1. FT-IR, ^{1}H-NMR, ^{13}C-NMR and MS spectra of selected products (4a-q)

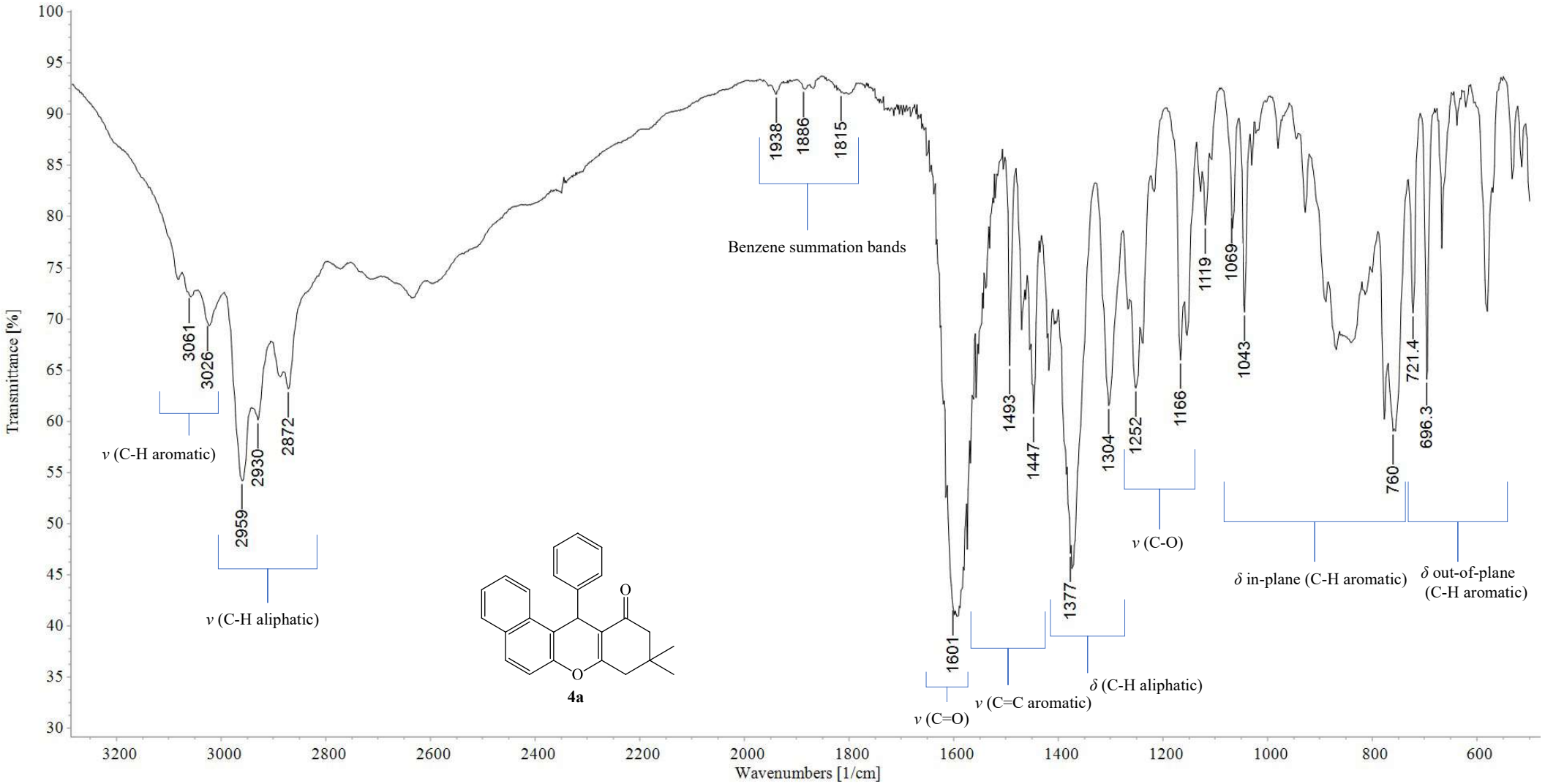

Figure S1 – FT-IR spectrum of 9,9-dimethyl-12-phenyl-8,9,10,12-tetrahydro-11H-benzo[a]xanthen-11-one (4a), NaCl

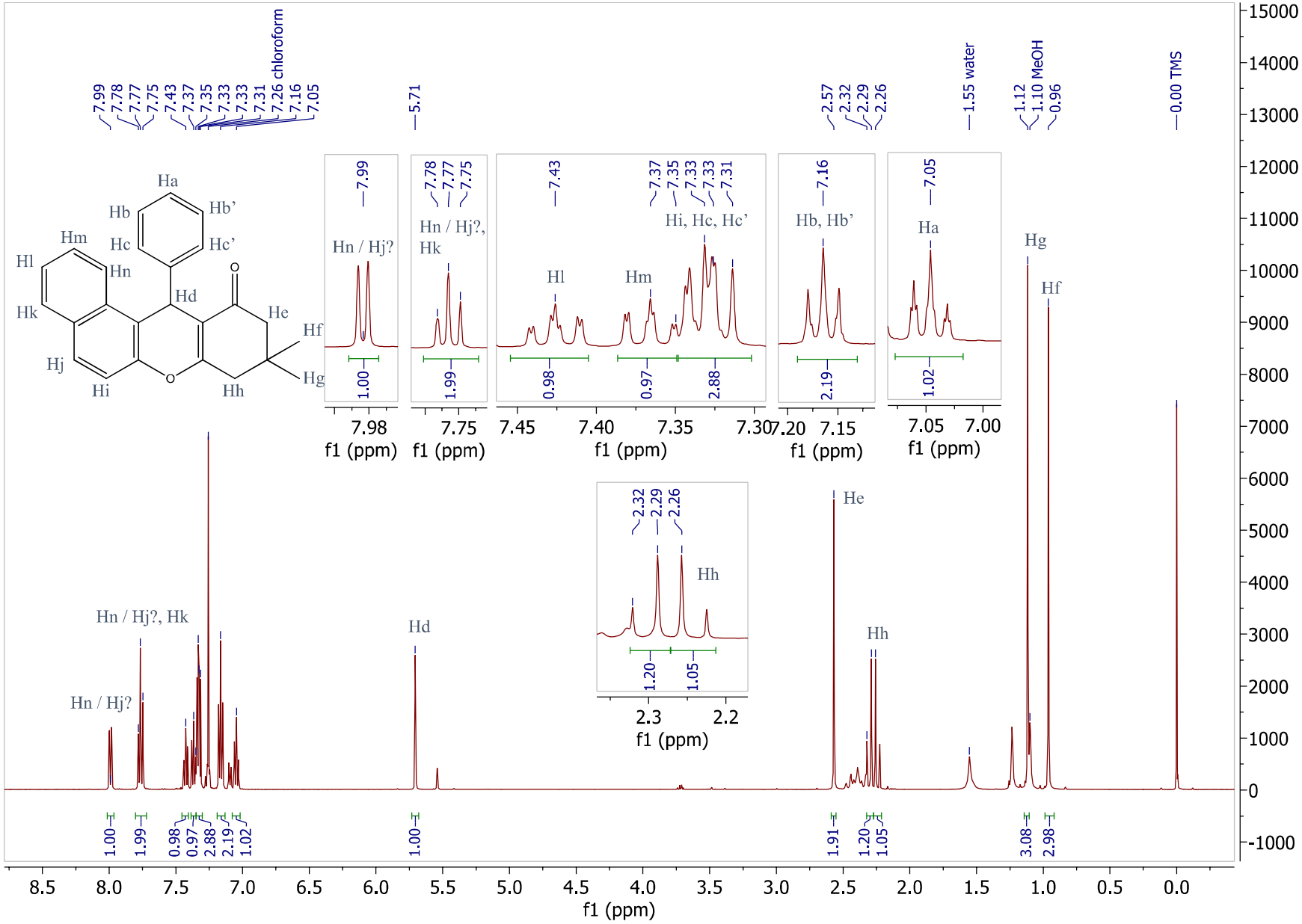

Figure S2 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-phenyl-8,9,10,12-tetrahydro-11H-benzo[a]xanthen-11-one (4a), in chloroform-d

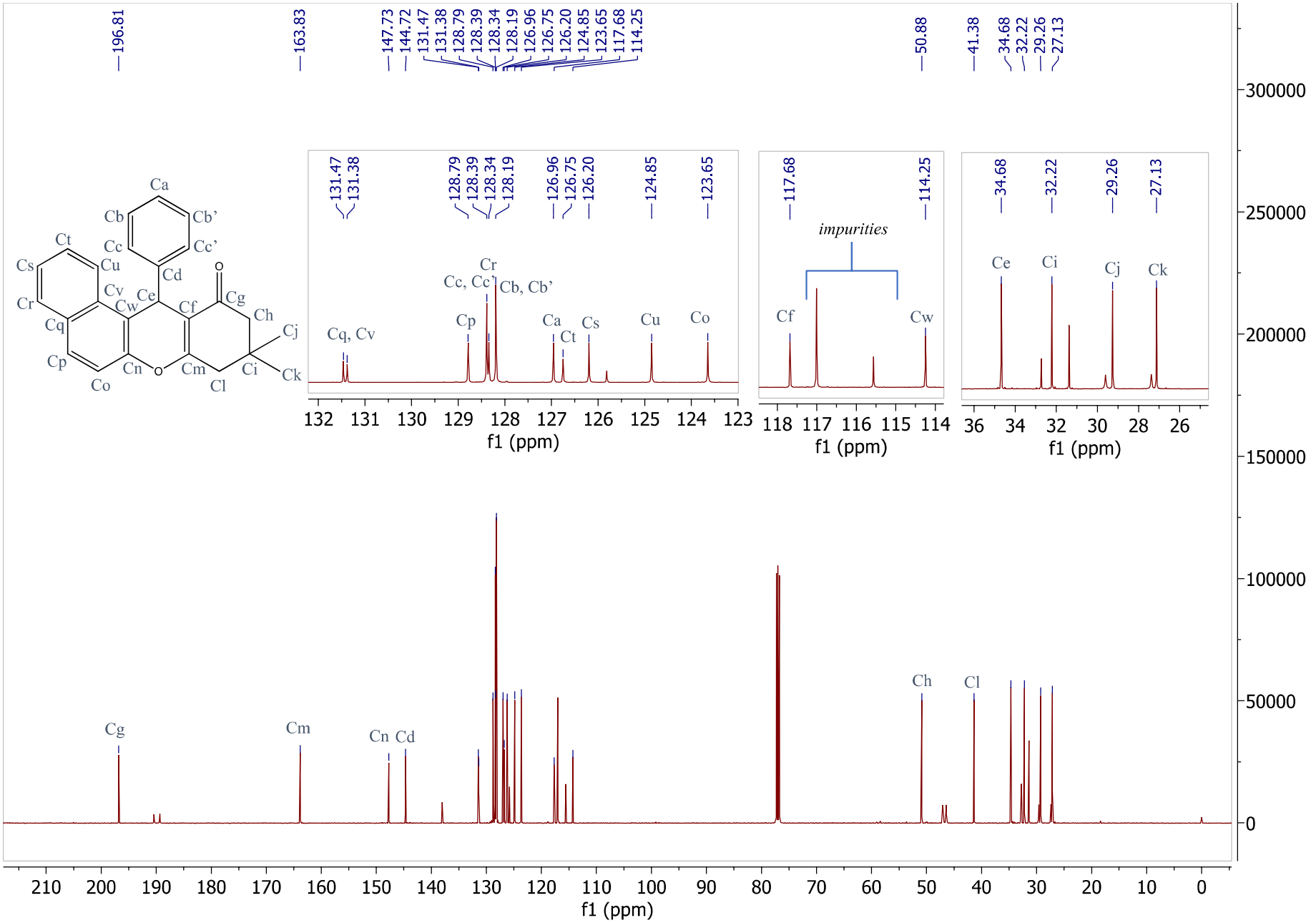

Figure S3 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-phenyl-8,9,10,12-tetrahydro-11H-benzo[a]xanthen-11-one (4a), in chloroform-d

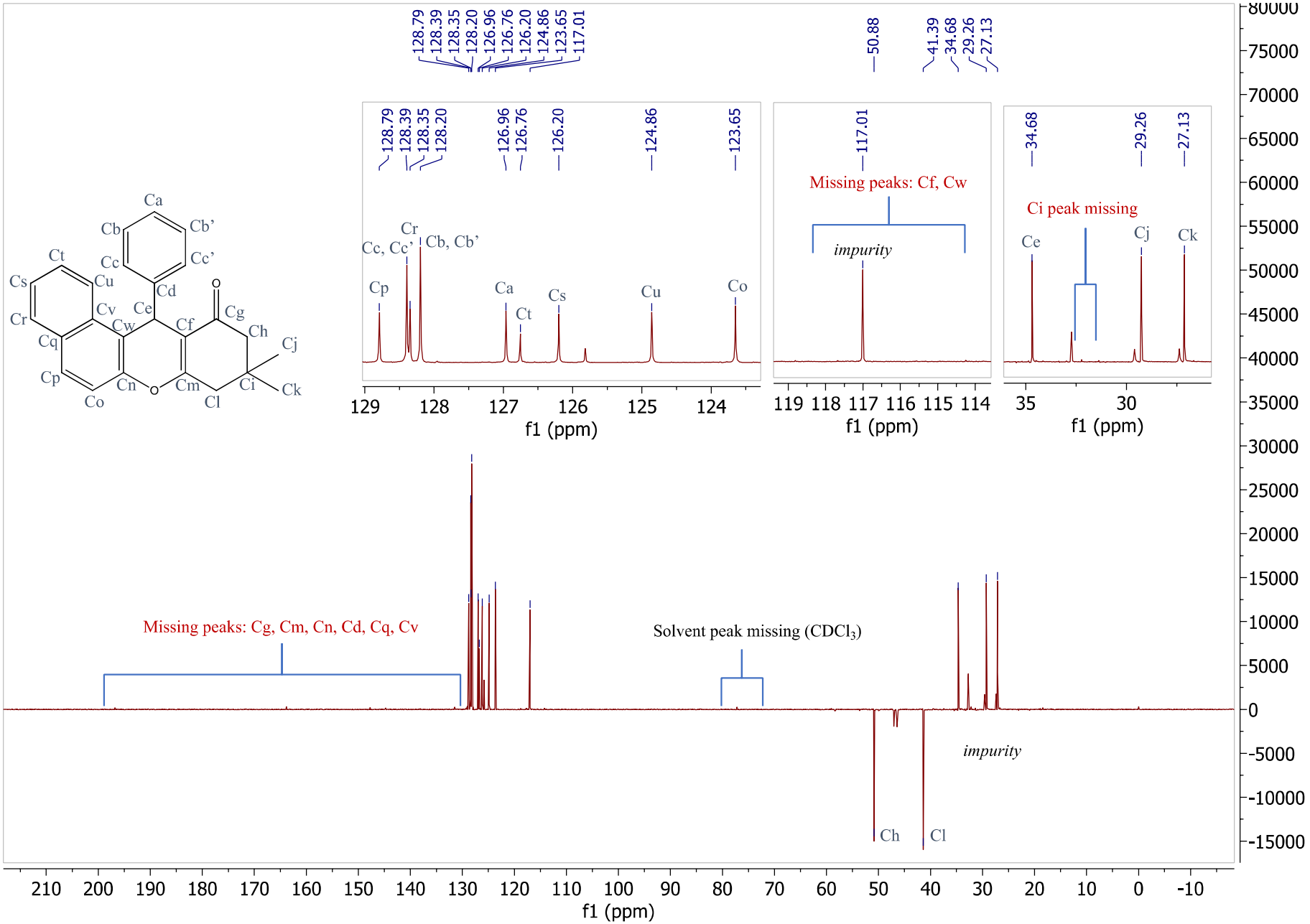

Figure S4 - DEPT spectrum of product 9,9-dimethyl-12-phenyl-8,9,10,12-tetrahydro-11H-benzo[a]xanthen-11-one (4a) in $CDCl_3$, showing missing peaks compared to ^{13}C-NMR spectrum

