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Study of the Conformational Equilibria of Some 2-(2′-Hydroxyphenyl)-4-aryl-3*H*-1,5-benzodiazepines using ^{1}H, ^{13}C, and ^{15}N NMR Spectroscopy

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Summary. Variable temperature ^{1}H NMR experiments of 2-(2′-hydroxyphenyl)-4-phenyl-3*H*-1,5-benzodiazepine (**5a**) and its derivatives **5d** and **5e** were carried out in order to investigate the conformational behaviour of these compounds. The ΔG^* values for the ring inversion barriers of **5a** and **5d** are *ca.* 52 kJ/mol, *i.e.* they do not differ significantly as compared to analogous compounds without phenolic OH group(s). This indicates that the hydrogen bond has not to be opened during the inversion process. In **5e** the barrier is about 2–3 kJ/mol higher which can be explained by steric interference between the additional methoxy group and the H-3 atoms during ring inversion. ^{15}N NMR data which can be discussed in terms of hydrogen bond strength support this interpretation.

Keywords. 3*H*-1,5-Benzodiazepines; Variable temperature ^{1}H NMR; ^{13}C NMR; ^{15}N NMR; Ring inversion; Intramolecular hydrogen bond.

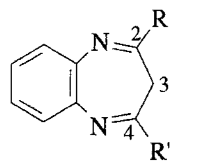
Untersuchung des Konformationsgleichgewichtes einiger 2-(2′-Hydroxyphenyl)-4-aryl-3*H*-1,5-benzodiazepine mit Hilfe der ^{1}H- und ^{15}N-NMR-Spektroskopie

Zusammenfassung. Es wurden ^{1}H-NMR-Experimente mit 2-(2′-Hydroxyphenyl)-4-aryl-3*H*-1,5-benzodiazepin (**5a**) und seinen Derivaten **5d** und **5e** bei unterschiedlichen Temperaturen durchgeführt. Die ΔG^*-Werte für die Ring-Inversion von **5a** und **5d** betragen *ca.* 52 kJ/mol, d.h. sie sind gegenüber Verbindungen ohne eine phenolische OH-Gruppe kaum verändert. Das zeigt an, daß die Wasserstoffbrückenbindung während der Inversion nicht geöffnet werden muß. In **5e** ist die Barriere um ungefähr 2–3 kJ/mol höher, was durch eine sterische Wechselwirkung zwischen der zusätzlichen Methoxygruppe und den H-3-Atomen während der Inversion erklärt werden kann. ^{15}N-NMR-Daten können als Hinweise auf die Stärke der Wasserstoffbrückenbindung interpretiert werden.

Introduction

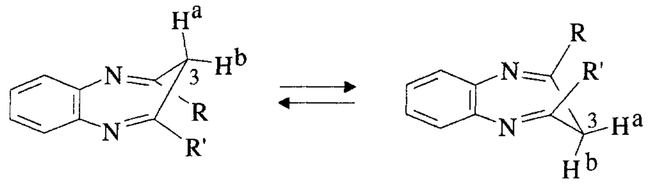
The structures of 1,5-benzodiazepines and the corresponding mono- and dications have been assigned on the basis of spectroscopic data. NMR spectra were particularly informative [1–3]. Some 2,4-disubstitued-3*H*-1,5-benzodiazepines (**1–3**,

(a)



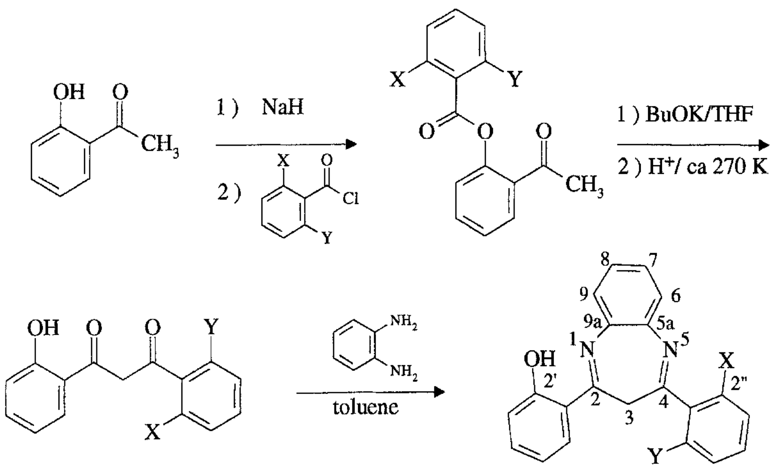
	R	R'
1	CH_3	CH_3
2	CH_3	Ph
3	Ph	Ph

(b)



