

Supporting Information

to the article

Title: Synthesis, structure, and stereospecific cross-[2 + 2] photocycloaddition of pseudodimeric complexes based on ammonioalkyl derivatives of styryl dyes

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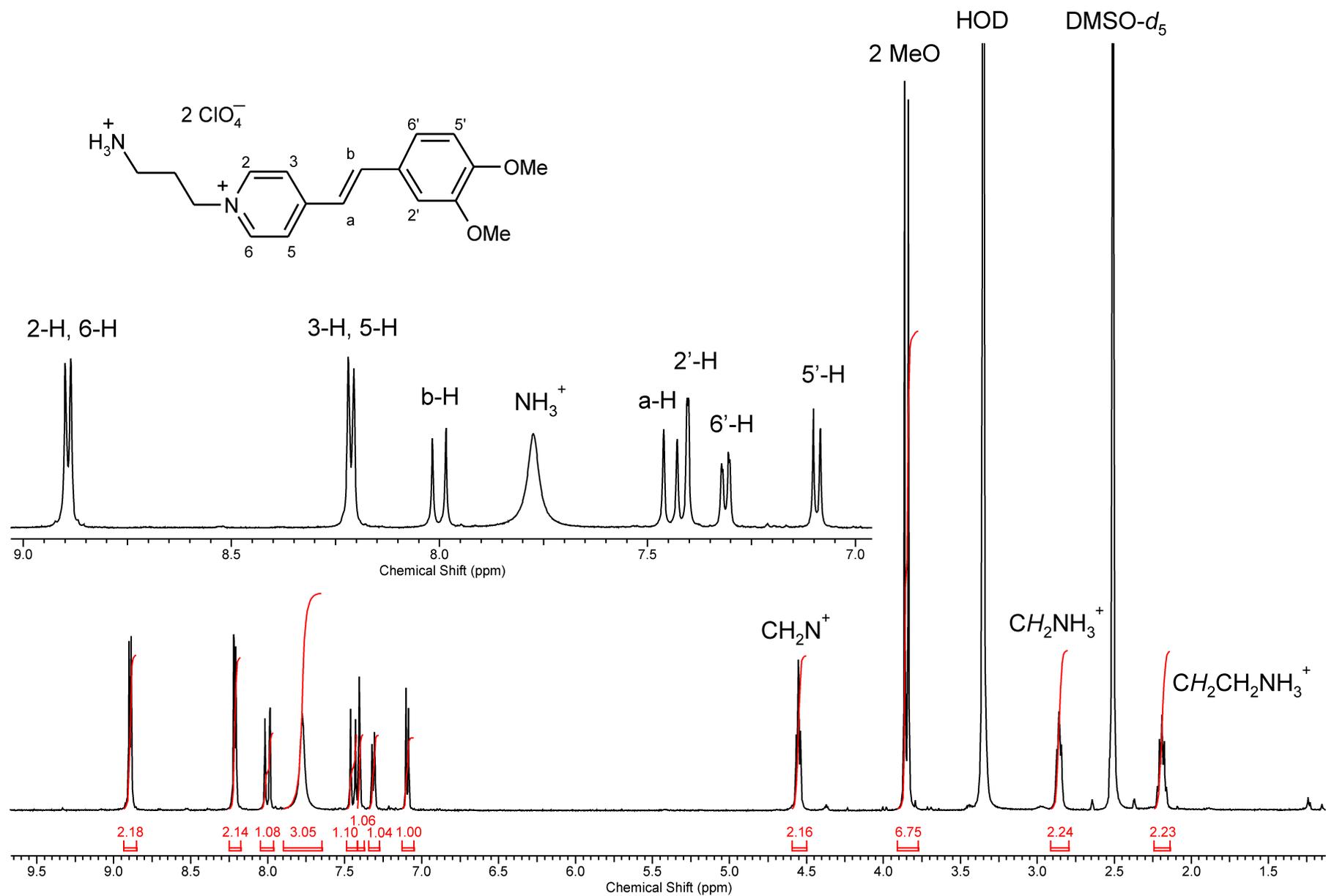


Fig. S1 ^1H NMR spectrum of dye **1a** (500.13 MHz, $\text{DMSO-}d_6$, 23 °C).

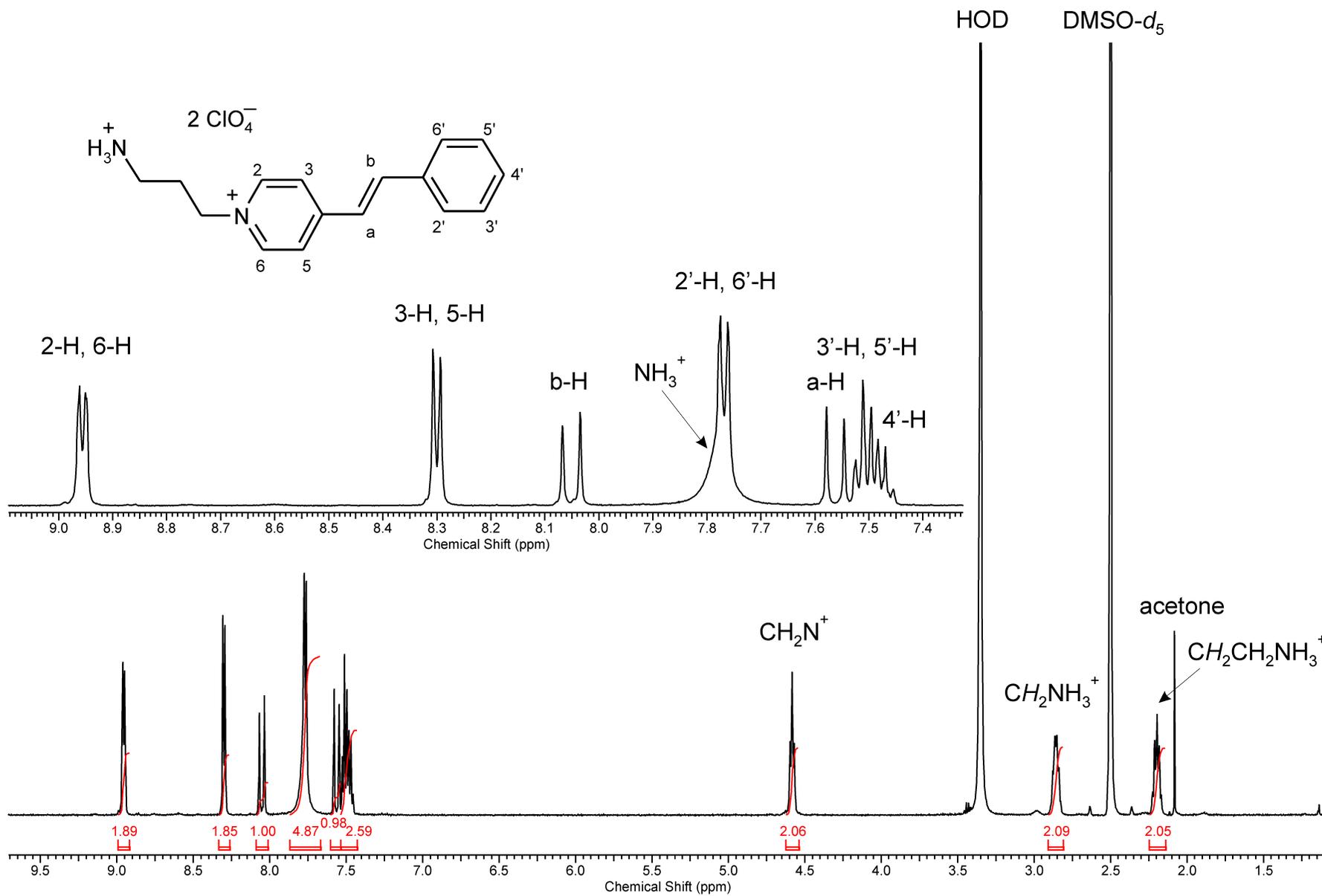


Fig. S2 ^1H NMR spectrum of dye **1b** (500.13 MHz, $\text{DMSO-}d_6$, 25 °C).

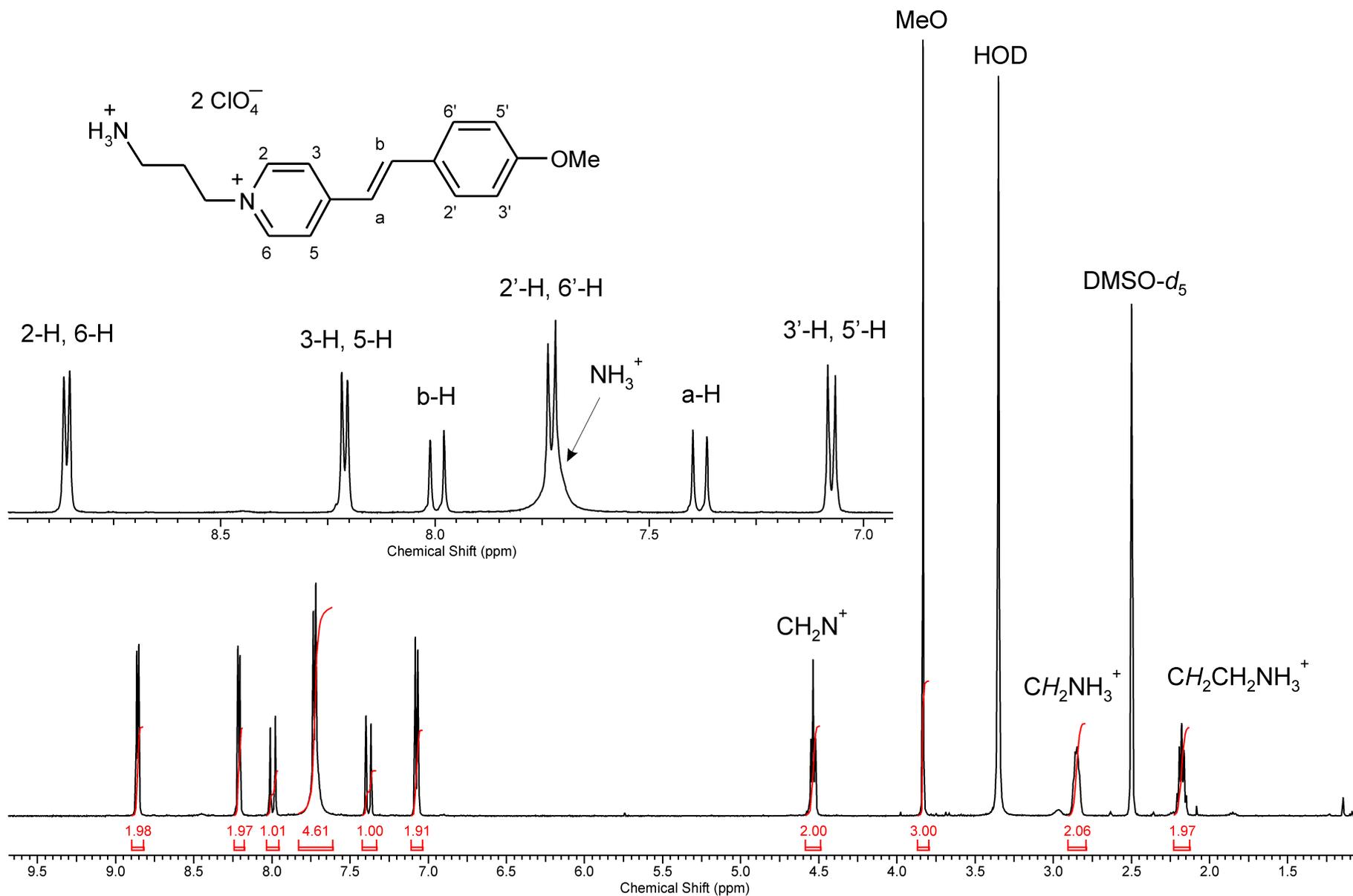


Fig. S3 ^1H NMR spectrum of dye **1c** (500.13 MHz, $\text{DMSO-}d_6$, 25 °C).

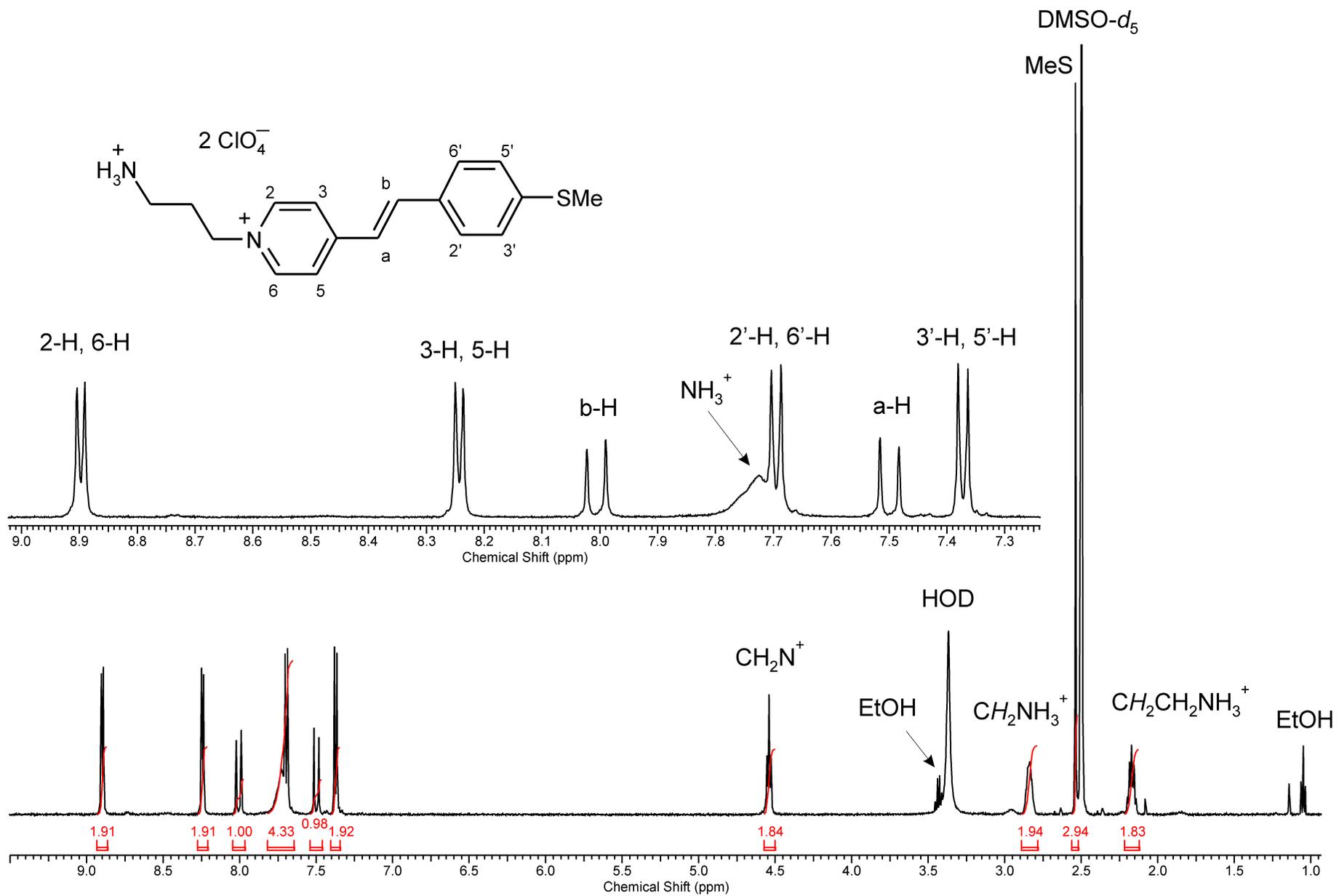


Fig. S4 ¹H NMR spectrum of dye **1d** (500.13 MHz, DMSO-*d*₆, 25 °C).

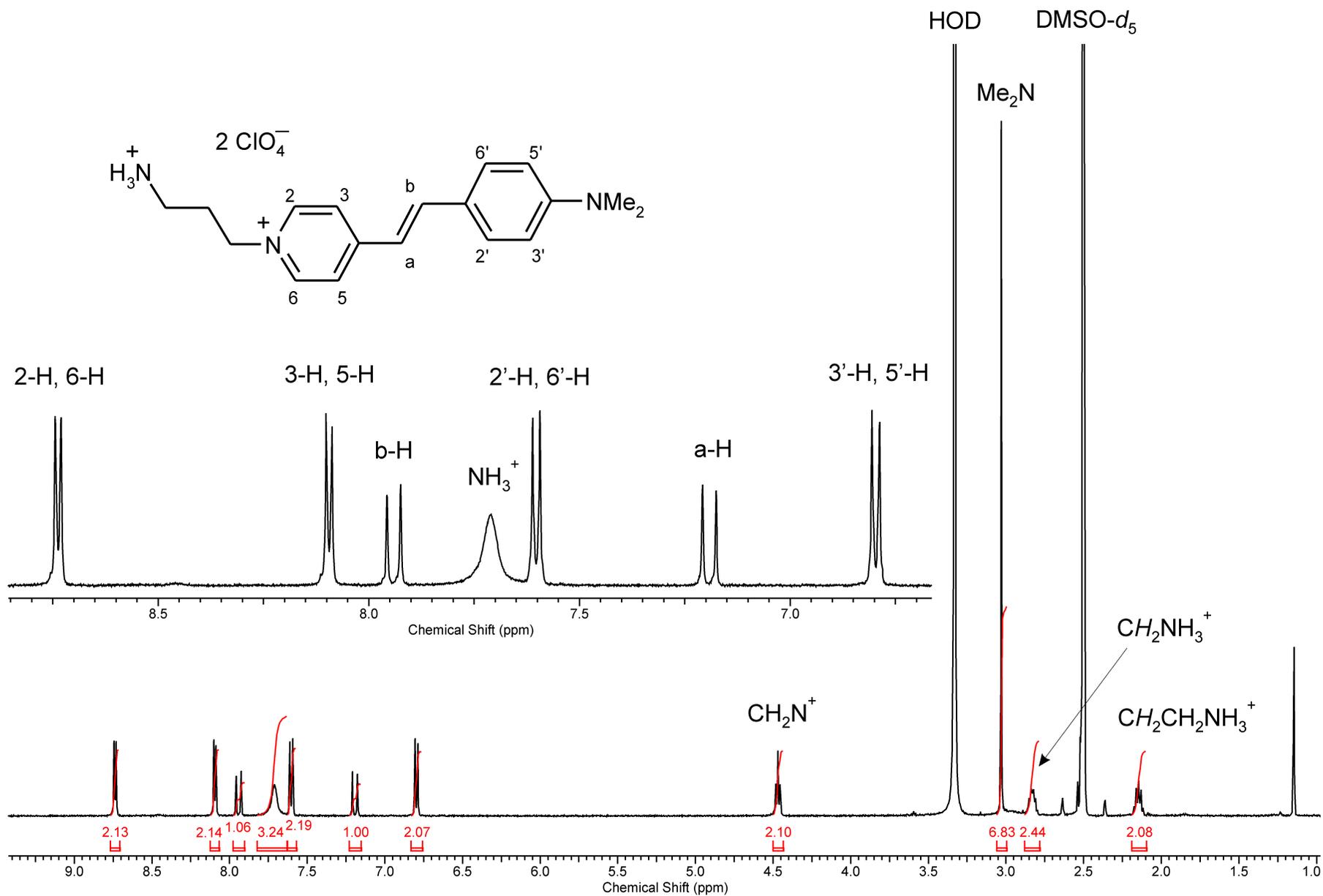


Fig. S5 ^1H NMR spectrum of dye **1e** (500.13 MHz, $\text{DMSO-}d_6$, 24 °C).

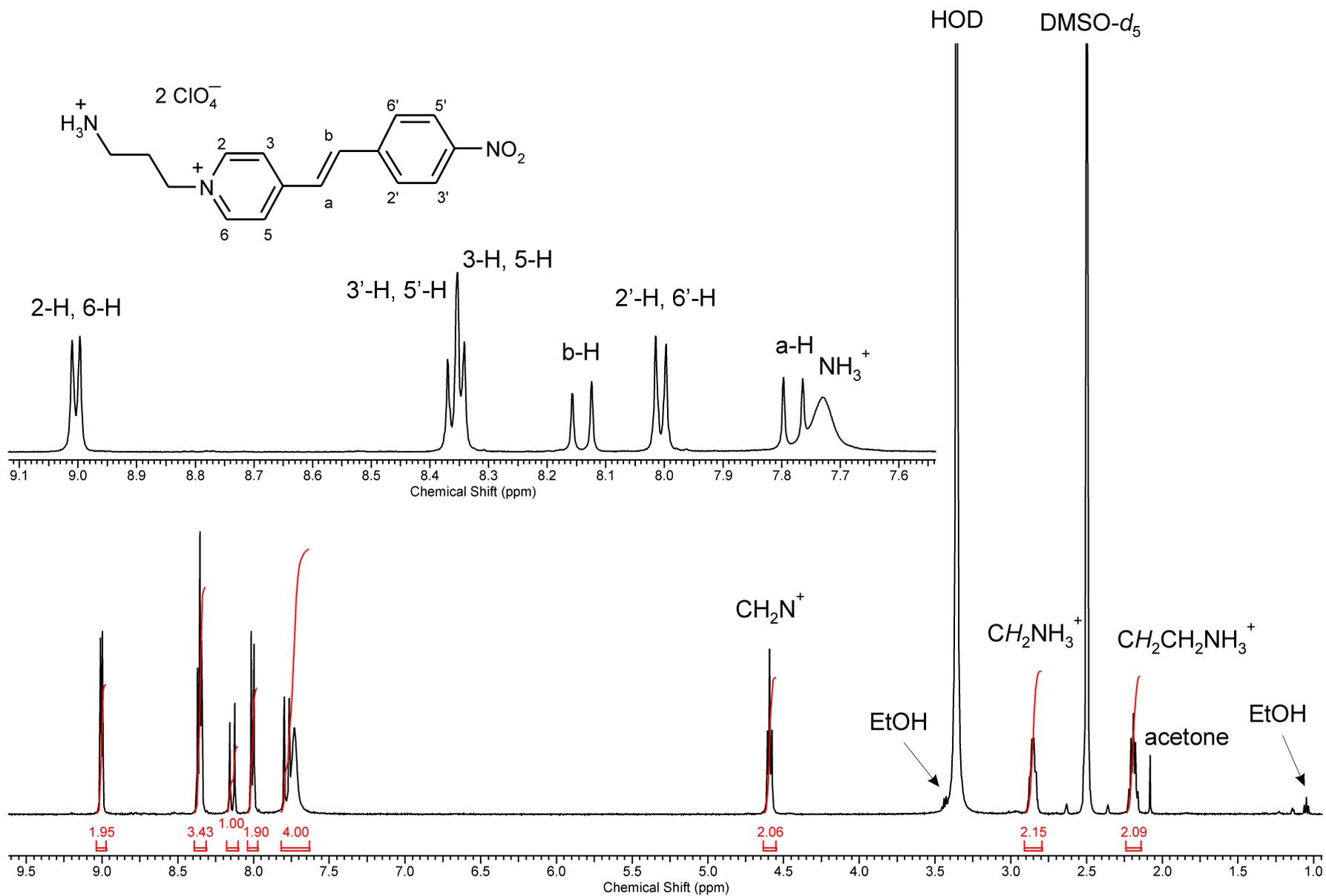


Fig. S6 ¹H NMR spectrum of dye **1f** (500.13 MHz, DMSO-*d*₆, 25 °C).

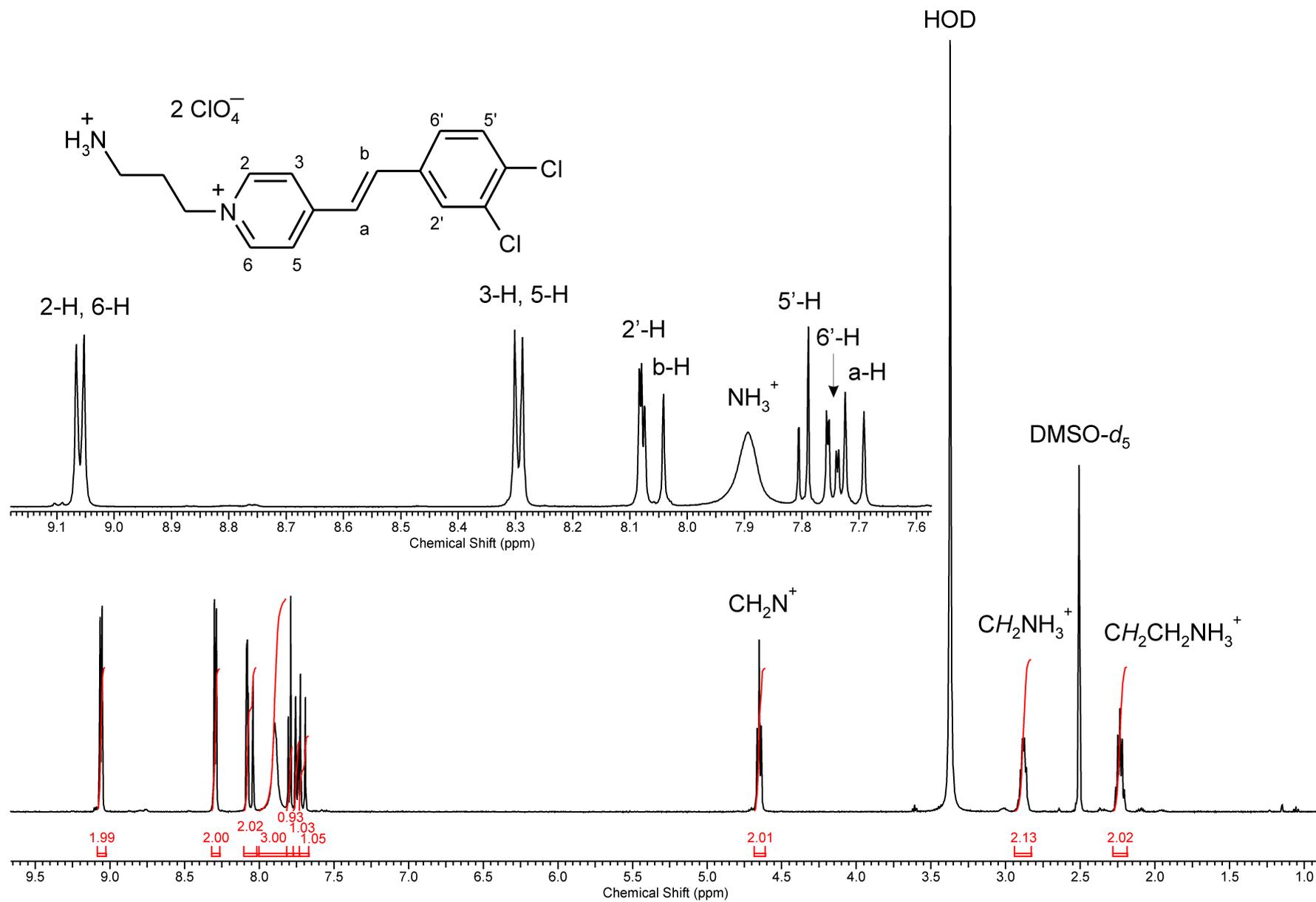


Fig. S7 ¹H NMR spectrum of dye **1g** (500.13 MHz, DMSO-*d*₆, 25 °C).