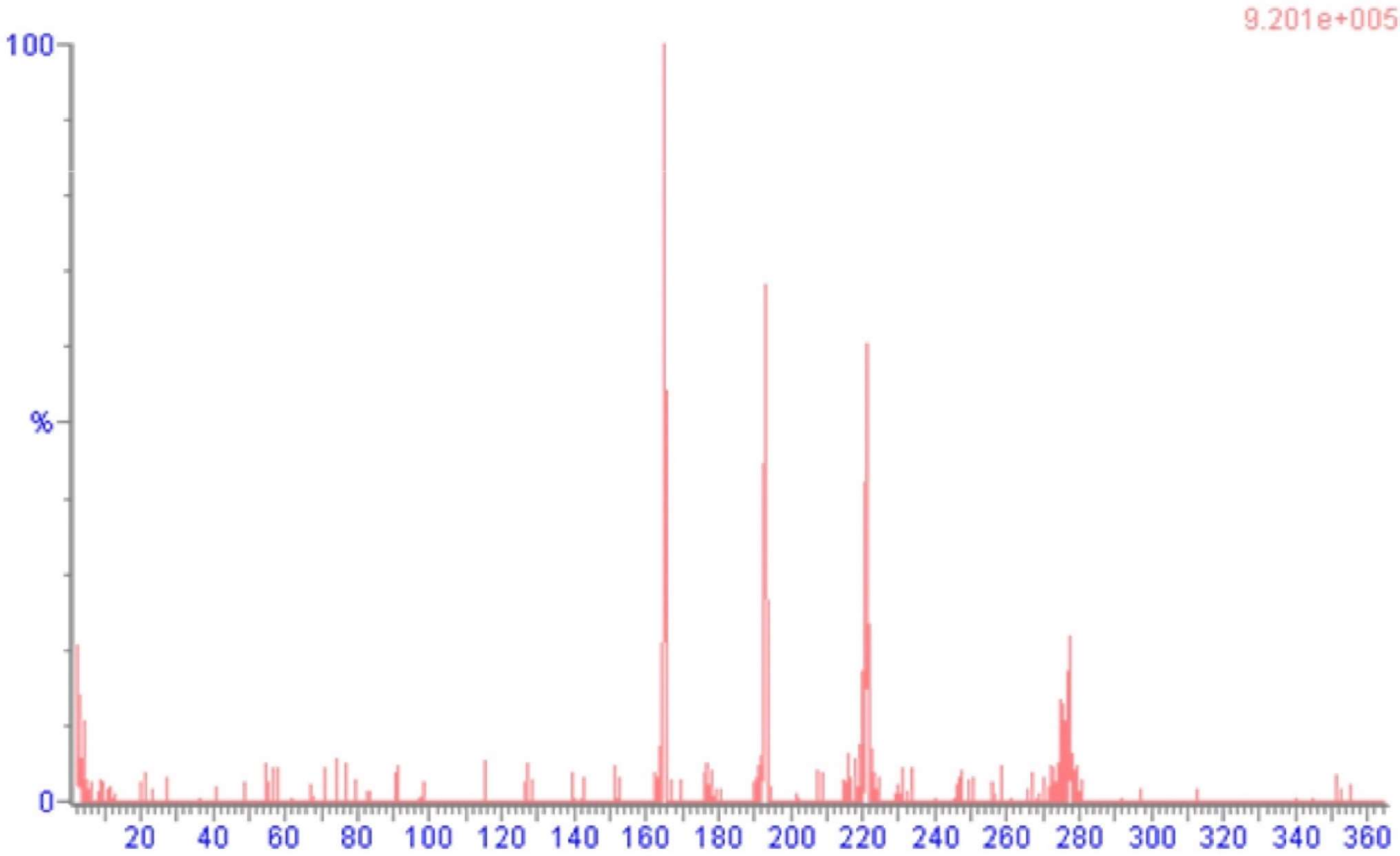

Figure S5 - MS (ES+) spectrum of 9,9-dimethyl-12-phenyl-8,9,10,12-tetrahydro-11H-benzo[a]xanthen-11-one (4a)

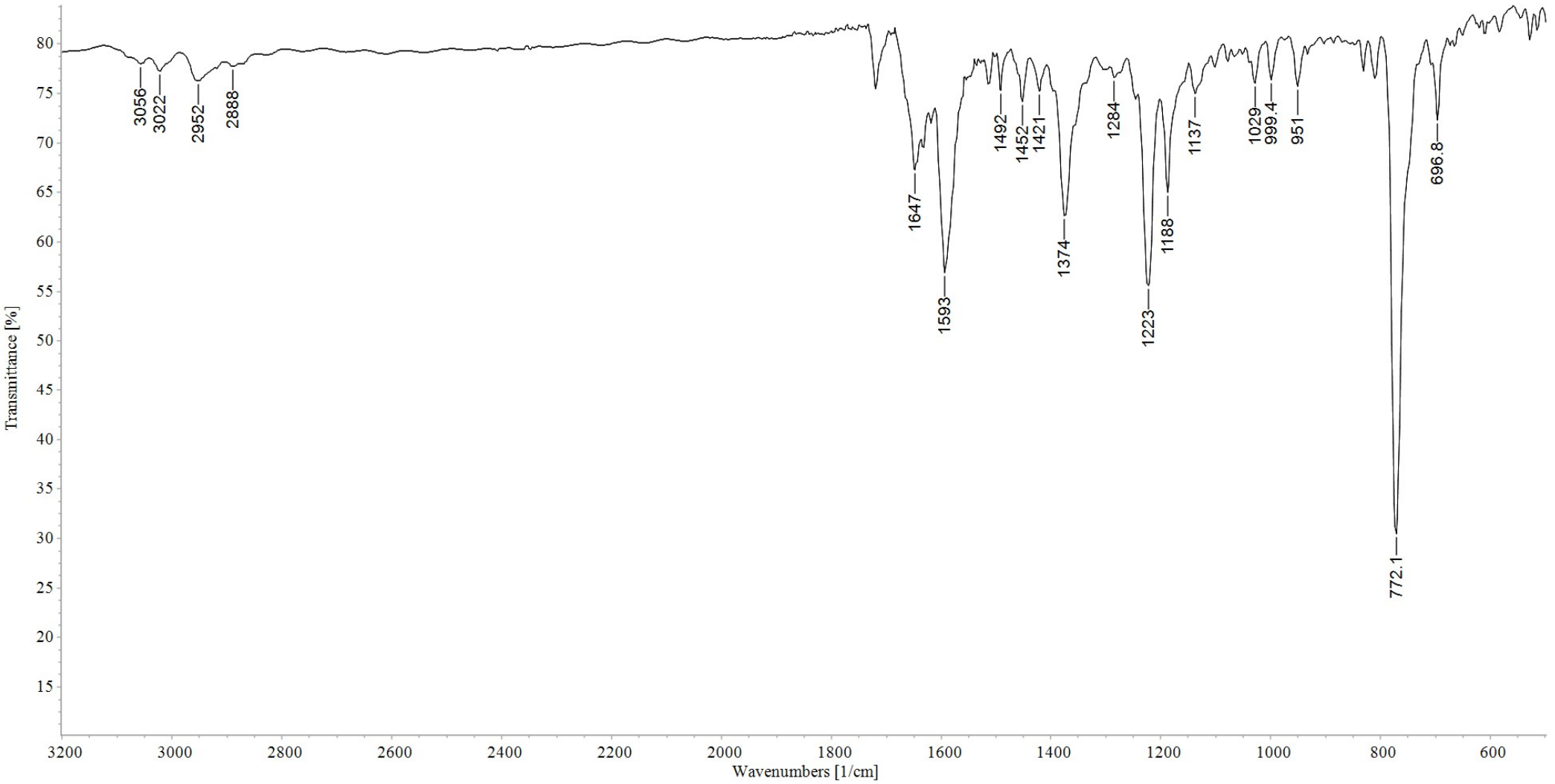

Figure S6 - FT-IR spectrum of 12-phenyl-9,10-dihydro-8H-benzo[a]xanthen-11(12H)-one (4b), NaCl

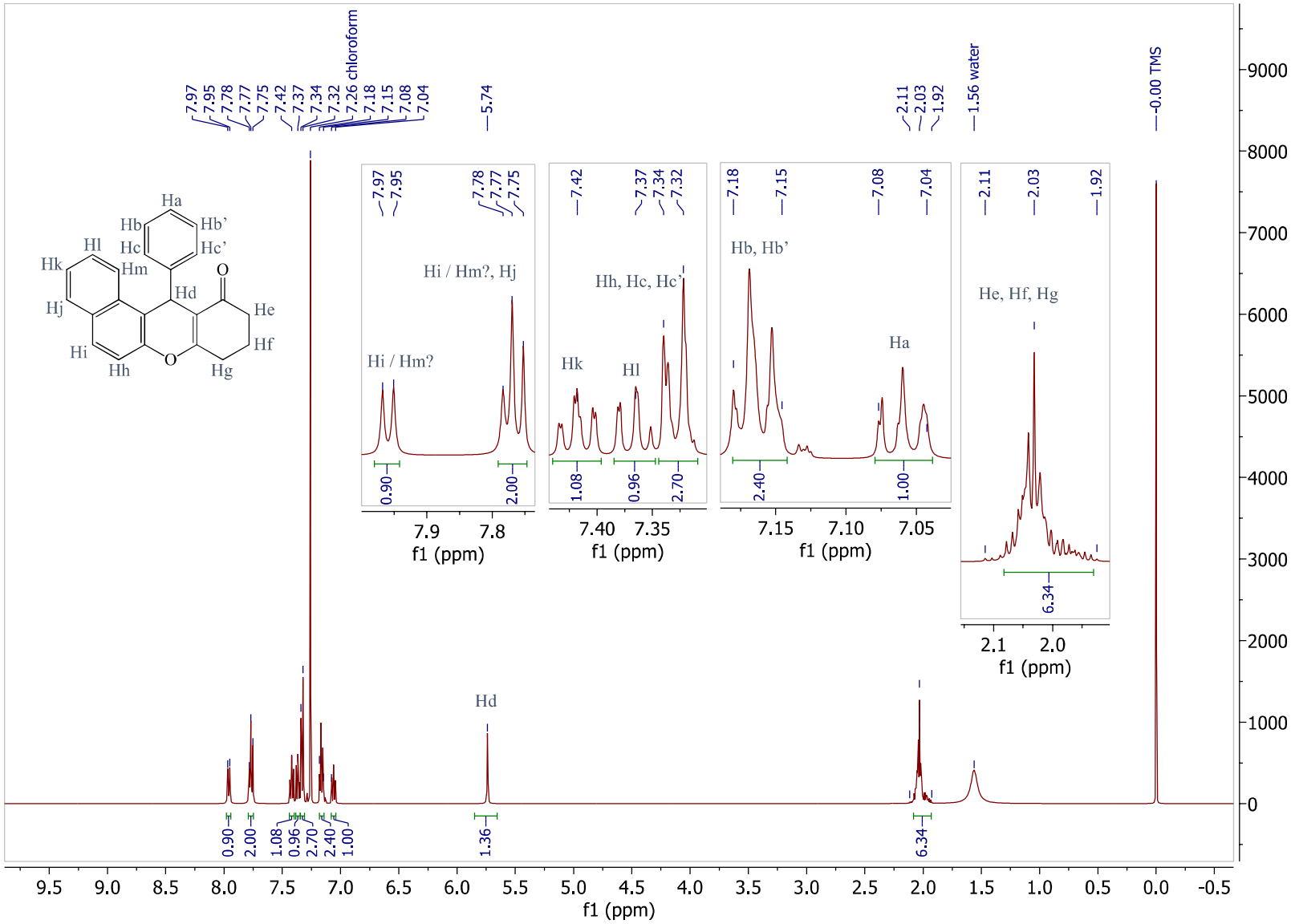

Figure S7 - ^{1}H-NMR spectrum of 12-phenyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4b), in chloroform-d

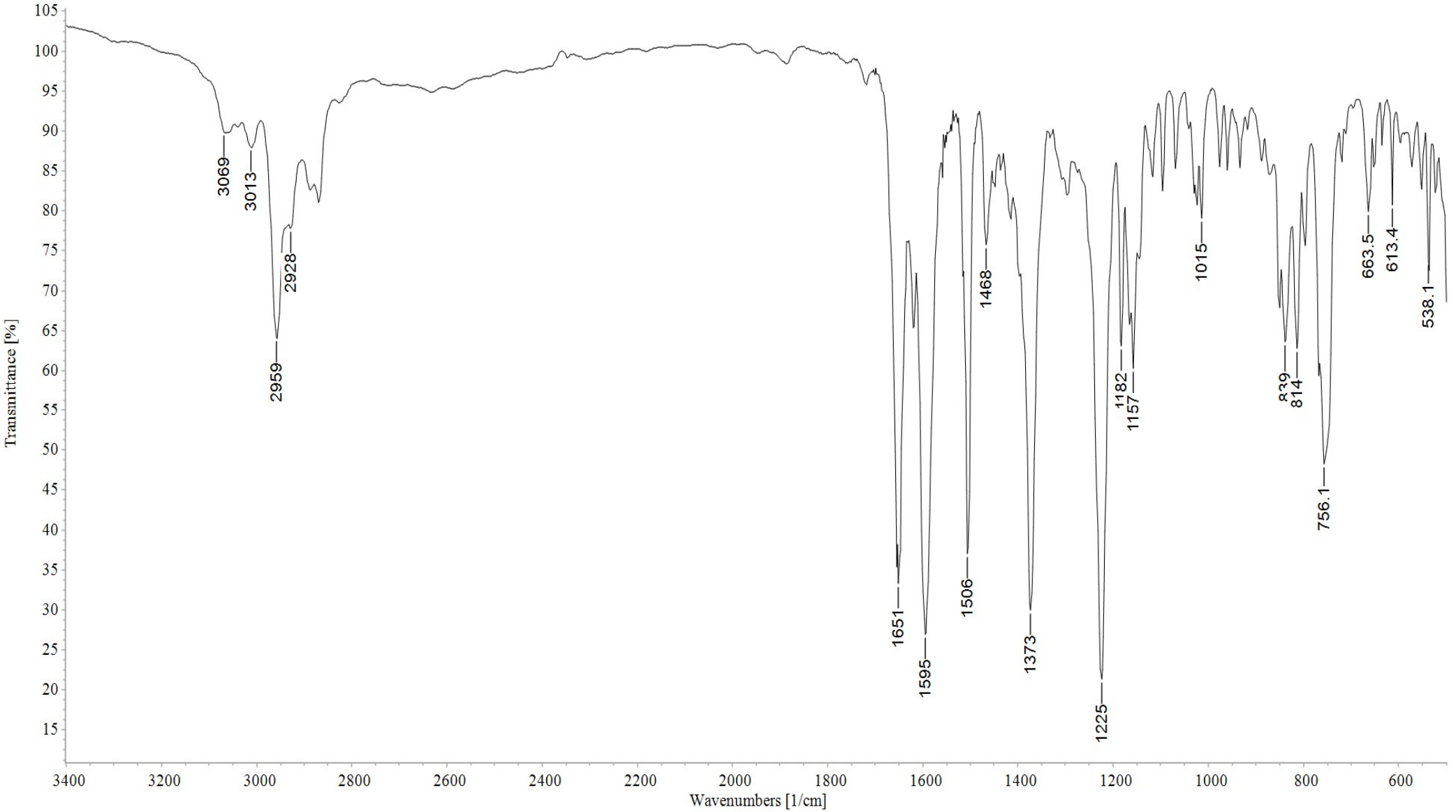

Figure S8 - FT-IR spectrum of 9,9-dimethyl-12-(4-fluorophenyl)-8,9,10,12-tetrahydro-11H-benzo[a]xanthen-11-one (4c), NaCl