Scheme 1. (a) Structures of 3*H*-1,5-benzodiazepines **1–3**; (b) conformational interconversion of **1–3**

(a)



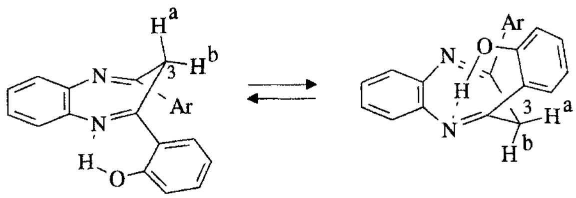
4a - 4c **5a - 5c**

5b $\xrightarrow{BBr_3 / S(CH_3)_2}$ **5d**

5c $\xrightarrow{BBr_3 / S(CH_3)_2}$ **5e**

	a	b	c	d	e
X	H	OCH_3	OCH_3	OH	OH
Y	H	H	OCH_3	H	OCH_3

(b)



Scheme 2. (a) Synthesis of 3*H*-1,5-benzodiazepines **5a–e**; (b) conformational interconversion of **5a–e**

Scheme 1a) have shown distinct singlet peaks for the methylene protons. An early report describes the conformational equilibria of azepines and diazepines detected by ^{1}H NMR spectroscopy at different temperatures [4]. Generally, the protons at C-3 appeared as singlets at ambient operating temperatures; at low temperatures AB systems have been observed. This result demonstrated the non-equivalence of the hydrogens at C-3 (H^{a}-H^{b}) due to the fact that the 3*H*-1,5-benzodiazepine ring is non-planar (boat-type conformation) and that there is an interconversion between two structurally equivalent conformations **A** and **B** of **1–3** (Scheme 1b) [4]. The energy barriers have been determined as 48.9 ± 1.7 kJ/mol for **1**, 51.0 ± 1.7 kJ/mol for **2**, and 52.7 ± 2.1 kJ/mol for **3** [4].

In the course of our studies, we have synthesized the analogous phenolic compounds **5a–5e** (Scheme 2a) in order to see whether hydrogen bond formation affects the conformational interconversion process (Scheme 2b). For this purpose, ^{1}H NMR spectra were recorded at different temperatures between 233 and 323 K.

Results and Discussion

400 MHz ^{1}H NMR spectra of **5a**, **5d**, and **5e** were recorded in $CDCl_3$ at temperatures ranging from 233 K to 333 K; as an example, Fig. 1 depicts these spectra for **5d**. Whereas the signals of the H-3 atoms (H^{a} and H^{b}) were barely visible at room temperature, they appeared at temperatures higher than 323 K as singlets at $\delta = 3.70$ (**5a**), 3.85 (**5d**), and 4.20 (**5e**) ppm, respectively, indicating an interconversion of the seven-membered rings [4]. Lowering the temperature resulted in broadening and splitting of this signal, and at temperatures below 250 K the signals were split into two doublets ($^{2}J \approx 11.5$ Hz) at $\delta = 2.38$ and 5.25 (**5a**), 2.39 and 5.26 (**5d**), and 2.36 and 6.03 (**5e**) ppm, respectively. We believe that the enormous differences in the ^{1}H chemical shifts of these two geminal protons are due to their relative orientation with respect to the π-electron systems and, consequently, we assigned the low-frequency signal (*e.g.*, $\delta = 2.39$ ppm of **5d** in Fig. 1) to the hydrogen in *quasi*-axial position where it is much more exposed to shielding aromatic ring current effect of the benzodiazepine moiety.

