine, and dried over anhydrous sodium sulfate. After removal of solvent, the crude product was recrystallized from ethyl acetate–petroleum ether (3:1, unless stated otherwise) to give 0.418 g (91%) of the pure oxime **1a**. Other oximes **1b**–**1m** were prepared by following this procedure. For known

Table 3 Synthesis of various isoxazoles

Entry	Substrates	Products	Yields[a,b](%)
1	 **4a**	 **5a**	79 (88)
2	 **4b**	 **5b**	76 (90)
3	 **4c**	 **5c**	77 (90)
4	 **4d**	 **5d**	65 (82)

[a] Deoximation product (less than 5%) was isolated

[b] Improved yields by using 2 Eq of NaOCl are given in parenthesis

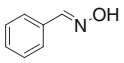
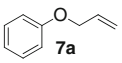
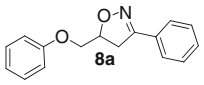
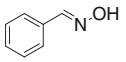
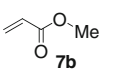
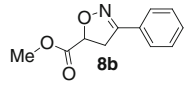


Scheme 2

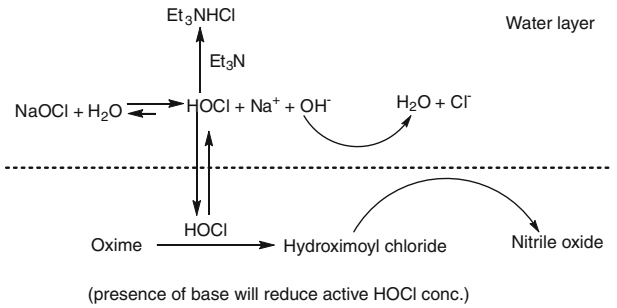
compounds **1f** [19], **1g** [28], **1h** [28], **4b** [21], **4d** [22], **2f** [19], **2g** [28], **2h** [28], **5b** [21], **5d** [22] data were in agreement with reported values given in references.

(E)-2-((E)-2-Butenyloxy)benzaldehyde oxime
(**1a**, $C_{11}H_{13}NO_2$)
Light yellow solid, yield 91%; m.p.: 88–89 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 1.75$ (d, 3H, $J = 6.1$ Hz, CH_3), 4.49 (d, 2H, $J = 5.5$ Hz, OCH_2), 5.67–5.74 (m, 1H, CH=CH), 5.80–5.87 (m, 1H, CH=CH), 6.89 (d, 1H, $J = 8.3$ Hz, ArH), 6.93 (t, 1H, $J = 7.5$ Hz, ArH), 7.32 (t, 1H, $J = 7.5$ Hz, ArH), 7.71 (d, 1H, $J = 7.2$ Hz, ArH), 7.79 (bs, 1H, OH), 8.54 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 17.72$, 69.02, 112.35, 120.71, 120.83, 125.68, 126.47, 130.27, 131.06, 146.54, 156.74 ppm; IR (KBr): $\bar{\nu} = 3{,}250$, 2,919, 1,618, 1,599 cm^{-1}; HRMS: calcd. for $C_{11}H_{13}NO_2$: 192.0946 [$M^+ + H$], found: 192.0945 [$M^+ + H$].

Table 4 Intermolecular nitrile oxide cycloaddition reaction

Entry	Substrate	Dipolarophiles	Products	Yields[a](%)
1	 **6**	 **7a**	 **8a**	82
2	 **6**	 **7b**	 **8b**	85

[a] 7–8% Deoximation product was also isolated



Scheme 3

(E)-2-(2-Chloro-2-propenyloxy)benzaldehyde oxime
(**1b**, $C_{10}H_{10}ClNO_2$)
White solid, yield 89%; m.p.: 57–58 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 4.63$ (s, 2H, OCH_2), 5.45 (s, 1H, $=CH_2$), 5.54 (s, 1H, $=CH_2$), 6.87 (d, 1H, $J = 8.4$ Hz, ArH), 7.00 (t, 1H, $J = 7.4$ Hz, ArH), 7.33 (t, 1H, $J = 7.5$ Hz, ArH), 7.73 (d, 1H, $J = 7.4$ Hz, ArH), 8.41(bs, 1H, OH), 8.55 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 70.41$, 112.73, 114.10, 121.08, 121.86, 127.11, 131.30, 135.89, 146.27, 155.83 ppm; IR (KBr): $\bar{\nu} = 3{,}256$, 2,926, 1,640, 1,600 cm^{-1}; MS: $m/z = 211$ (M^+).