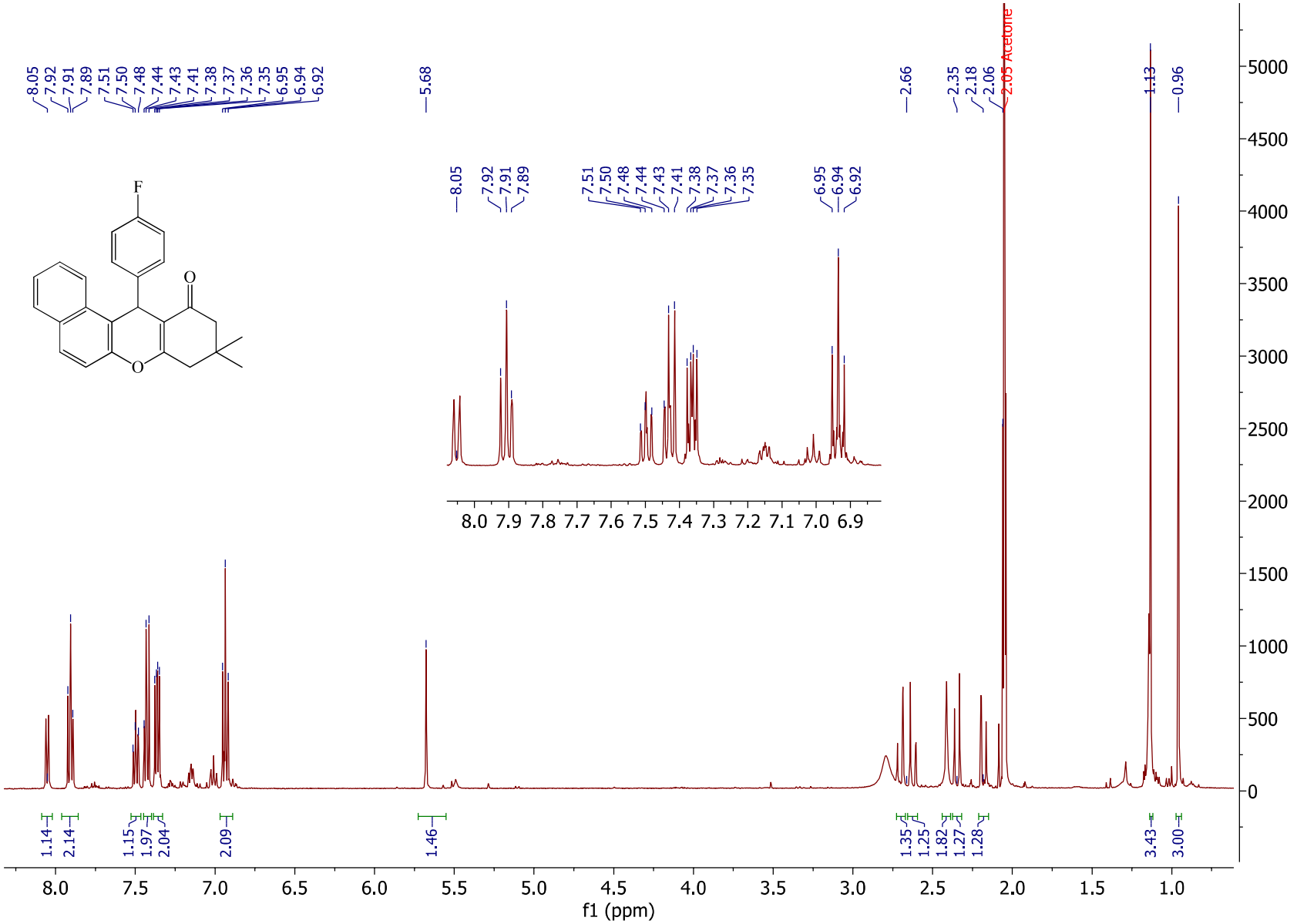

Figure S9 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(4-fluorophenyl)-9,10-dihydro-8H-benzo[a]xanthen-11(12H)-one (4c), in acetone-d_6

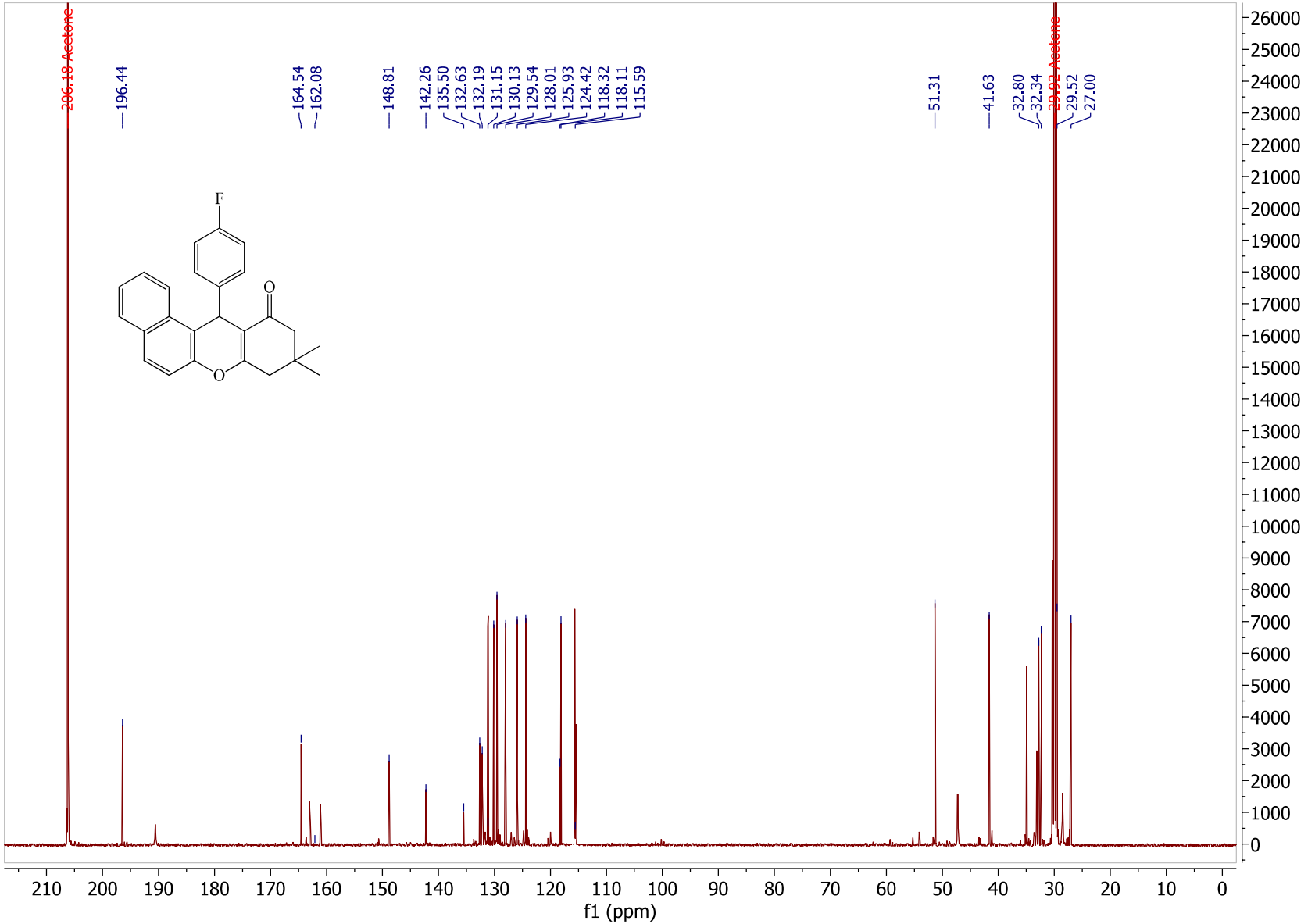

Figure S10 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-(4-fluorophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4c), in acetone-d$_6$

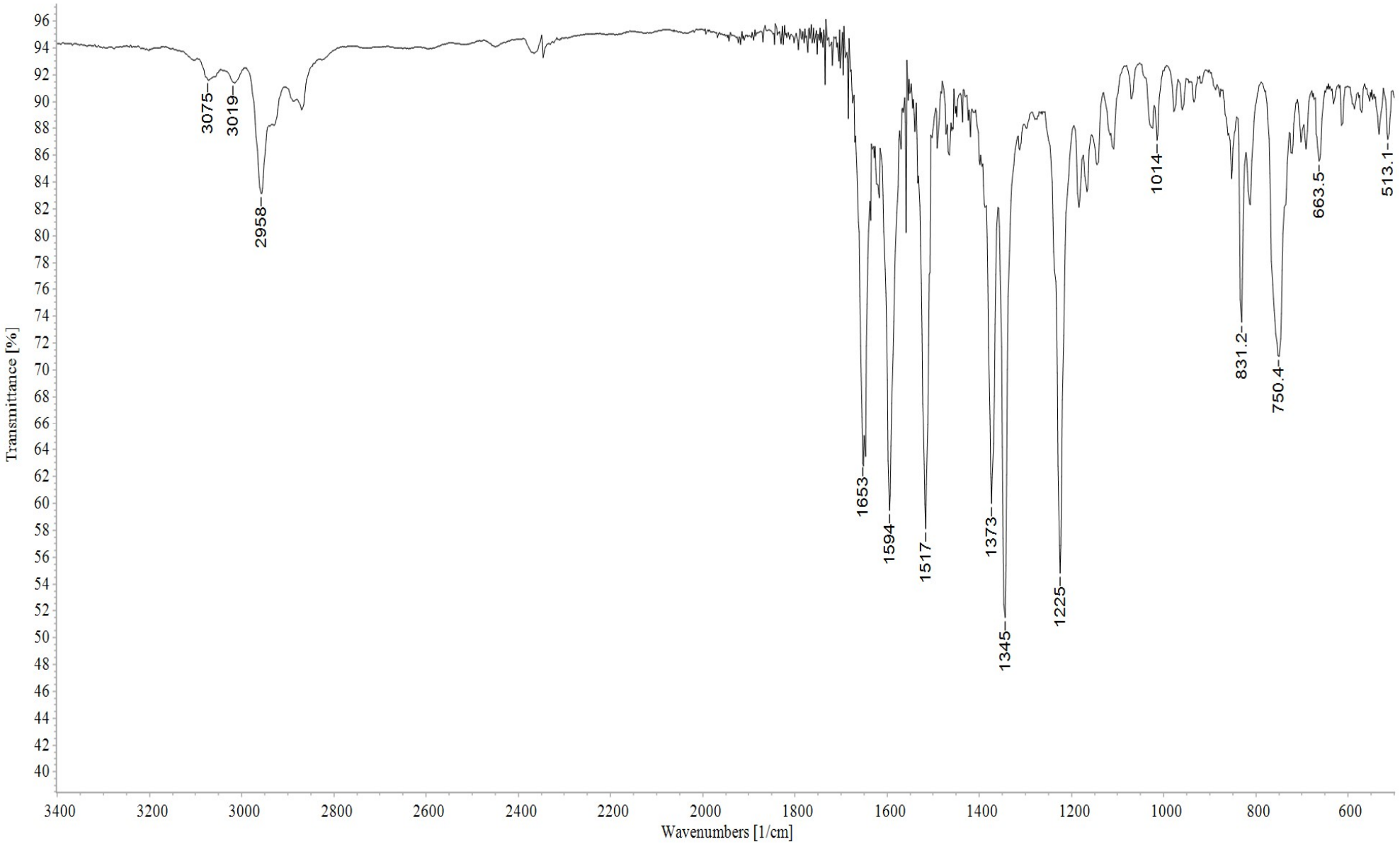

Figure S11 - FT-IR spectrum of 9,9-dimethyl-12-(4-chlorophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4d), NaCl

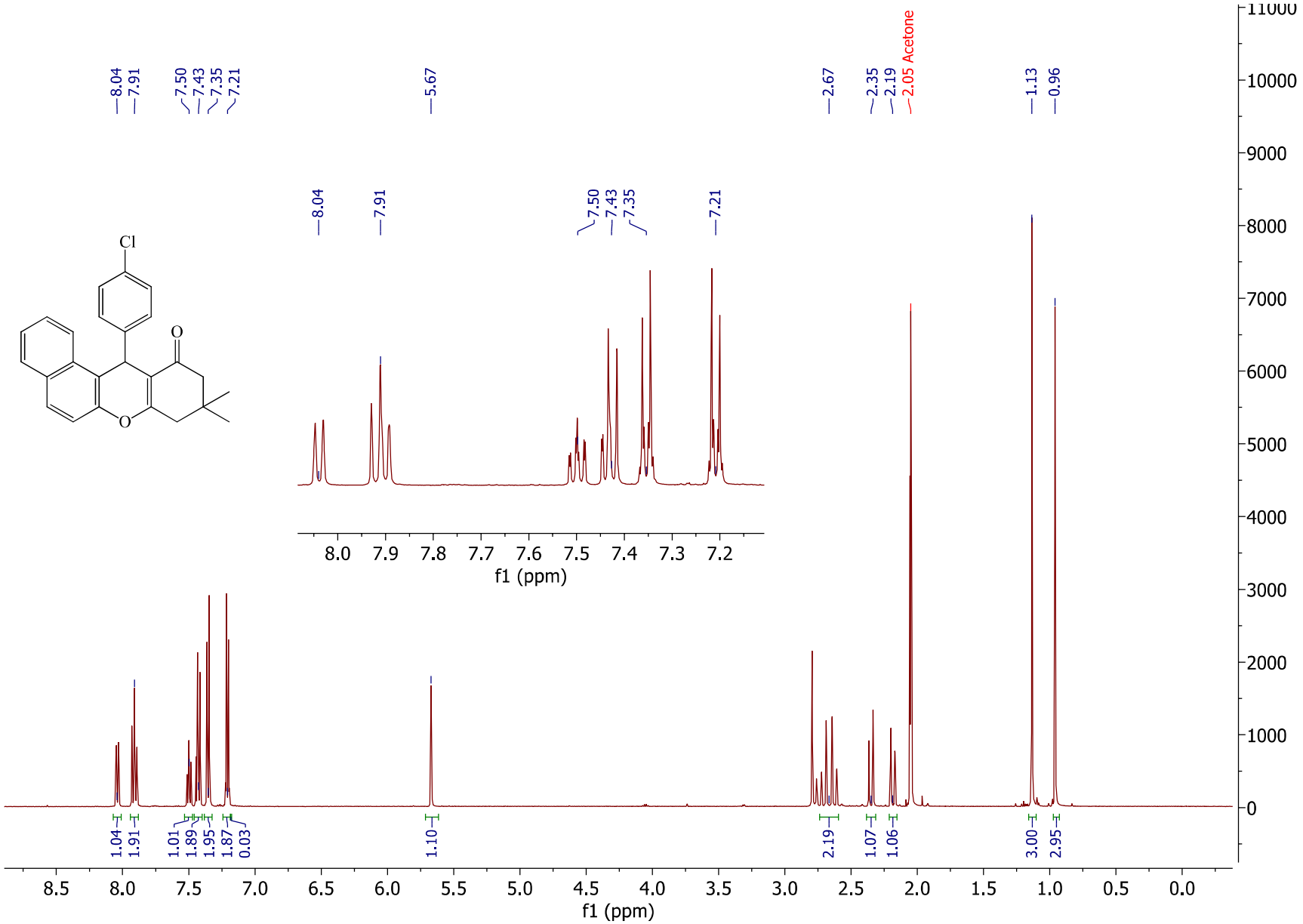

Figure S12 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(4-chlorophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4d), in acetone-d_6