The evaluation of the kinetics from the ^{1}H NMR spectra followed well-established procedures [4, 5], and the ring inversion barriers ΔG^{*} were found to be 52.3 kJ/mol for **5a**, 51.5 kJ/mol for **5d**, and 54.2 kJ/mol for **5e**; estimated error limits are ca ± 1 kJ/mol. An inspection of the values for **5a** and **5d** reveals very similar magnitudes to those reported for **1–3** [4], apparently showing that the introduction of *ortho*-hydroxy groups leading to hydrogen bond formation does not affect the ring inversion barrier significantly. Therefore, we have to assume that the hydrogen bond has not to be opened during the passage of the molecule through the transition state. However, things are a little different for **5e** with an additional methoxy group in 6″-position where the barrier is about 2–3 kJ/mol higher. A reasonable explanation can be found when viewing *Dreiding* models. During the ring inversion process, the *quasi*-equatorial H-3 atom – on its way to *quasi*-axial – has to pass and thereby to come close to the 6″-OCH_3 group. This group is kept in an inward-oriented position by the hydrogen bond formation which is not opened during this transition; therefore, an additional steric congestion is created.

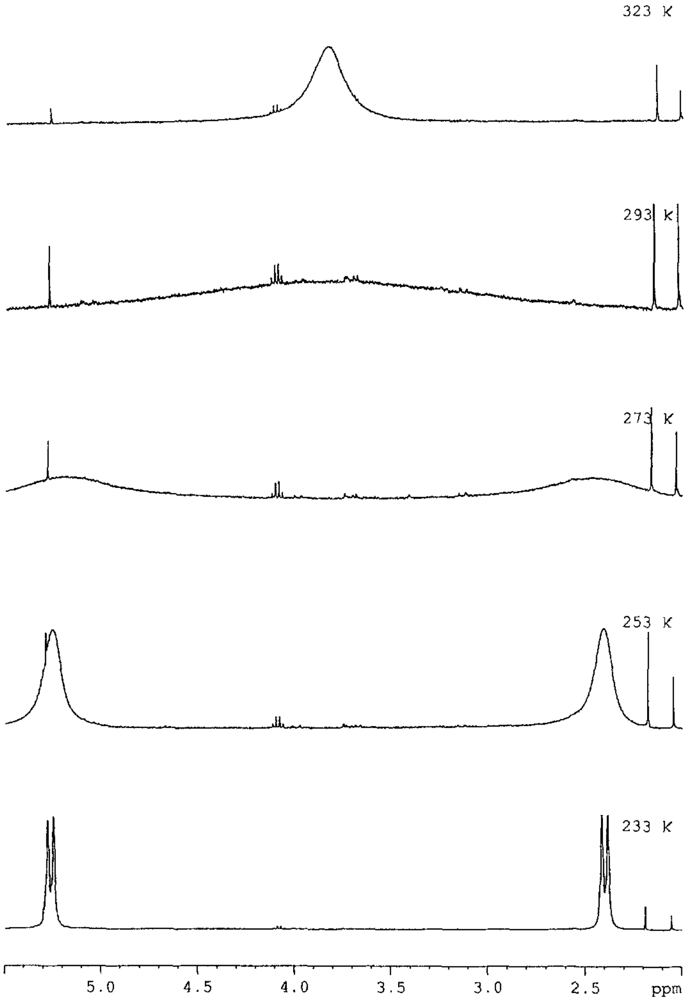


Fig. 1. ^{1}H NMR spectra of **5d** at variable temperature

This, however, is reasonable only if that steric interaction is less severe in the ground state conformation requiring a distortion of the H–O–C-2″–C-1″–C-4–N-5 moiety away from coplanarity. Such a distortion is associated with a weakening of the strength of the hydrogen bond strength for which experimental evidences are present in the NMR spectra.