(E)-2-((E)-2-Butenyloxy)-5-methylbenzaldehyde oxime
(**1c**, $C_{12}H_{15}NO_2$)
Yellow solid, yield 92%; m.p.: 68 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 1.74$ (d, 3H, $J = 6.4$ Hz, $CHCH_3$), 2.27 (s, 3H, CH_3), 4.46 (d, 2H, $J = 5.7$ Hz, OCH_2), 5.66–5.72 (m, 1H, CH=CH), 5.78–5.85 (m, 1H, CH=CH), 6.79 (d, 1H, $J = 8.4$ Hz, ArH), 7.10 (d, 1H, $J = 8.6$ Hz, ArH), 7.51 (s, 1H, ArH), 8.12 (bs, 1H, OH), 8.51 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 17.85$, 20.46, 69.37, 112.70, 120.56, 125.97, 127.01, 130.12, 130.27, 131.73, 146.74, 154.90 ppm; IR (KBr): $\bar{\nu} = 3{,}400$, 2,917, 1,638, 1,610 cm^{-1}; MS: $m/z = 205$ (M^+).

(E)-2-((E)-2-Butenyloxy)-5-methoxybenzaldehyde oxime
(**1d**, $C_{12}H_{15}NO_3$)
Yellow solid, yield 87%; m.p.: 62–63 °C; $R_f = 0.50$ (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 1.74$

(d, 3H, $J = 6.0$ Hz, $CHCH_3$), 3.77 (s, 3H, OCH_3), 4.43 (d, 2H, $J = 5.8$ Hz, OCH_2), 5.66–5.70 (m, 1H, CH=CH), 5.75–5.88 (m, 1H, CH=CH), 6.82–6.89 (m, 2H, ArH), 7.26 (d, 1H, $J = 8.9$ Hz, ArH), 8.11 (bs, 1H, OH), 8.51 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 17.84$, 55.74, 70.20, 110.17, 114.61, 117.68, 121.62, 126.02, 130.37, 146.58, 151.38, 153.67 ppm; IR (KBr): $\bar{\nu} = 3{,}396$, 2,917, 1,630, 1,582 cm^{-1}; MS: $m/z = 221$ (M^+).

(E)-2-((E)-2-Butenyloxy)-5-chlorobenzaldehyde oxime (**1e**, $C_{11}H_{12}ClNO_2$)
Yellow solid, yield 88%; m.p.: 83–84 °C; $R_f = 0.40$ (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 1.75$ (d, 3H, $J = 5.8$ Hz, CH_3), 4.46 (d, 2H, $J = 5.8$ Hz, OCH_2), 5.63–5.70 (m, 1H, CH=CH), 5.79–5.87 (m, 1H, CH=CH), 6.81 (d, 1H, $J = 8.9$ Hz, ArH), 7.23–7.26 (m, 1H, ArH), 7.62 (bs, 1H, OH), 7.70 (s, 1H, ArH), 8.45 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 17.85$, 69.56, 113.90, 122.33, 125.36, 126.00, 126.28, 130.67, 130.91, 145.63, 155.32 ppm; IR (KBr): $\bar{\nu} = 3{,}277$, 2,916, 1,680, 1,594 cm^{-1}; MS: $m/z = 225$ (M^+).

(E)-4,6-Dimethyl-2-(2-propenyloxy)benzaldehyde oxime (**1i**, $C_{12}H_{15}NO_2$)
Yellow solid, yield 84%; m.p.: 77–78 °C; $R_f = 0.50$ (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 2.29$ (s, 3H, CH_3), 2.43 (s, 3H, CH_3), 4.54 (d, 2H, $J = 4.9$ Hz, OCH_2), 5.26 (d, 1H, $J = 10.2$ Hz, $=CH_2$), 5.38 (d, 1H, $J = 16.7$ Hz, $=CH_2$), 5.99–6.08 (m, 1H, OCH_2CH), 6.56 (s, 1H, ArH), 6.65 (s, 1H, ArH), 8.21 (bs, 1H, OH), 8.56 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 21.71$, 21.93, 69.33, 110.78, 116.79, 117.50, 124.60, 133.20, 138.75, 140.32, 147.41, 157.55 ppm; IR (KBr): $\bar{\nu} = 3{,}300$, 2,919, 1,649, 1,609, cm^{-1}; MS: $m/z = 205$ (M^+).