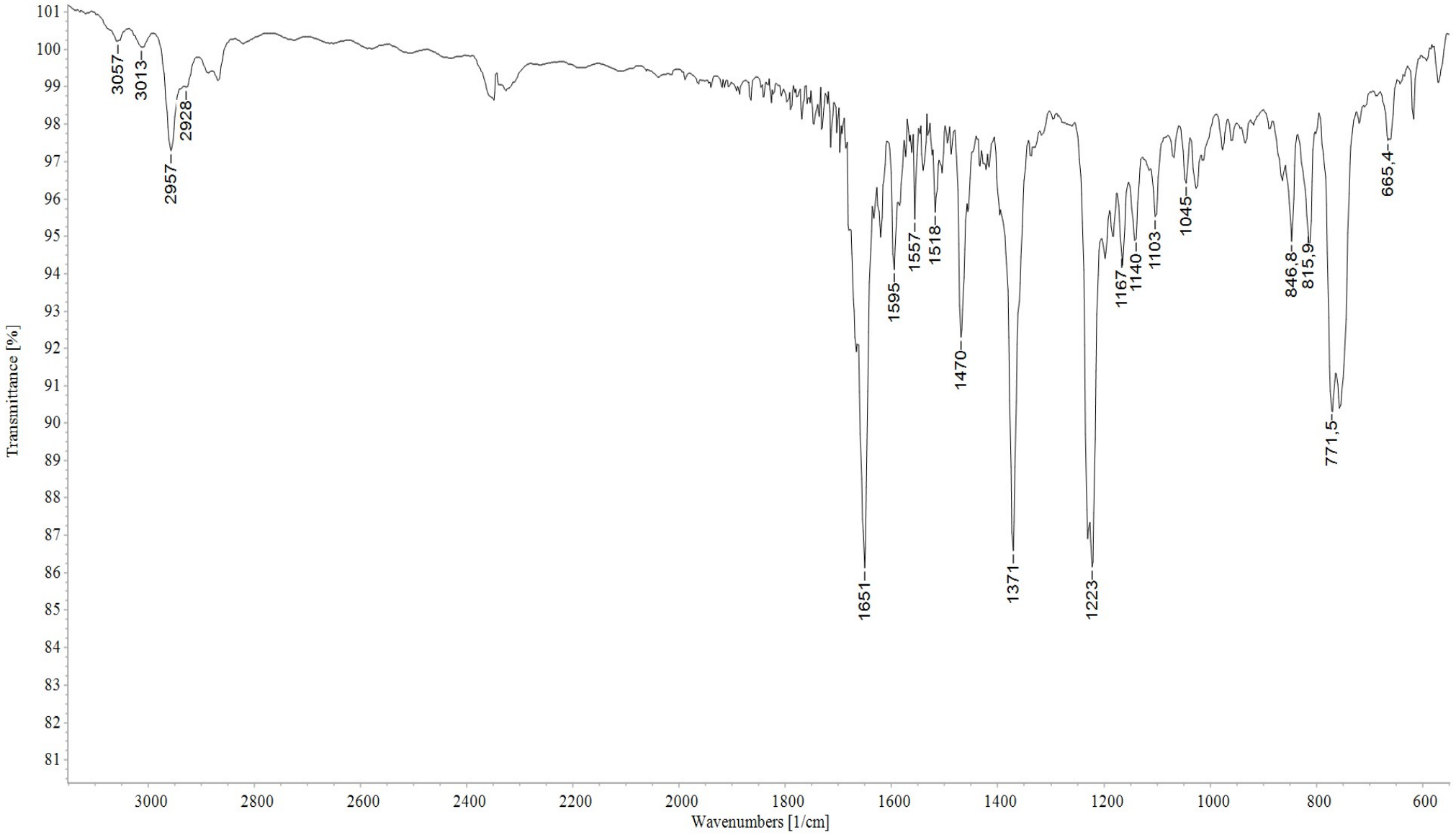

Figure S13 - FT-IR spectrum of 9,9-dimethyl-12-(2,4-dichlorophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4e), NaCl

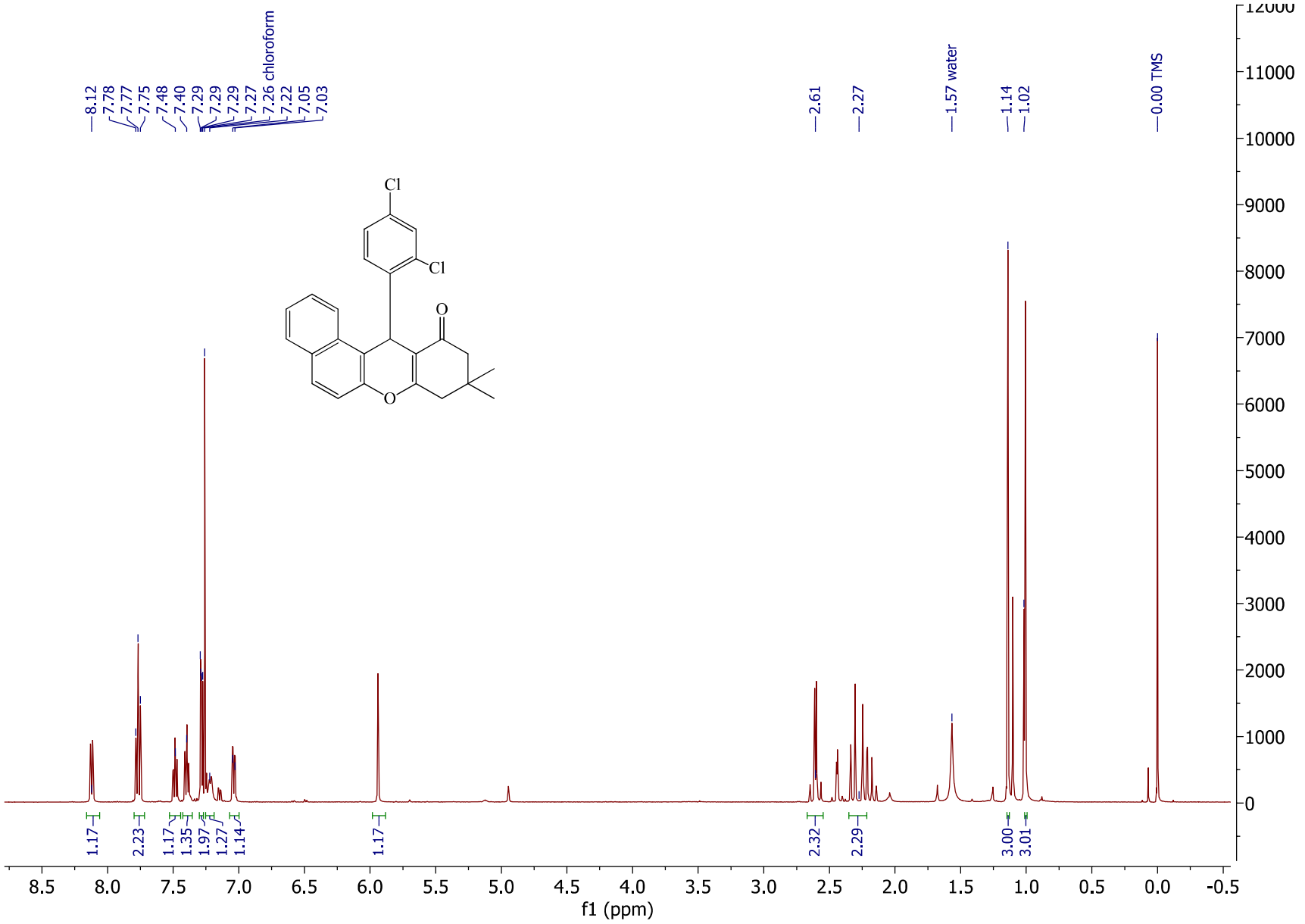

Figure S14 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(2,4-dichlorophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4e), in chloroform-d

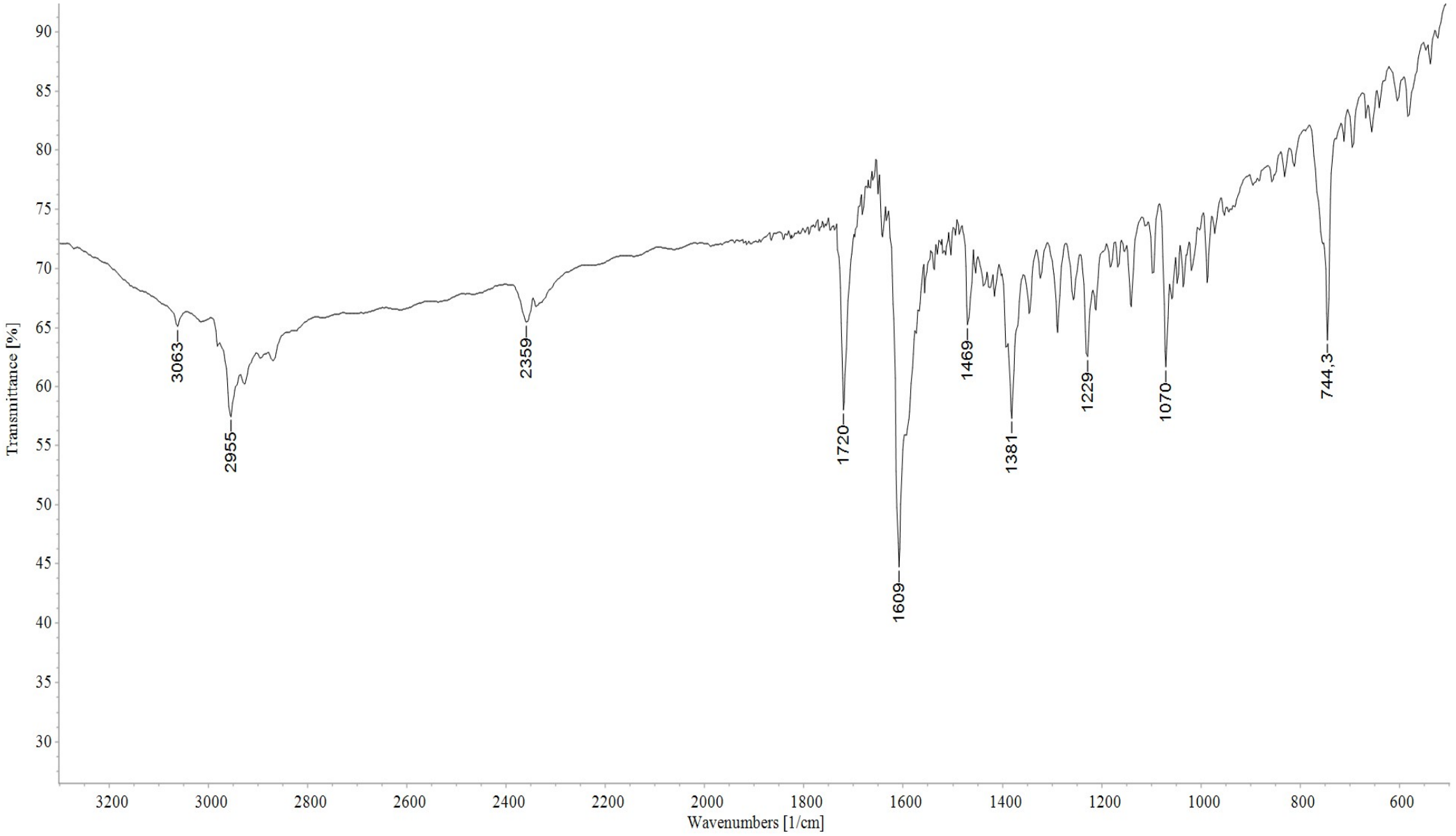

Figure S15 - FT-IR spectrum of 9,9-dimethyl-12-(2-chlorophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4f), NaCl

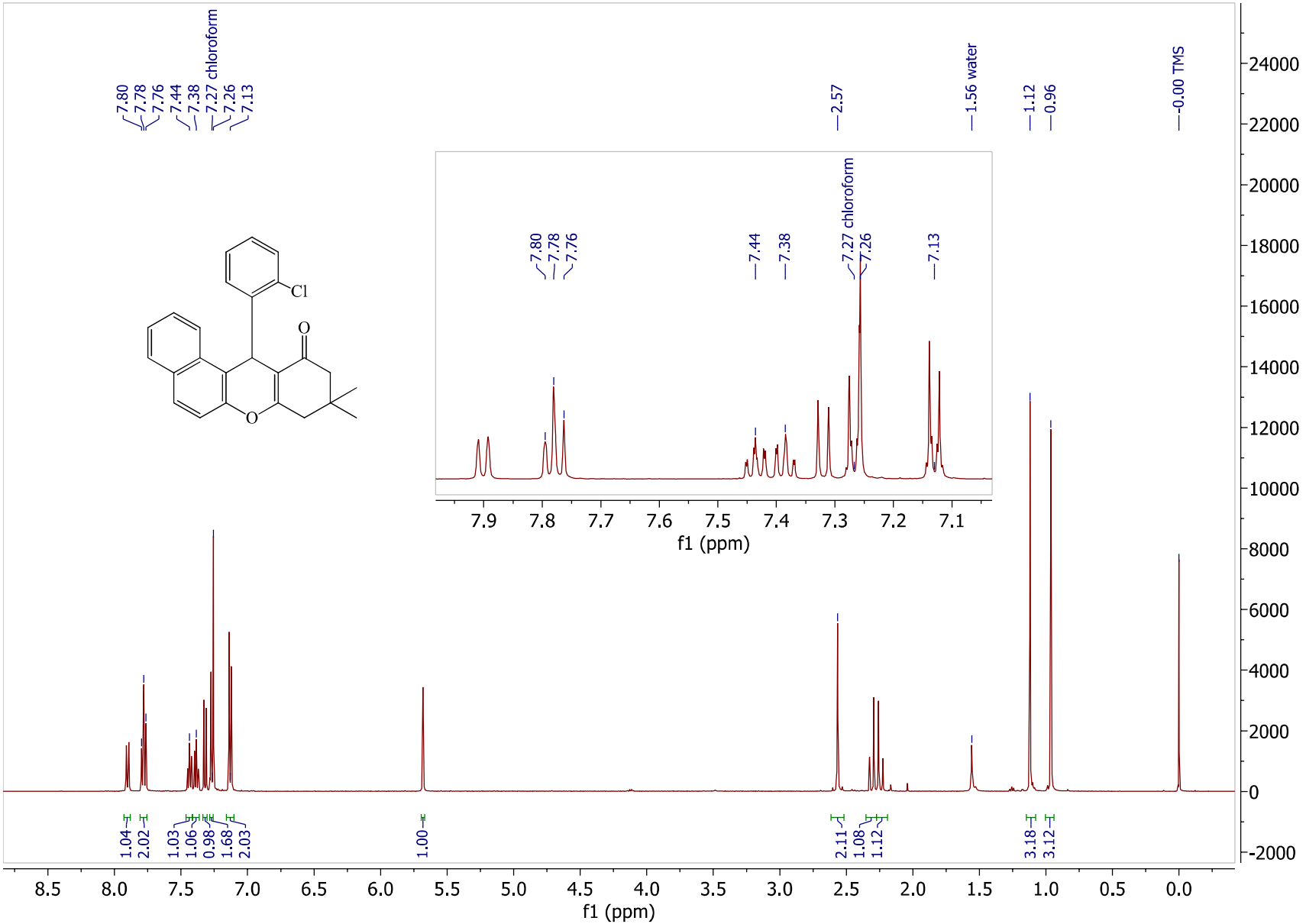

Figure S16 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(2-chlorophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4f), in chloroform-d