The ^{15}N chemical shift of the symmetrical compound **5d** is −101.1 ppm (N-1/5), whereas $\delta = -102.0$ (N-1) and −96.9 (N-5) ppm were observed for **5e**. The assignment of the two ^{15}N signals of **5e** was performed by a simple comparison. Typically, shielding of ^{15}N nuclei is observed when the nitrogen atom is exposed to a N···OH hydrogen bond, and ^{15}N chemical shifts of a benzodiazepine without hydrogen bridges are expected to be in the range of −60 to −70 ppm [6]. This leads to the conclusion that one hydrogen bond in **5e** is weaker than the other, and that can only be the bridge between 2″-OH and N-5. An additional argument arises when looking at the chemical shifts of the hydroxy protons. Whereas they are between 14.1 and 14.6 ppm in **5a–d**, two quite different values

(14.3 and 13.7 ppm) were found for **5e**. Since the latter again points to a weaker hydrogen bridge, we assigned it to the proton of the OH group at C-2″.

That there is indeed some steric interference between the 6″-methoxy group and the *quasi*-equatorial H-3 in the ground state is reflected by the chemical shift of the H-3 atom ($\delta = 6.03$ ppm), a value which is approximately 0.8 ppm higher than any other δ value of *quasi*-equatorial protons. Such deshieldings are typical for steric compression situations [7].

Experimental

The ^{1}H NMR spectra were recorded in $CDCl_3$ at 400 MHz (Bruker AM-400 and DRX-400 spectrometers), those of **5a**, **5d**, and **5e** also at temperatures ranging from 233 K to 333 K. ^{13}C NMR spectra ($CDCl_3$, room temperature) were obtained at 100.6 MHz (Bruker AM-400); ^{15}N NMR spectra ($CDCl_3$, room temperature) were recorded at 50.7 MHz (Bruker AMX-500) using the INEPT pulse sequence optimized to a ^{15}N, ^{1}H coupling constant of 2.5 Hz with a relaxation delay of 6 s and an aquisition time of 1.6 s. The chemical shifts of ^{1}H were referred to internal $CHCl_3$ ($\delta = 7.24$ ppm), those of ^{13}C to internal $CDCl_3$ ($\delta = 77.0$ ppm), and those of ^{15}N to external CH_3NO_2 ($\delta = 0.0$ ppm). ^{1}H and ^{13}C chemical shifts assignments are based exclusively on ^{1}H multiplicity patterns and comparisons with structurally related compounds. Therefore, some ambiguities arose in signal assignments which, however, did not affect the verification of the structures. IR spectra were recorded as KBr pellets on a Bruker IFS 25 spectrophotometer. Mass spectra were obtained with electronimpact ionisation on a Finnigan MAT 312 instrument.

The β-diketones **4a–c** were prepared by a method reported earlier for **4a** [8]. Purifications were performed by column chromatography on silica gel using petrol ether and ethyl acetate (in gradient polarity ratios) as eluent.

1-(2′-Hydroxyphenyl)-3-phenyl-propane-1,3-dione (**4a**)

M.p.: 120–121°C (Ref. [8]: 120–122°C).

1-(2′-Hydroxyphenyl)-3-(2″-methoxyphenyl)-propane-1,3-dione (**4b**)

M.p.: 79.5–80°C; yield: 64%; IR (V_{max}, KBr, cm^{-1}): 3188, 3080, 3048, 3008, 2976, 2940, 2880, 2836, 1684, 1604, 1568, 1508, 1492, 1456, 1432, 1388, 1352, 1324, 1292, 1236, 1200, 1180, 1160, 1076, 1056, 1028, 900, 844, 820, 760, 704, 668, 616, 524; ^{1}H NMR ($CDCl_3$): $\delta = 3.65$ (s, *ca.* 0.8H, CH_2), 3.98 (s, 3H, OCH_3), 4.62 (s, *ca.* 0.4H, -CH=), 6.81–7.13 (m, 4H, Ar-H), 7.42–7.53 (m, 2H, Ar-H), 7.72 (dd, 1H, ArH), 7.98 (d, 1H, Ar-H), 12.22 (s, 1H, phenolic, exchangeable with D_2O) 15.60 (s, 1H, enolic, exchangeable with D_2O) ppm; MS: *m/z* (%) = 270 (M^+, 12), 269 (1), 252 (4), 240 (2), 239 (9), 237 (2), 223 (1), 205 (1.5), 177 (2), 163 (3), 151 (3), 149 (3), 137 (4), 136 (11), 135 (100), 123 (4), 121 (21), 120 (5), 97 (6), 92 (8), 82 (7), 77 (4), 76 (16), 68 (11).