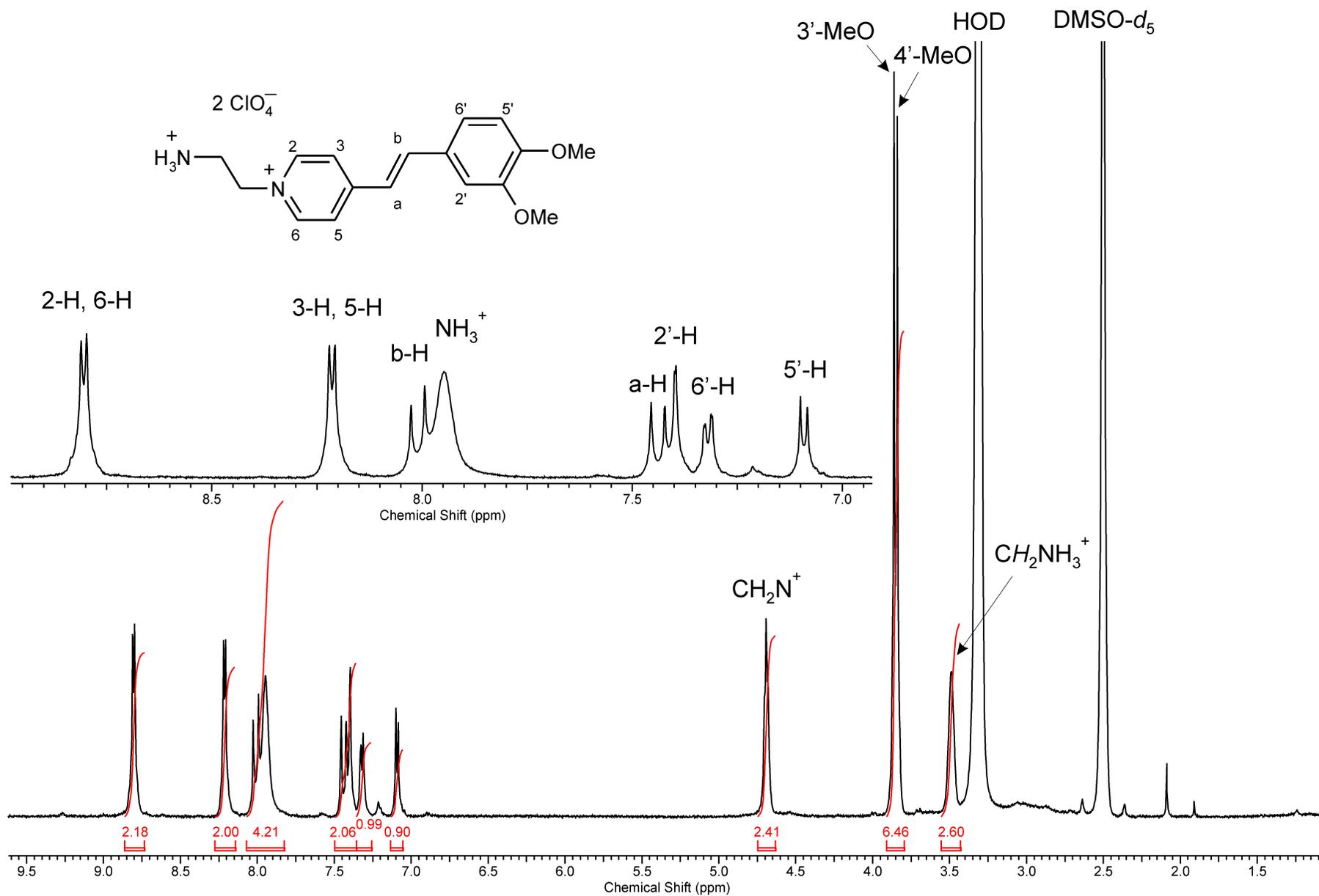


Fig. S8 ^1H NMR spectrum of dye **1h** (500.13 MHz, DMSO- d_6 , 30 °C).

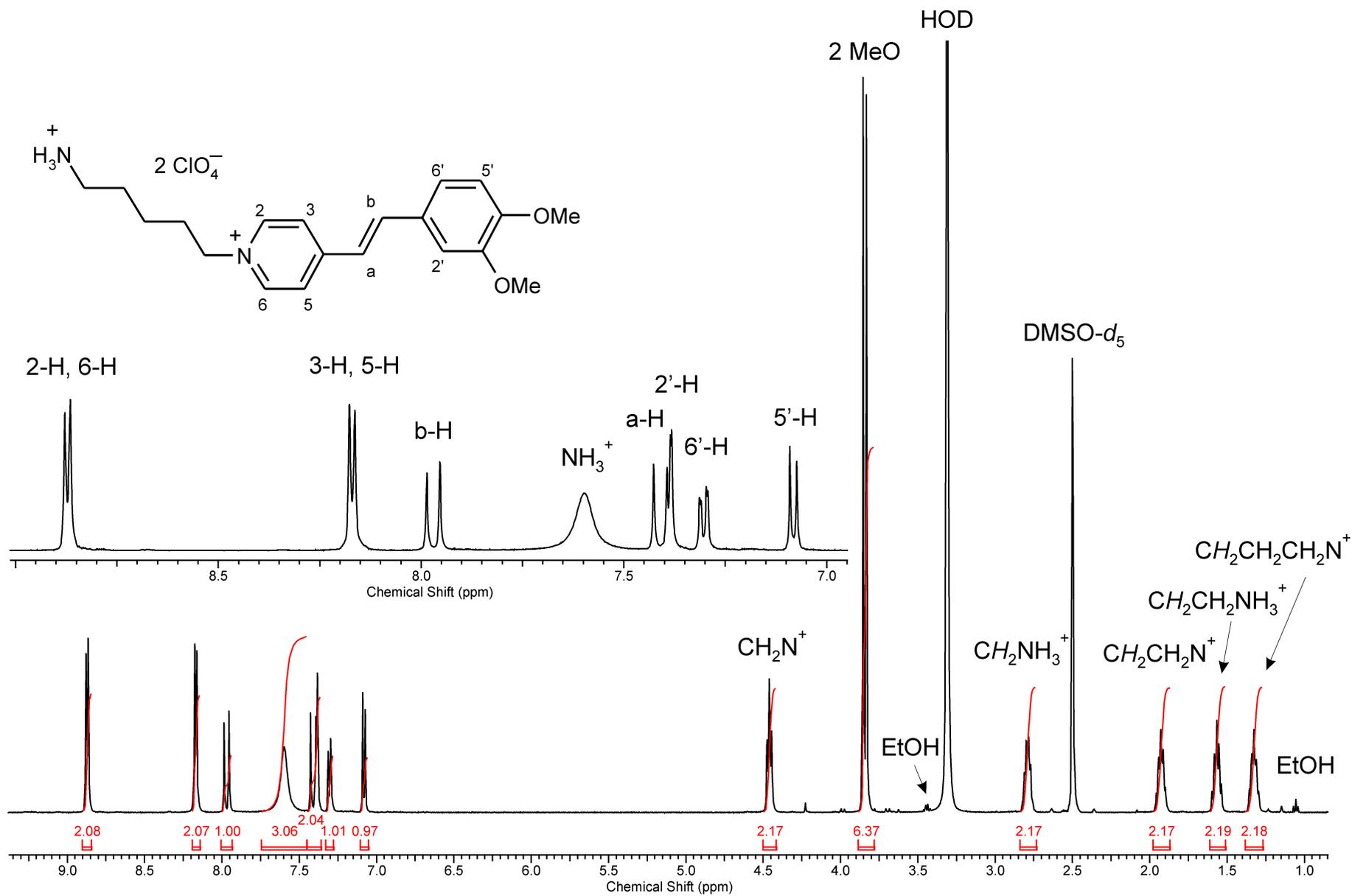


Fig. S9 ¹H NMR spectrum of dye **1i** (500.13 MHz, DMSO-*d*₆, 25 °C).

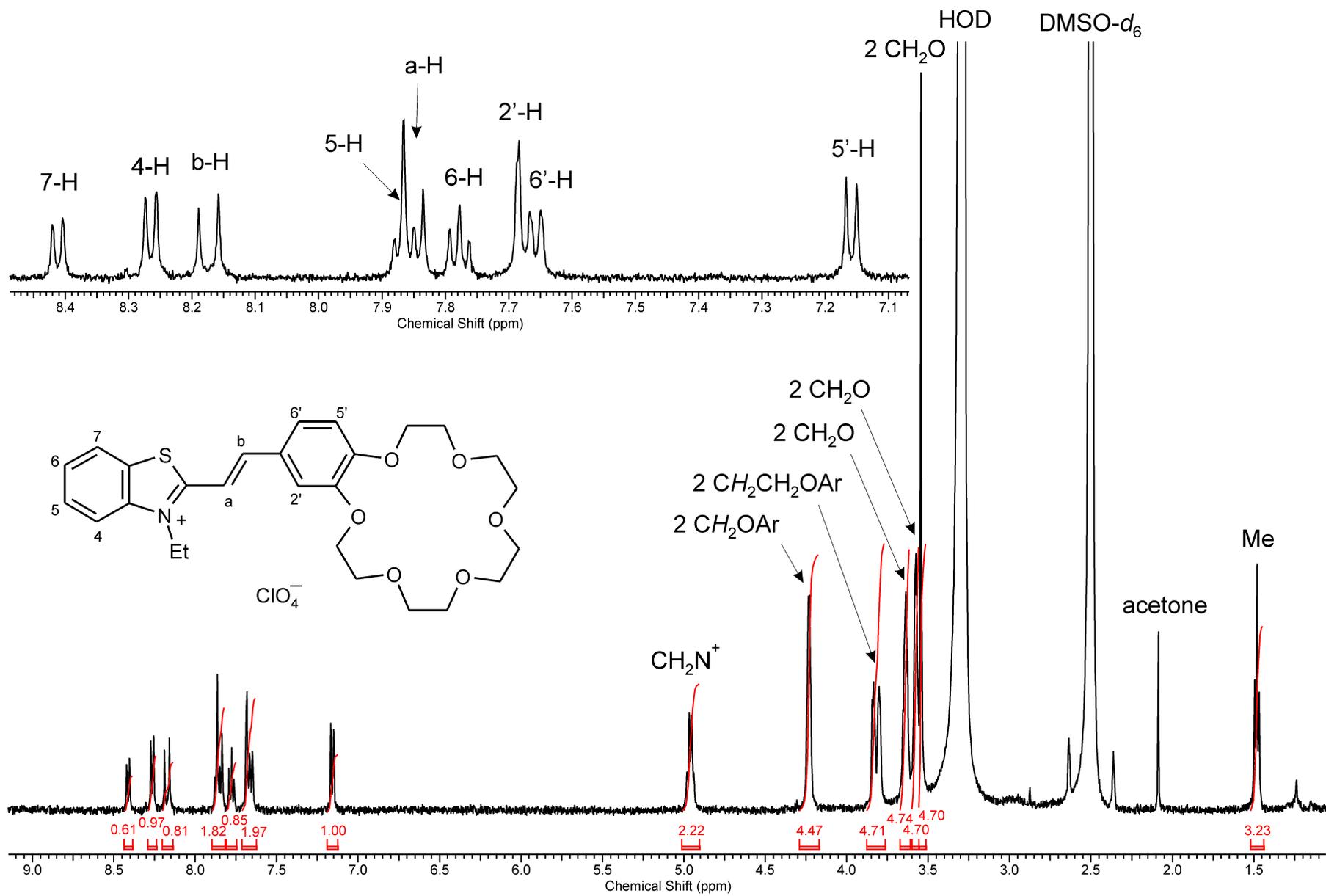


Fig. S10 ^1H NMR spectrum of dye **2c** (500.13 MHz, $\text{DMSO-}d_6$, $30\text{ }^\circ\text{C}$).

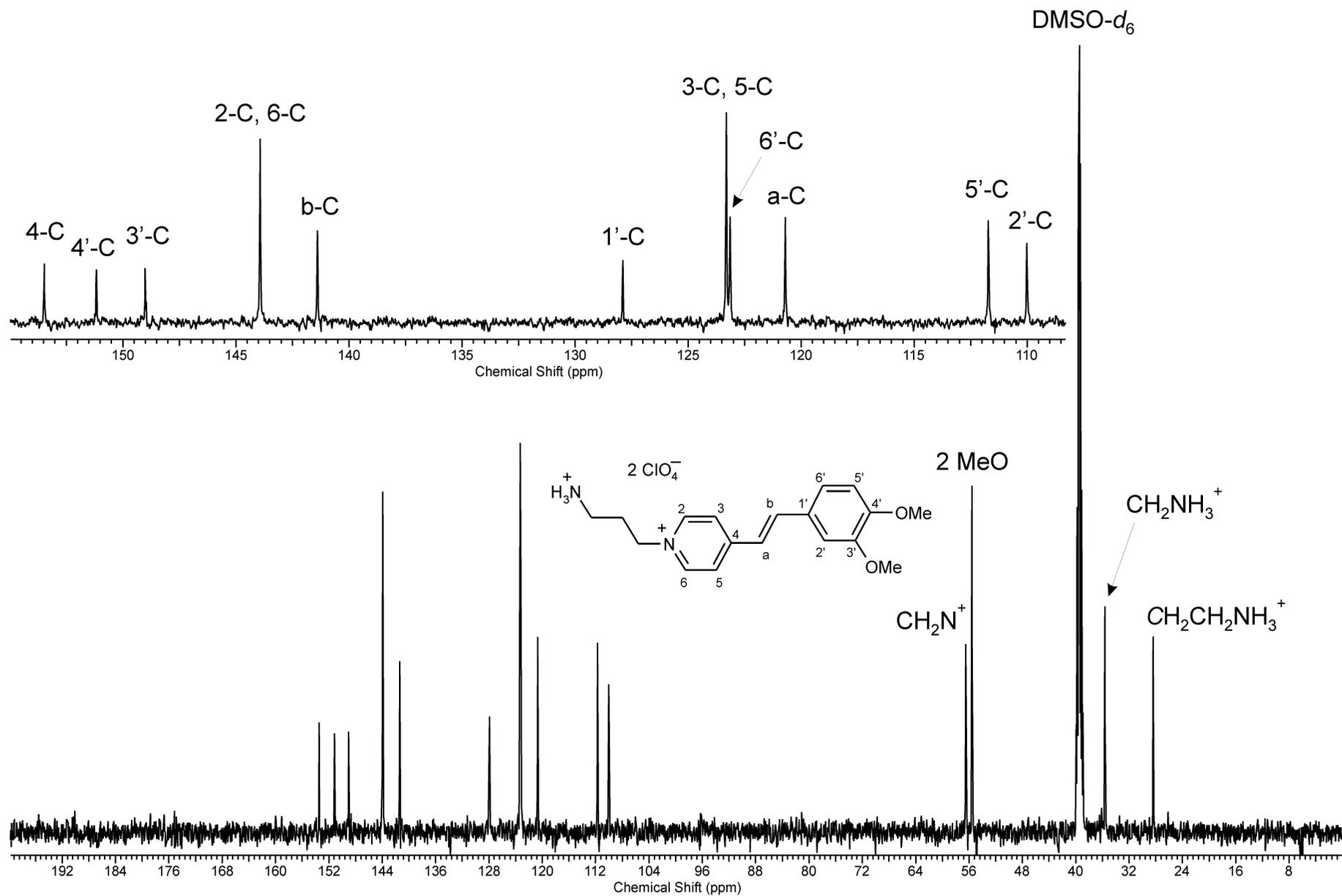


Fig. S11 ^{13}C NMR spectrum of dye **1a** (125.76 MHz, $\text{DMSO-}d_6$, 30 °C).

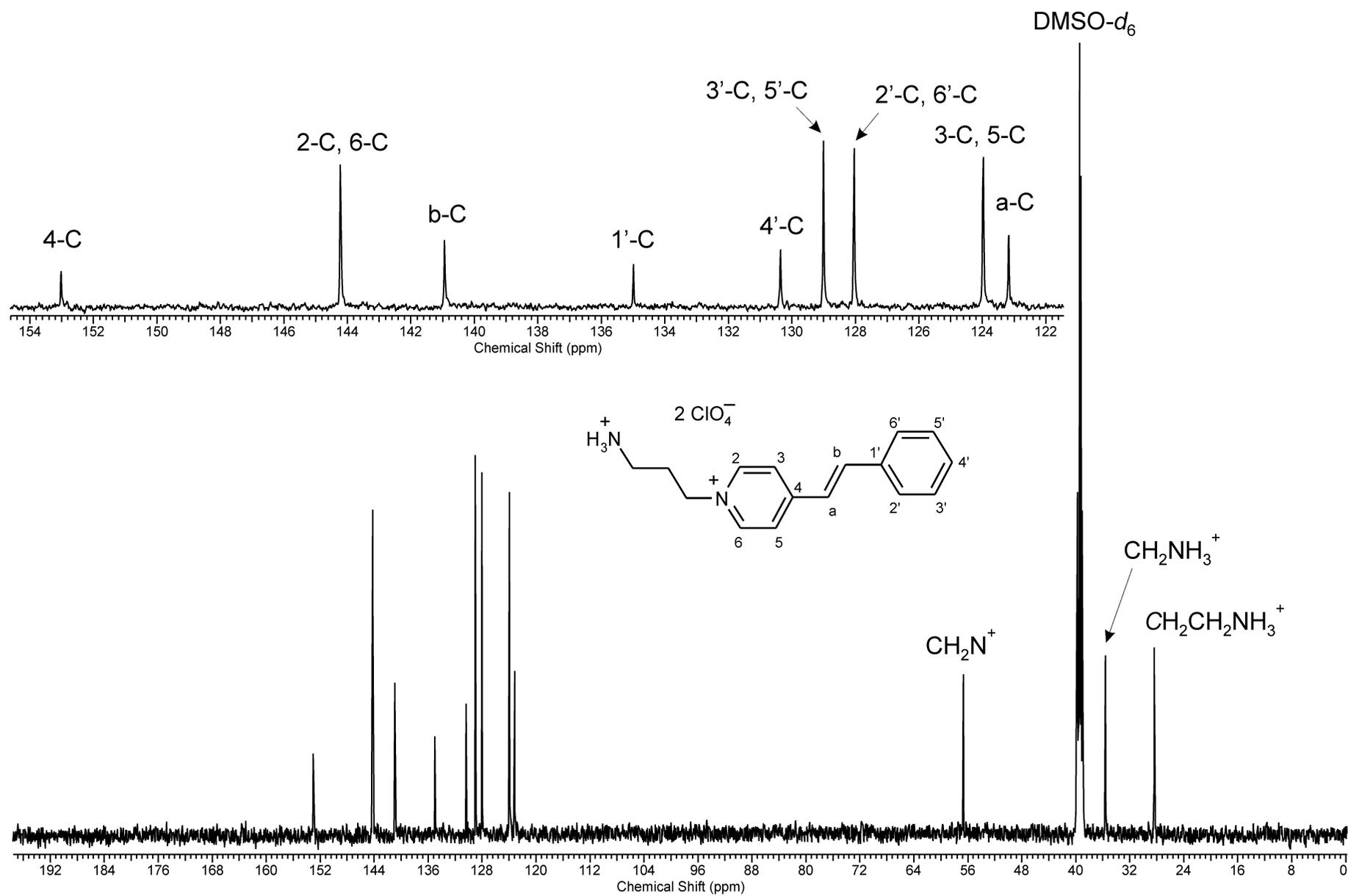


Fig. S12 ^{13}C NMR spectrum of dye **1b** (125.76 MHz, $\text{DMSO-}d_6$, 30°C).

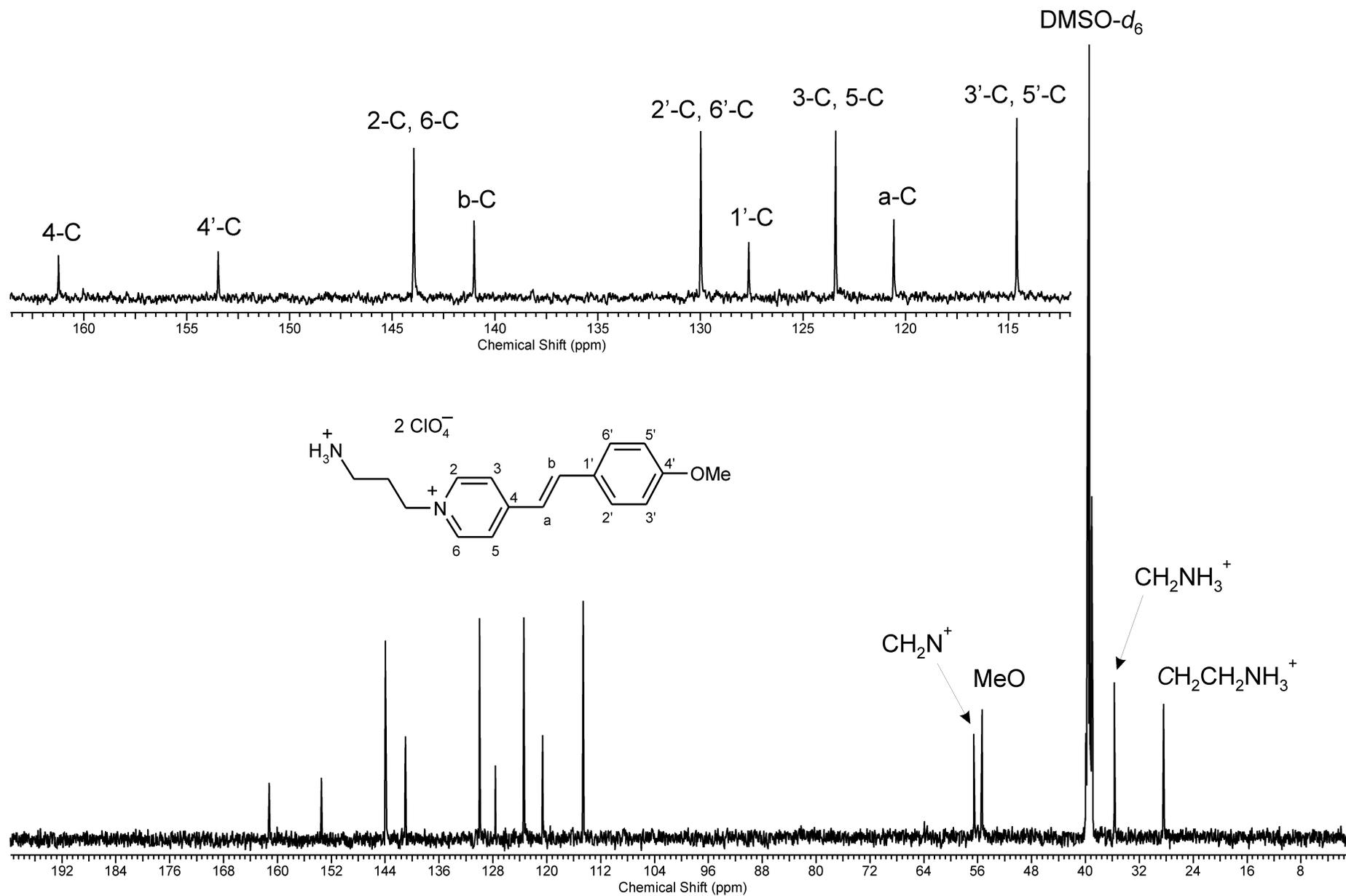


Fig. S13 ^{13}C NMR spectrum of dye **1c** (125.76 MHz, $\text{DMSO-}d_6$, $30\text{ }^\circ\text{C}$).

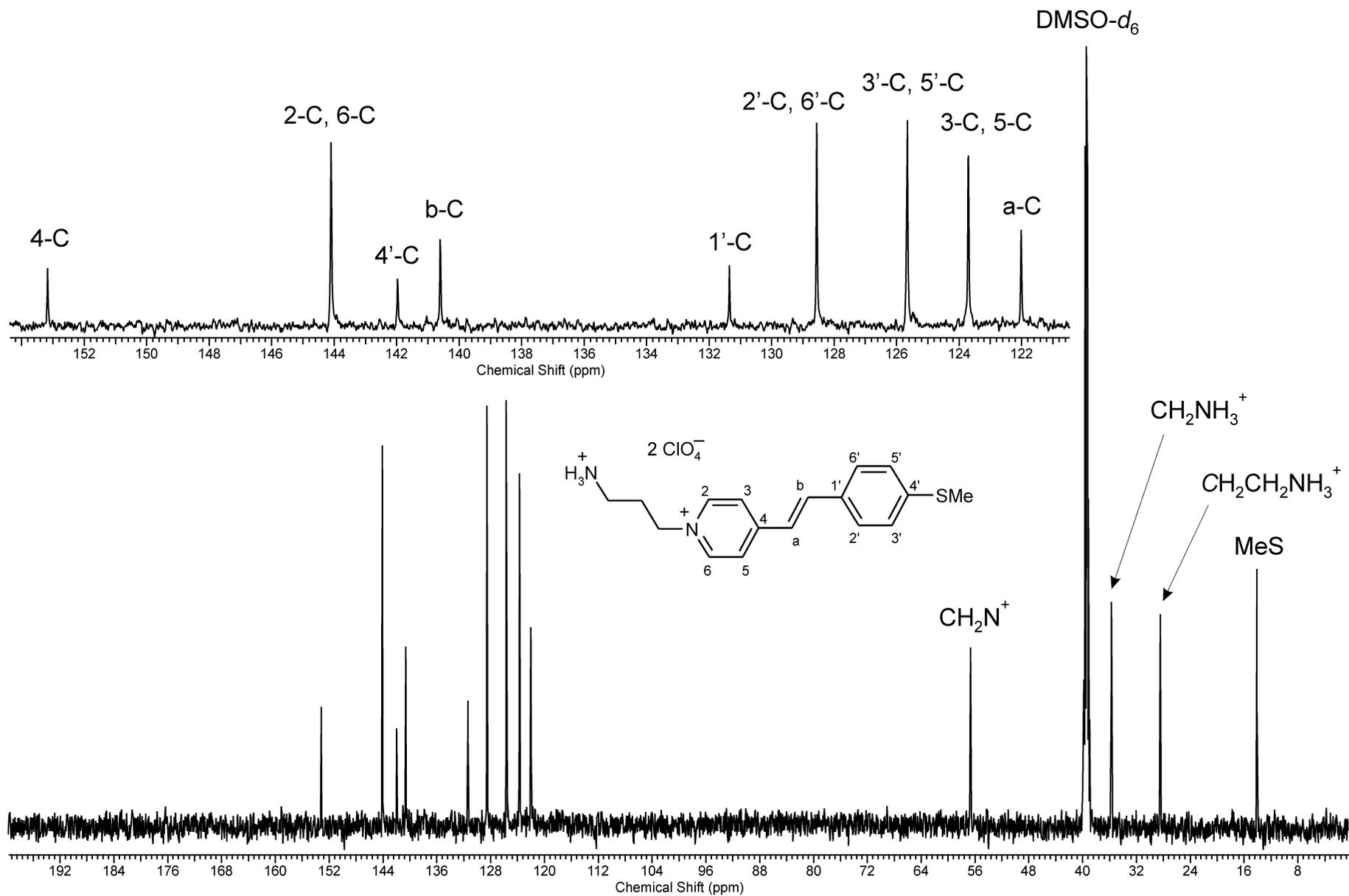


Fig. S14 ^{13}C NMR spectrum of dye **1d** (125.76 MHz, $\text{DMSO-}d_6$, 27 °C).

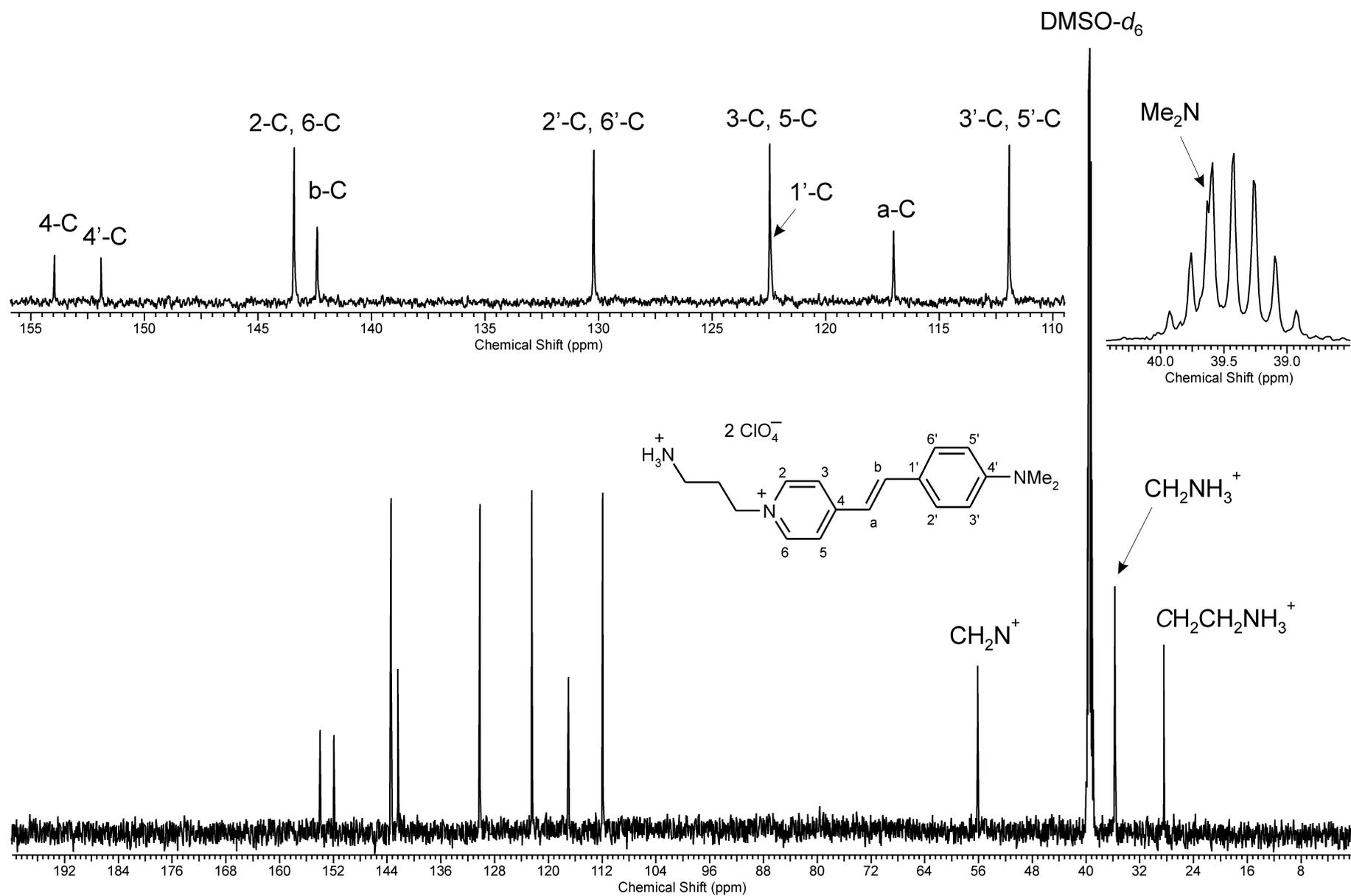


Fig. S15 ^{13}C NMR spectrum of dye **1e** (125.76 MHz, DMSO- d_6 , 25 °C).