(E)-6-Methyl-2-(2-propenyloxy)benzaldehyde oxime (**1j**, $C_{11}H_{13}NO_2$)
Yellow gummy liquid, yield 93%; $R_f = 0.40$ (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 2.46$ (s, 3H, CH_3), 4.56 (d, 2H, $J = 8.8$ Hz, OCH_2), 5.27 (d, 1H, $J = 10.8$ Hz, $=CH_2$), 5.39 (d, 1H, $J = 17.6$ Hz, $=CH_2$), 5.99–6.07 (m, 1H, OCH_2CH), 6.74 (d, 1H, $J = 8.2$ Hz, ArH), 6.82 (d, 1H, $J = 7.6$ Hz, ArH), 7.18 (t, 1H, $J = 7.9$ Hz, ArH), 8.11 (bs, 1H, OH), 8.59 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 21.52$, 69.38, 114.54, 115.09, 117.29, 124.01, 132.90, 133.23, 138.64, 147.41, 155.70 ppm; IR (KBr): $\bar{\nu} = 3{,}315$, 2,924, 1,596, 1,577 cm^{-1}; MS: $m/z = 191$ (M^+).

(E)-4-Methyl-2-(2-propenyloxy)benzaldehyde oxime (**1k**, $C_{11}H_{13}NO_2$)
Yellow solid, yield 85%; m.p.: 68–70 °C; $R_f = 0.40$ (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 2.32$ (s, 3H, CH_3), 4.56 (d, 2H, $J = 4.8$ Hz, OCH_2), 5.28 (d, 1H, $J = 10.4$ Hz, $=CH_2$), 5.40 (d, 1H, $J = 17.0$ Hz, $=CH_2$), 6.04 (m, 1H, OCH_2CH), 6.70 (s, 1H, ArH), 6.76 (d, 1H, $J = 7.8$ Hz, ArH), 7.59 (d, 1H, $J = 7.8$ Hz, ArH), 8.33 (bs, 1H, OH), 8.51 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 21.48$, 69.44, 112.20, 115.91, 117.08, 122.06, 129.92, 133.10, 143.82, 147.54, 156.57 ppm; IR (KBr): $\bar{\nu} = 3{,}279$, 2,920, 2,850, 1,608 cm^{-1}; MS: $m/z = 191$ (M^+).

(E)-2-(2-Propenyloxy)-1-naphthalenecarbaldehyde oxime (**1l**, $C_{14}H_{13}NO_2$)
Recrystallized from ethyl acetate. White solid, yield 89%; m.p.: 98–99 °C; $R_f = 0.50$ (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 4.71$ (d, 2H, $J = 5.3$ Hz, OCH_2), 5.30 (d, 1H, $J = 10.6$ Hz, $=CH_2$), 5.43 (d, 1H, $J = 17.8$ Hz, $=CH_2$), 6.03–6.13 (m, 1H, OCH_2CH), 7.23 (d, 1H, $J = 9.2$ Hz, ArH), 7.37 (t, 1H, $J = 7.4$ Hz, ArH), 7.52 (t, 1H, $J = 7.4$ Hz, ArH), 7.75 (d, 1H, $J = 5.6$ Hz, ArH), 7.83 (d, 1H, $J = 9.1$ Hz, ArH), 8.32 (bs, 1H, OH), 8.82 (d, 1H, $J = 8.7$ Hz, ArH), 8.91 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 70.28$, 114.06, 114.19, 117.97, 124.22, 125.42, 127.89, 128.38, 129.24, 131.72, 131.94, 133.03, 147.63, 155.97 ppm; IR (KBr): $\bar{\nu} = 3{,}243$, 2,921, 1,644, 1,618 cm^{-1}; MS: $m/z = 227$ (M^+).