1-(2′-Hydroxyphenyl)-3-(2″,6″-dimethoxyphenyl)-propane-1,3-dione (**4c**)

M.p.: 118–121°C; yield: 69%; IR (V_{max}, KBr, cm^{-1}): 3092, 3072, 3004, 2964, 2940, 2840, 1700, 1616, 1588, 1572, 1512, 1472, 1432, 1324, 1296, 1252, 1200, 1180, 1156, 1116, 1028, 916, 864, 828, 776, 744, 716, 660, 620, 600, 580, 544; ^{1}H NMR ($CDCl_3$): $\delta = 3.72$ (s, *ca.* 0.7H, CH_2), 3.81 (s, 6H, 2 OCH_3), 4.42 (s, *ca.* 0.4H, –CH=), 6.51–6.64 (m, 2H, Ar-H), 6.85 (t, 1H, H-4″), 7.63 (dd, 1H, J = 8 Hz, H-6′), 12.18 (s, 1H, phenolic, exchangeable with D_2O), 15.25 (s, 1H, enolic, exchangeable with D_2O) ppm; MS: *m/z* (%) = 300 (M^+, 6), 270 (6), 269 (31), 254 (2), 167 (2), 166 (15), 165

(100), 151 (4), 150 (11), 138 (6), 135 (2), 122 (6), 121 (13), 107 (9), 93 (4), 92 (3), 91 (3), 77 (5), 76 (2), 69 (3), 65 (7).

General method for the preparation of 1,5-benzodiazepines **5a–c**

Benzodiazepines **5a–c** were prepared in analogy to a reported method [9]. The β-diketones **4a–c** (0.01 mol) were dissolved in toluene. Glacial acetic acid (5 ml) was added, followed by *o*-phenylendiamine (0.01 mole), and the mixture was refluxed for 6 h. The solvent was removed under reduced pressure, and the mixture was treated with a small quantity of methanol. The precipitates obtained were recrystallized using isopropanol. Purification of **5a–c** was performed using silica gel and ethylacatate-petrol ether as eluent.

2-(2′-Hydroxyphenyl)-4-phenyl-1,5-benzodiazepine (**5a**)

The synthesis of **5a** has been described before [8]; ^{1}H NMR ($CDCl_3$): δ = *ca.* 3.7 (very broad s, 2H, H-3); 6.77 (t, 1H, H-5′), 6.89 (t, 1H, H-5′), *ca.* 7.25 (m, 2H, H-7, H-8), 7.30 (t, 1H, H-4′), 7.36 (m, 3H, H-3″, H-4″, H-5″), 7.45, 7.53 (dd, each 1H, H-6, H-9), 7.92 (m, 2H, H-2″, H-6″), 14.5 (broad s, 1H, OH exchangeable with D_2O) ppm; ^{13}C NMR ($CDCl_3$): δ = 33.3 (C-3), 117.9 (C-1′, C-1″), 118.6, 118.4 (C-3′, C-5′, C-3″, C-5″), 126.3, 125.8 (C-7, C-8), 127.9 (C-9), 128.3 (C-4′), 128.8, 128.3 (C-2″/6″, C-3″/5″), 129.0 (C-4″), 131.0 (C-6), 133.5 (C-6′), 137.2, 136.8 (C-9a, C-1″), 141.6 (C-5a), 155.2 (C-4), 158.5 (C-2), 162.5 (C-2′) ppm.

2-(2′-Hydroxyphenyl)-4-(2″-methoxyphenyl)-1,5-benzodiazepine (**5b**)