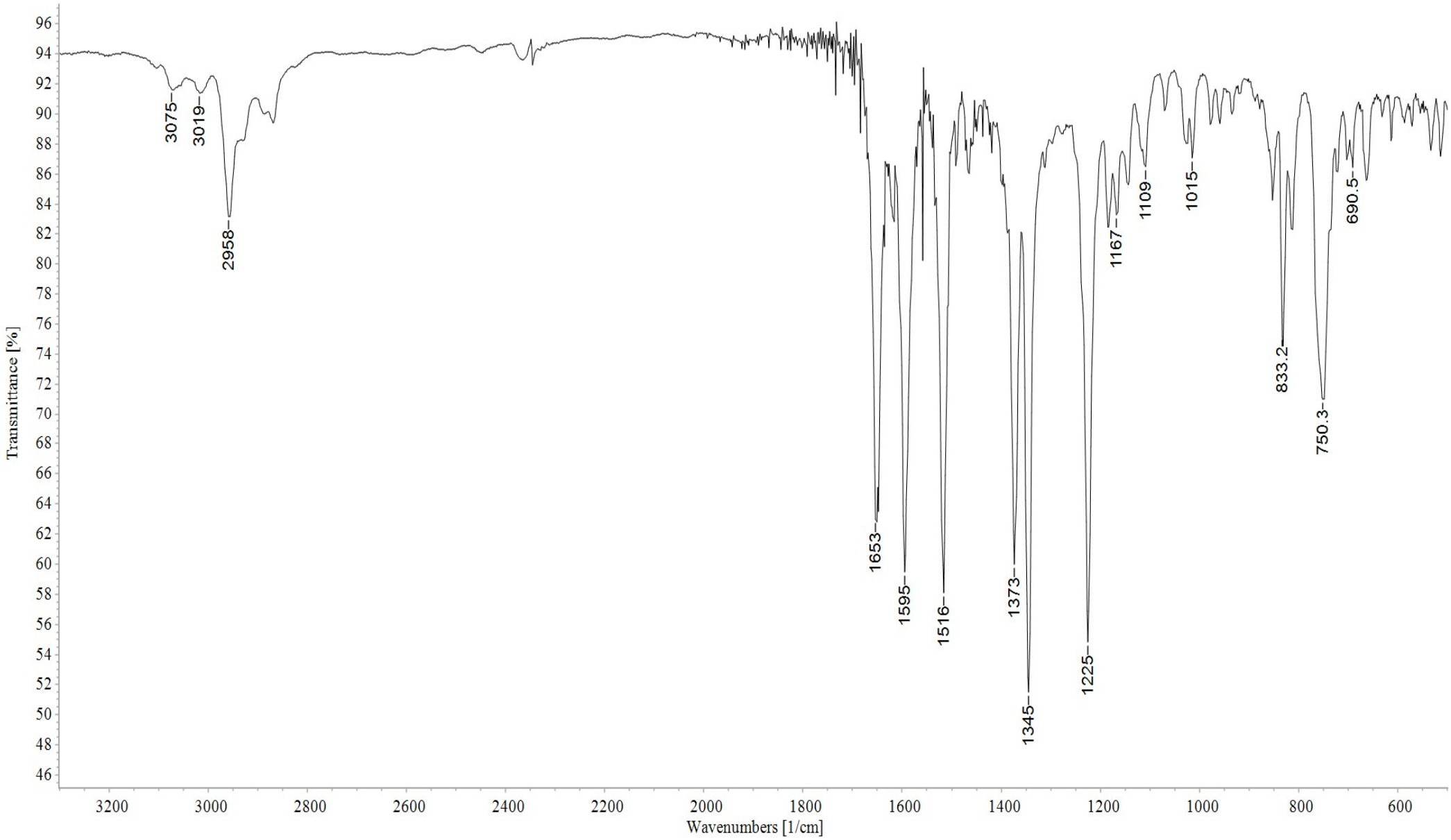

Figure S17 - FT-IR spectrum of 9,9-dimethyl-12-(4-nitrophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4g), NaCl

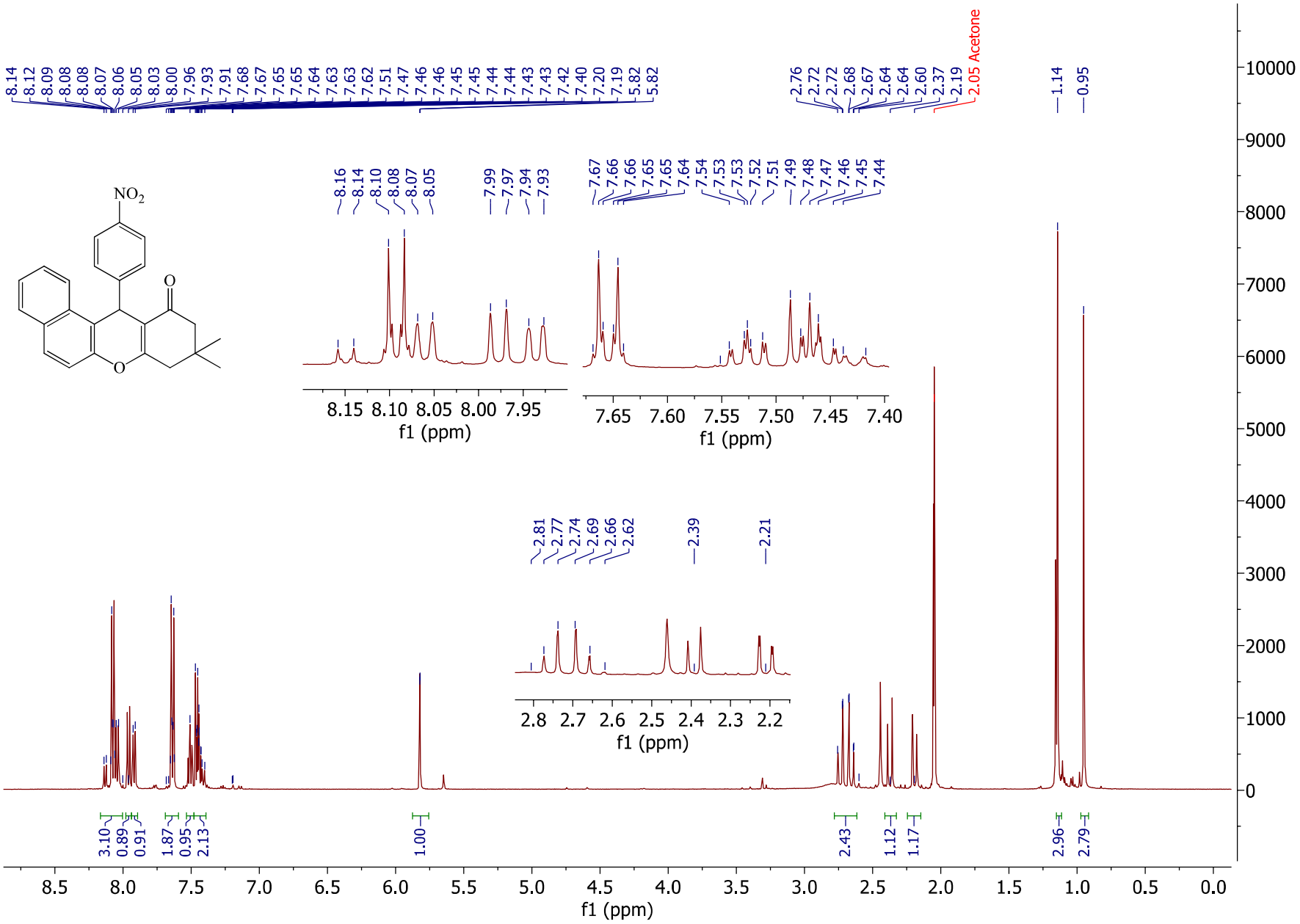

Figure S18 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(4-nitrophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4g), in acetone-d_6

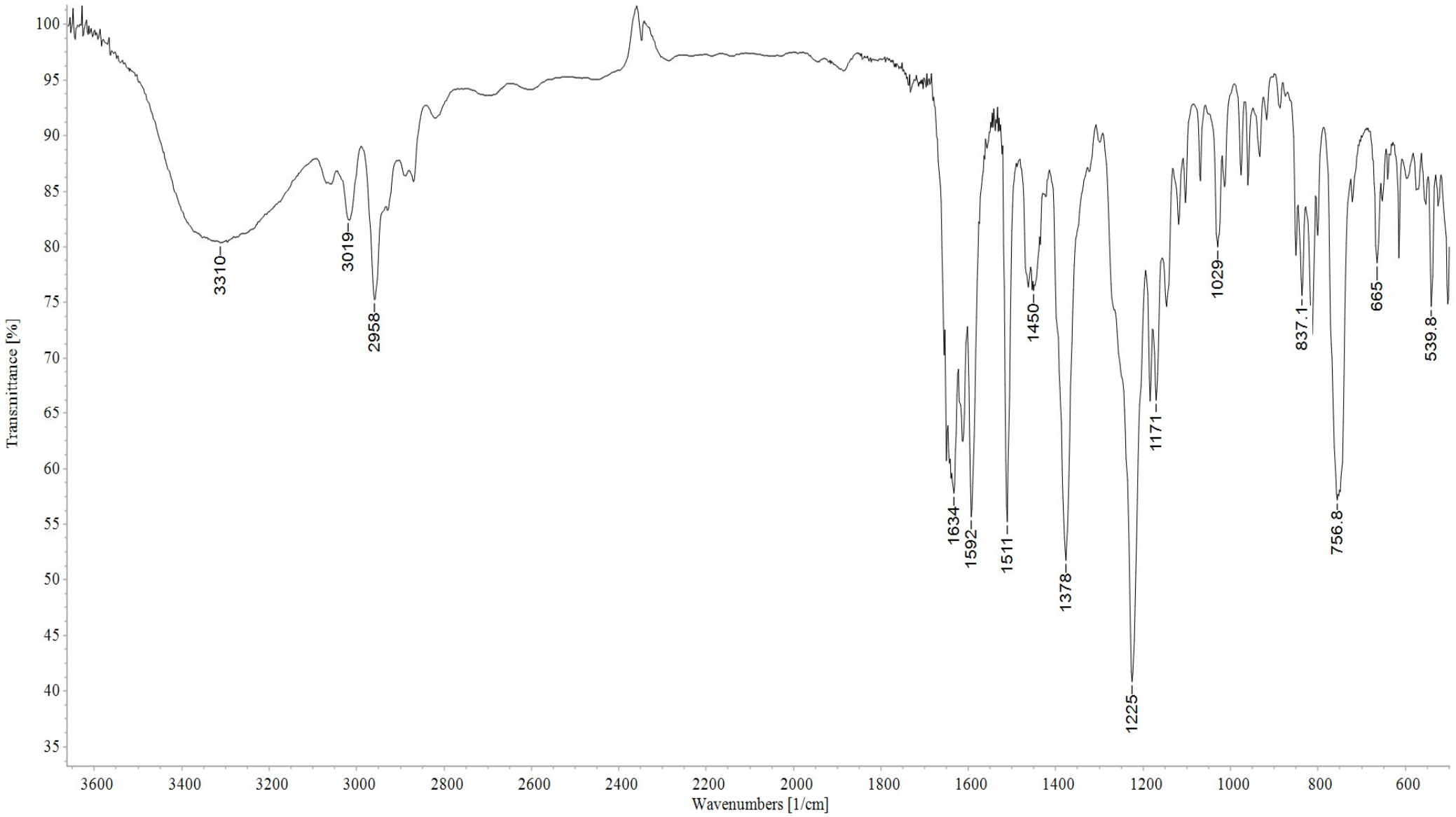

Figure S19 - FT-IR spectrum of 9,9-dimethyl-12-(4-hydroxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4h), NaCl

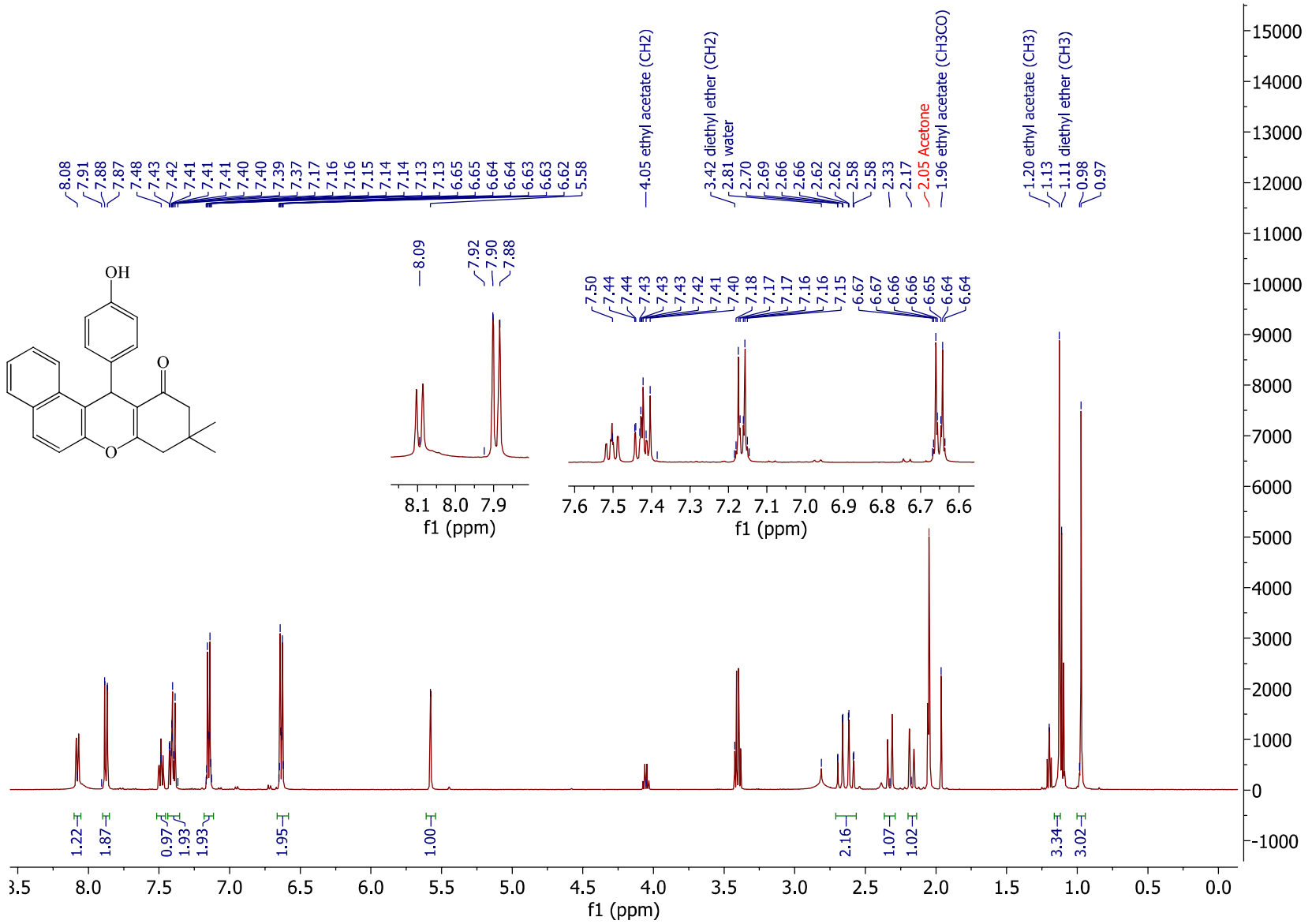

Figure S20 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(4-hydroxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4h), in acetone-d_6

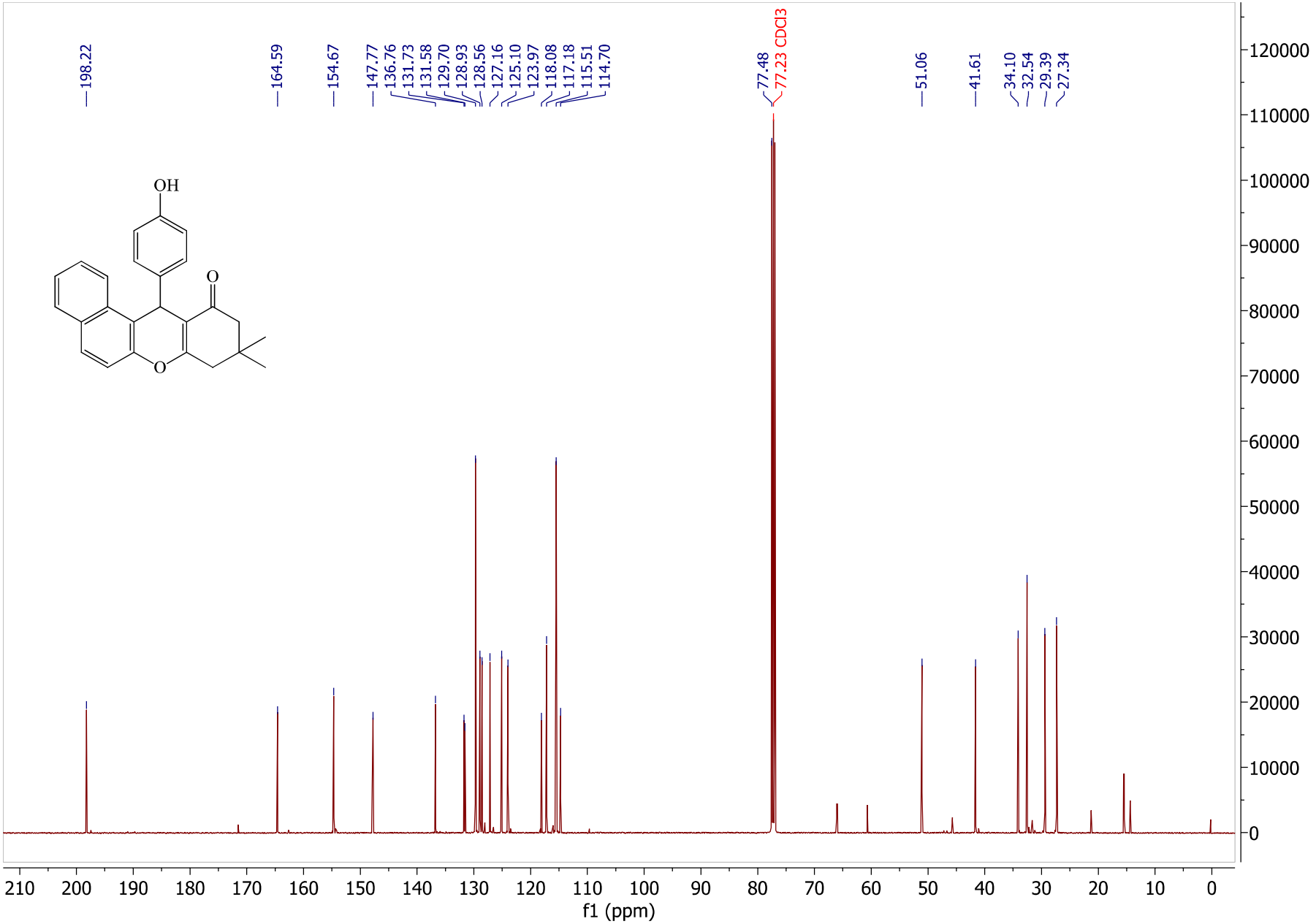

Figure S21 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-(4-hydroxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4h), in chloroform-d