(E)-2-(2-Chloro-2-propenyloxy)-1-naphthalenecarbaldehyde oxime (**1m**, $C_{14}H_{12}ClNO_2$)
White solid, yield 88%; m.p.: 57–58 °C; $R_f = 0.60$ (20% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 4.75$ (s, 2H, OCH_2), 5.46 (s, 1H, $=CH_2$), 5.57 (s, 1H, $=CH_2$), 7.19 (d, 1H, $J = 9.2$ Hz, ArH), 7.40 (t, 1H, $J = 7.6$ Hz, ArH), 7.54 (d, 1H, $J = 7.7$ Hz, ArH), 7.79 (d, 1H, $J = 8.1$ Hz, ArH), 7.84 (d, 1H, $J = 9.1$ Hz, ArH), 8.11 (bs, 1H, OH), 8.77 (d, 1H, $J = 8.6$ Hz, ArH), 8.91 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 71.49$, 114.05, 114.37, 114.64, 124.60, 125.71, 128.03, 128.38, 129.62, 131.62, 132.10, 136.10, 147.54, 155.09 ppm; IR (KBr): $\bar{\nu} = 3{,}214$, 2,943, 1,638, 1,617 cm^{-1}; MS: $m/z = 261$ (M^+).

*Representative procedure for preparation of aldoximes **4a–4d***

Aldoximes **4a–4d** were prepared following the procedure for preparation of **1a**. The crude compound was purified by column chromatography using 15% ethyl acetate–petroleum ether as eluent and recrystallized from chloroform–petroleum ether 4:1 to give the pure oximes **4a–4d**.

(E)-5-Methyl-2-(2-propynyloxy)benzaldehyde oxime (**4a**, $C_{11}H_{11}NO_2$)
White solid, yield 86%; m.p.: 107–108 °C; $R_f = 0.50$ (15% EtOAc/pet ether); ^{1}H NMR (300 MHz, $CDCl_3$): $\delta = 2.3$ (s, 3H, $-CH_3$), 2.51 (s, 1H, alkyne proton), 4.71 (s, 2H, $-OCH_2$), 6.94 (d, 1H, $J = 8.4$ Hz, ArH), 7.16 (d, 1H, $J = 8.4$ Hz, ArH), 7.54 (s, 1H, ArH), 7.83 (bs, 1H, OH), 8.49 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$):

δ = 20.48, 56.53, 75.83, 78.35, 113.05, 121.10, 127.28, 131.25, 131.63, 146.56, 153.62 ppm, IR (KBr): $\bar{\nu}$ = 3,291, 2,853, 2,122, 1,495 cm^{-1}; MS: m/z = 189 (M^+).

(E)-5-Methoxy-2-(2-propynyloxy)benzaldehyde oxime (**4c**, $C_{11}H_{11}NO_3$)

Yellow solid, yield 90%; m.p.: 110–111 °C; R_f = 0.50 (15% EtOAc/pet ether); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.51 (s, 1H, alkyne proton), 3.78 (s, 3H, –OCH_3), 4.68 (s, 2H, OCH_2), 6.89–6.93 (m, 2H, ArH), 6.99 (d, 1H, J = 9 Hz, ArH), 7.89 (bs, 1H, OH), 8.49 (s, 1H, N=CH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): δ = 55.74, 57.41, 75.90, 78.44, 110.55, 115.20, 117.48, 122.36, 146.36, 150.08, 154.41 ppm; IR (KBr): $\bar{\nu}$ = 3,288, 2,837, 2,121, 1,496 cm^{-1}; MS: m/z = 205 (M^+).

*Representative procedure for the preparation of **2a** from **1a***

To a solution of 0.191 g **1a** (1.0 mmol) in 5 cm^3 dichloromethane at room temperature was added dropwise 3.6 cm^3 4% aqueous sodium hypochlorite solution (2 Eq) within 2 min. After completion of the addition the reaction mixture was stirred for 7 min. Water (10 cm^3) was added to the reaction mixture and it was extracted with dichloromethane (3 × 15 cm^3). The organic layer was dried over anhydrous sodium sulfate, filtered, and concentrated, and the residue was subjected to flash column chromatography using an eluent of petroleum ether–ethyl acetate (4:1) to afford 0.174 g (92%) **2a**. The product was recrystallized from ethanol (unless stated otherwise). Compounds **2b**–**2m** were prepared similarly.