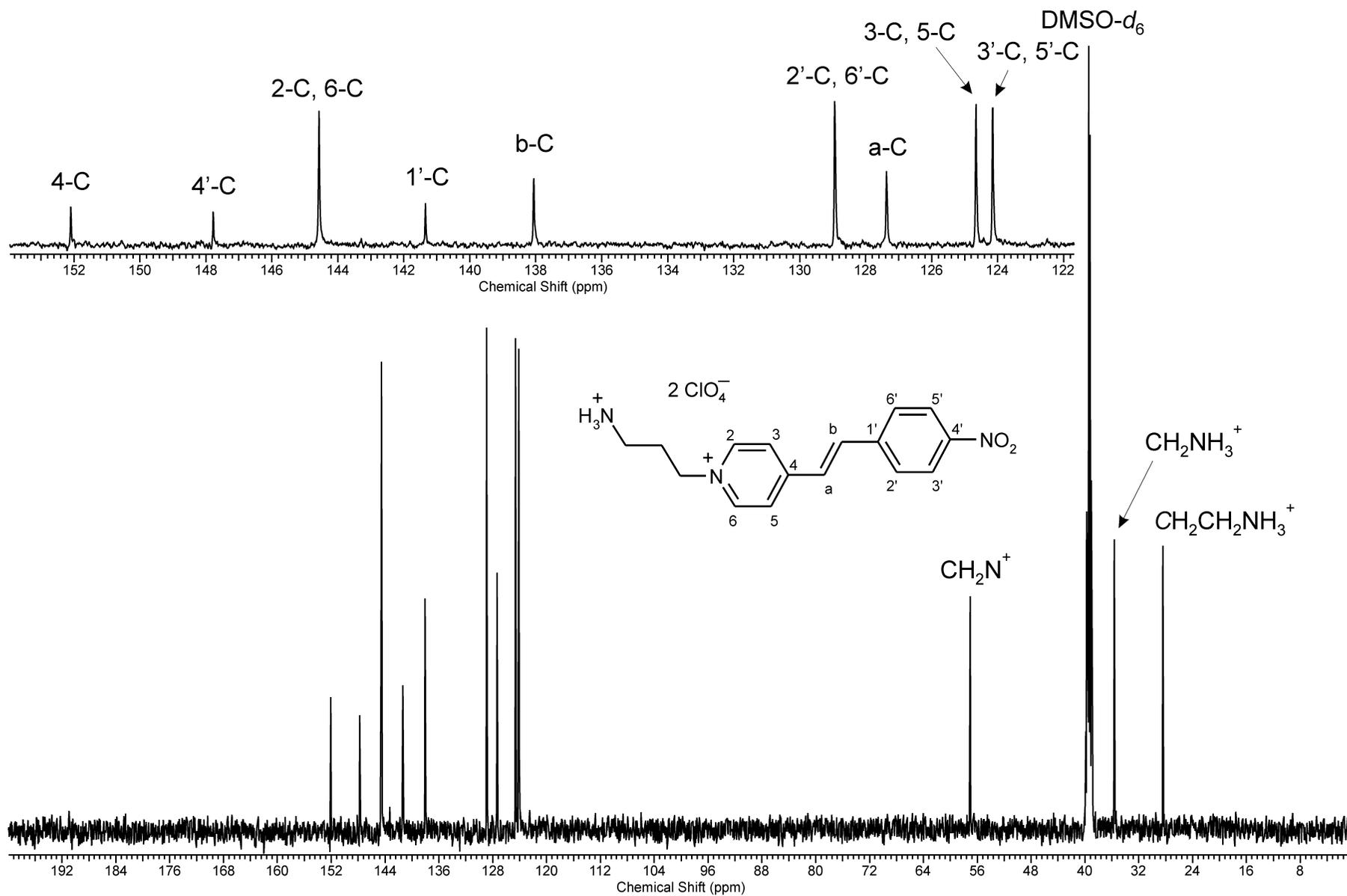


Fig. S16 ^{13}C NMR spectrum of dye **1f** (125.76 MHz, $\text{DMSO-}d_6$, 30°C).

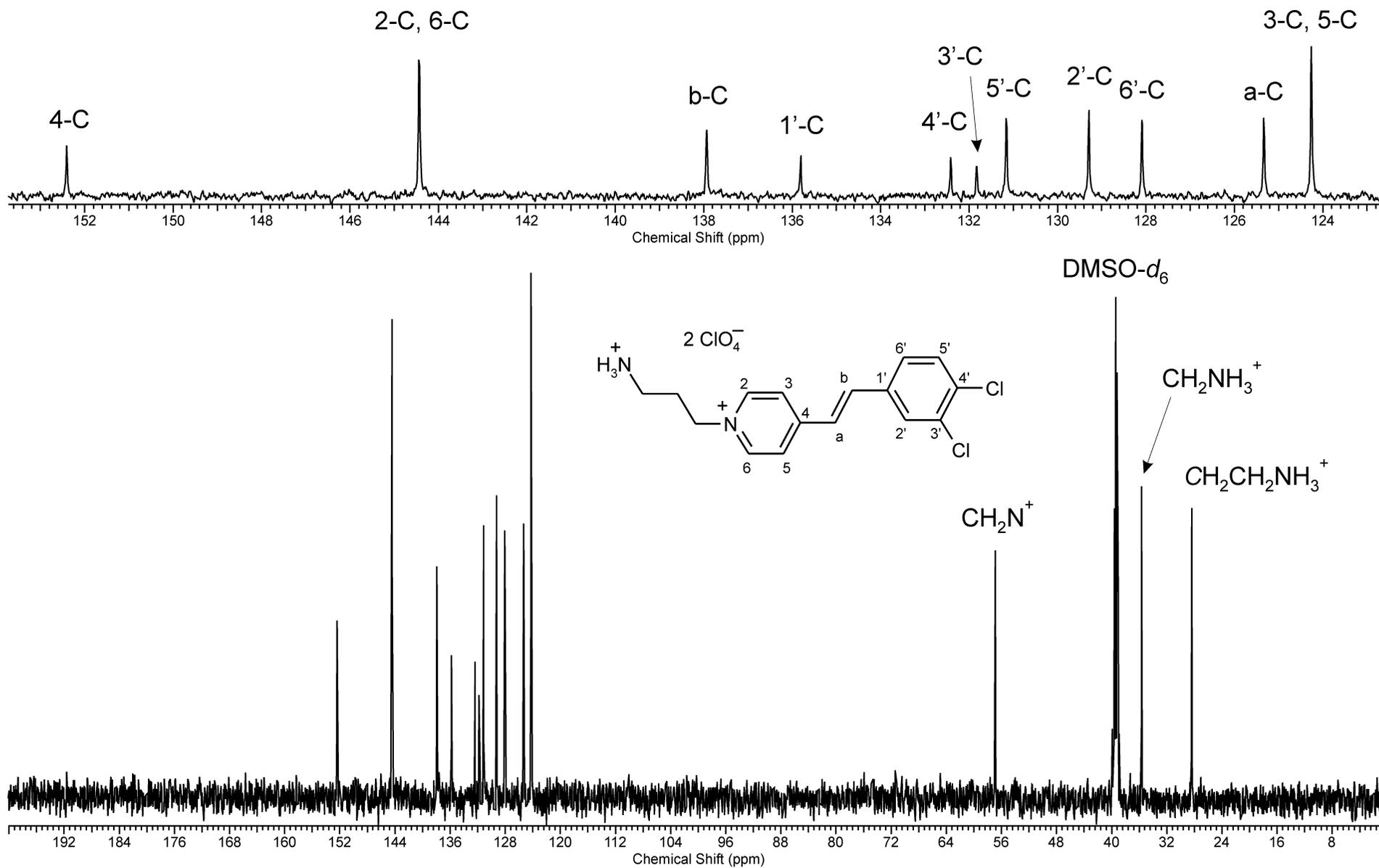


Fig. S17 ^{13}C NMR spectrum of dye **1g** (125.76 MHz, $\text{DMSO-}d_6$, 30 °C).

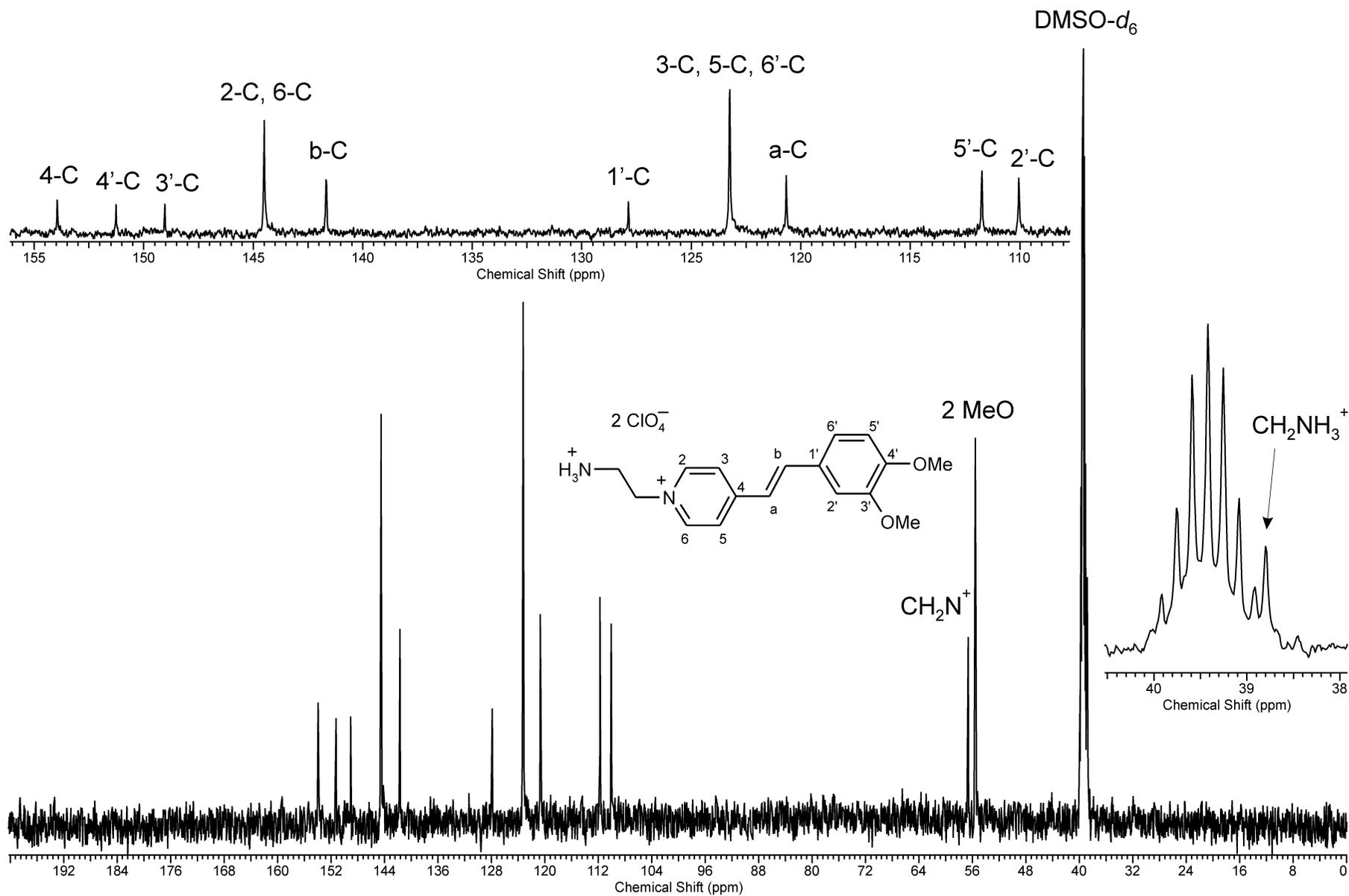


Fig. S18 ^{13}C NMR spectrum of dye **1h** (125.76 MHz, $\text{DMSO-}d_6$, 30 °C).

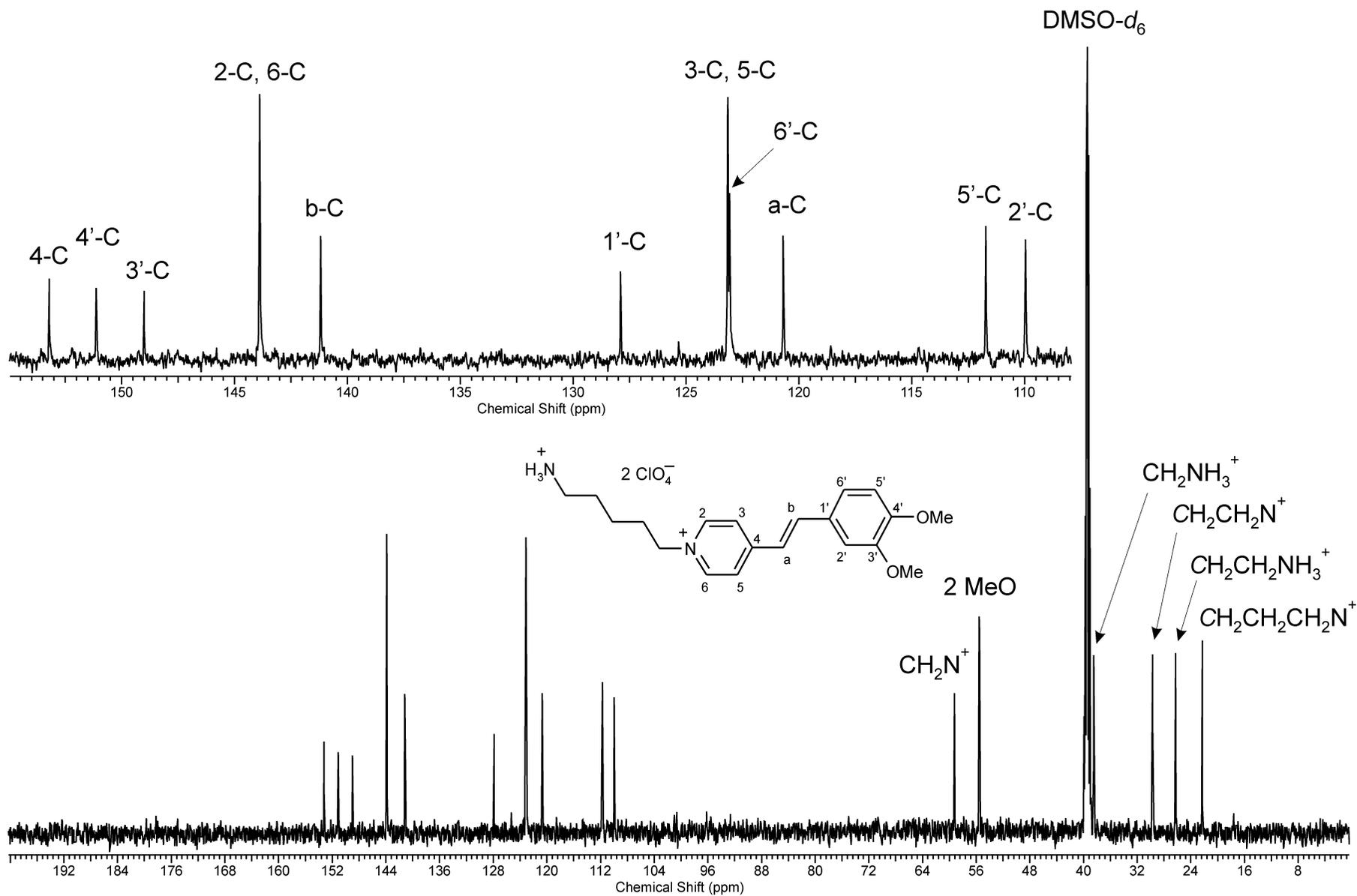


Fig. S19 ^{13}C NMR spectrum of dye **1i** (125.76 MHz, DMSO- d_6 , 30 °C).

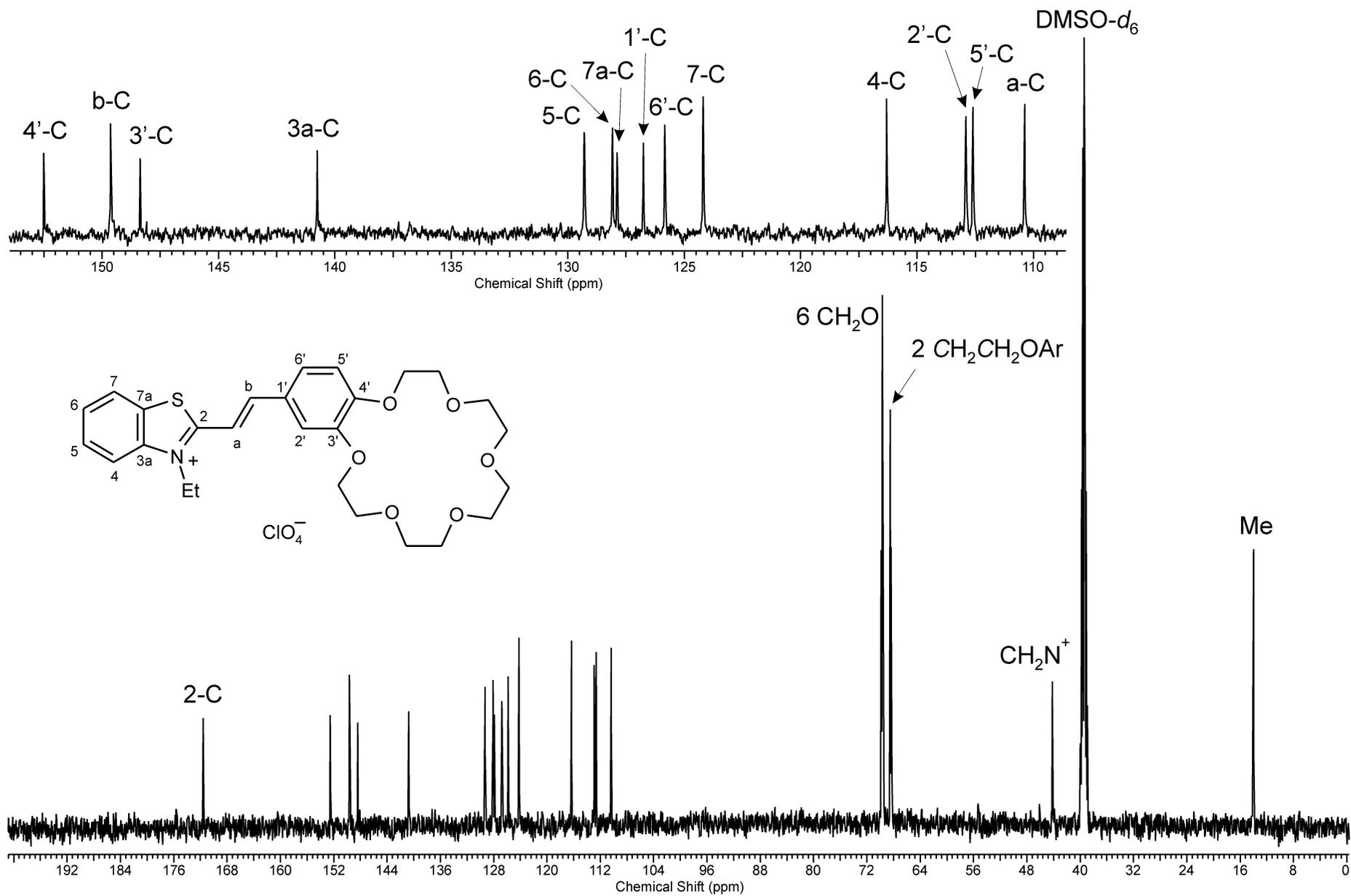


Fig. S20 ^{13}C NMR spectrum of dye **2c** (125.76 MHz, $\text{DMSO-}d_6$, 30 $^\circ\text{C}$).

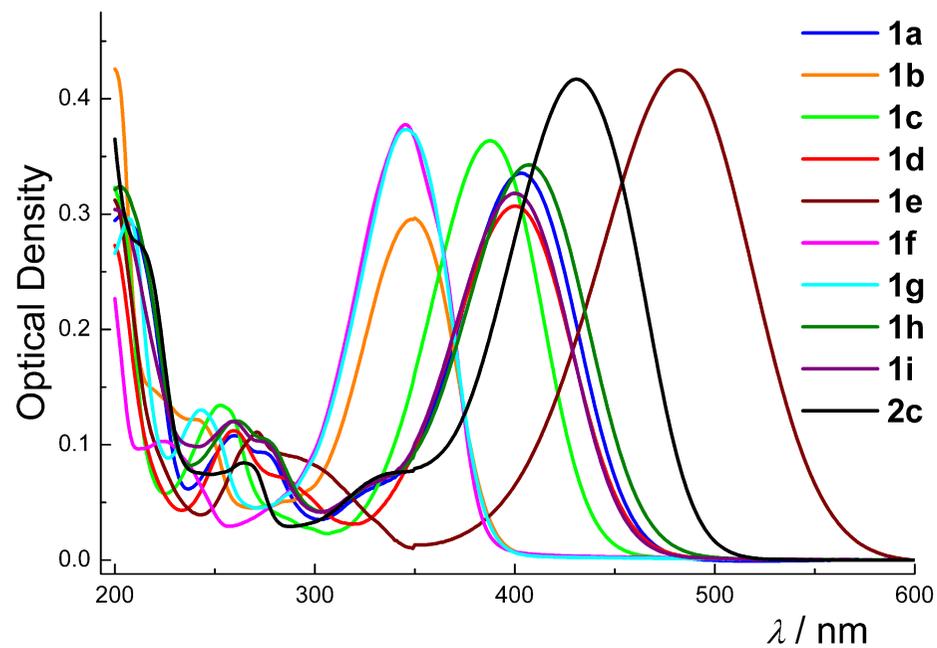


Fig. S21 Absorption spectra of dyes **1a–i** and **2c** (MeCN, $C_{\text{dye}} = 1 \times 10^{-5}$ M, 1-cm quartz cell, ambient temperature).

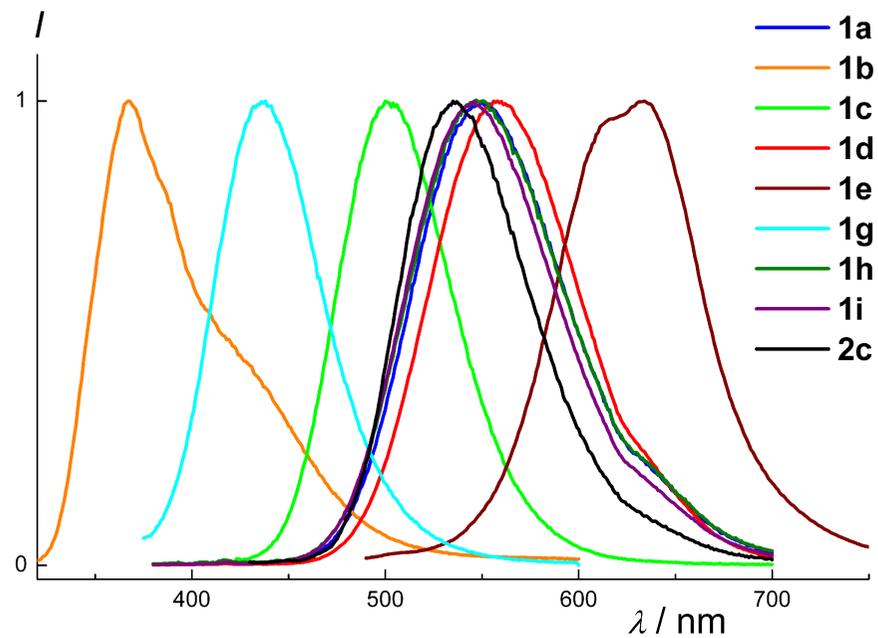


Fig. S22 Normalized emission spectra of dyes **1a–e,g–i** and **2c** (MeCN, $C_{\text{dye}} = 1 \times 10^{-5}$ M, 1-cm quartz cell, ambient temperature). Excitation 310 nm (**1b**), 330 nm (**1g**), 370 nm (**1a,c,d,h,i**), 420 nm (**2c**), and 470 nm (**1e**). Dye **1f** does not fluoresce.

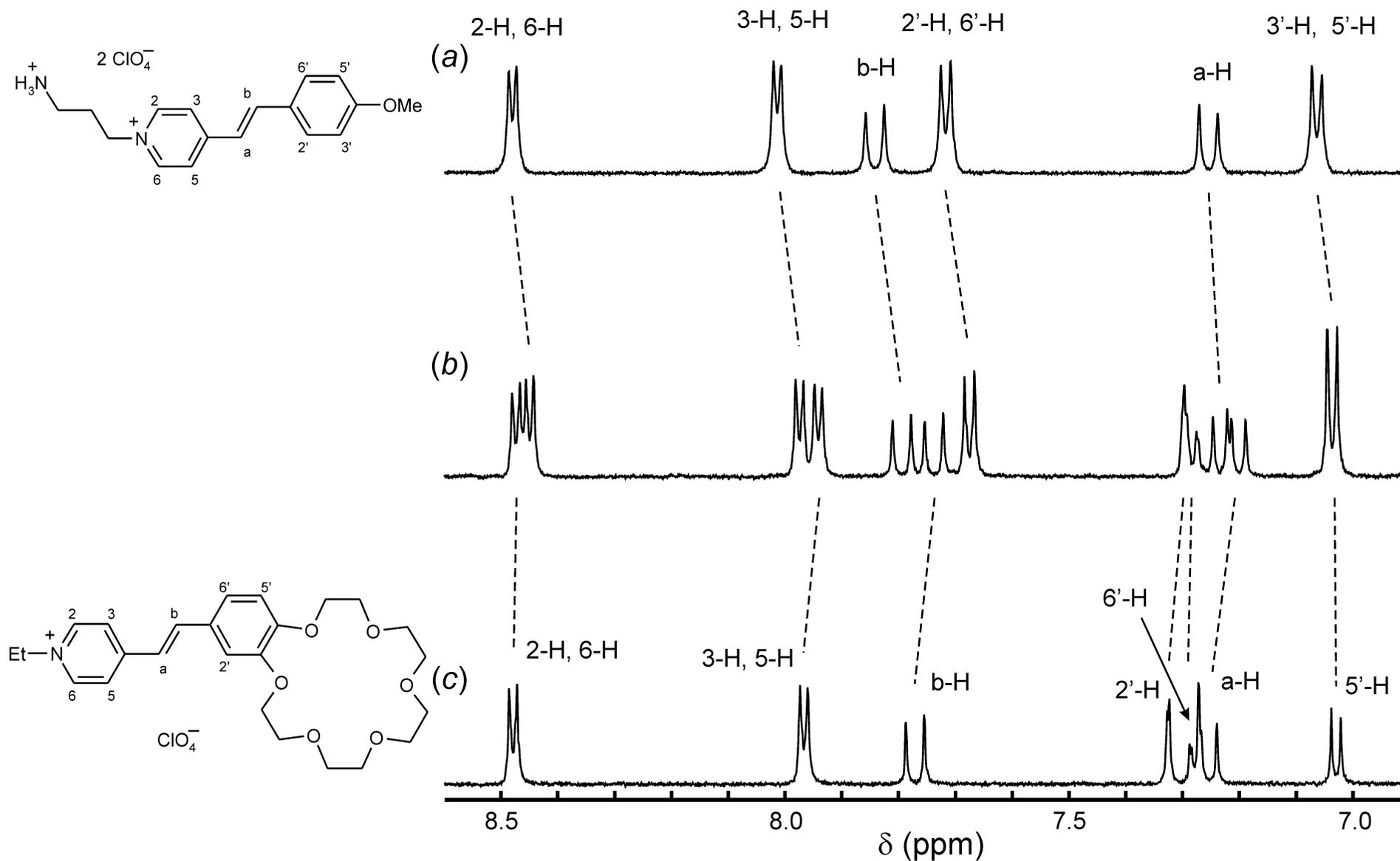


Fig. S23 ¹H NMR spectra (aromatic proton region) of (a) dye **1c**, (b) a 1:1 mixture of dyes **1c** and **2a**, and (c) dye **2a** ($C_{\text{dye}} = 1 \times 10^{-3}$ M) (500.13 MHz, MeCN-*d*₃, 30 °C).