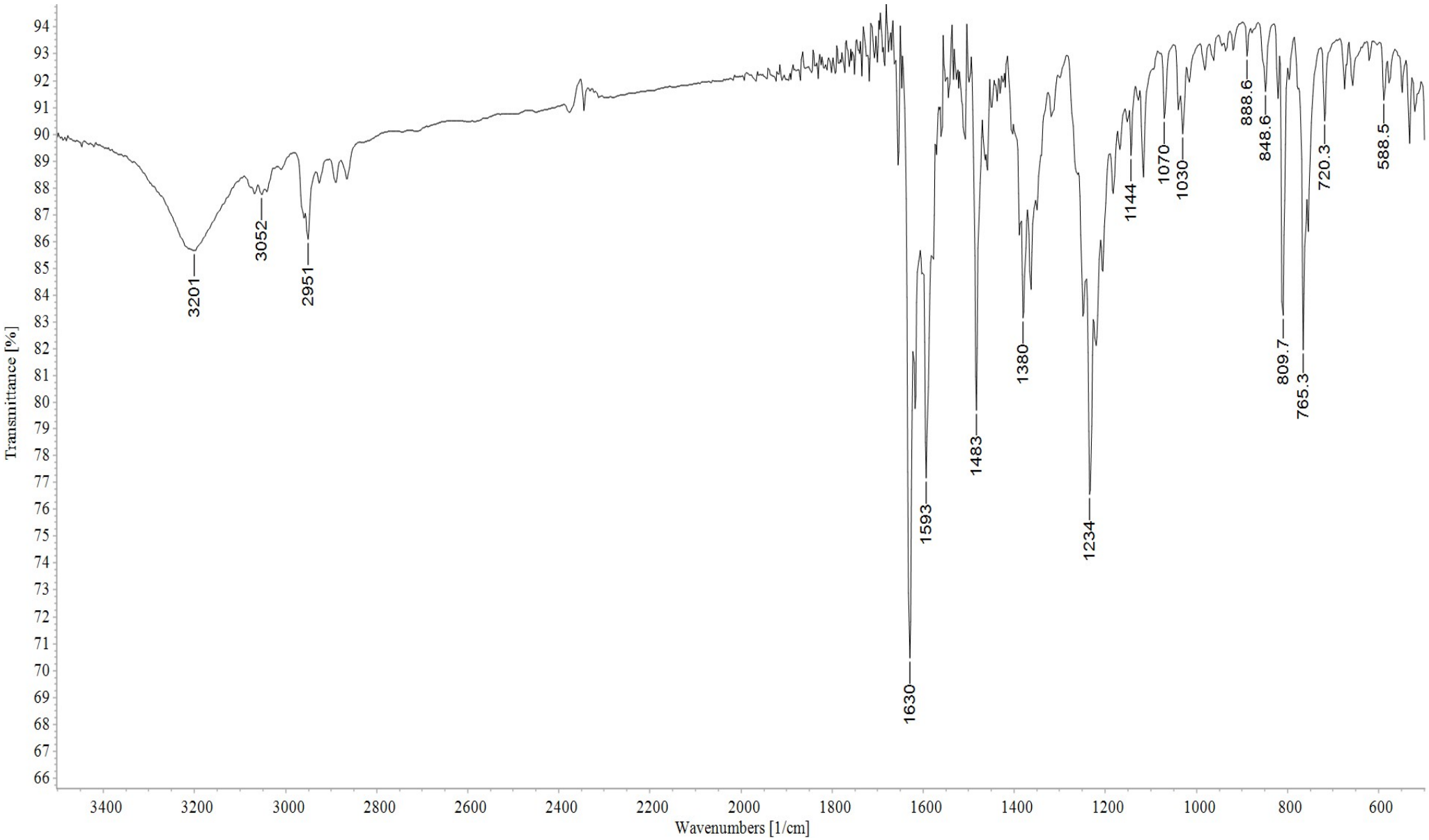

Figure S22 - FT-IR spectrum of 9,9-dimethyl-12-(2-hydroxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4i), NaCl

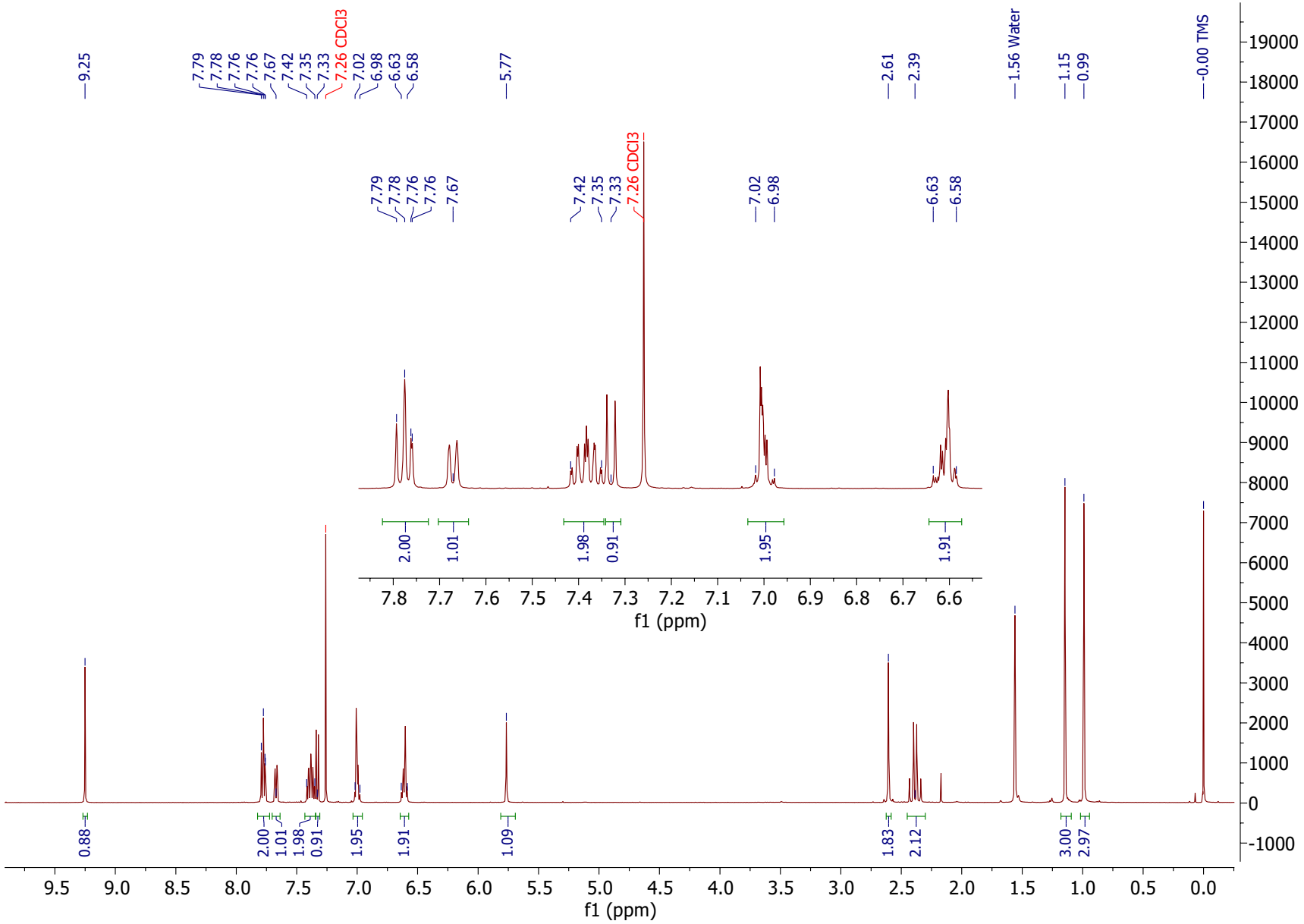

Figure S23 - ^{1}H NMR spectrum of 9,9-dimethyl-12-(2-hydroxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4i), in DMSO-d$_6$

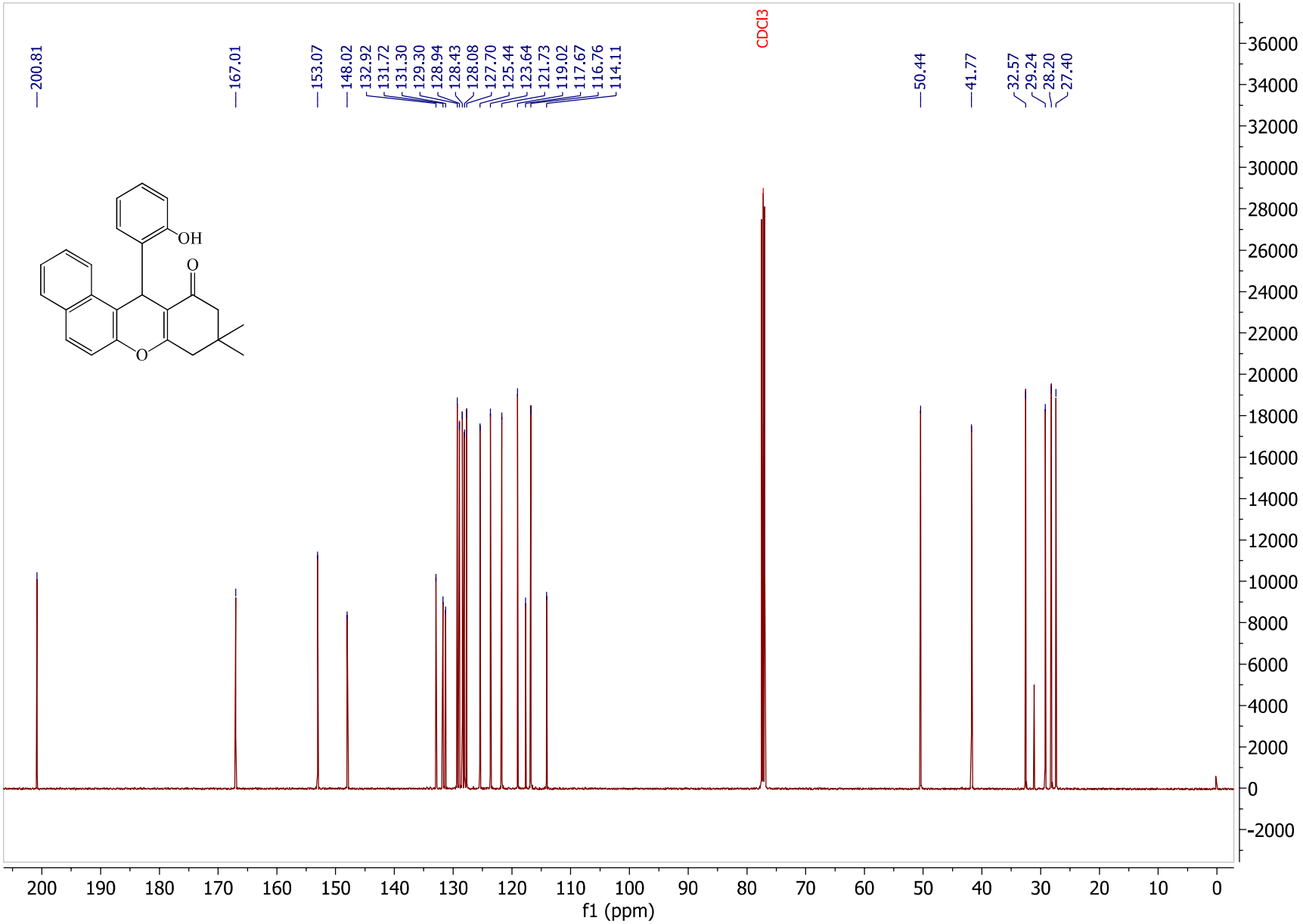

Figure S24 -^{13}C-NMR spectrum of 9,9-dimethyl-12-(4-hydroxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one one (4i), in chloroform-d_6

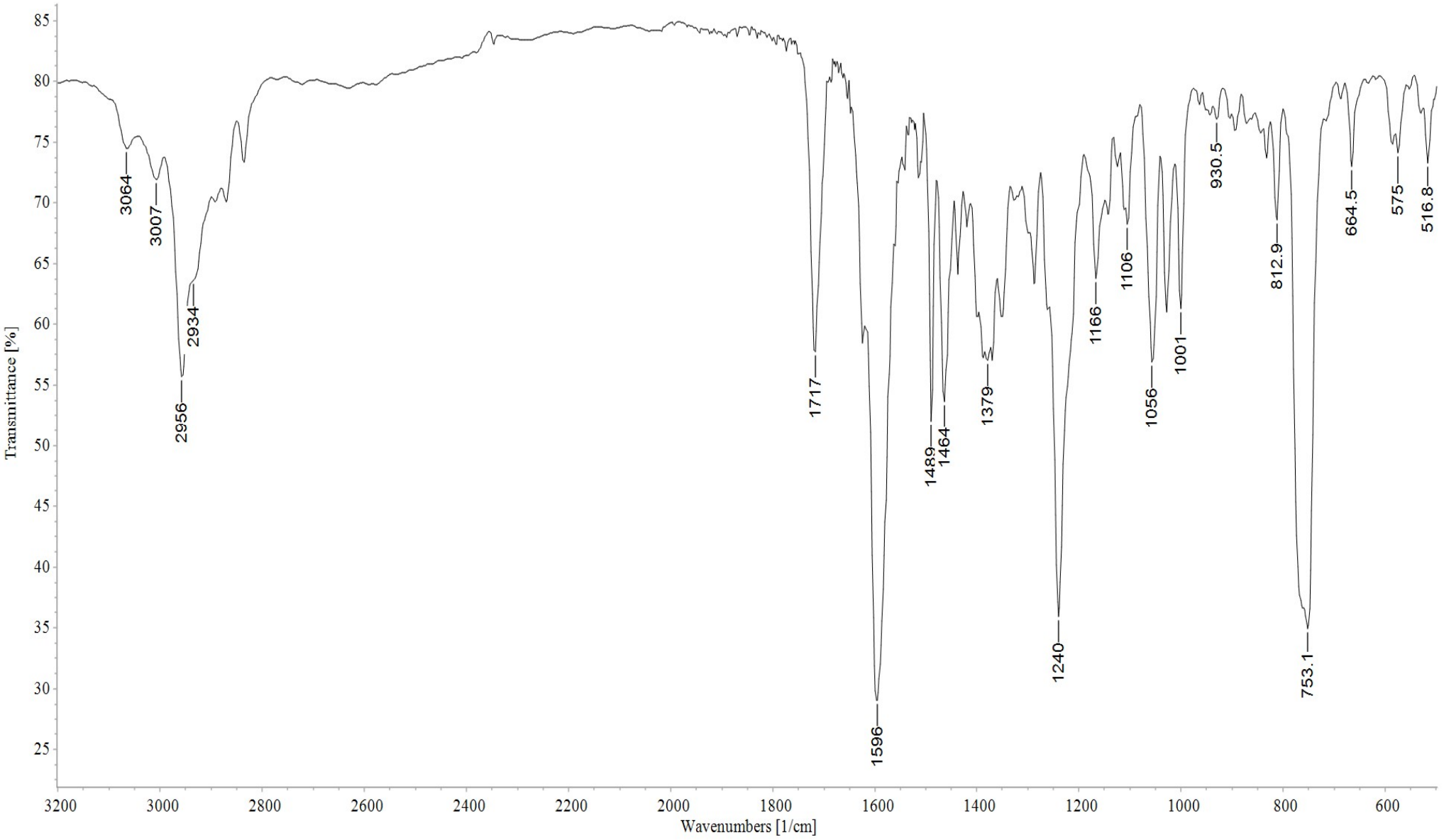

Figure S25 - FT-IR spectrum of 9,9-dimethyl-12-(2-methoxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4j), NaCl