3a,4-Dihydro-3-methyl-3H-[1]benzopyrano[4,3-c]isoxazole (**2a**, $C_{11}H_{11}NO_2$)

White solid, yield 92%; m.p.: 99 °C; R_f = 0.50 (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.59 (d, 3H, J = 6.0 Hz, CH_3), 3.48 (dt, 1H, J = 5.8, 12.0 Hz, ring juncture H), 4.07 (dd, 1H, J = 10.7, 12.0 Hz, OCH_2), 4.39 (qd, 1H, J = 6.0, 12.2 Hz, $CHCH_3$), 4.57 (dd, 1H, J = 5.8, 10.4 Hz, OCH_2), 6.92 (d, 1H, J = 8.3 Hz, ArH), 6.98 (t, 1H, J = 7.4 Hz, ArH), 7.31 (t, 1H, J = 7.2 Hz, ArH), 7.76 (d, 1H, J = 7.1 Hz, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): δ = 18.22, 51.50, 68.83, 80.10, 113.30, 117.29, 121.73, 125.30, 132.28, 153.75, 155.39 ppm; IR (KBr): $\bar{\nu}$ = 2,895, 2,883, 1,600, 1,611 cm^{-1}; HRMS: calcd. for $C_{11}H_{11}NO_2$: 190.0868 [M^+ + H]; found: 190.0867 [M^+ + H].

3a-Chloro-3a,4-dihydro-3H-[1]benzopyrano[4,3-c]isoxazole (**2b**, $C_{10}H_8ClNO_2$)

White solid, yield 86%; m.p.: 122 °C; R_f = 0.60 (15% EtOAc/pet ether); ^{1}H NMR (500 MHz, $CDCl_3$): δ = 4.32 (d, 1H, J = 11.0 Hz, OCH_2), 4.39 (d, 1H, J = 12.0 Hz, OCH_2), 4.78 (d, 1H, J = 12.0 Hz, OCH_2), 4.85 (d, 1H, J = 11.0 Hz, OCH_2), 7.03–7.05 (m, 1H, ArH), 7.08 (dd, 1H, J = 1.0, 7.8 Hz, ArH), 7.38–7.42 (m, 1H, ArH), 7.84 (dd, 1H, J = 1.5, 7.8 Hz, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): δ = 71.26, 72.93, 78.77, 110.77, 117.69, 122.67, 126.24, 133.17, 152.05, 154.19 ppm; IR (KBr): $\bar{\nu}$ = 2,936, 2,916, 1,610, 1,593 cm^{-1}; MS: m/z = 209 (M^+).

3a,4-Dihydro-3,8-dimethyl-3H-[1]benzopyrano[4,3-c]isoxazole (**2c**, $C_{12}H_{13}NO_2$)

Recrystallized from petroleum ether. Yellow solid, yield 95%; m.p.: 44 °C; R_f = 0.60 (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.58 (d, 3H, J = 5.5 Hz, $CHCH_3$), 2.28 (s, 3H, CH_3), 3.45 (dt, 1H, J = 5.0, 12.4 Hz, ring juncture H), 4.03 (dd ≈ t, J = 11.3 Hz, OCH_2), 4.37 (qd, 1H, J = 6.1, 12.1 Hz, $CHCH_3$), 4.50–4.56 (m, 1H, OCH_2), 6.82 (d, 1H, J = 8.4 Hz, ArH), 7.11 (d, 1H, J = 7.8 Hz, ArH), 7.57 (s, 1H, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): δ = 18.27, 20.47, 51.69, 68.93, 80.14, 112.97, 117.10, 125.17, 131.23, 133.34, 153.49, 154.06 ppm; IR (KBr): $\bar{\nu}$ = 2,973, 2,929, 1,621, 1,603 cm^{-1}; MS: m/z = 203 (M^+).

3a,4-Dihydro-8-methoxy-3-methyl-3H-[1]benzopyrano[4,3-c]isoxazole (**2d**, $C_{12}H_{13}NO_3$)

Recrystallized from petroleum ether. Yellow solid, yield 94%; m.p.: 59–60 °C; R_f = 0.50 (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.59 (d, 3H, J = 5.9 Hz, $CHCH_3$), 3.46 (dt, 1H, J = 5.8, 12.4 Hz, ring juncture H), 3.77 (s, 3H, OCH_3), 4.02 (dd ≈ t, 1H, J = 11.4 Hz, OCH_2), 4.39 (qd, 1H, J = 6.1, 12.3 Hz, $CHCH_3$), 4.51–4.55 (m, 1H, OCH_2), 6.85 (d, 1H, J = 9.0 Hz, ArH), 6.9 (d, 1H, J = 8.4 Hz, ArH), 7.23 (s, 1H, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): δ = 18.25, 51.71, 55.79, 68.98, 80.39, 106.96, 107.13, 118.59, 120.92, 146.47, 149.95, 154.32 ppm; IR (KBr): $\bar{\nu}$ = 2,976, 2,905, 1,600, 1,581 cm^{-1}; MS: m/z = 219 (M^+).