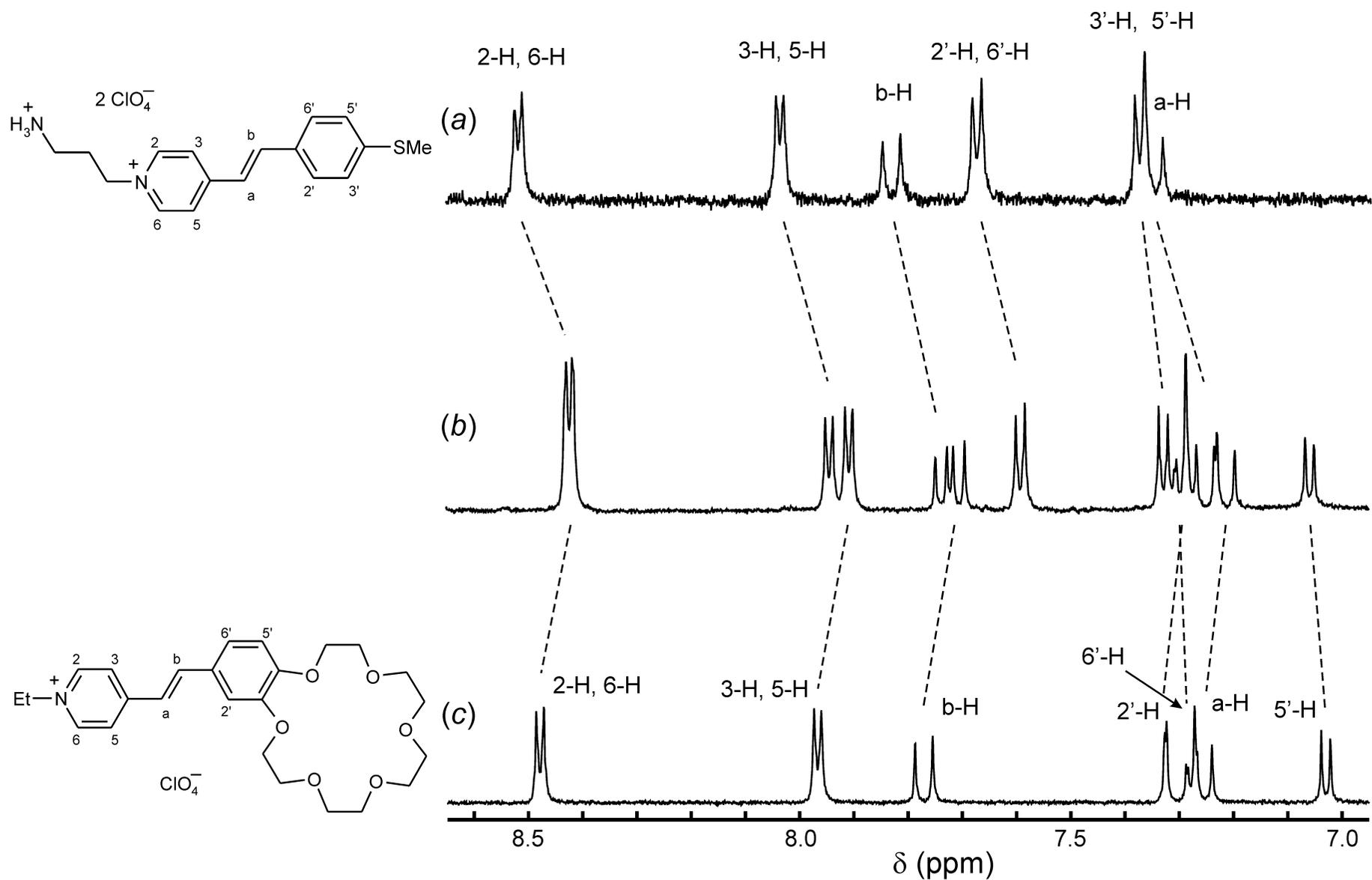


Fig. S24 ^1H NMR spectra (aromatic proton region) of (a) dye **1d**, (b) a 1:1 mixture of dyes **1d** and **2a**, and (c) dye **2a** ($C_{\text{dye}} = 1 \times 10^{-3}$ M) (500.13 MHz, $\text{MeCN-}d_3$, 30 °C).

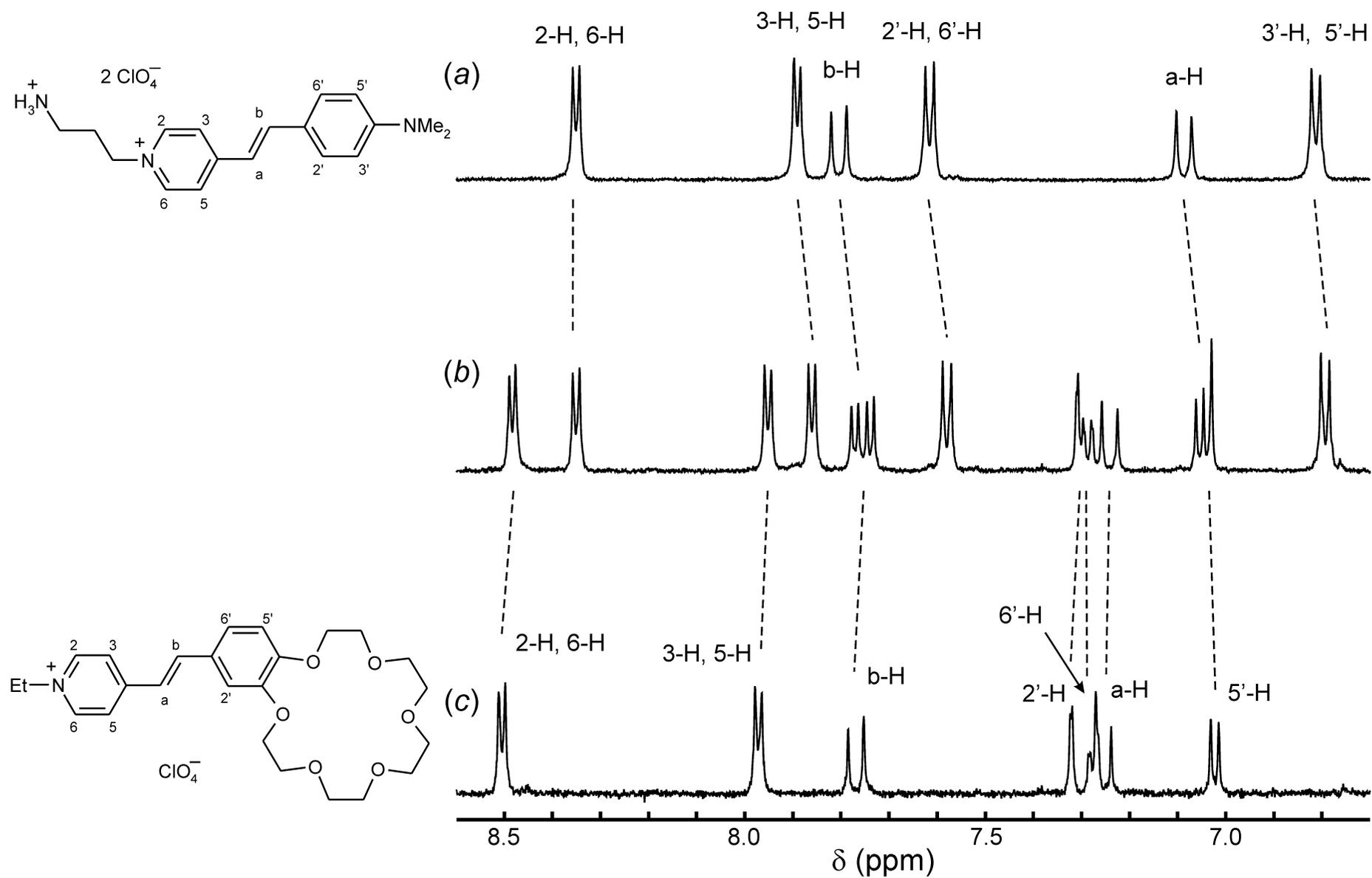


Fig. S25 ^1H NMR spectra (aromatic proton region) of (a) dye **1e**, (b) a 1:1 mixture of dyes **1e** and **2a**, and (c) dye **2a** ($C_{\text{dye}} = 1 \times 10^{-3} \text{ M}$) (500.13 MHz, $\text{MeCN-}d_3$, 30°C).

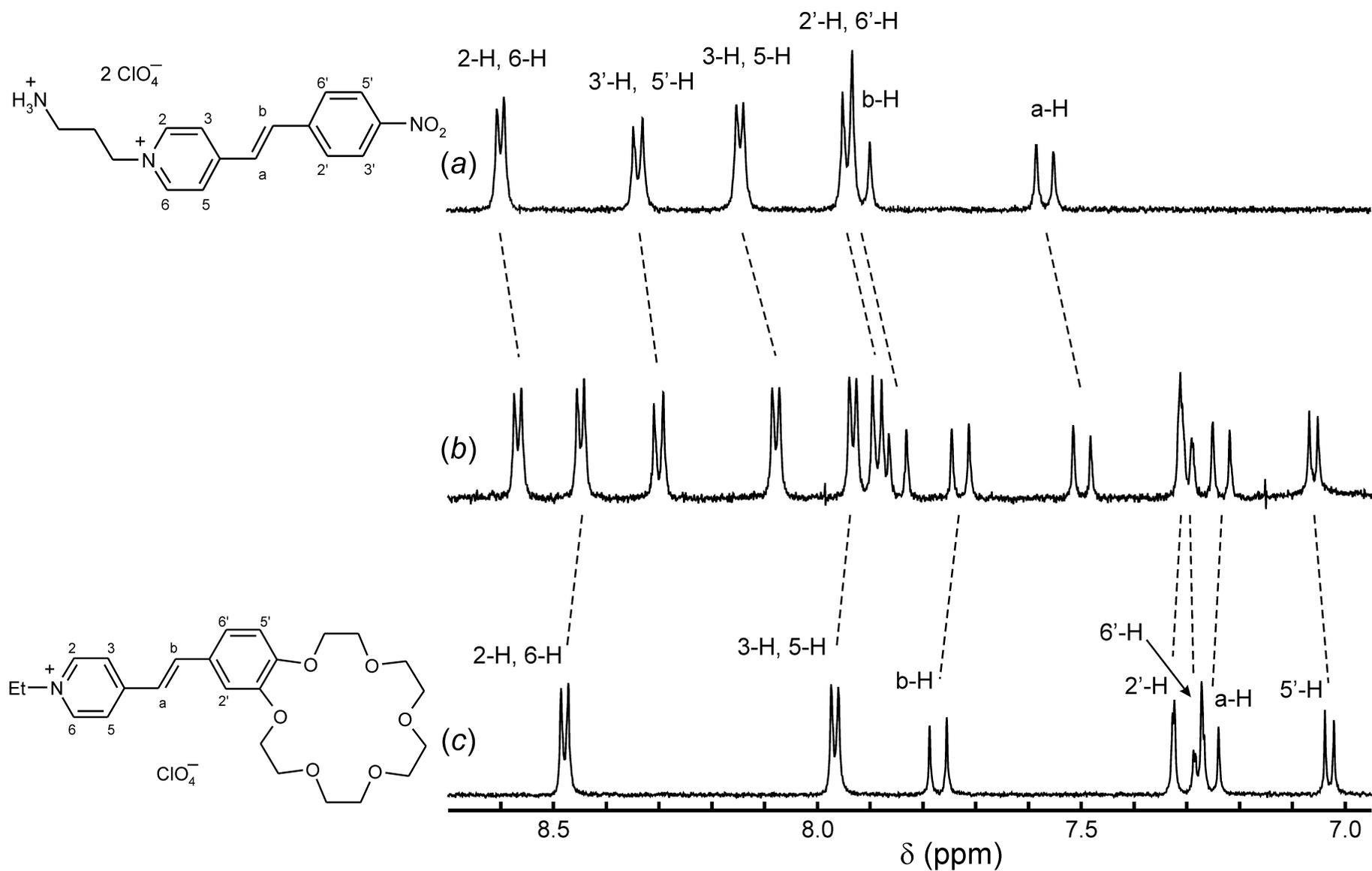


Fig. S26 ^1H NMR spectra (aromatic proton region) of (a) dye **1f**, (b) a 1:1 mixture of dyes **1f** and **2a**, and (c) dye **2a** ($C_{\text{dye}} = 1 \times 10^{-3}$ M) (500.13 MHz, $\text{MeCN-}d_3$, 30 $^\circ\text{C}$).

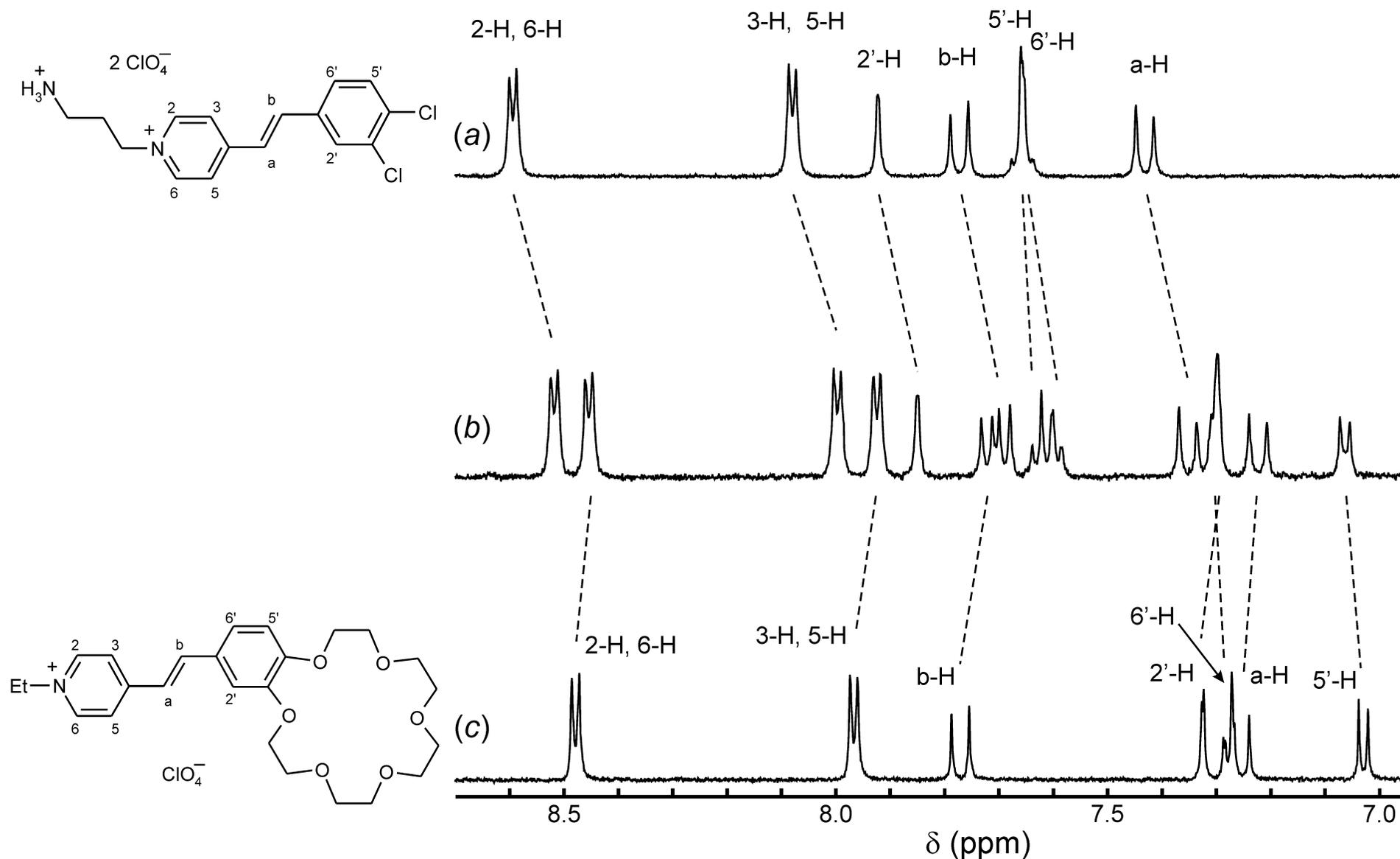


Fig. S27 ^1H NMR spectra (aromatic proton region) of (a) dye **1g**, (b) a 1:1 mixture of dyes **1g** and **2a**, and (c) dye **2a** ($C_{\text{dye}} = 1 \times 10^{-3}$ M) (500.13 MHz, $\text{MeCN-}d_3$, 30 °C).

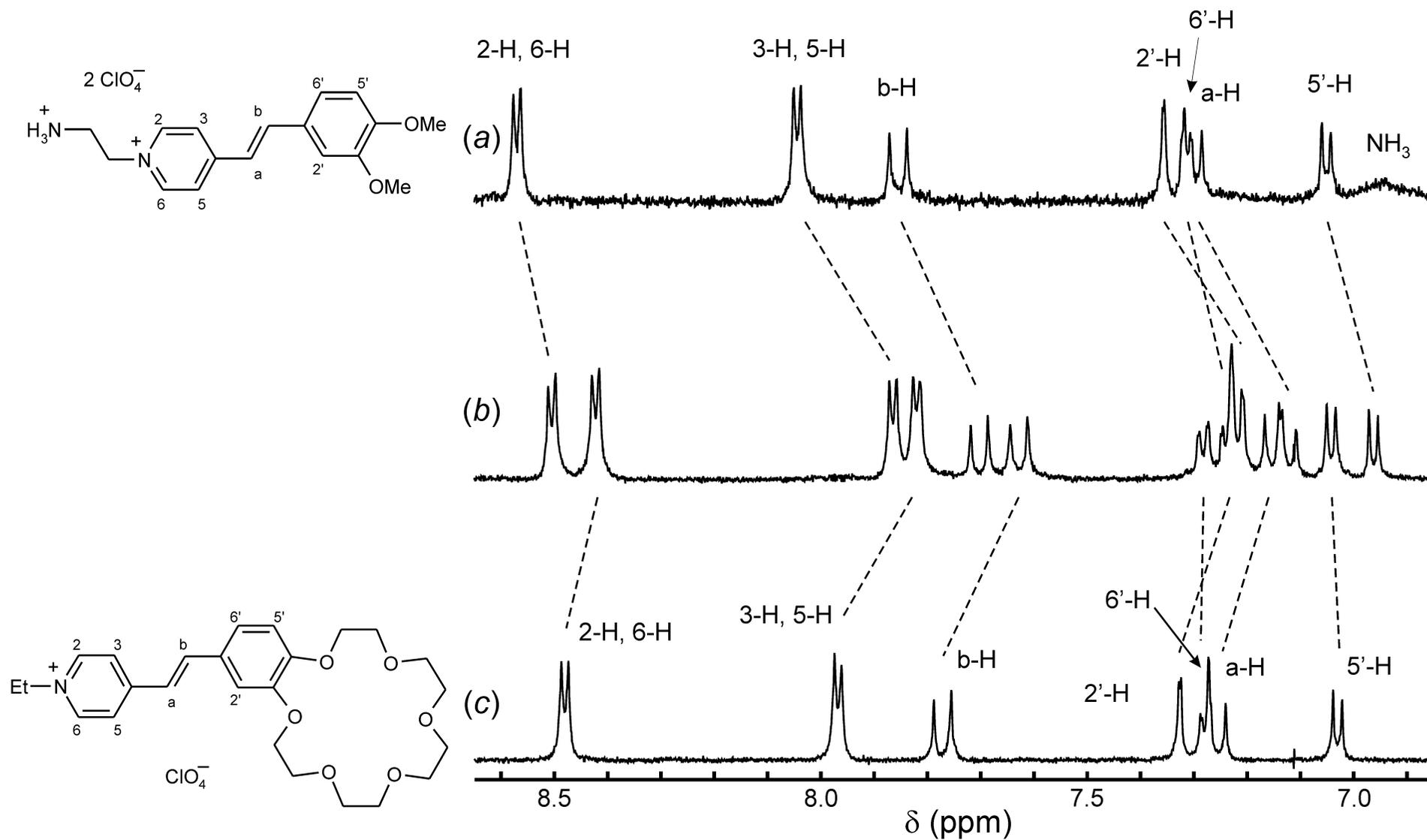


Fig. S28 ^1H NMR spectra (aromatic proton region) of (a) dye **1h**, (b) a 1:1 mixture of dyes **1h** and **2a**, and (c) dye **2a** ($C_{\text{dye}} = 1 \times 10^{-3}$ M) (500.13 MHz, $\text{MeCN-}d_3$, 30 °C).

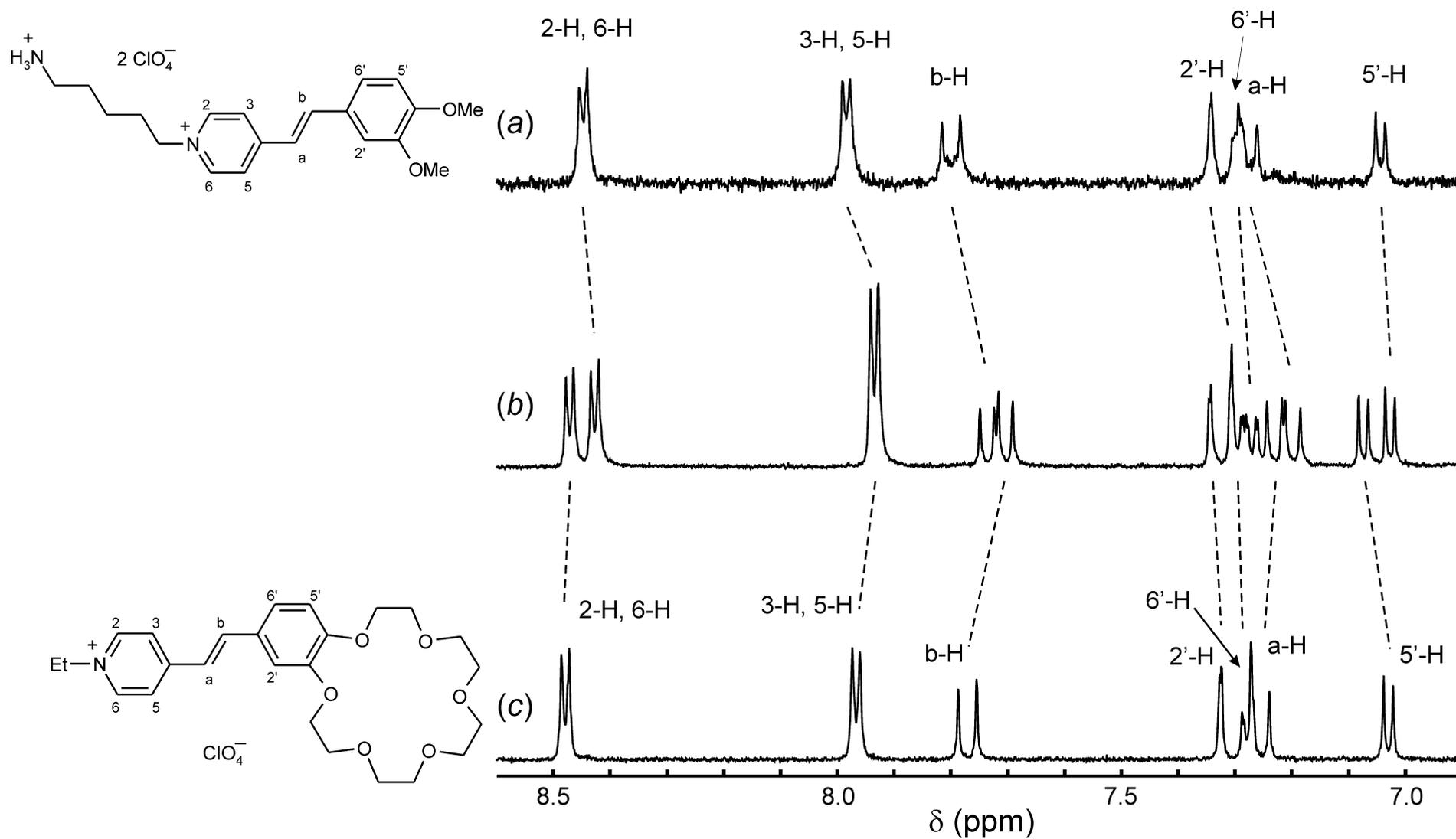


Fig. S29 ^1H NMR spectra (aromatic proton region) of (a) dye **1i**, (b) a 1:1 mixture of dyes **1i** and **2a**, and (c) dye **2a** ($C_{\text{dye}} = 1 \times 10^{-3}$ M) (500.13 MHz, $\text{MeCN-}d_3$, 30 $^\circ\text{C}$).

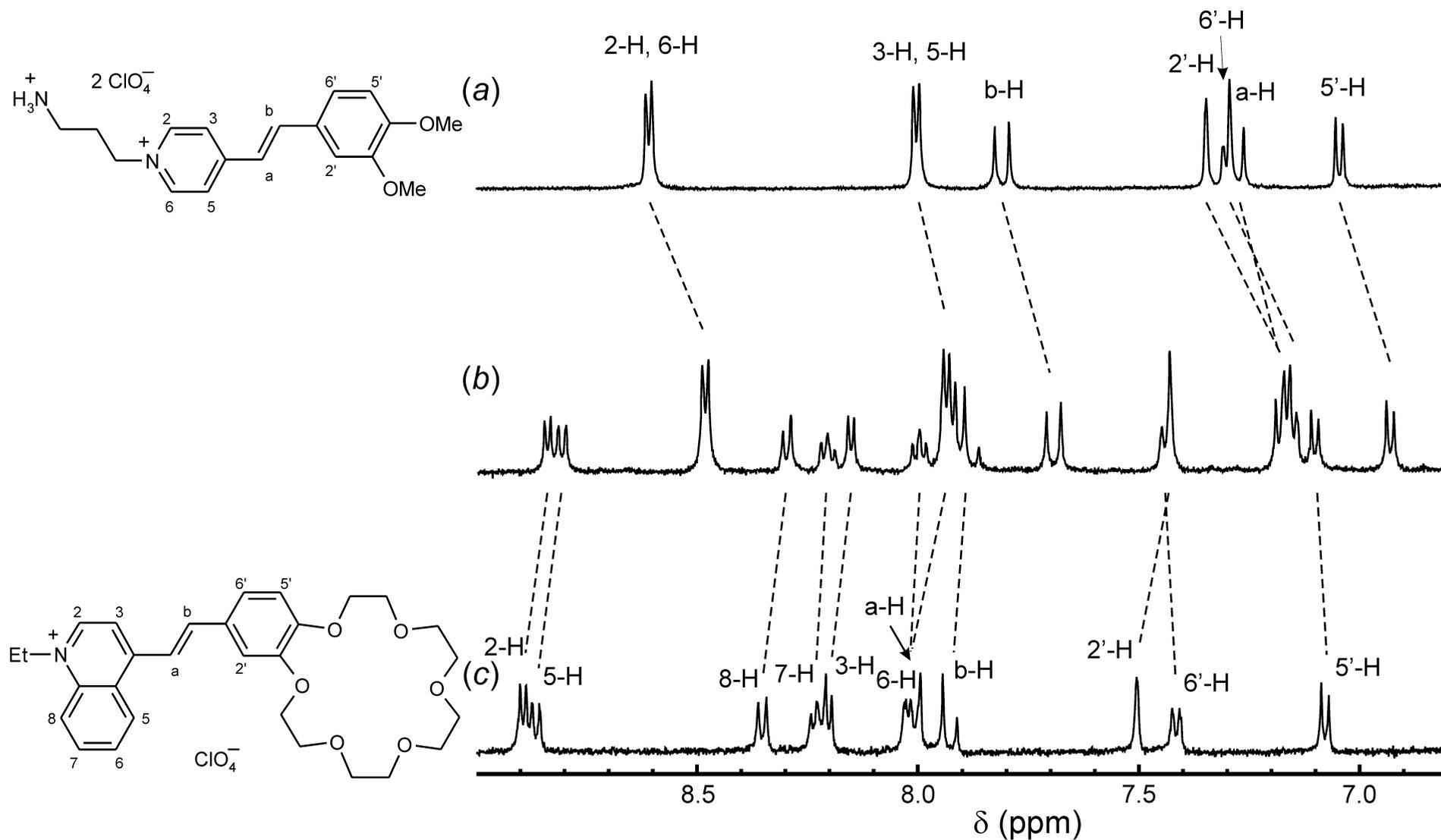


Fig. S30 ^1H NMR spectra (aromatic proton region) of (a) dye **1a**, (b) a 1:1 mixture of dyes **1a** and **2b**, and (c) dye **2b** ($C_{\text{dye}} = 1 \times 10^{-3}$ M) (500.13 MHz, $\text{MeCN-}d_3$, 30 °C).