M.p.: 131.5–132°C; yield: 70%; IR (V_{max}, KBr, cm^{-1}): 3668, 3428, 3368, 3336, 3080, 3000, 2956, 2872, 2836, 2464, 1924, 1748, 1716, 1672, 1620, 1596, 1508, 1472, 1432, 1412, 1388, 1356, 1304, 1260, 1228, 1176, 1148, 1124, 1112, 1092, 1060, 1032, 1008, 976, 952, 920, 896, 864, 828, 656, 632, 616, 596, 572, 532; ^{1}H NMR ($CDCl_3$): H-3 signal not observed due to coalescence; δ = 3.90 (s, 3H, OCH_3), 6.72 (t, 1H, H-5″), 6.91 (t, 1H, H-5′), 6.97 (dd, 1H, H-3′), 7.02 (dd, 1H, H-3″), 7.27 (2 t, 2H, H-4′, H-4″), *ca.* 7.4 (m, 4H, H-7, H-8, H-6′, H-6″), 7.53, 7.62 (2 m, each 1H, H-6, H-9), 14.5 (broad s, 1H, OH, exchangeable with D_2O) ppm; ^{13}C NMR ($CDCl_3$): δ = 36.9 (C-3), 55.7 (OCH_3), 111.2 (C-3″), 118.5, 118.1 (C-3′, C-5′), 121.1 (C-5′), 126.1, 125.8 (C-7, C-8), 129.1, 128.8, 127.7 (C-6, C-9, C-4″), 131.9, 131.7 (C-4″, C-6″), 133.2 (C-6′), 137.3 (C-9a), 141.7 (C-5a), 157.5, 158.1 (C-2, C-4), 159.5 (C-2″), 162.4 (C-2′) ppm; MS: *m/z* (%) = 342 (M^+, 88), 327 (53), 311 (100), 298 (7), 270 (5), 249 (20), 235 (12), 223 (31), 219 (23), 210 (32).

2-(2′-Hydroxyphenyl)-4-(2″,6″-dimethoxyphenyl)-1,5-benzodiazepine (**5c**)

M.p.: 163–164°C; yield: 72%; IR (ν_{max}, KBr, cm^{-1}): 3505, 3448, 3420, 3052, 3008, 2964, 2936, 2904, 2836, 2768, 2708, 1588, 1560, 1500, 1472, 1432, 1400, 1332, 1300, 1256, 1212, 1160, 1108, 1028, 1004, 880, 856, 840, 824, 764, 656, 592, 528, 508, 480; ^{1}H NMR ($CDCl_3$): H-3 signal not observed due to coalescence; δ = 3.47 (s, 6H, 2 OCH_3), 6.51 (d, 2H, H-3″, H-5″), 6.68 (td, 1H, H-5′), 6.97 (dd, 1H, H-3′), 7.27 (2 t, 2H, H-4′, H-4″), 7.3–7.35 (m, 4H, H-7, H-8, H-6′, H-6″), 7.53, 7.62 (2 m, each 1H, H-6, H-9), 14.6 (s, 1H, OH, exchangeable with D_2O) ppm; ^{13}C NMR ($CDCl_3$): δ = 38.9 (C-3), 55.6 (2 OCH_3), 104.0 (C-3″, C-5″), 118.2, 118.0 (C-3′, C-5′), (C-1′), 118.8 (C-1′), 125.8, 125.7 (C-7, C-8), 128.8, 128.7, 127.8 (C-6, C-9, C-4′), 130.7 (C-4″), 133.0 (C-6′), 137.3 (C-9a), 141.5 (C-5a), 154.2 (C-2″, C-6″), 157.8, 158.4 (C-4, C-2), 162.4 (C-2′) ppm; MS: *m/z* (%) = 372 (M^+, 44), 358, (32), 341 (67), 326 (22), 311 (3), 279 (13), 269 (15), 249 (18), 223 (8), 210 (21), 179 (13), 165 (100), 150 (13), 121 (17), 84 (21), 77 (22), 65 (11).

General method for the demethylation of compounds **5b** *and* **5c** [10]

To 1 mmol of methoxybenzodiazepines **5b** and **5c** dissolved in 20 ml of 1,2-dichloroethane, a solution of $BBr_3 \cdot (CH_3)_2S$ (1.25 g, 4 mmol) in 20 ml of dichloroethane was added dropwise under an argon atmosphere. The mixture was heated at 356 K for 12 h for **5b** and 6 h for **5c**. After completion, the mixture was worked up as reported [10]; **5b** gave **5d**, **5c** gave **5e**.