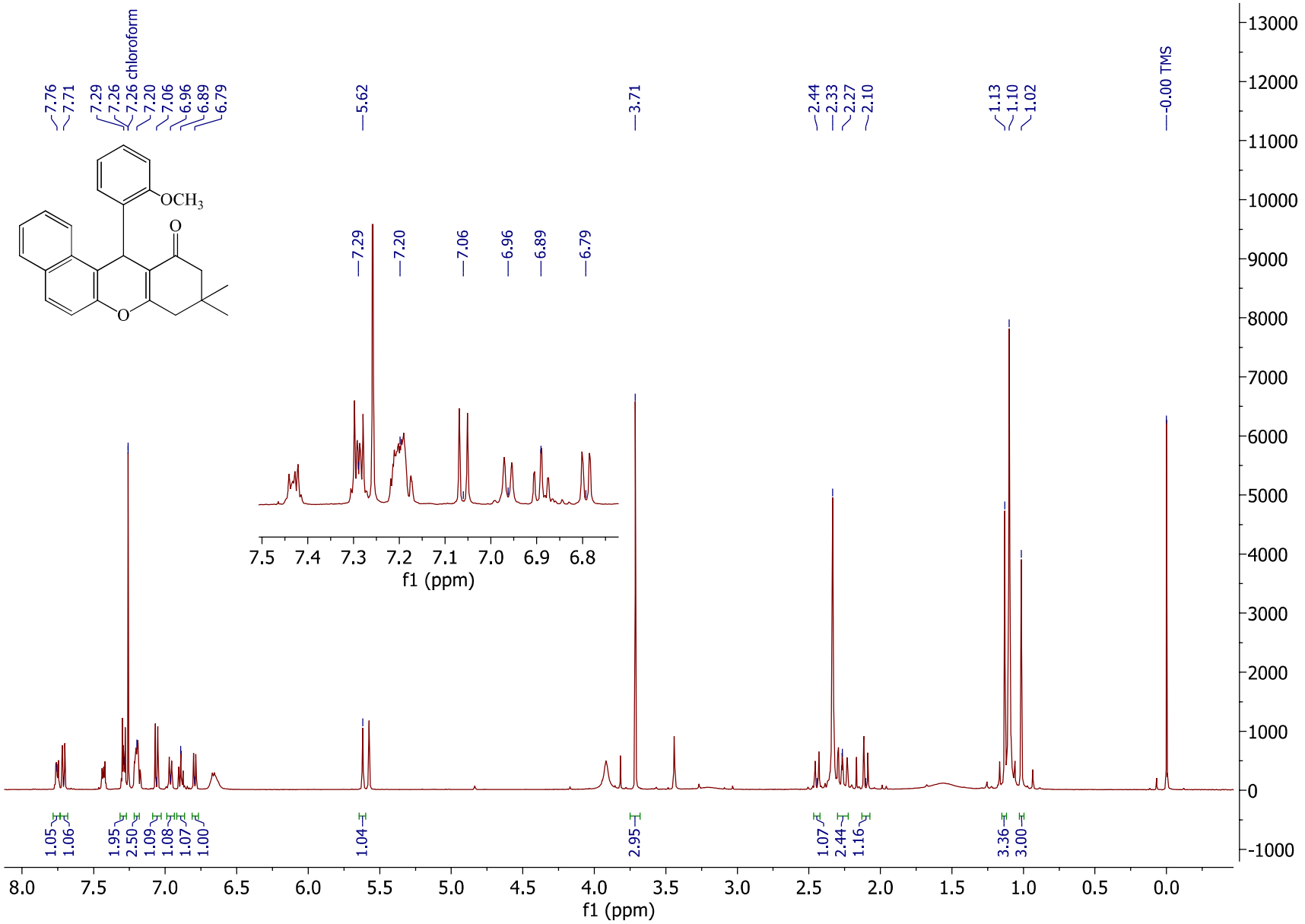

Figure S26 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(2-methoxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4j), in chloroform-d

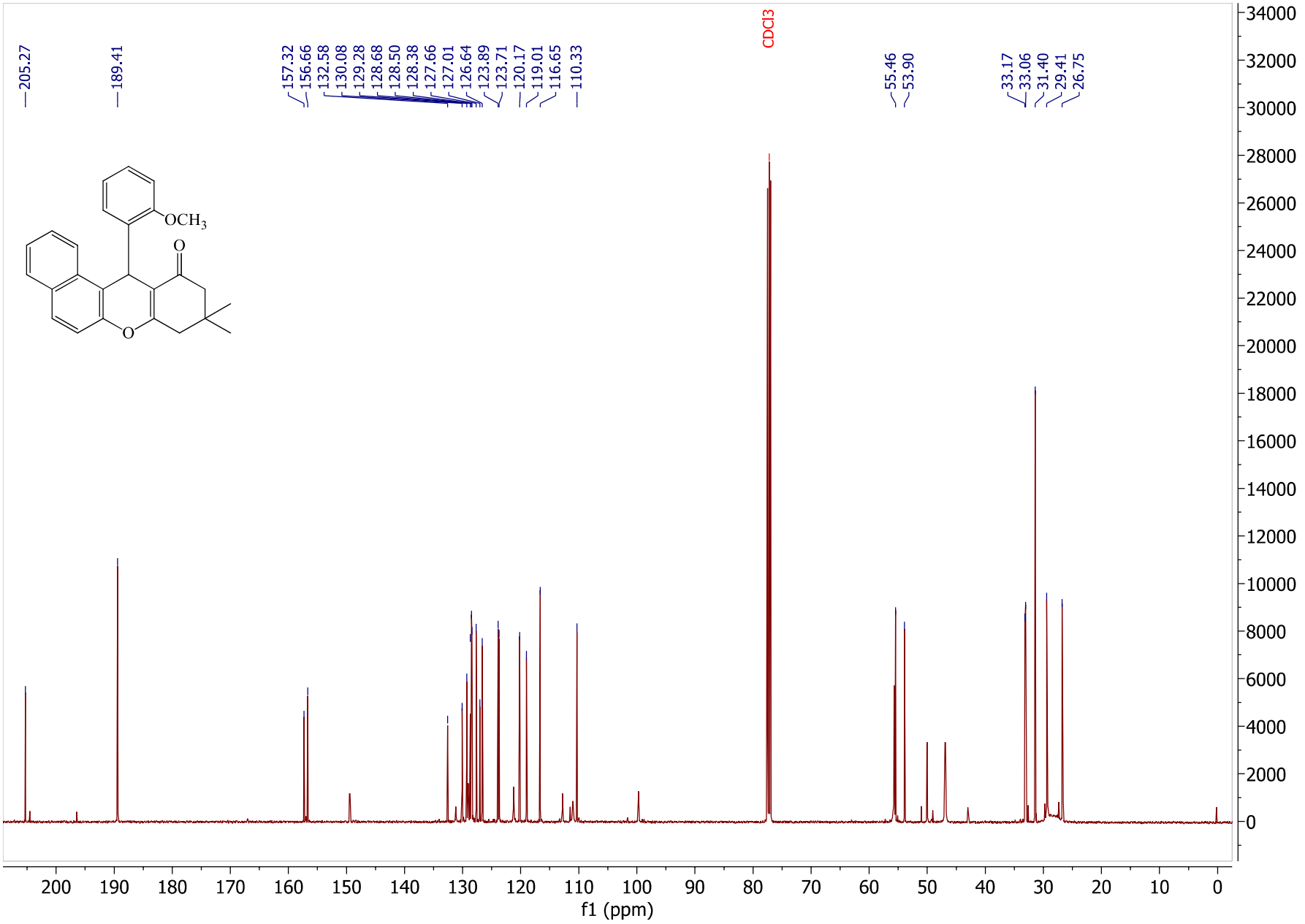

Figure S27 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-(2-methoxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4j), in chloroform-d

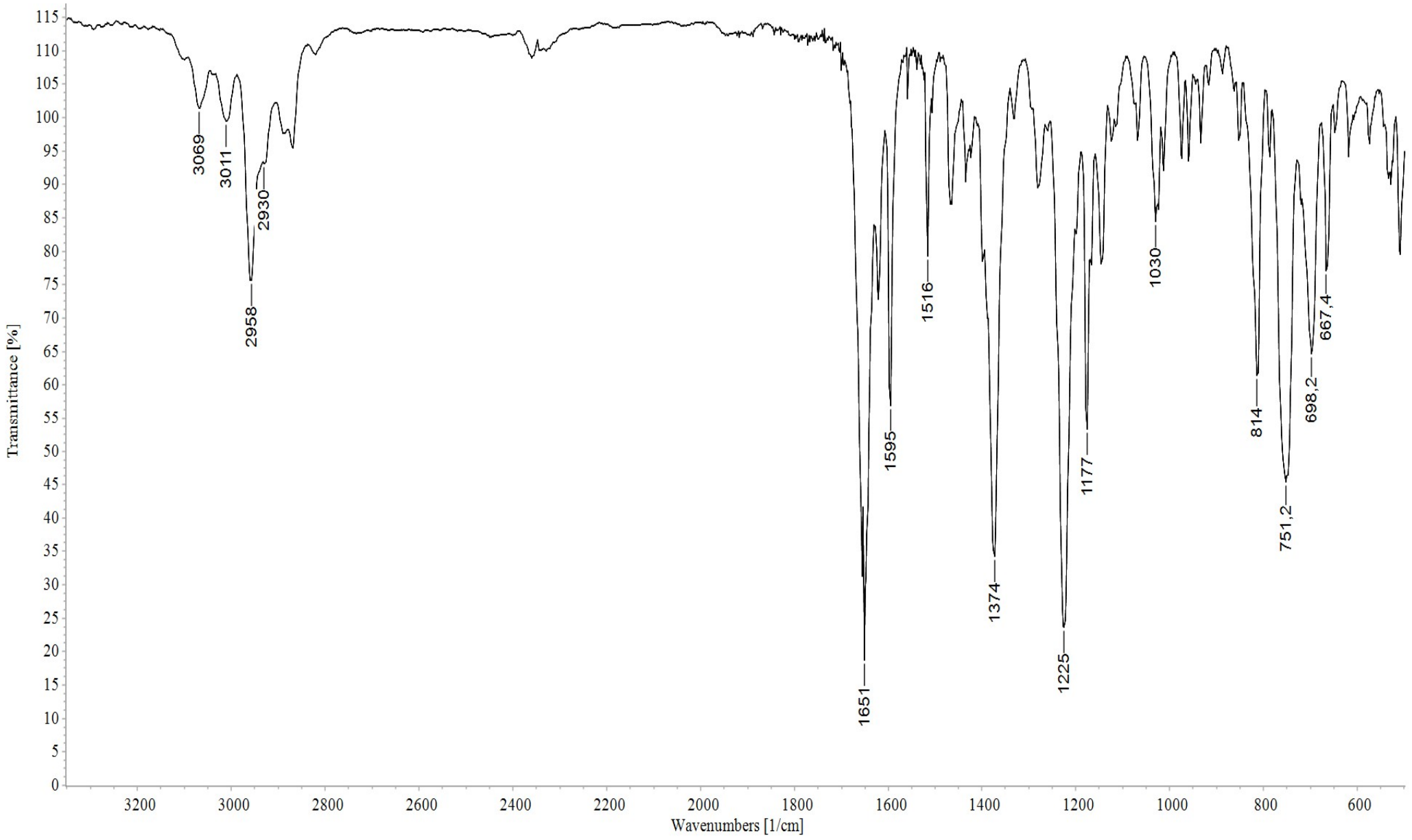

Figure S28 - FT-IR spectrum of 9,9-dimethyl-12-(furan-2-yl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4k), NaCl

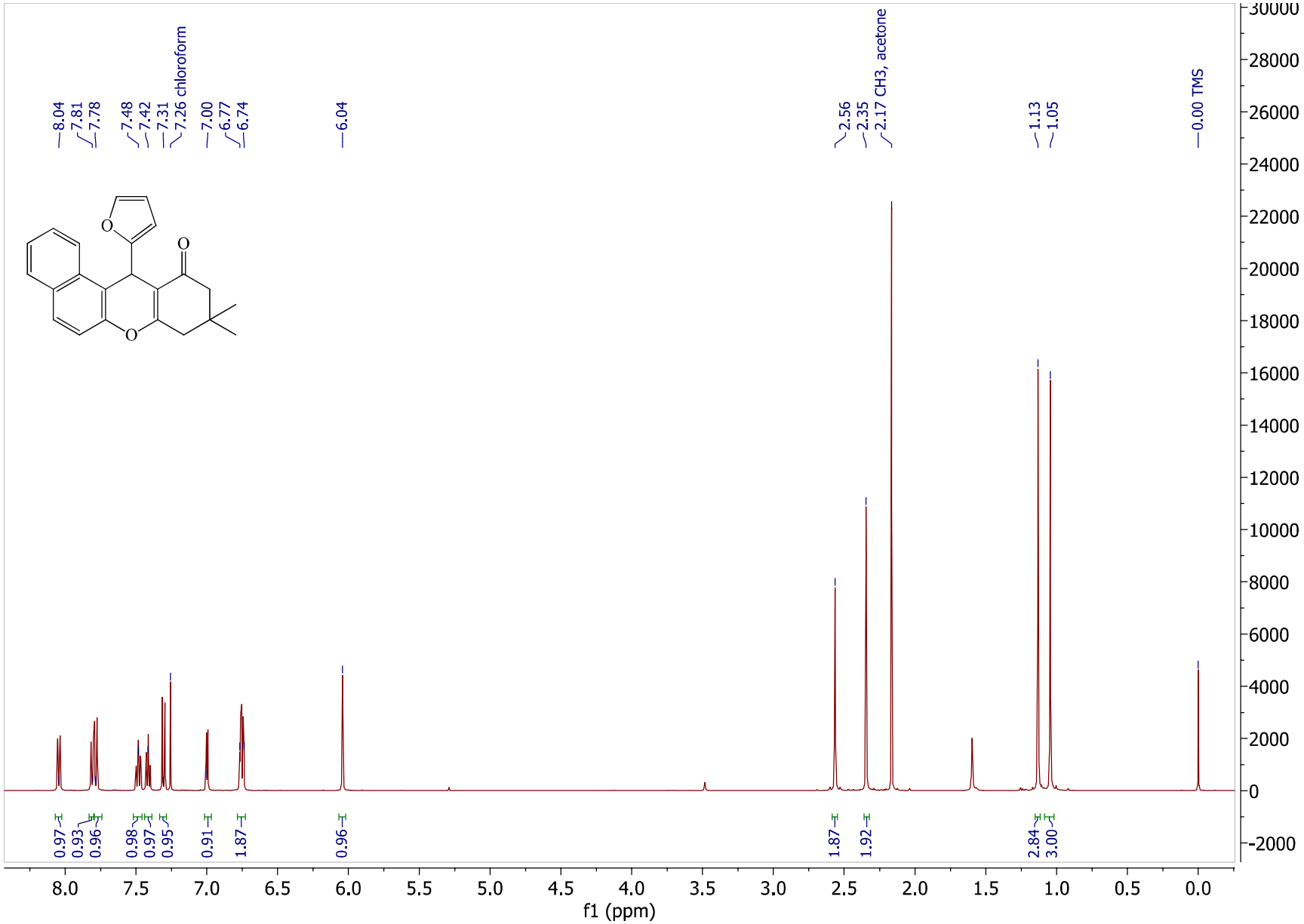

Figure S29 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(furan-2-yl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4k), in chloroform-d