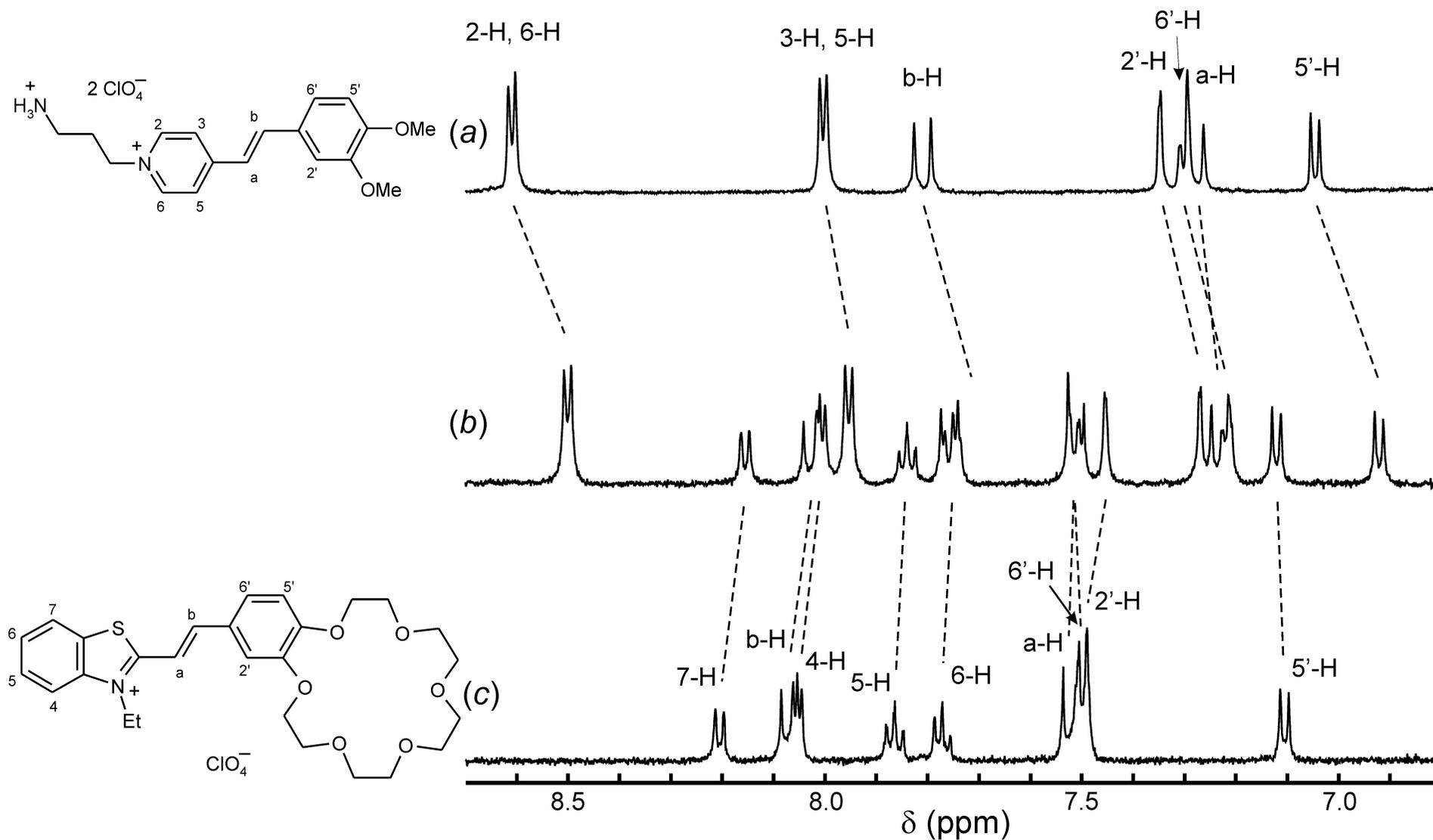


Fig. S31 ^1H NMR spectra (aromatic proton region) of (a) dye **1a**, (b) a 1:1 mixture of dyes **1a** and **2c**, and (c) dye **2c** ($C_{\text{dye}} = 1 \times 10^{-3}$ M) (500.13 MHz, $\text{MeCN-}d_3$, 30 $^\circ\text{C}$).

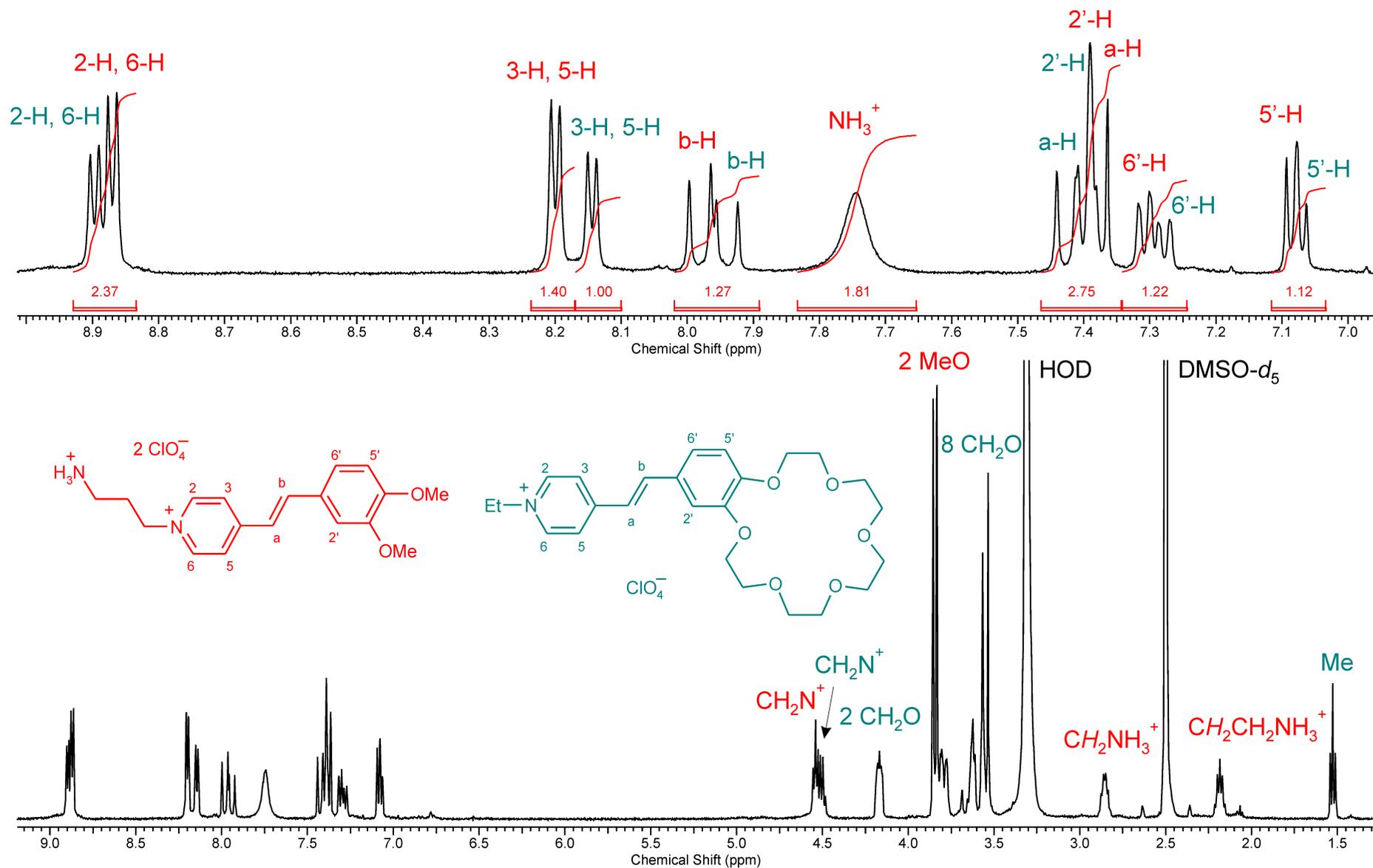


Fig. S32 ¹H NMR spectrum of complex (1a)_{1.5}·2a, which was obtained by crystallization (500.13 MHz, DMSO-d₆, 30 °C). In DMSO-d₆, the complex is destroyed to form a mixture of free dyes 1a and 2a.

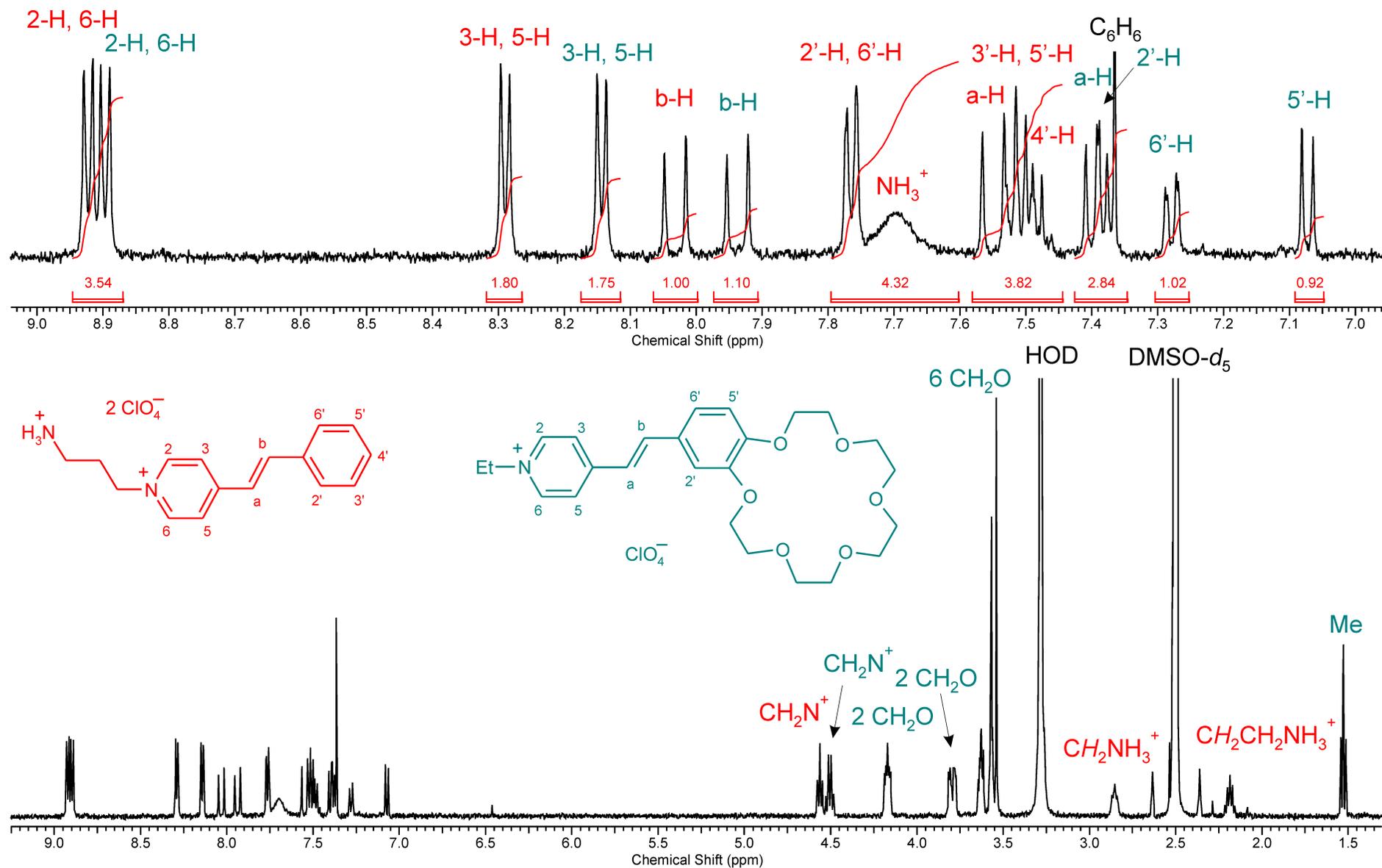


Fig. S33 ^1H NMR spectrum of complex **1b-2a**, which was obtained by crystallization (500.13 MHz, $\text{DMSO-}d_6$, 30 °C). In $\text{DMSO-}d_6$, the complex is destroyed to form a mixture of free dyes **1b** and **2a**.

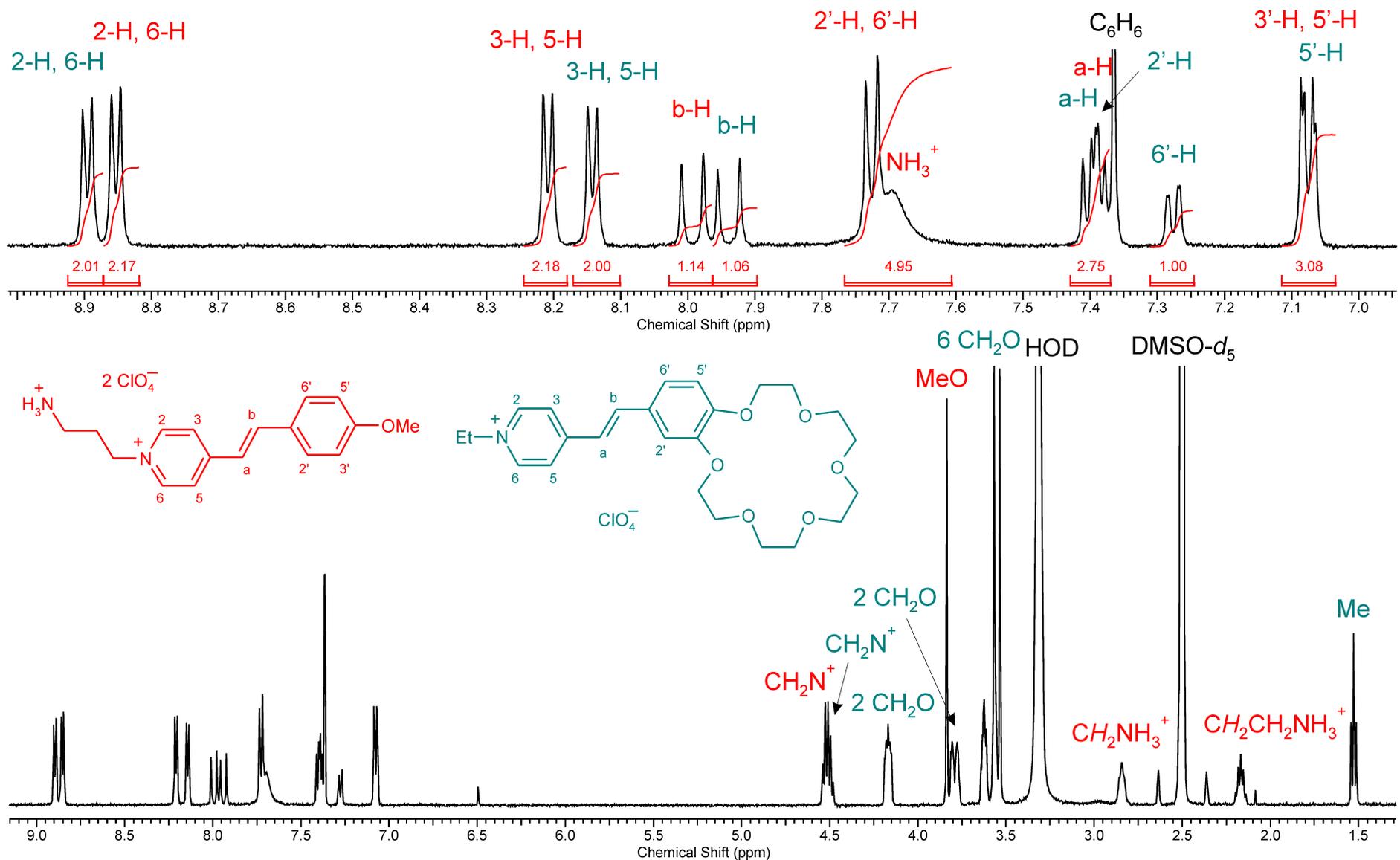


Fig. S34 ¹H NMR spectrum of complex **1c·2a**, which was obtained by crystallization (500.13 MHz, DMSO-*d*₆, 29 °C). In DMSO-*d*₆, the complex is destroyed to form a mixture of free dyes **1c** and **2a**.

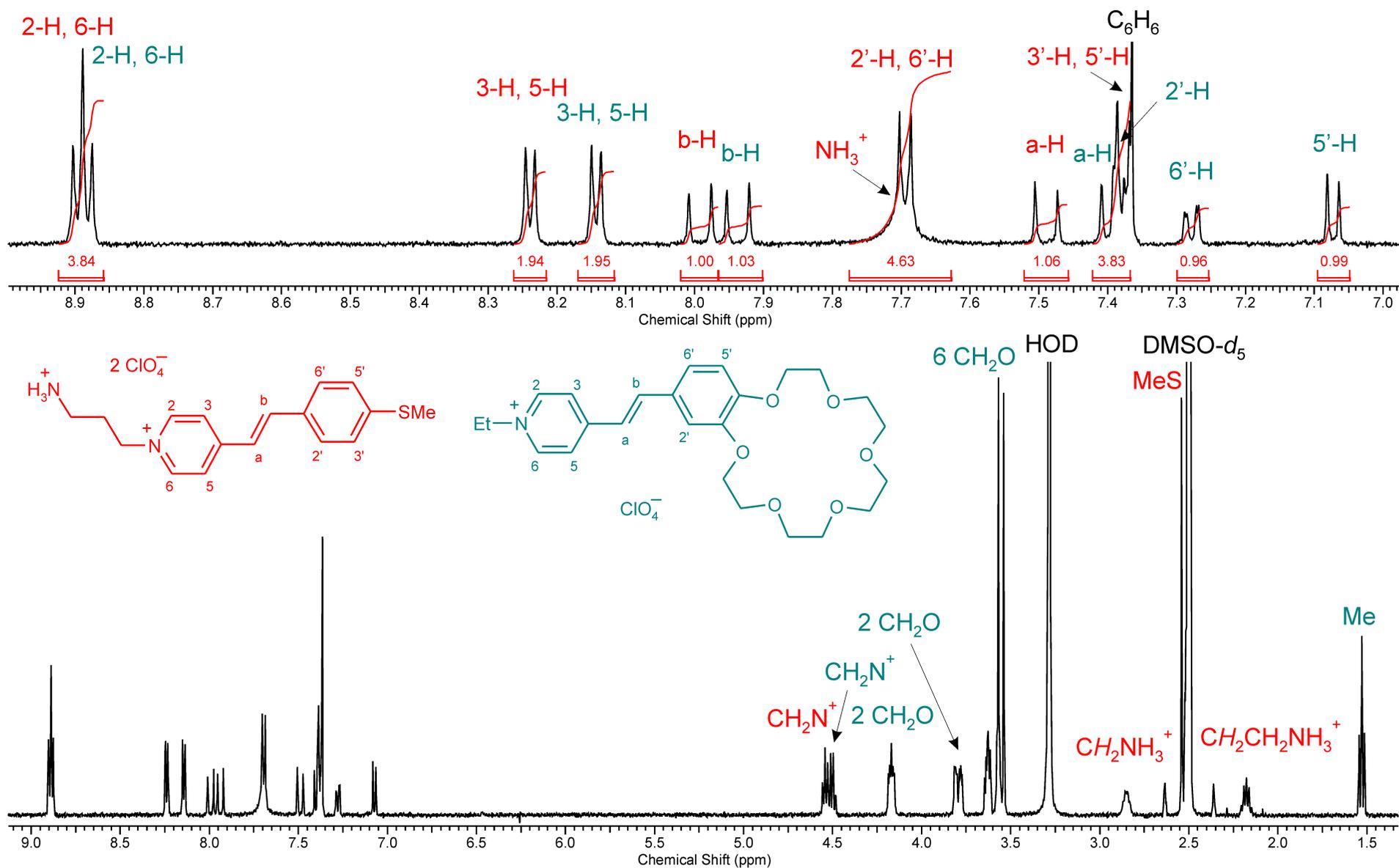


Fig. S35 ¹H NMR spectrum of complex **1d-2a**, which was obtained by crystallization (500.13 MHz, DMSO-*d*₆, 30 °C). In DMSO-*d*₆, the complex is destroyed to form a mixture of free dyes **1d** and **2a**.

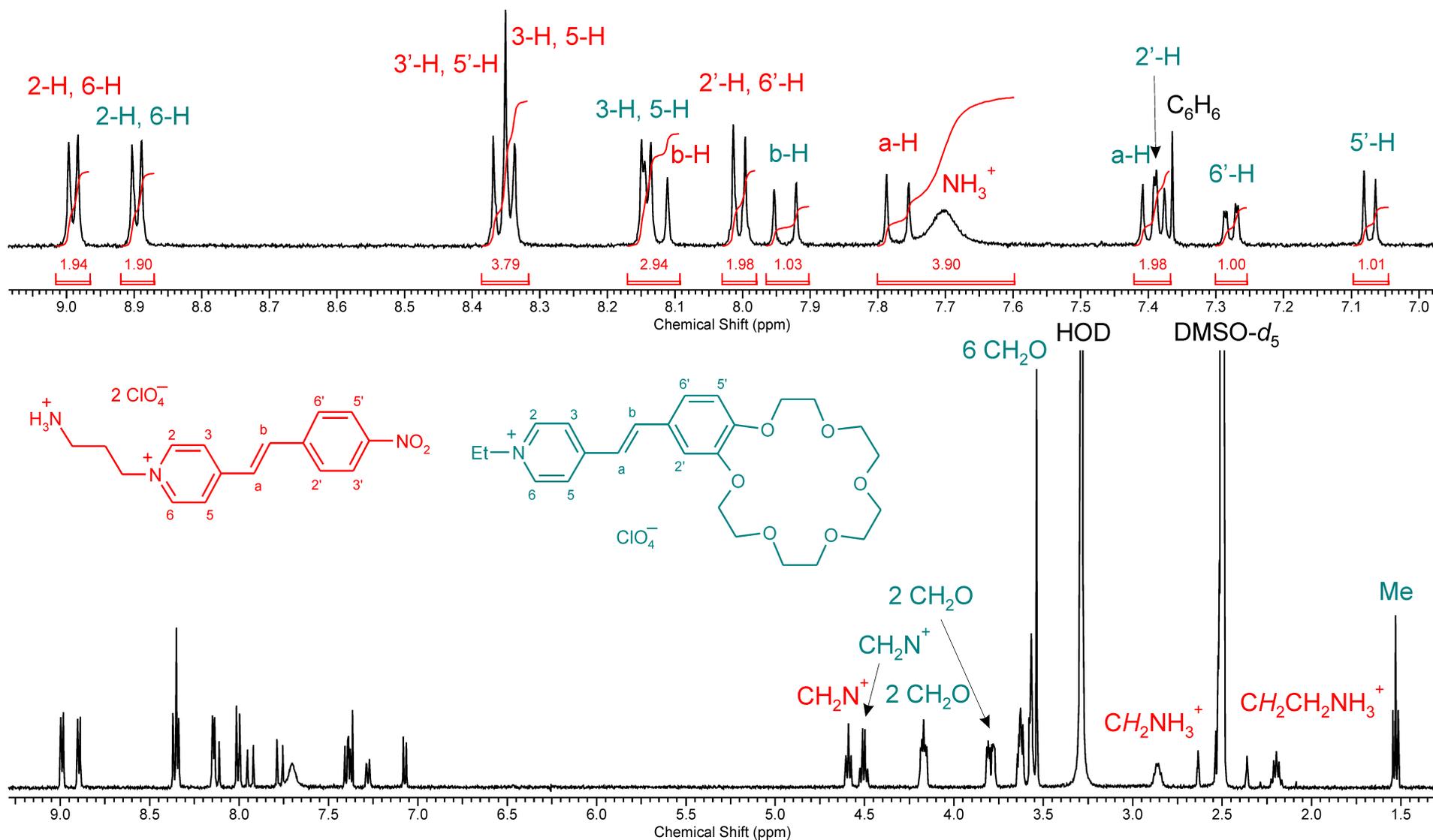


Fig. S36 ^1H NMR spectrum of complex **1f·2a**, which was obtained by crystallization (500.13 MHz, $\text{DMSO-}d_6$, 30 °C). In $\text{DMSO-}d_6$, the complex is destroyed to form a mixture of free dyes **1f** and **2a**.

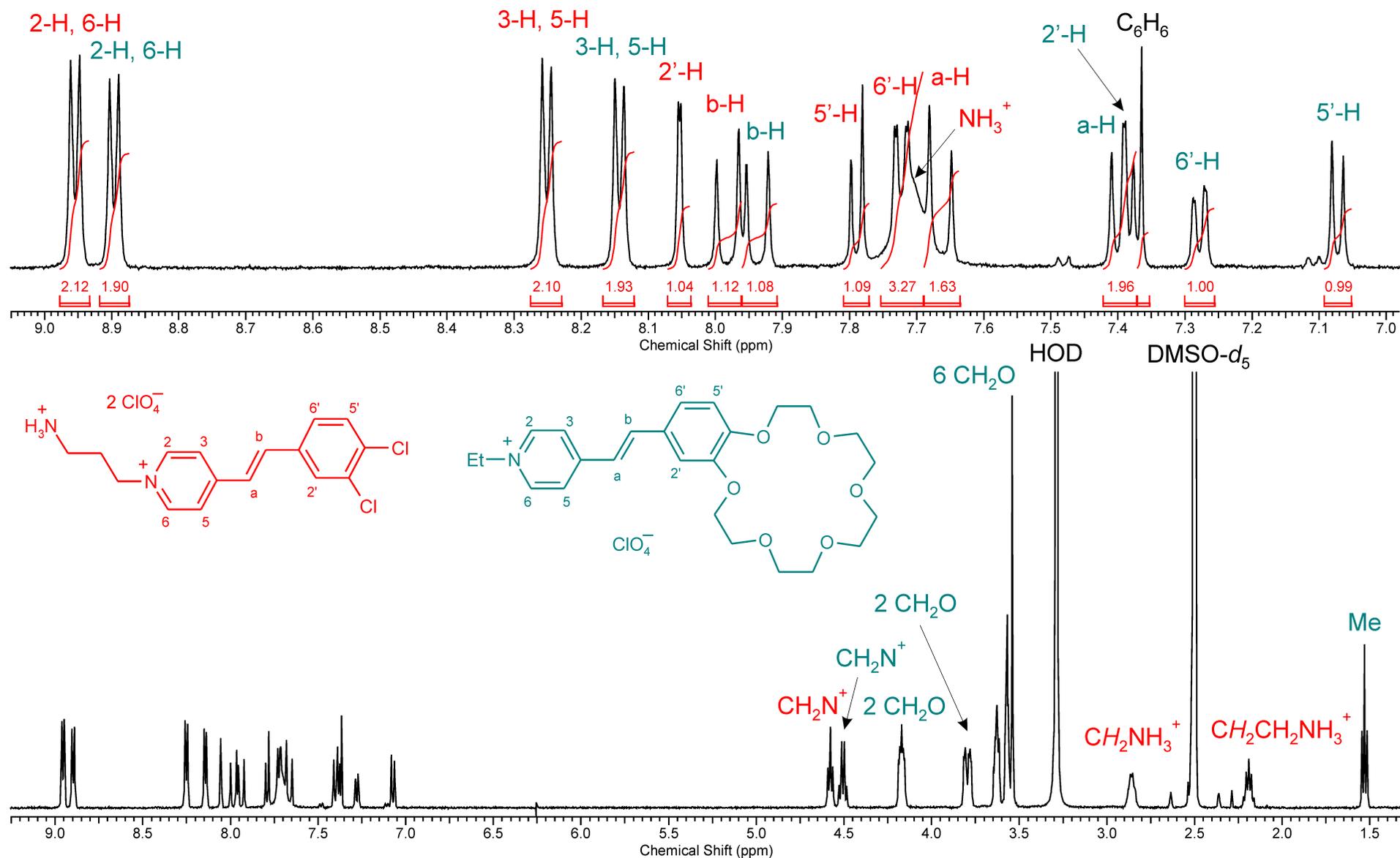


Fig. S37 ¹H NMR spectrum of complex **1g·2a**, which was obtained by crystallization (500.13 MHz, DMSO-*d*₆, 30 °C). In DMSO-*d*₆, the complex is destroyed to form a mixture of free dyes **1g** and **2a**.