8-Chloro-3a,4-dihydro-3-methyl-3H-[1]benzopyrano-[4,3-c]isoxazole (**2e**, $C_{11}H_{10}ClNO_2$)

Light yellow solid, yield 88%; m.p.: 91–92 °C; R_f = 0.40 (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.59 (d, 3H, J = 5.7 Hz, $CHCH_3$), 3.45 (dt, 1H, J = 5.7, 12.4 Hz, ring juncture H), 4.04 (dd ≈ t, 1H, J = 11.3 Hz, OCH_2), 4.41 (qd, 1H, J = 6.1, 12.2 Hz, $CHCH_3$), 4.59 (dd, 1H, J = 6.0, 10.1 Hz, OCH_2), 6.85–6.91 (m, 1H, ArH), 7.21–7.26 (m, 1H, ArH), 7.74 (s, 1H, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): δ = 18.31, 51.14, 69.04, 80.59, 114.58, 118.91, 124.71, 126.94, 132.23, 152.95, 153.94 ppm; IR (KBr): $\bar{\nu}$ = 2,928, 1,605, 1,480 cm^{-1}; MS: m/z = 223 (M^+).

3a,4-Dihydro-7,9-dimethyl-3H-[1]benzopyrano[4,3-c]isoxazole (**2i**, $C_{12}H_{13}NO_2$)

Yellow solid, yield 89%; m.p.: 119–120 °C; R_f = 0.50 (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$):

$\delta = 2.28$ (s, 3H, CH_3), 2.53 (s, 3H, CH_3), 3.82 (dd, 1H, $J = 7.6$, 13.0 Hz, OCH_2), 3.87–3.95 (m, 1H, ring juncture H), 4.03 (dd $\approx$ t, 1H, $J = 11.0$ Hz, OCH_2), 4.59–4.63 (m, 2H, OCH_2), 6.61 (s, 1H, ArH), 6.68 (s, 1H, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 21.46$, 22.96, 47.09, 68.63, 69.41, 109.73, 115.07, 125.10, 139.25, 142.18, 153.57, 156.19 ppm; IR (KBr): $\bar{\nu} = 2{,}921$, 2,861, 1,613, 1,586 cm^{-1}; MS: $m/z = 203$ (M^+).

3a,4-Dihydro-9-methyl-3H-[1]benzopyrano[4,3-c]isoxazole (**2j**, $C_{11}H_{11}NO_2$)
White solid, yield 87%; m.p.: 122 °C; $R_f = 0.40$ (15% EtOAc/pet ether); ^{1}H NMR (500 MHz, $CDCl_3$): $\delta = 2.58$ (s, 3H, CH_3), 3.86 (dd, 1H, $J = 7.7$, 12.8 Hz, OCH_2), 3.90–3.98 (m, 1H, ring juncture H), 4.07 (dd, 1H, $J = 10.2$, 12.0 Hz, OCH_2), 4.63–4.66 (m, 2H, OCH_2), 6.80 (d, 1H, $J = 8.3$ Hz, ArH), 6.86 (d, 1H, $J = 7.5$ Hz, ArH), 7.20 (t, 1H, $J = 7.9$ Hz, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 23.08$, 46.89, 68.64, 69.54, 112.47, 114.76, 123.86, 131.40, 139.54, 153.51, 156.21 ppm; IR (KBr): $\bar{\nu} = 2{,}921$, 2,850, 1,600 cm^{-1}; MS: $m/z = 189$ (M^+).