2-(2′-Hydroxyphenyl)-4-(2″-hydroxyphenyl)-1,5-benzodiazepine (**5d**)

M.p.: 240.5–241°C; yield: 91%; IR (ν_{max}, KBr, cm^{-1}): 3896, 3804, 3768, 3748, 3732, 3672, 3644, 3624, 3592, 3572, 3564, 3440, 3060, 2996, 2908, 2848, 2716, 2588, 2352, 2276, 1608, 1592, 1492, 1444, 1416, 1332, 1312, 1248, 1200, 1160, 1124, 1036, 996, 948, 888, 848, 824, 748, 664, 588, 572, 544, 504; ^{1}H NMR ($CDCl_3$): $\delta = ca.$ 3.7 (very broad s, 2H, H-3), 6,89 (tm, 2H, H-5′, H-5″), 6.94 (dm, 2H, H-3′, H-3″), 7.32 (tm, 2H, H-4′, H-4″), 7.34 (m, 2H, H-7, H-8), 7.48 (m, 2H, H-6, H-9), 7.90 (dd, 2H, H-6′, H-6″), 14.1 (s, 2H, OH, exchangeable with D_2O) ppm; ^{13}C NMR ($CDCl_3$): $\delta = 32.0$ (C-3), 117.9 (C-1′ and C-1″), 118.8 (C-3′, C-5′, C-3″, C-5″), 126.8 (C-7, C-8), 128.5, 128.2 (C-C-6, C-9, C-4′, C-4″), 134.1 (C-6′, C-6″), 138.4 (C-5a, C-9a), 159.8 (C-2, C-4), 162.6 (C-2′, C-2″) ppm; ^{15}N NMR ($CDCl_3$): $\delta = -101.1$ ppm; MS: *m/z* (%) = 328 (M^+, 100), 311 (32), 283 (4), 235 (38, M^+-C_6H_4OH), 221 (39), 210 (65), 182 (23), 181 (27), 155 (16), 140 (12), 119 (14), 91 (28), 89 (9), 77 (10), 65 (14).

2-(2′-Hydroxyphenyl)-4-(2″-hydroxy-6″-methoxyphenyl)-1,5-benzodiazepine (**5e**)

M.p.: 172–174 yield: 89%; IR (ν_{max}, KBr, cm^{-1}): 3436, 3104, 3056, 2992, 2964, 2940, 2912, 2836, 2708, 2620, 2568, 2536, 2212, 2184, 1592, 1540, 1500, 1452, 1332, 1308, 1240, 1212, 1160, 1116, 1088, 1032, 1008, 936, 892, 864, 836, 788, 760, 728, 648, 584, 564, 516, 476; ^{1}H NMR ($CDCl_3$): H-3 signal not observed due to coalescence; $\delta = 4.04$ (s, 3H, OCH_3), 6.50, 6.58 (2 dd, each 1H, H-3″, H-5″), 6.81 (td, 1H, H-5′), 6.98 (dd, 1H, H-3′), 7.26 (t, 1H, H-4″), 7.29 (td, 1H, H-4′), 7.38 (m, 2H, H-7, H-8), 7.38 (m, 2H, H-6, H-9), 7.70 (dd, 1H, H-6′), 14.2, 13.7 (broad s, 1H, 2′-OH), 13.7 (broad s, 1H, 2″-OH) ppm; ^{13}C NMR ($CDCl_3$): $\delta = 35.9$ (C-3), 56.0 (OCH_3), 101.8 (C-5″), 109.3 (C-1″), 111.5 (C-3″), 118.6, 118.3 (C-3′, C-5′), 118.8 (C-1′), 126.5, 126.4 (C-7, C-8); 129.1, 128.2, 127.9, (C-6, C-9, C-4′), 133.6, 133.4 (C-4′, C-6′), 138.5, 138.2 (C-5a, C-9a), 160.2 (C-2, C-4), 161.1 (C-6″), 162.5 (C-2′), 163.0 (C-2″) ppm; ^{15}N NMR ($CDCl_3$): $\delta = -102.0$ (N-1), -96.9 (N-5) ppm; MS: *m/z* (%) = 358 (M^+, 100), 341 (41), 327 (18), 265 (36), 251 (60), 239 (25), 210 (47), 181 (19), 149 (21), 119 (21), 91 (18), 77 (26), 65 (11).

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