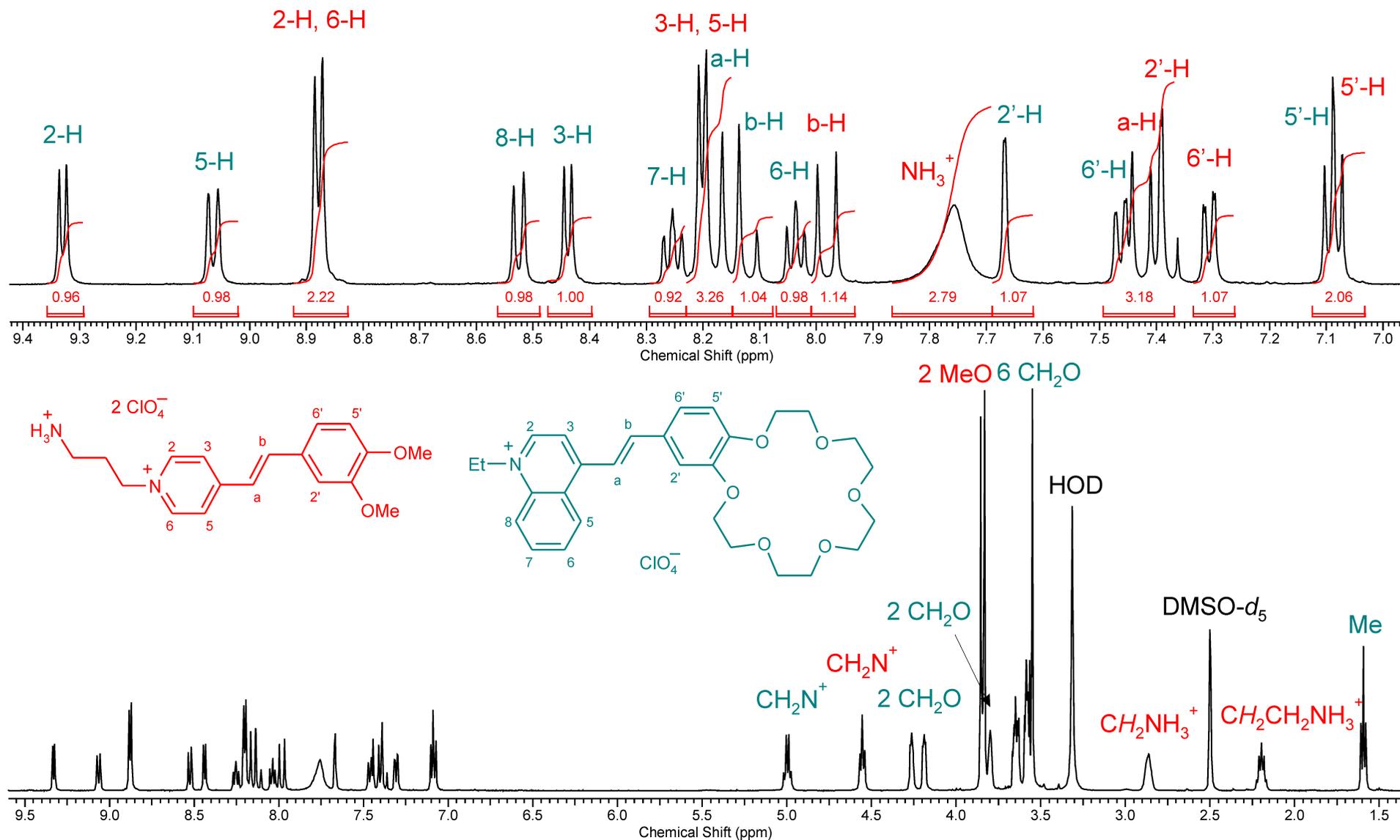


Fig. S38 ¹H NMR spectrum of complex **1a-2b**, which was obtained by crystallization (500.13 MHz, DMSO-*d*₆, 30 °C). In DMSO-*d*₆, the complex is destroyed to form a mixture of free dyes **1a** and **2b**.

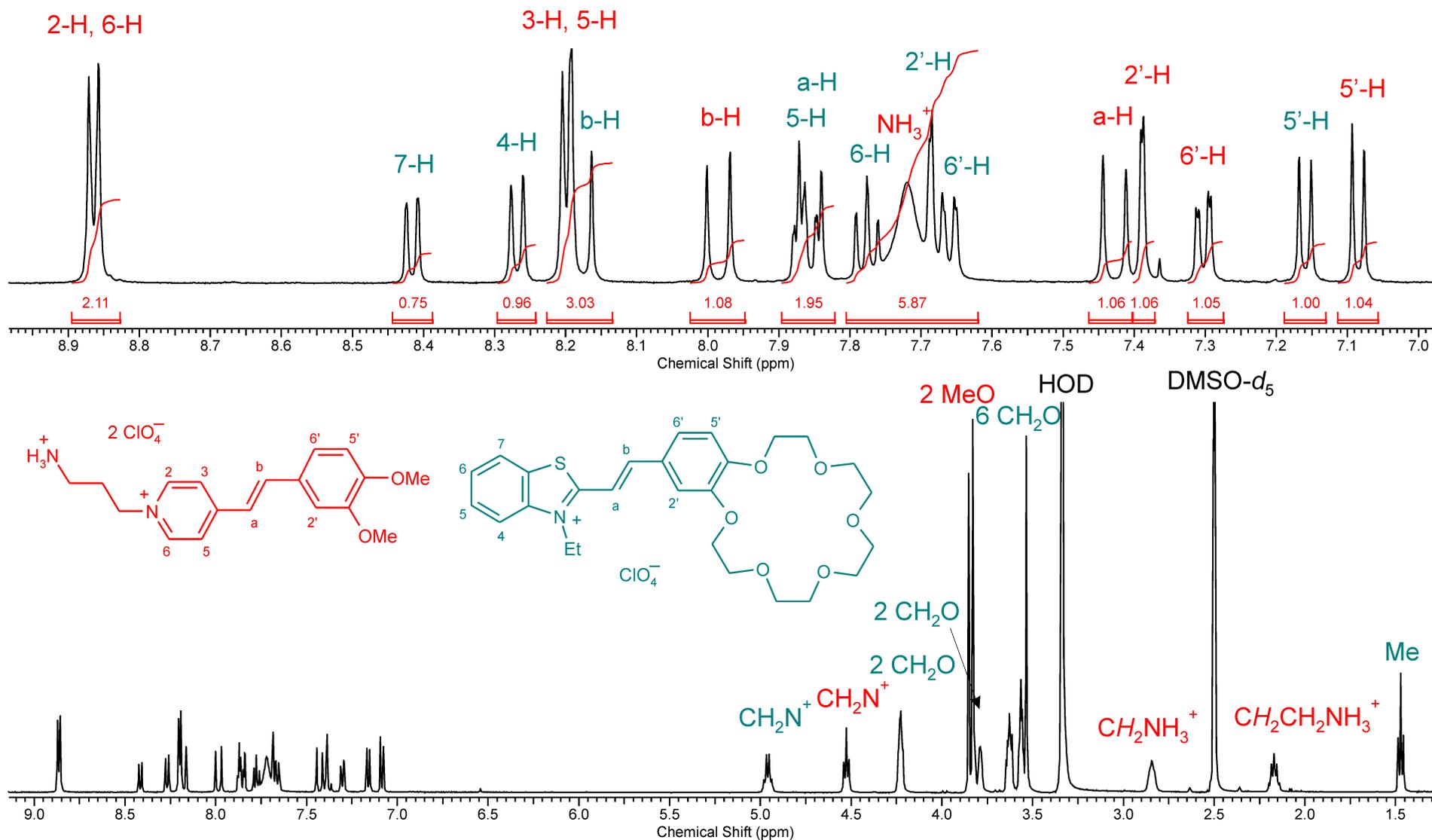


Fig. S39 ¹H NMR spectrum of complex **1a-2c**, which was obtained by crystallization (500.13 MHz, DMSO-*d*₆, 30 °C). In DMSO-*d*₆, the complex is destroyed to form a mixture of free dyes **1a** and **2c**.

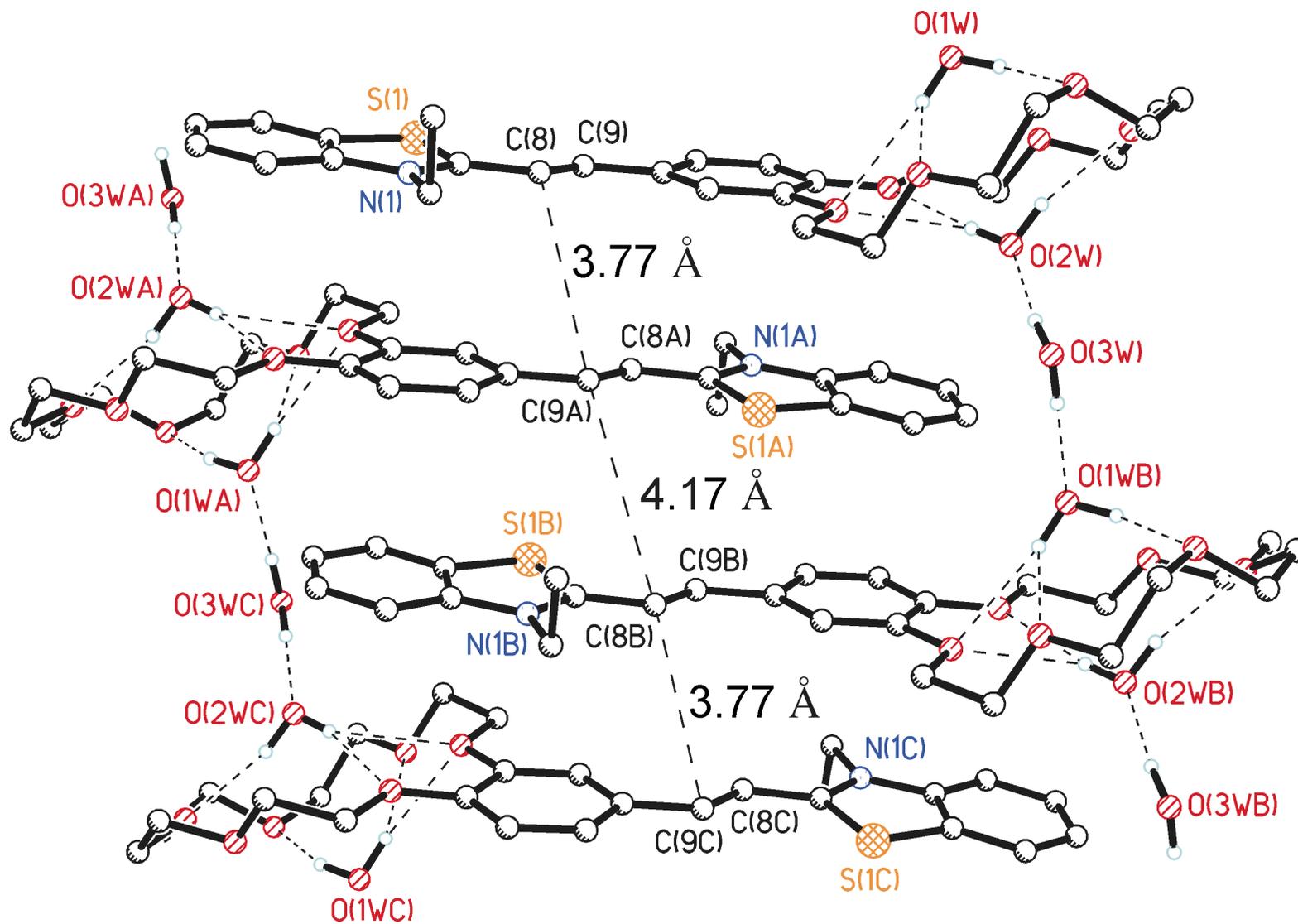


Fig. S40 Head-to-tail stack of dye cations in structure $2c \cdot C_4H_8O_2 \cdot 3H_2O$.

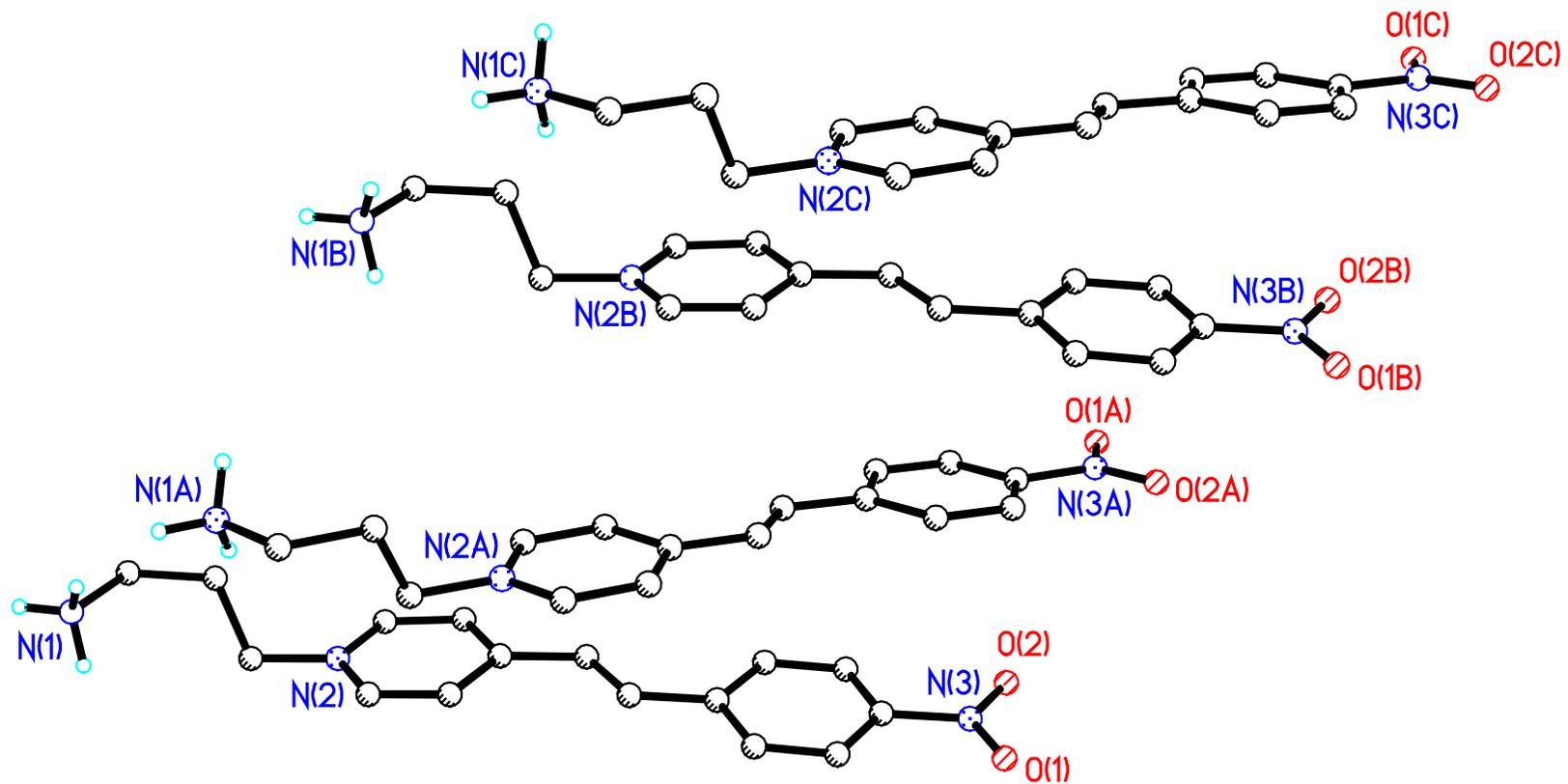


Fig. S41 Head-to-head stack of dye cations in structure **1f**·0.25H₂O.

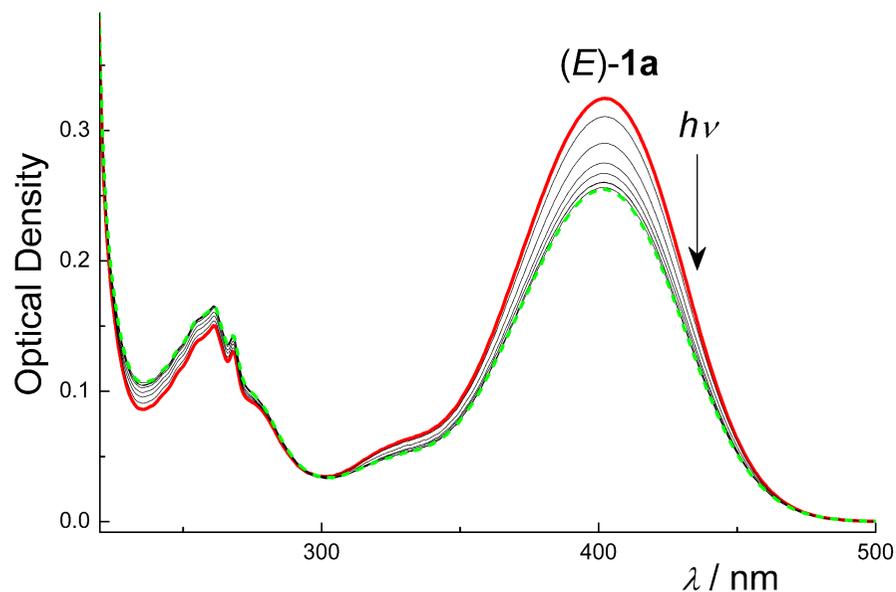


Fig. S42 Photolysis of solution of dye **1a** for 0, 1, 3, 6, 10, 20, 30, and 50 min (MeCN, $C_{1a} = 9.66 \times 10^{-5}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **1a**.

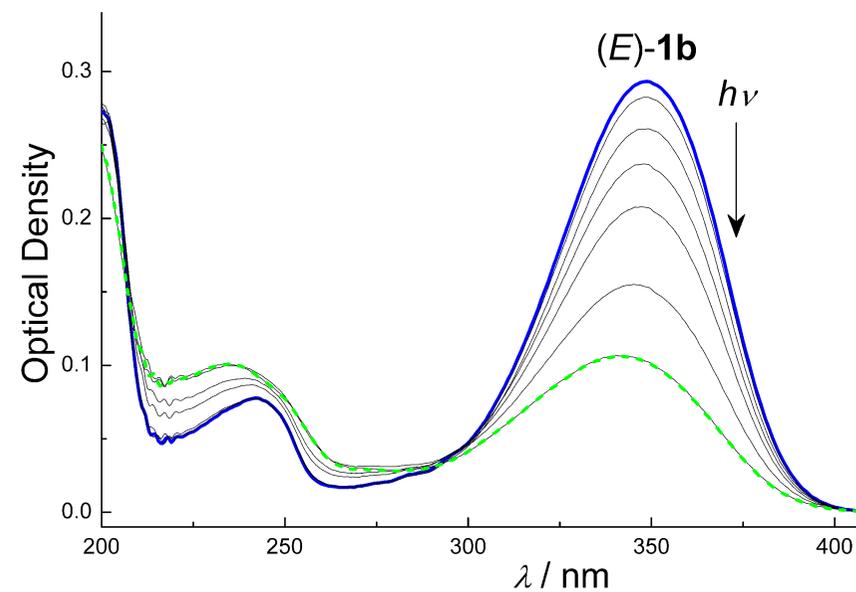


Fig. S43 Photolysis of solution of dye **1b** for 0, 1, 3, 6, 10, 20, and 80 min (MeCN, $C_{1b} = 9.86 \times 10^{-5}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **1b**.

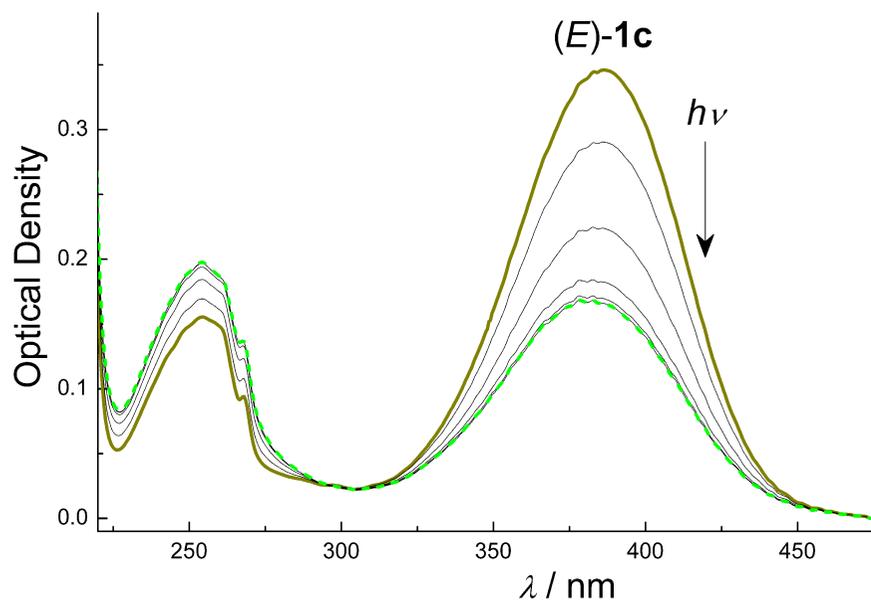


Fig. S44 Photolysis of solution of dye **1c** for 0, 1, 3, 6, 10, and 20 min (MeCN, $C_{1c} = 9.50 \times 10^{-5}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **1c**.

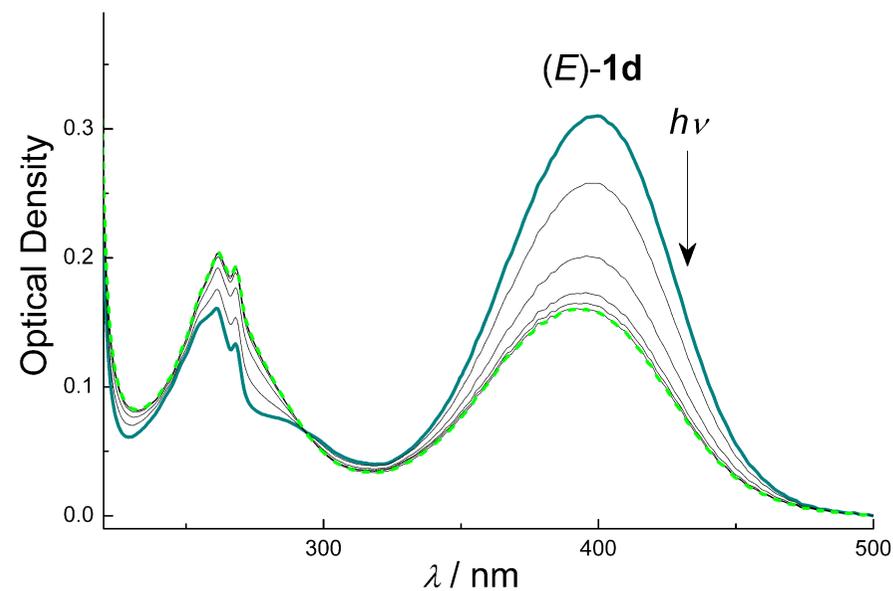


Fig. S45 Photolysis of solution of dye **1d** for 0, 1, 3, 6, 10, and 20 min (MeCN, $C_{1d} = 1.01 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **1d**.

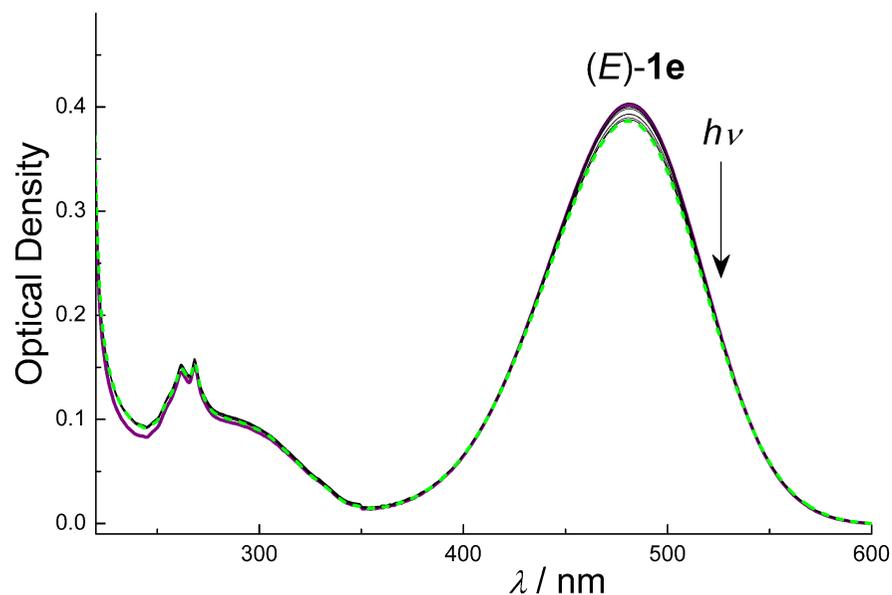


Fig. S46 Photolysis of solution of dye **1e** for 0, 1, 3, 6, 10, 20, 30, 50, 80, and 110 min (MeCN, $C_{1e} = 1.03 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **1e**.

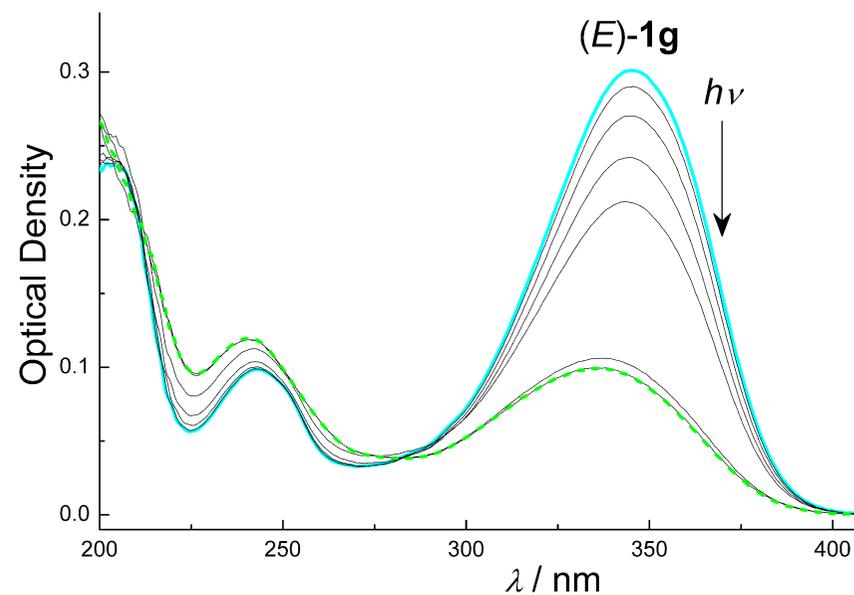


Fig. S47 Photolysis of solution of dye **1g** for 0, 1, 3, 6, 10, 50, and 80 min (MeCN, $C_{1g} = 8.05 \times 10^{-5}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **1g**.

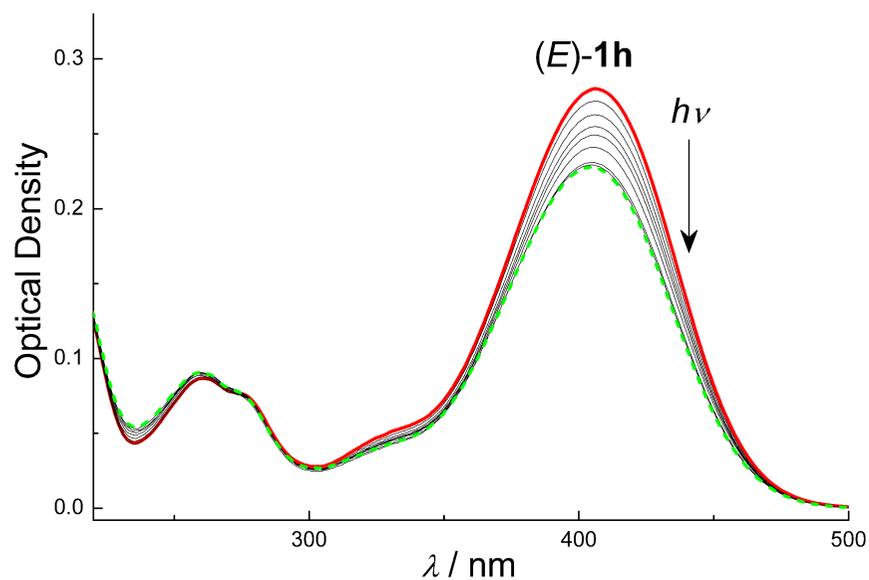


Fig. S48 Photolysis of solution of dye **1h** for 0, 1, 3, 6, 10, 20, 50, and 80 min (MeCN, $C_{1h} = 8.16 \times 10^{-5}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **1h**.

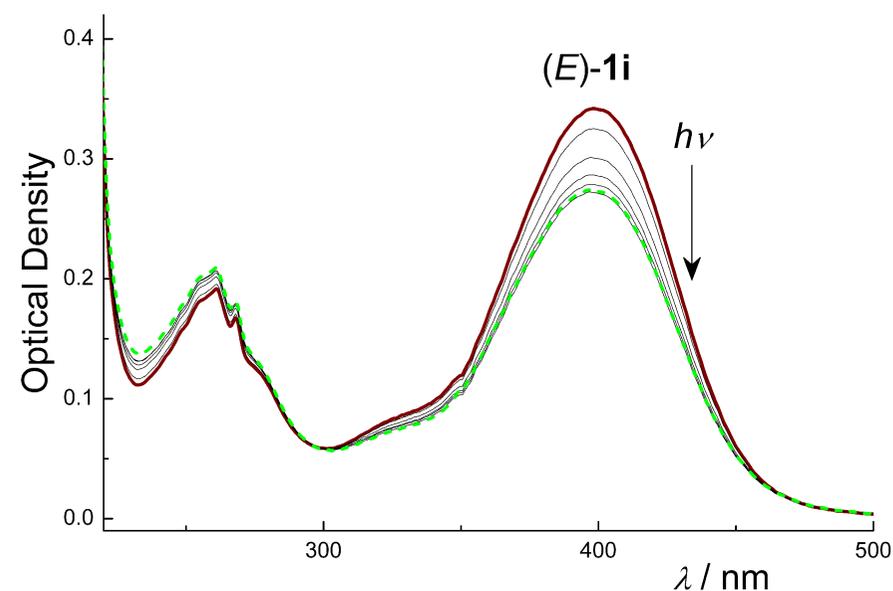


Fig. S49 Photolysis of solution of dye **1i** for 0, 1, 3, 6, 10, and 20 min (MeCN, $C_{1i} = 1.07 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **1i**.