3a,4-Dihydro-7-methyl-3H-[1]benzopyrano[4,3-c]isoxazole (**2k**, $C_{11}H_{11}NO_2$)
White solid, yield 91%; m.p.: 144 °C; $R_f = 0.40$ (15% EtOAc/pet ether); ^{1}H NMR (500 MHz, $CDCl_3$): $\delta = 2.33$ (s, 3H, CH_3), 3.87–3.95 (m, 2H, OCH_2), 4.06–4.08 (m, 1H, ring juncture H), 4.65–4.69 (m, 2H, OCH_2), 6.76 (s, 1H, ArH), 6.82 (d, 1H, $J = 8.0$ Hz, ArH), 7.67 (d, 1H, $J = 8.0$ Hz, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 21.74$, 46.07, 69.23, 70.43, 110.22, 117.58, 123.10, 125.46, 143.42, 152.85, 155.56 ppm; IR (KBr): $\bar{\nu} = 2{,}924$, 2,870, 1,619, 1,605 cm^{-1}; MS: $m/z = 189$ (M^+).

3a,4-Dihydro-3H-naphtho[1′,2′:5,6]pyrano[4,3-c]isoxazole (**2l**, $C_{14}H_{11}NO_2$)
Recrystallized from petroleum ether. White solid, yield 88%; m.p.: 64 °C; $R_f = 0.50$ (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 3.93$ (dd, 1H, $J = 8.0$, 12.7 Hz, OCH_2), 4.04–4.14 (m, 1H, ring juncture H), 4.23 (dd, 1H, $J = 10.4$, 12.1 Hz, OCH_2), 4.70 (dd, 1H, $J = 8.0$, 9.6 Hz, OCH_2), 4.76 (dd, 1H, $J = 5.4$, 10.1 Hz, OCH_2), 7.10 (d, 1H, $J = 8.9$ Hz, ArH), 7.42 (t, 1H, $J = 7.5$ Hz, ArH), 7.59 (t, 1H, $J = 7.7$ Hz, ArH), 7.75–7.80 (m, 2H, ArH), 9.02 (d, 1H, $J = 8.6$ Hz, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 46.89$, 69.19, 69.48, 106.09, 118.22, 124.72, 126.54, 128.37, 128.49, 129.26, 130.48, 133.41, 153.20, 155.65 ppm; IR (KBr): $\bar{\nu} = 2{,}938$, 2,887, 1,622, 1,597 cm^{-1}; MS: $m/z = 225$ (M^+).

3a-Chloro-3a,4-dihydro-3H-naphtho[1′,2′:5,6]pyrano[4,3-c]isoxazole (**2m**, $C_{14}H_{10}ClNO_2$)
White solid, yield 86%; m.p.: 151 °C; $R_f = 0.60$ (20% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 4.35$ (d, 1H, $J = 10.9$ Hz, OCH_2), 4.54 (d, 1H, $J = 11.9$ Hz, OCH_2), 4.87 (d, 1H, $J = 10.8$ Hz, OCH_2), 4.88 (d, 1H, $J = 11.8$ Hz, OCH_2), 7.17 (d, 1H, $J = 8.9$ Hz, ArH), 7.46 (t, 1H, $J = 7.6$ Hz, ArH), 7.61–7.65 (m, 1H, ArH), 7.79 (d, 1H, $J = 8.1$ Hz, ArH), 7.86 (d, 1H, $J = 9.0$ Hz, ArH), 8.98 (d, 1H, $J = 8.5$ Hz, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 72.38$, 72.92, 77.85, 104.12, 118.10, 125.14, 126.57, 128.60, 128.80, 129.64, 130.64, 134.21, 152.72, 154.51 ppm; IR (KBr): $\bar{\nu} = 2{,}926$, 1,618, 1,599 cm^{-1}; MS: $m/z = 259$ (M^+).

*Representative procedure for the preparation of **5a–5d** from **4a–4d***

Compounds **5a–5d** were prepared following the procedure for the preparation of **2a** described above. The products were purified by column chromatography using 10% ethyl acetate–petroleum ether as eluent and were recrystallized from petroleum ether.

8-Methyl-4H-[1]benzopyrano[4,3-c]isoxazole (**5a**, $C_{11}H_9NO_2$)
White solid, yield 88%; m.p.: 103–104 °C; $R_f = 0.60$ (10% EtOAc/pet ether); ^{1}H NMR (300 MHz, $CDCl_3$): $\delta = 2.34$ (s, 3H, $-CH_3$), 5.20 (s, 2H, $-OCH_2$), 6.92 (d, 1H, $J = 8.4$ Hz, ArH), 7.16 (d, 1H, $J = 8.4$ Hz, ArH), 7.68 (s, 1H, ArH), 8.19 (s, 1H, O–CH=) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 20.60$, 61.28, 111.39, 113.65, 117.59, 124.67, 131.92, 132.90, 150.54, 152.76, 153.89 ppm; IR (KBr): $\bar{\nu} = 2{,}852$, 1,621, 1,489 cm^{-1}; MS: $m/z = 187$ (M^+).