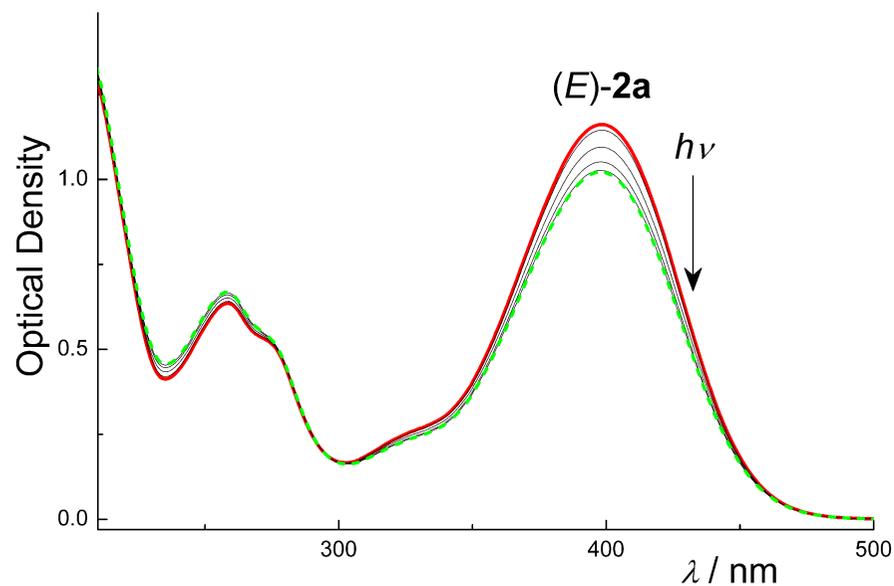


Fig. S50 Photolysis of solution of dye **2a** for 0, 1, 4, 9, and 20 min (MeCN, $C_{2a} = 3.75 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm). The green dash curve is a photostationary mixture consisting of *E* and *Z* isomers of **2a**.

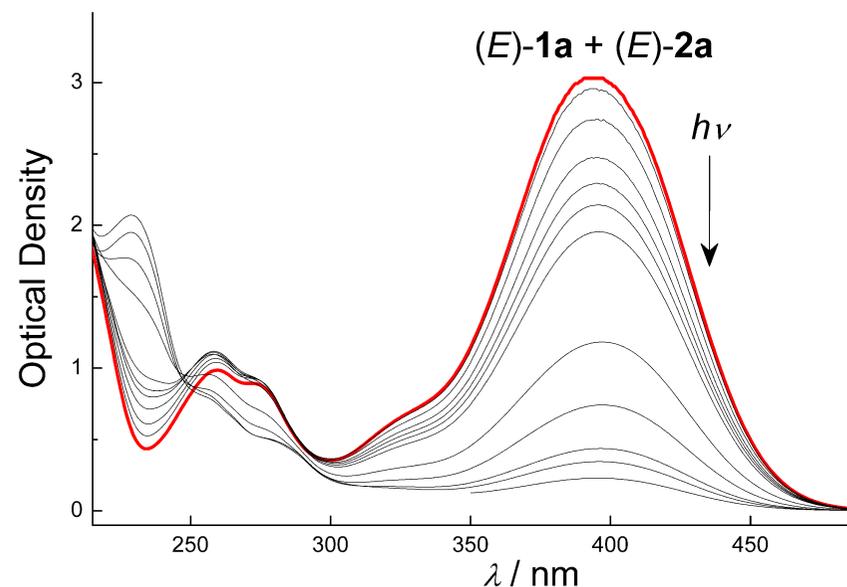


Fig. S51 Photolysis of solution of an equimolar mixture of dyes **1a** and **2a** for 0, 2, 5, 10, 15, 20, and 30 min and 2, 4, 8, 13, and 36 h (MeCN, $C_{1a} = C_{2a} = 5 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm).

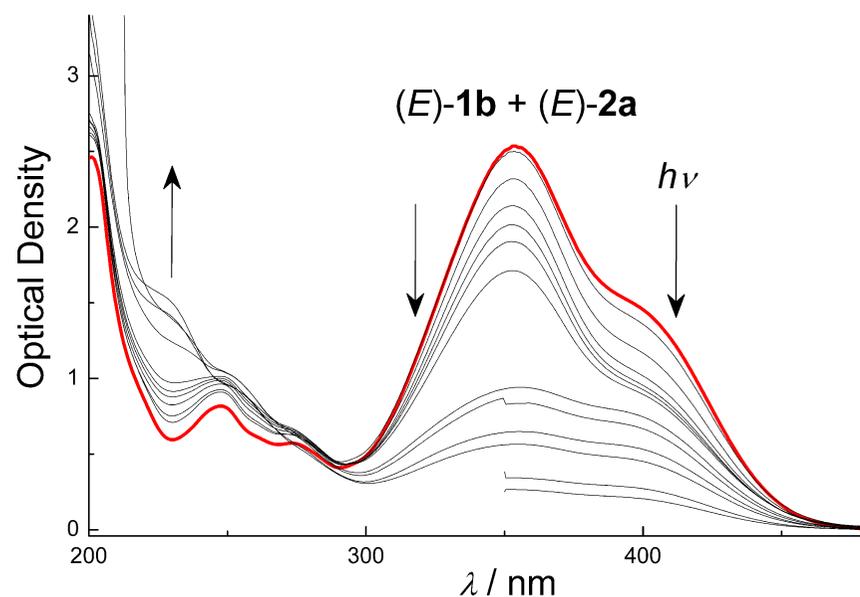


Fig. S52 Photolysis of solution of an equimolar mixture of dyes **1b** and **2a** for 0, 2, 5, 10, 15, 20, and 30 min and 2, 4, 8, 13, 36, and 60 h (MeCN, $C_{1b} = C_{2a} = 5 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm).

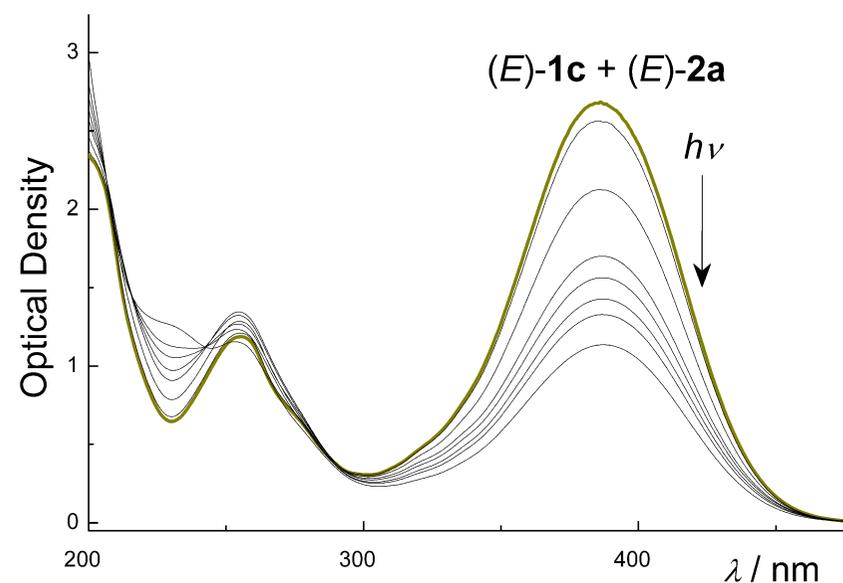


Fig. S53 Photolysis of solution of an equimolar mixture of dyes **1c** and **2a** for 0, 1, 6, 20, 40, and 80 min and 2 and 4 h (MeCN, $C_{1c} = C_{2a} = 5 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm).

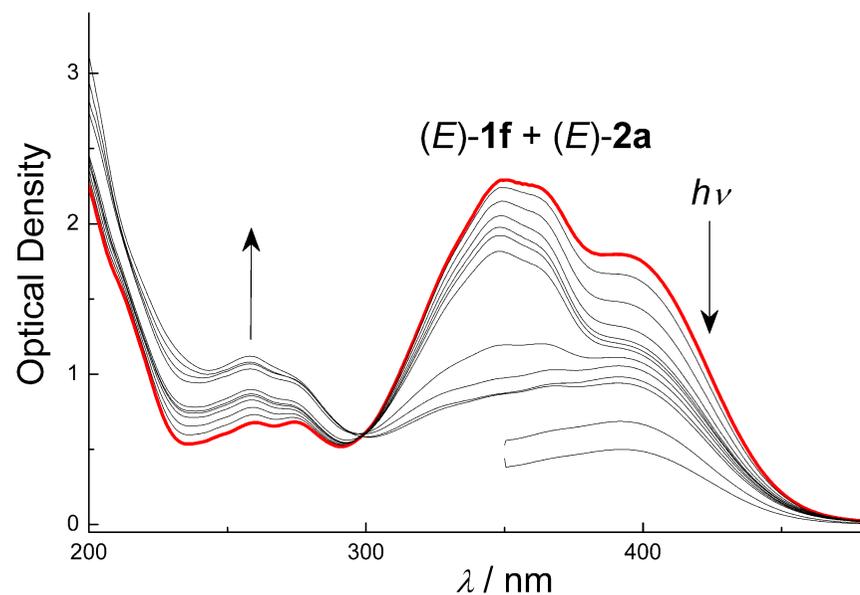


Fig. S54 Photolysis of solution of an equimolar mixture of dyes **1f** and **2a** for 0, 2, 5, 10, 15, 20, and 30 min and 2, 4, 8, 13, 36, and 60 h (MeCN, $C_{1f} = C_{2a} = 5 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm).

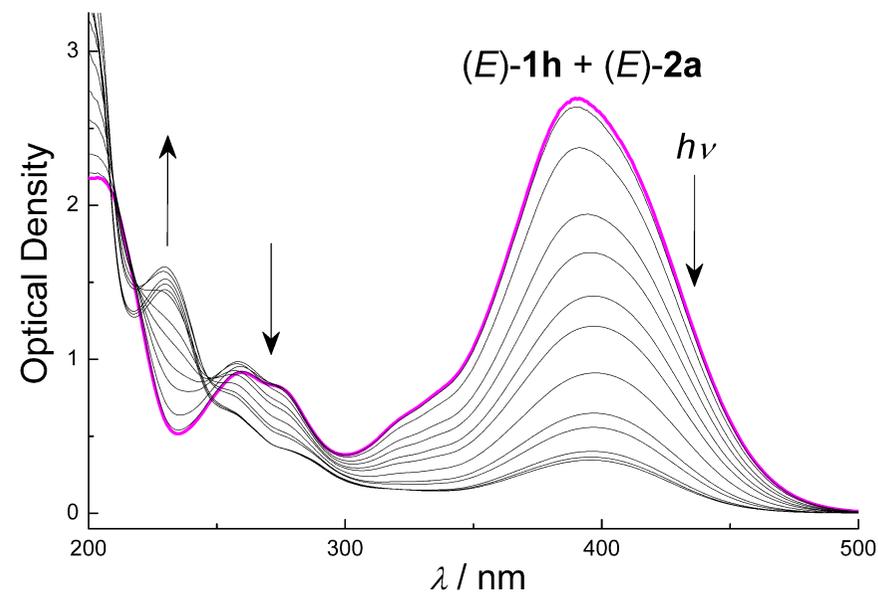


Fig. S55 Photolysis of solution of an equimolar mixture of dyes **1h** and **2a** for 0, 1, 6, 20, 40, and 80 min and 2, 4, 8, 16, 24, 34, and 52 h (MeCN, $C_{1h} = C_{2a} = 5 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm).

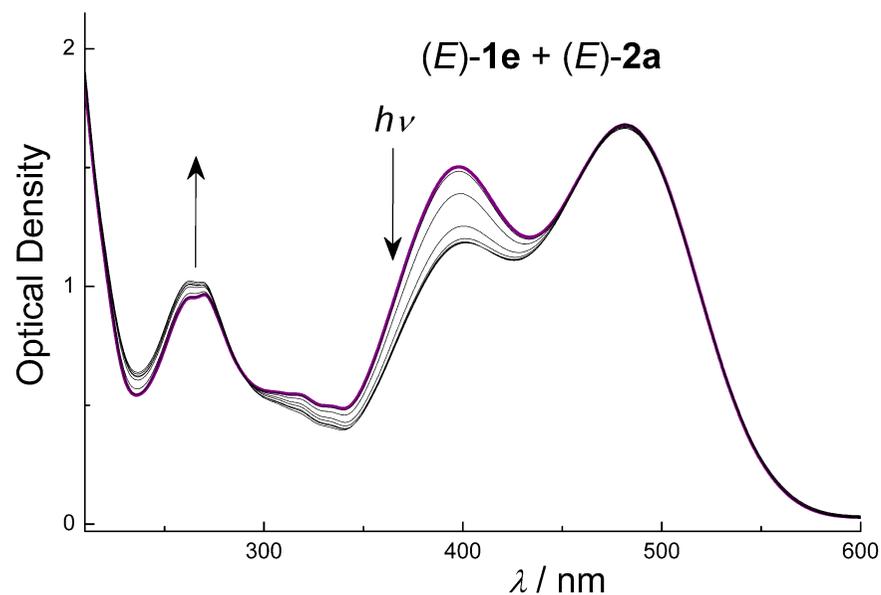


Fig. S56 Photolysis of solution of an equimolar mixture of dyes **1e** and **2a** for 0, 1, 6, 20, 40, and 80 min and 2 and 4 h (MeCN, $C_{1e} = C_{2a} = 5 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm).

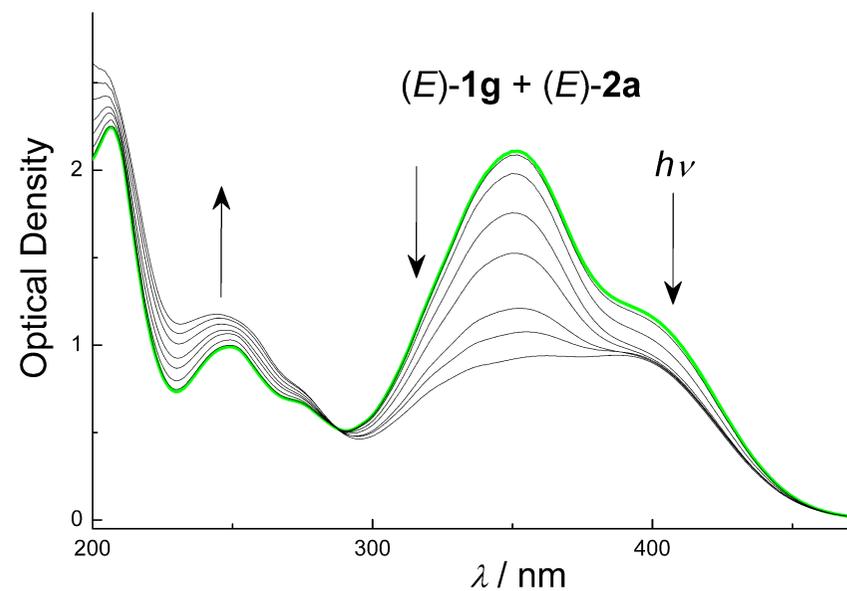


Fig. S57 Photolysis of solution of an equimolar mixture of dyes **1g** and **2a** for 0, 1, 6, 20, 40, and 80 min and 2 and 4 h (MeCN, $C_{1g} = C_{2a} = 5 \times 10^{-4}$ M, 0.1-cm quartz cell, unfiltered light from a 60 W incandescent lamp, distance to the light source ~ 15 cm).

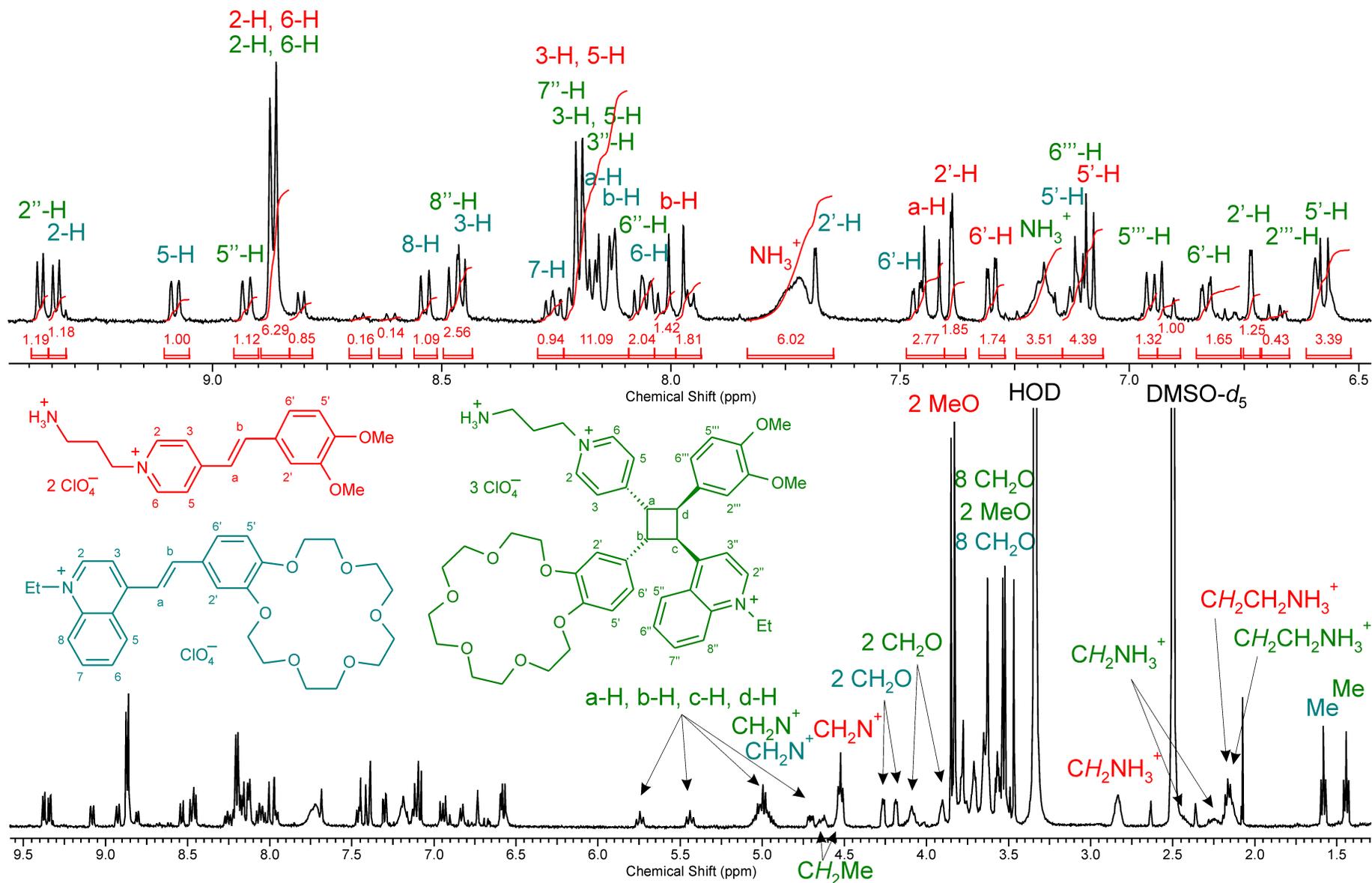


Fig. S58 ^1H NMR spectrum of a sample of 1:1 mixture of dyes **1a** and **2b**, which was irradiated with visible light for 290 h in MeCN and then redissolved in $\text{DMSO-}d_6$ (500.13 MHz, 30 °C).

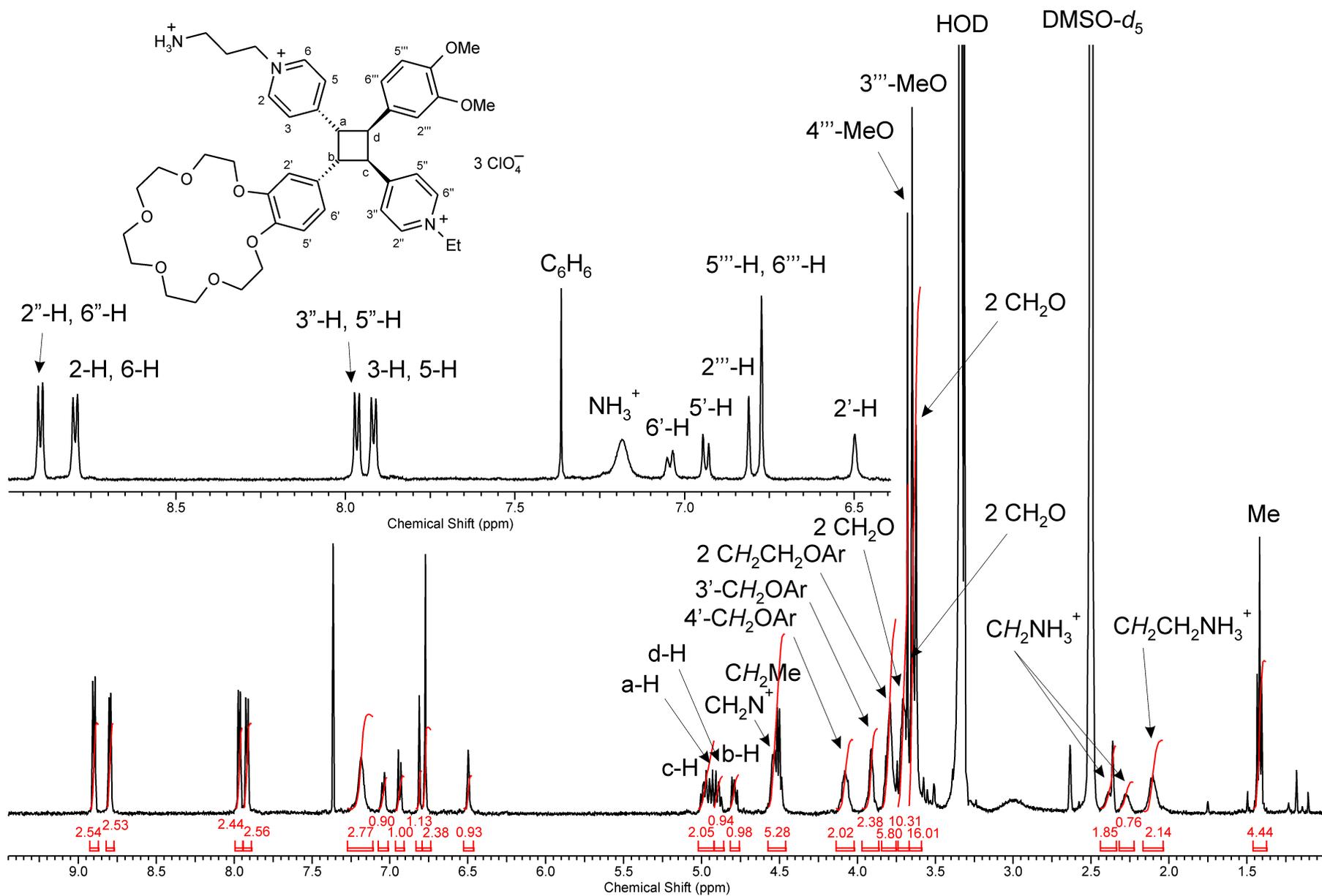


Fig. S59 ¹H NMR spectrum of cyclobutane *rctt*-**3a** (500.13 MHz, DMSO-*d*₆, 23 °C).

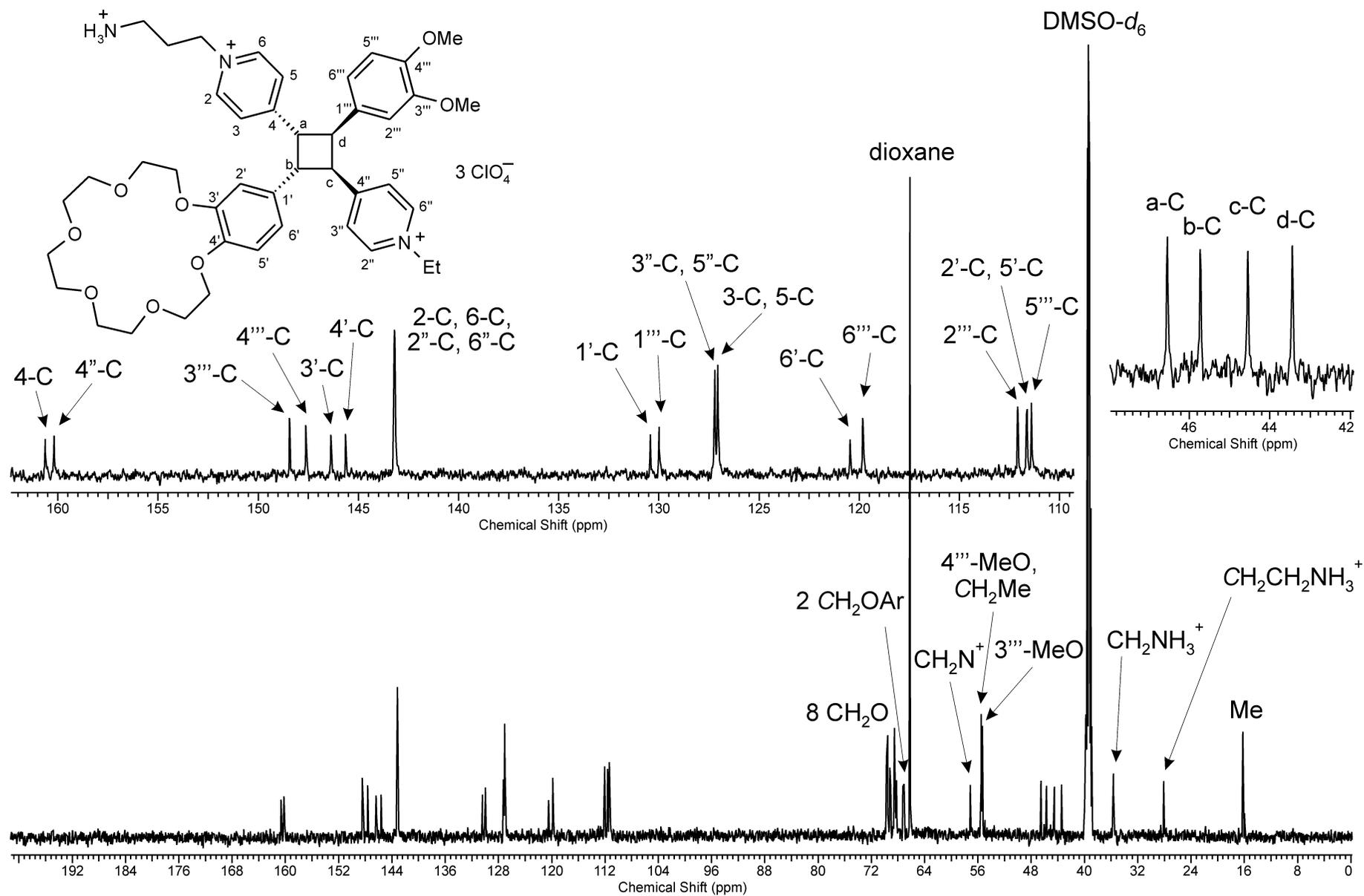


Fig. S60 ¹³C NMR spectrum of cyclobutane *rctt*-**3a** (125.76 MHz, DMSO-*d*₆, 30 °C).

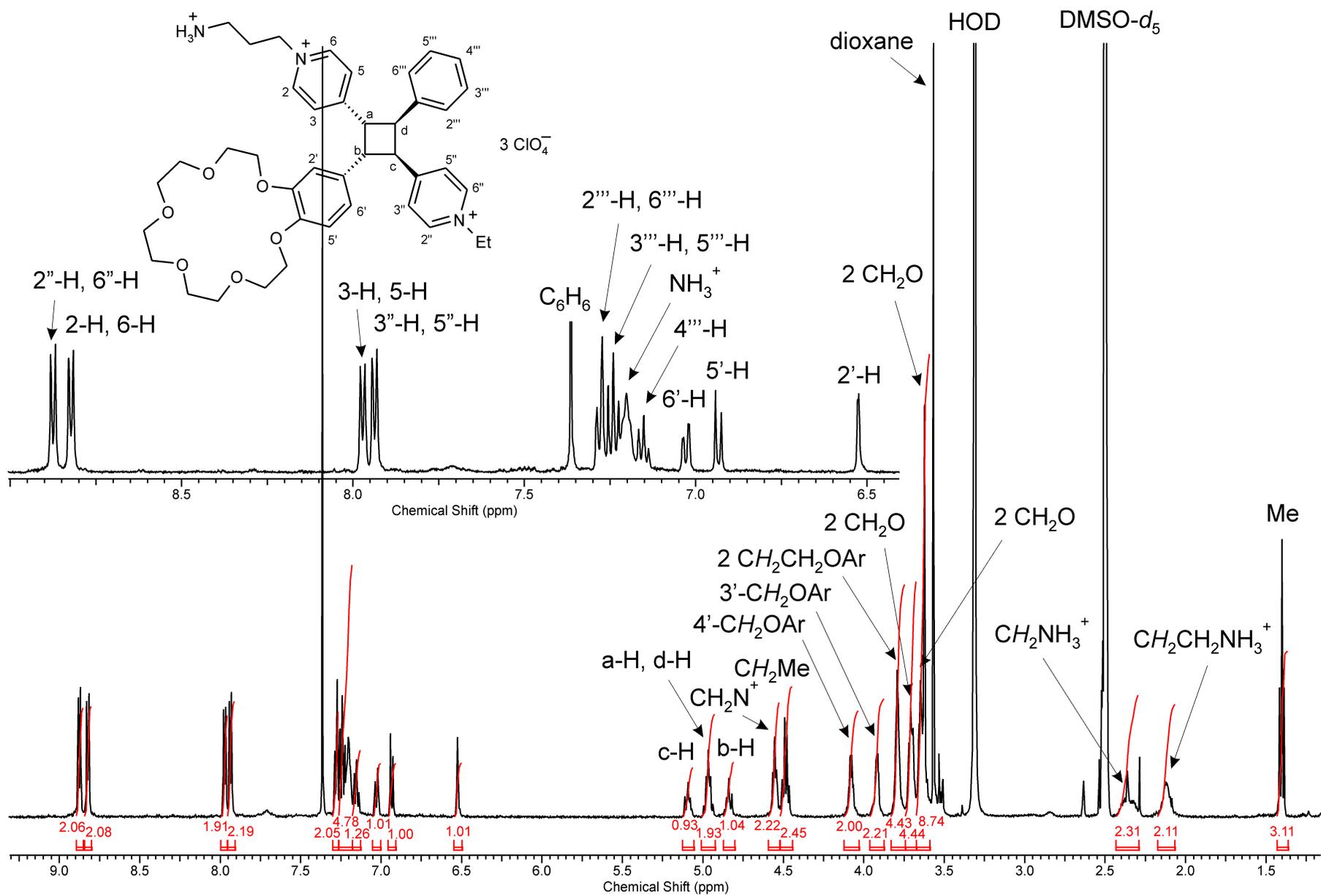


Fig. S61 ¹H NMR spectrum of of cyclobutane *rctt*-**3b** (500.13 MHz, DMSO-*d*₆, 25 °C).

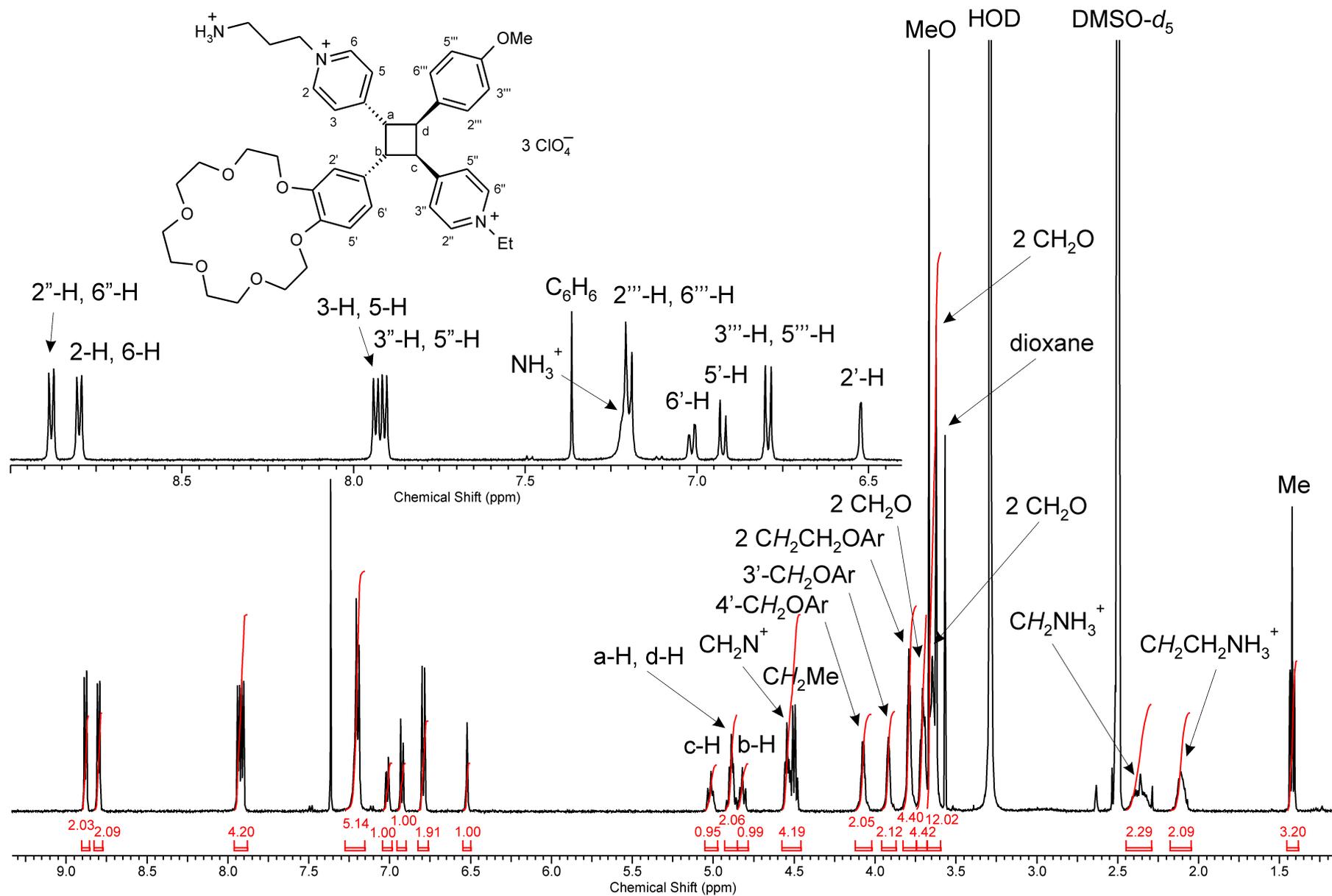


Fig. S62 ^1H NMR spectrum of cyclobutane *rctt*-**3c** (500.13 MHz, $\text{DMSO-}d_6$, 30°C).

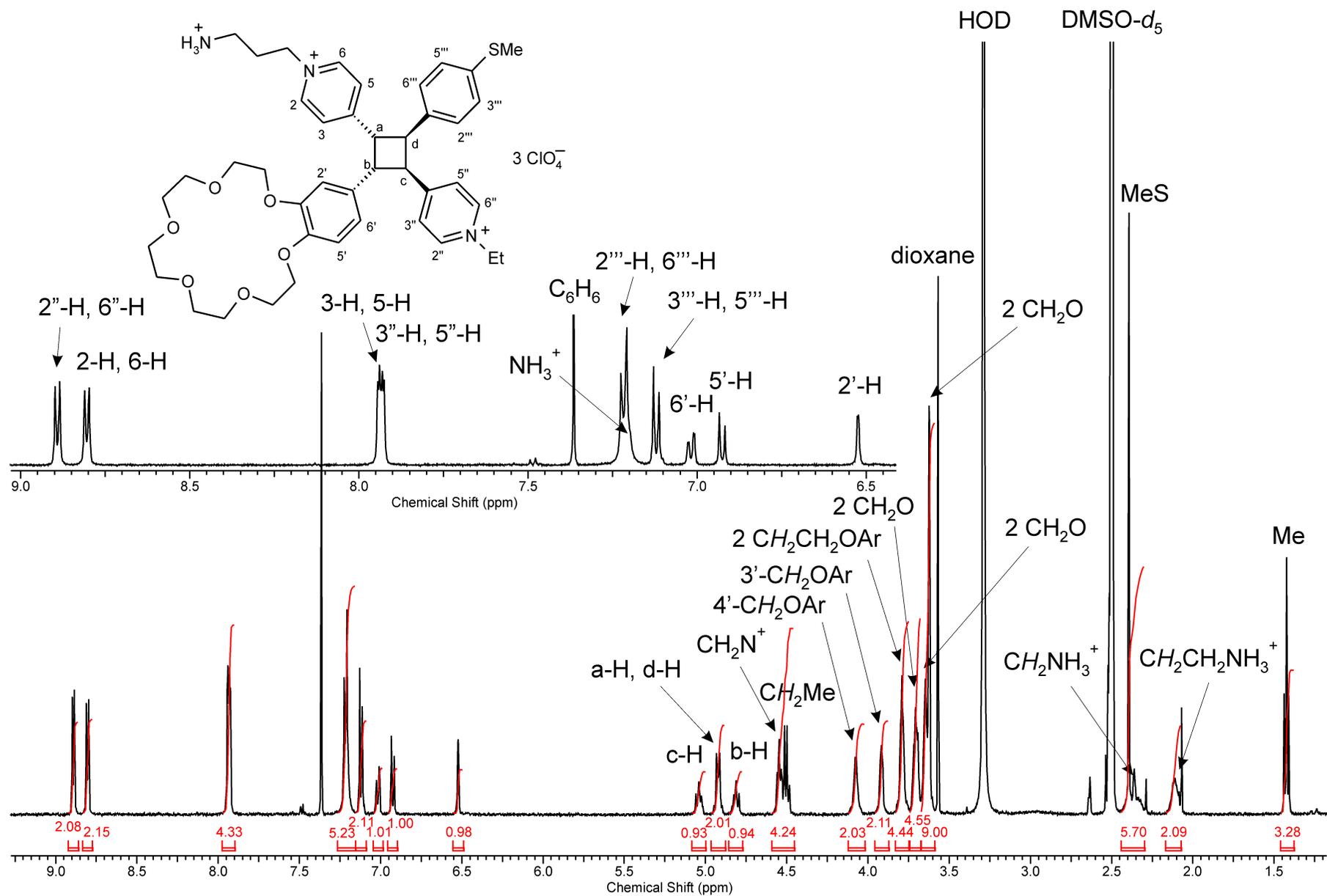


Fig. S63 ^1H NMR spectrum of cyclobutane *rctt*-**3d** (500.13 MHz, $\text{DMSO-}d_6$, 30 $^\circ\text{C}$).

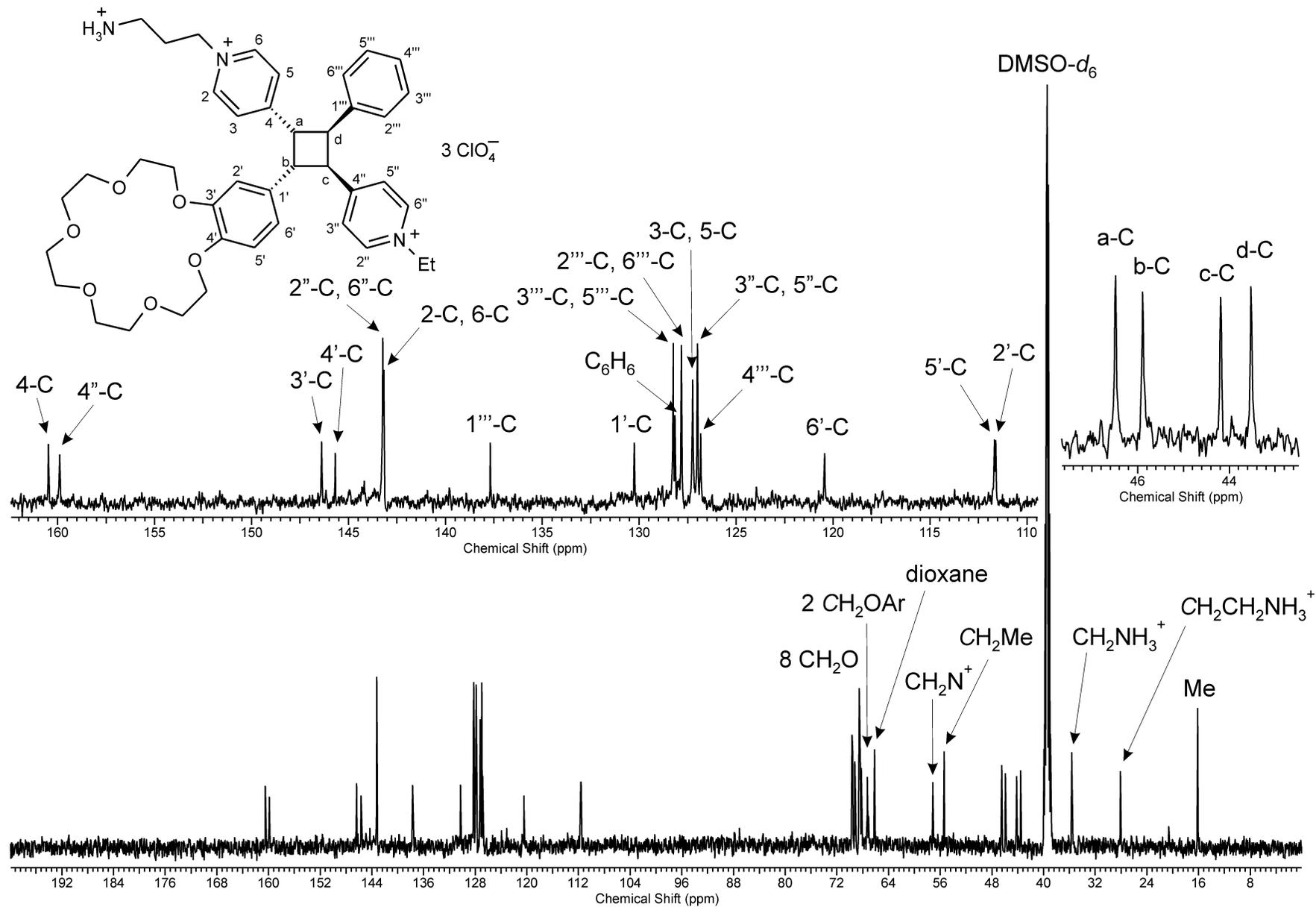


Fig. S64 ¹³C NMR spectrum of cyclobutane *rctt-3b* (125.76 MHz, DMSO-*d*₆, 30 °C).

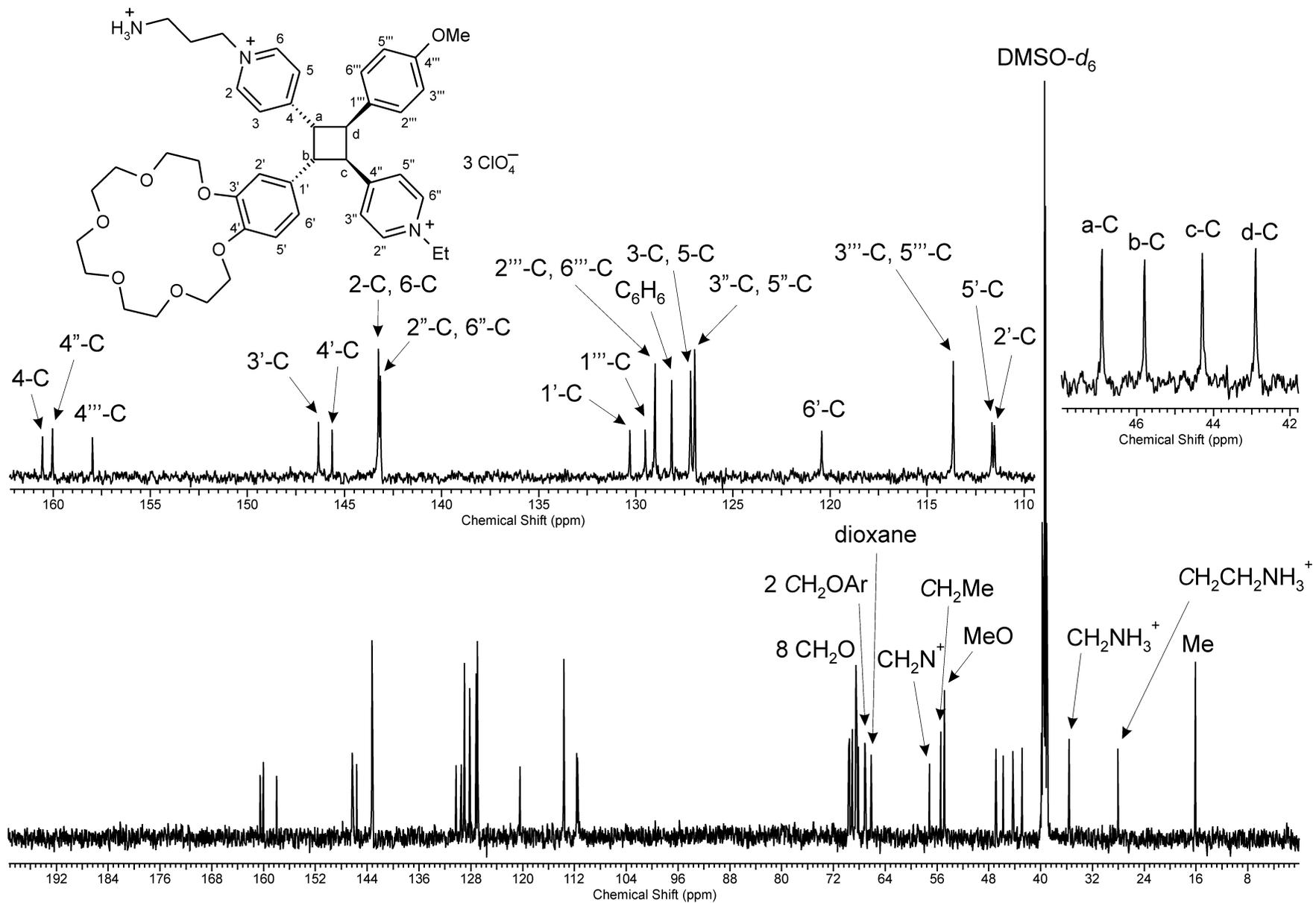


Fig. S65 ^{13}C NMR spectrum of cyclobutane *rctt*-**3c** (125.76 MHz, DMSO- d_6 , 30 °C).

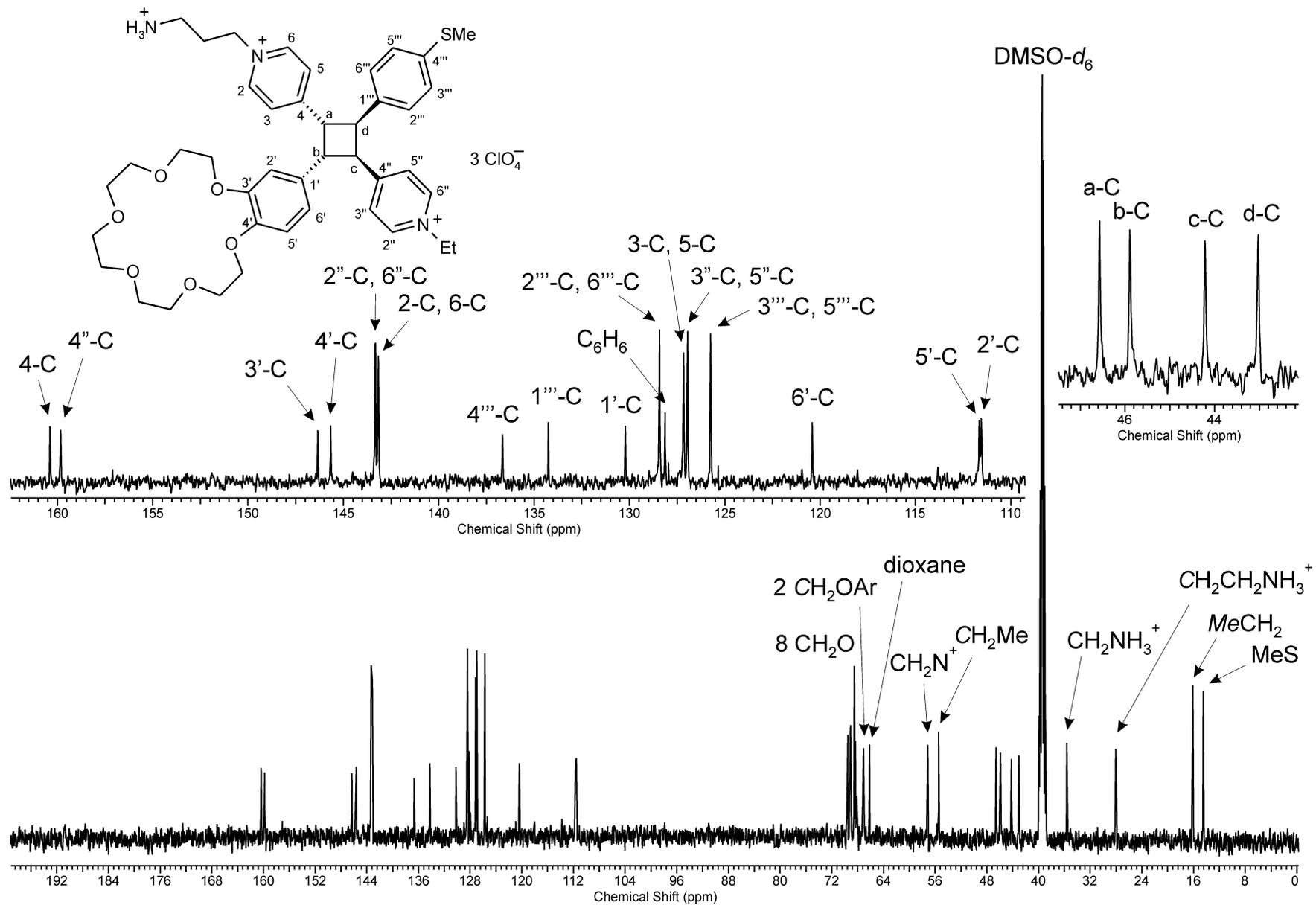


Fig. S66 ¹³C NMR spectrum of cyclobutane *rctt*-**3d** (125.76 MHz, DMSO-*d*₆, 30 °C).

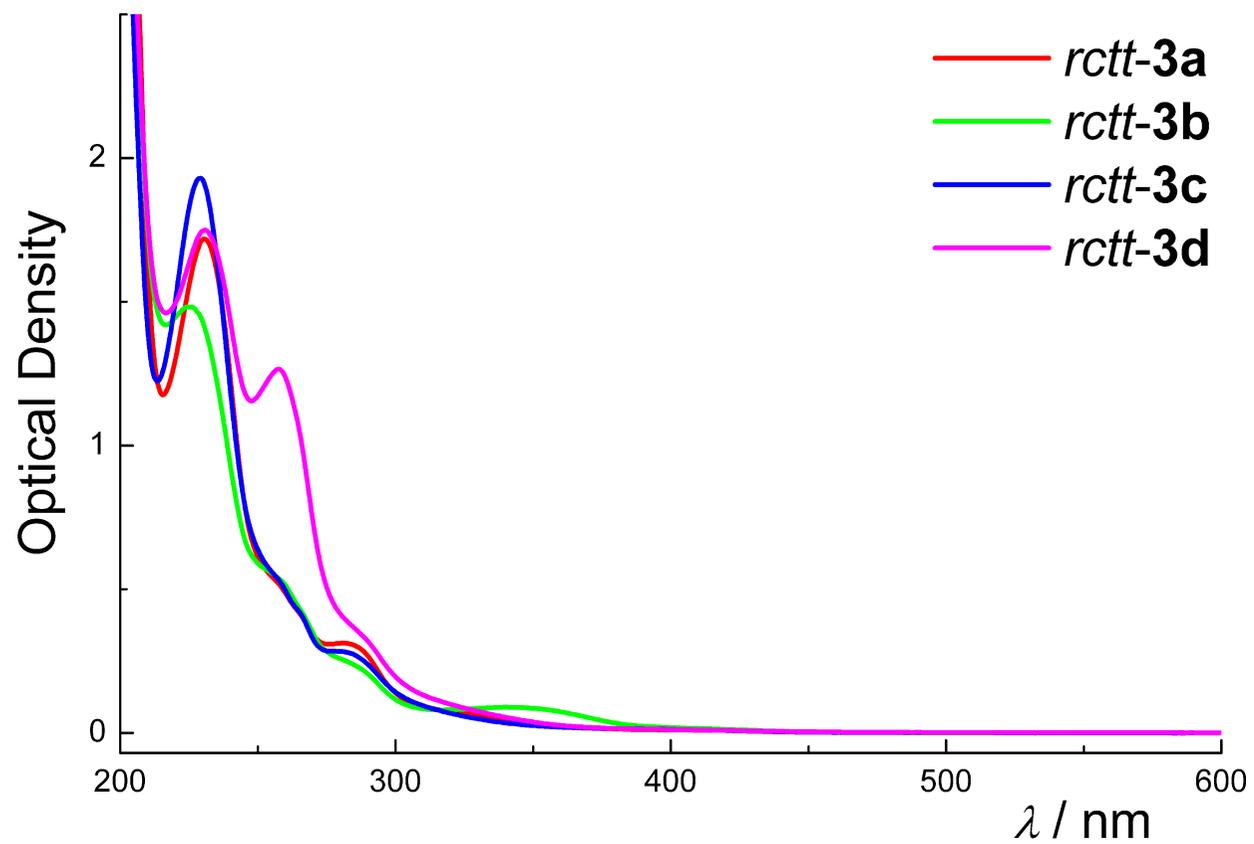


Fig. S67 Absorption spectra of cyclobutanes *rctt-3a-d* (MeCN, $C_{\text{cyclobutane}} = 5 \times 10^{-5}$ M, 1-cm quartz cell, ambient temperature).

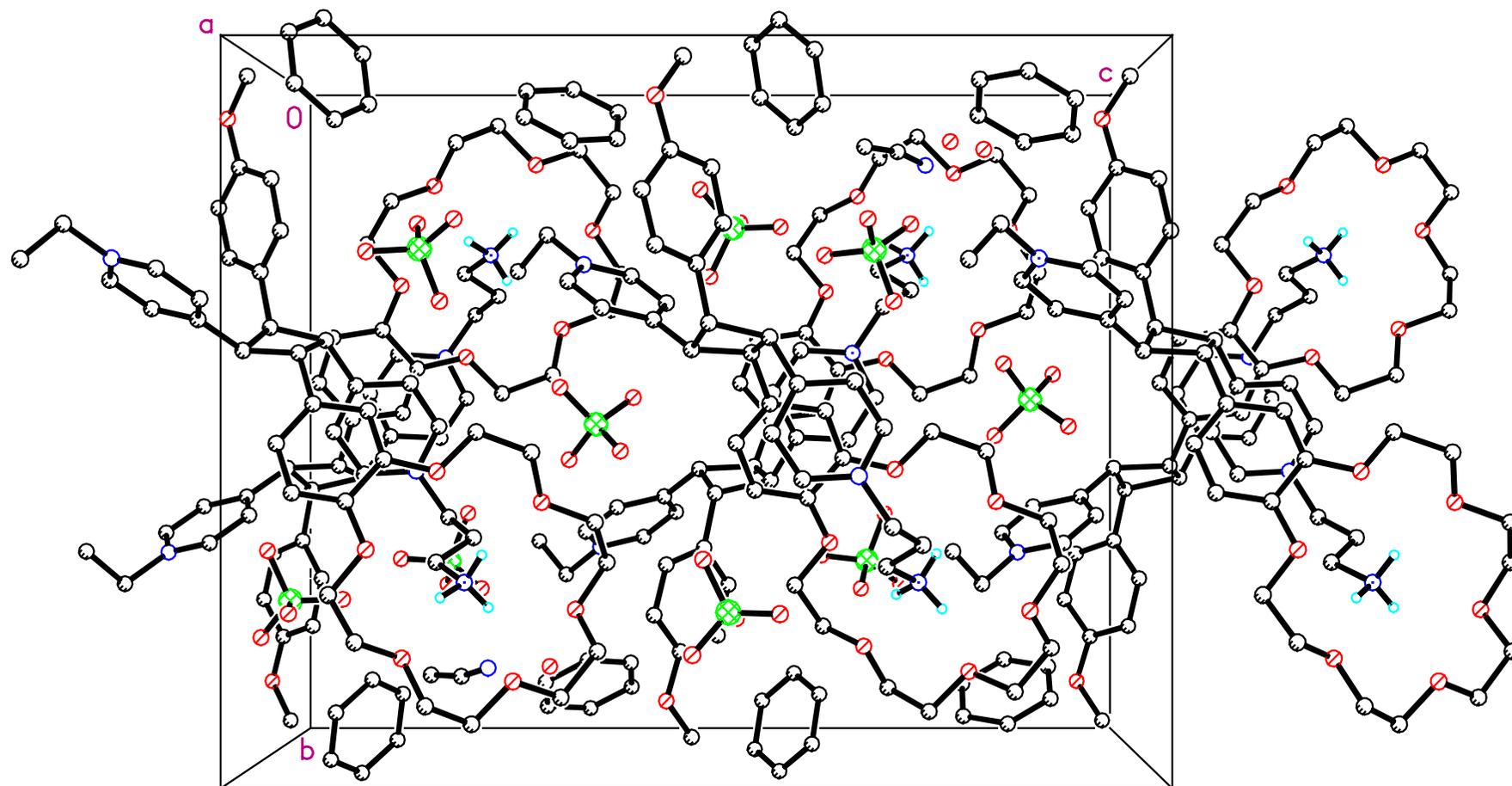


Fig. S68 Packing of *rctt-3c*·2C₆H₆·0.75MeCN·0.25H₂O.