8-Methoxy-4H-[1]benzopyrano[4,3-c]isoxazole (**5c**, $C_{11}H_9NO_3$)
Light yellow solid, yield 90%; m.p.: 57–58 °C; $R_f = 0.60$ (10% EtOAc/pet ether); ^{1}H NMR (300 MHz, $CDCl_3$): $\delta = 3.82$ (s, 3H, $-OCH_3$), 5.18 (s, 2H, $-OCH_2$), 6.93–6.98 (m, 2H, ArH), 7.36 (s, 1H, ArH), 8.20 (s, 1H, O–CH=) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 55.85$, 61.27, 107.41, 111.56, 114.24, 118.96, 119.61, 148.98, 150.62, 154.12, 154.77 ppm; IR (KBr): $\bar{\nu} = 2{,}917$, 1,620, 1,490 cm^{-1}; MS: $m/z = 203$(M^+).

*Representative procedure for the preparation of **8a** and **8b***

A 4% aqueous sodium hypochlorite solution (3.6 cm^3, 2.0 mmol) was added dropwise to a solution of 0.134 g allyloxybenzene **7a** (1.0 mmol) and 0.121 g benzaldehyde oxime **6** (1.0 mmol) in 10 cm^3 dichloromethane at room temperature within 2 min. After completion of the addition, the reaction mixture was stirred for 8 min. Water (10 cm^3) was added to the reaction mixture and it was extracted with dichloromethane (3 × 15 cm^3). The organic layer was dried over anhydrous sodium sulfate, filtered, and concentrated, and the residue was subjected to flash column

chromatography using an eluent of petroleum ether–ethyl acetate (5:1) to afford 0.207 g (82%) **8a**. The product was recrystallized from petroleum ether. Compound **8b** was prepared similarly.

5-(Phenoxymethyl)-3-phenyl-4,5-dihydroisoxazole (**8a**, $C_{16}H_{15}NO_2$)

White solid, yield 82%; m.p.: 79–80 °C; $R_f = 0.50$ (15% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 3.40$ (dd, 1H, $J = 6.8$, 16.8 Hz), 3.53 (dd, 1H, $J = 10.4$, 16.8 Hz), 4.05 (dd, 1H, $J = 6.0$, 9.6 Hz), 4.18 (dd, 1H, $J = 4.8$, 9.6 Hz), 5.09–5.16 (m, 1H), 6.91 (d, 2H, $J = 8.0$ Hz, ArH), 6.97 (t, 1H, $J = 7.6$ Hz, ArH), 7.28 (t, 2H, $J = 8.0$ Hz, ArH), 7.42–7.45 (m, 3H, ArH), 7.69–7.72 (m, 2H, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 37.69$, 68.42, 78.75, 114.61, 121.34, 126.79, 128.76, 129.35, 129.55, 130.23, 156.47, 158.42 ppm; IR (KBr): $\bar{\nu} = 2{,}918$, 1,599, 1,497 cm^{-1}; MS: $m/z = 253(M^+)$.

Methyl 3-phenyl-4,5-dihydroisoxazole-5-carboxylate (**8b**, $C_{11}H_{11}NO_3$)

White solid, yield 85%; m.p.: 61–62 °C; $R_f = 0.40$ (20% EtOAc/pet ether); ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 3.64$–3.72 (m, 2H), 3.82 (s, 3H, OCH_3), 5.20 (dd, 1H, $J = 7.6$, 10.4 Hz), 7.39–7.45 (m, 3H, ArH), 7.67–7.69 (m, 2H, ArH) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): $\delta = 38.92, 52.86, 77.95$, 126.95, 128.48, 128.80, 130.56, 156.09, 170.74 ppm; IR (KBr): $\bar{\nu} = 2{,}952$, 1,752, 1,603, 1,441 cm^{-1}; MS: $m/z = 205$ (M^+).

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