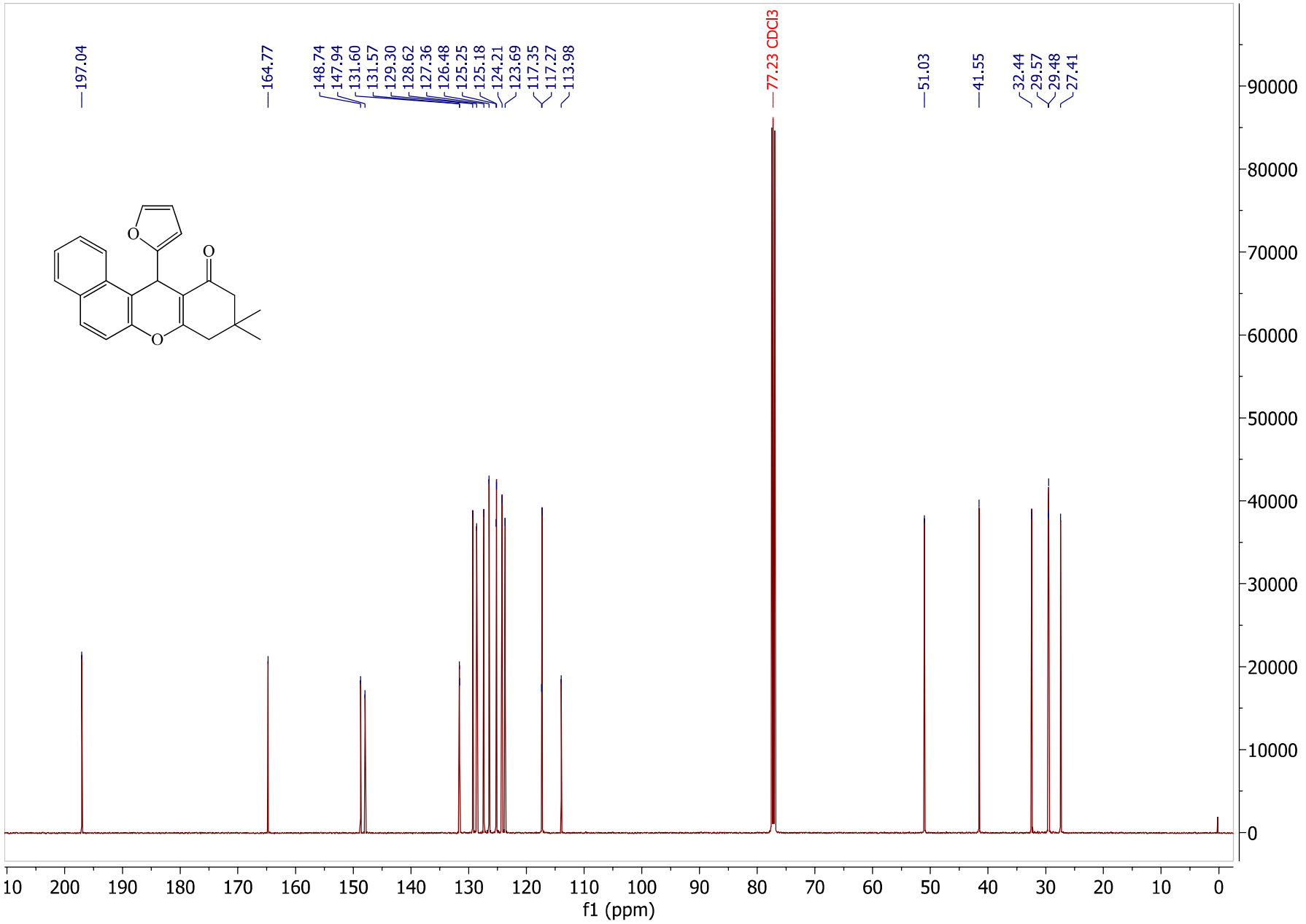

Figure S30 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-(furan-2-yl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4k), in chloroform-d

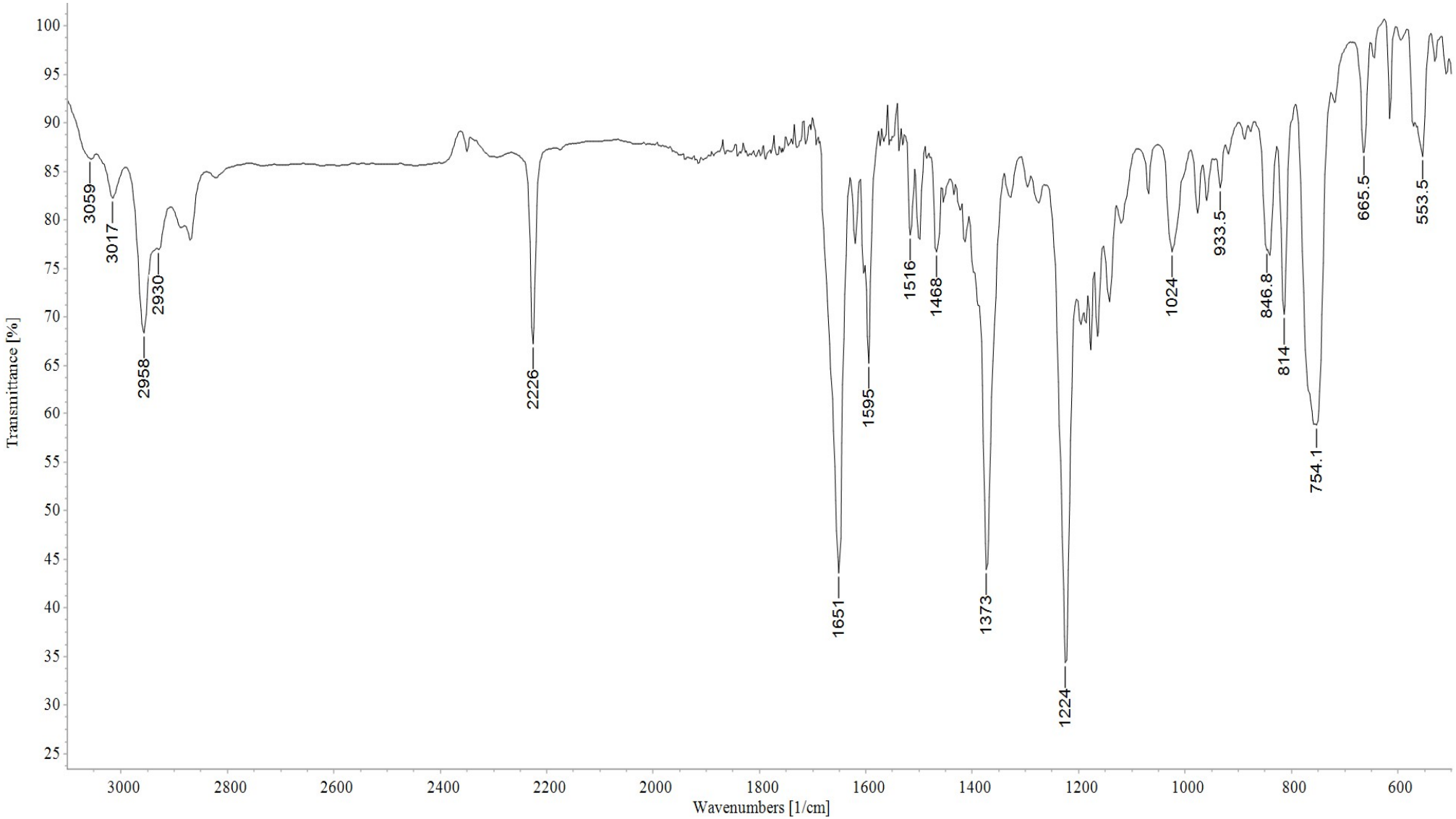

Figure S31 - FT-IR spectrum of 9,9-dimethyl-12-(4-cyanophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4l), NaCl

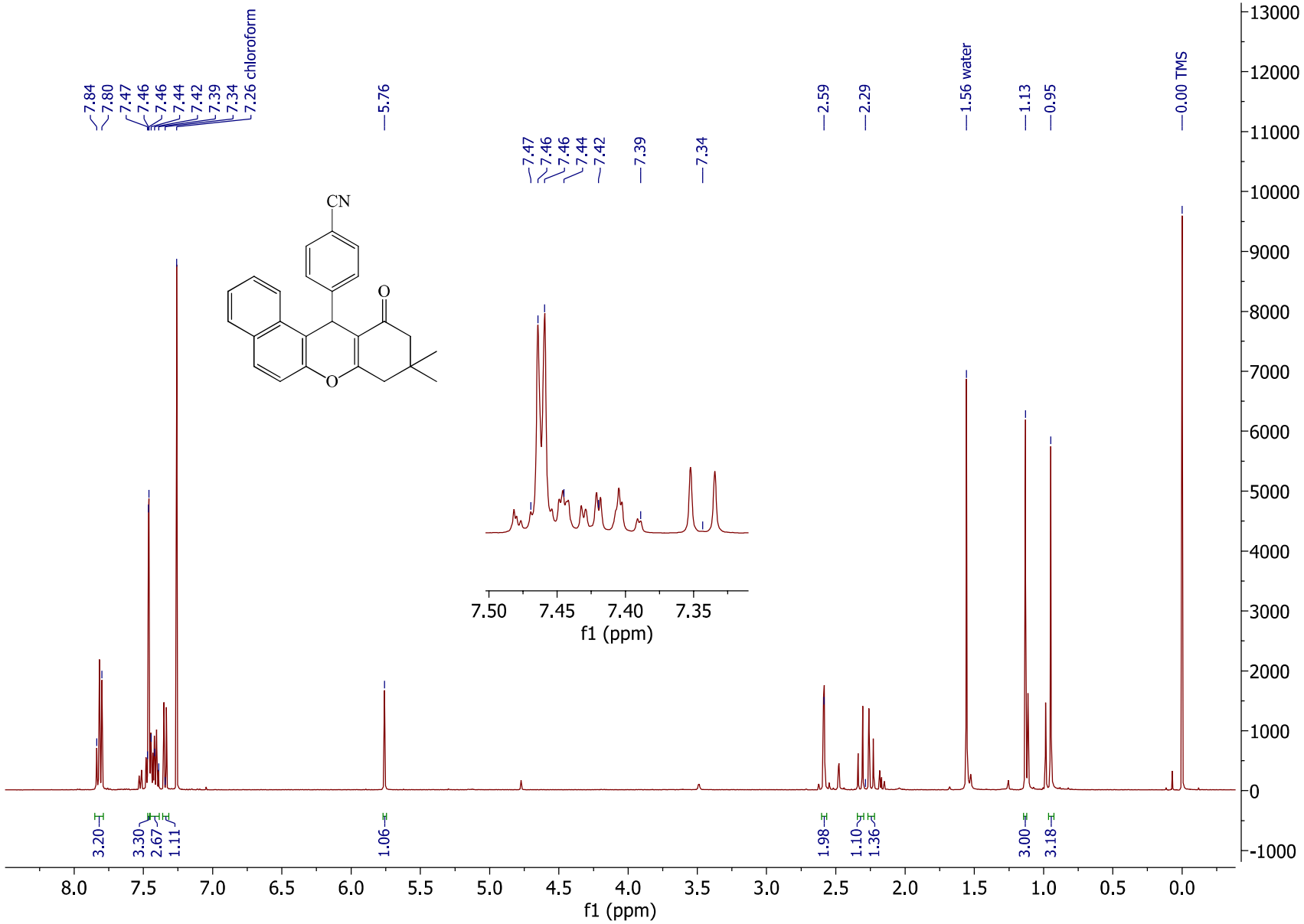

Figure S32 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(4-cyanophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4l), in chloroform-d

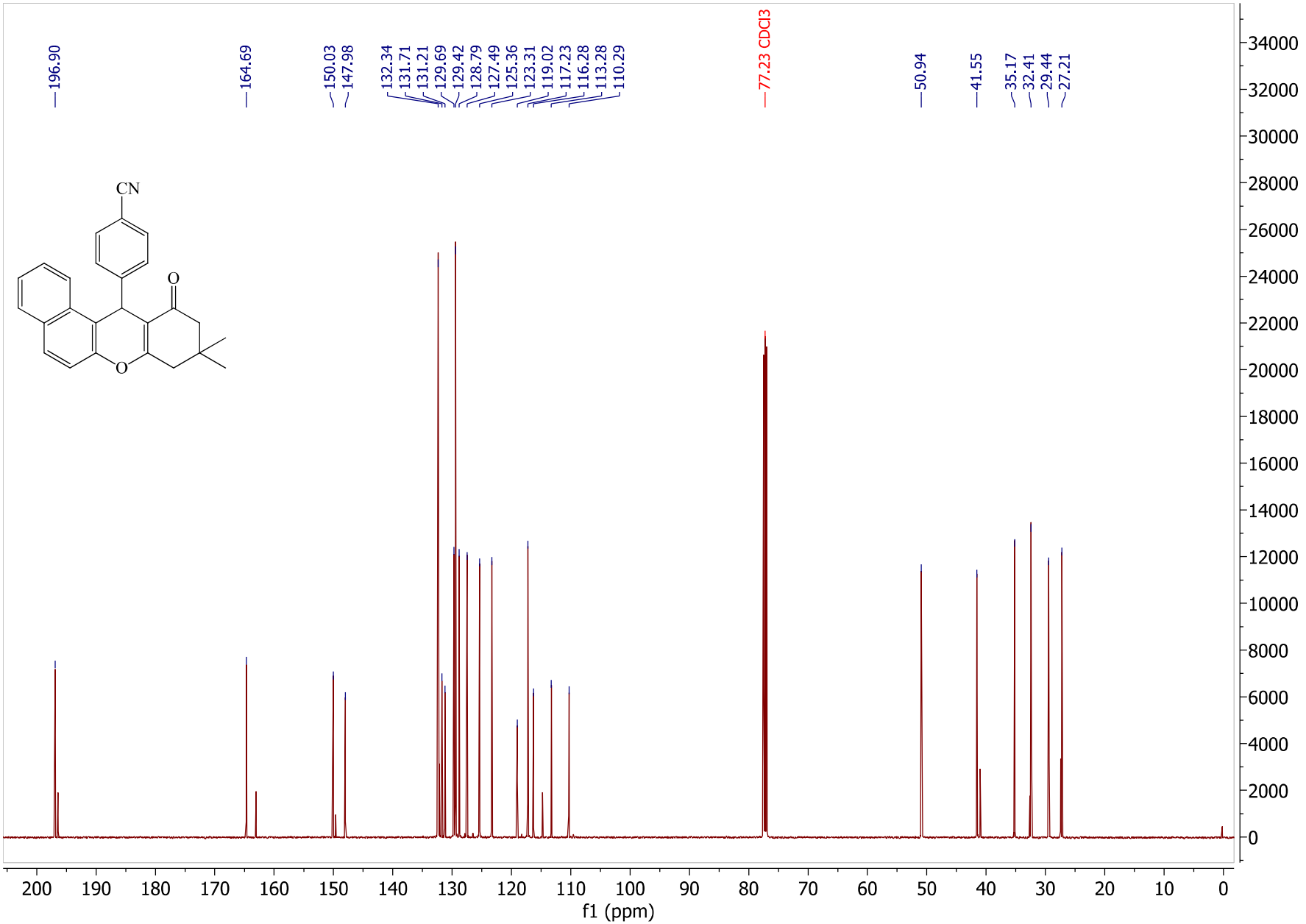

Figure S33 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-(4-cyanophenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4l), in chloroform-d

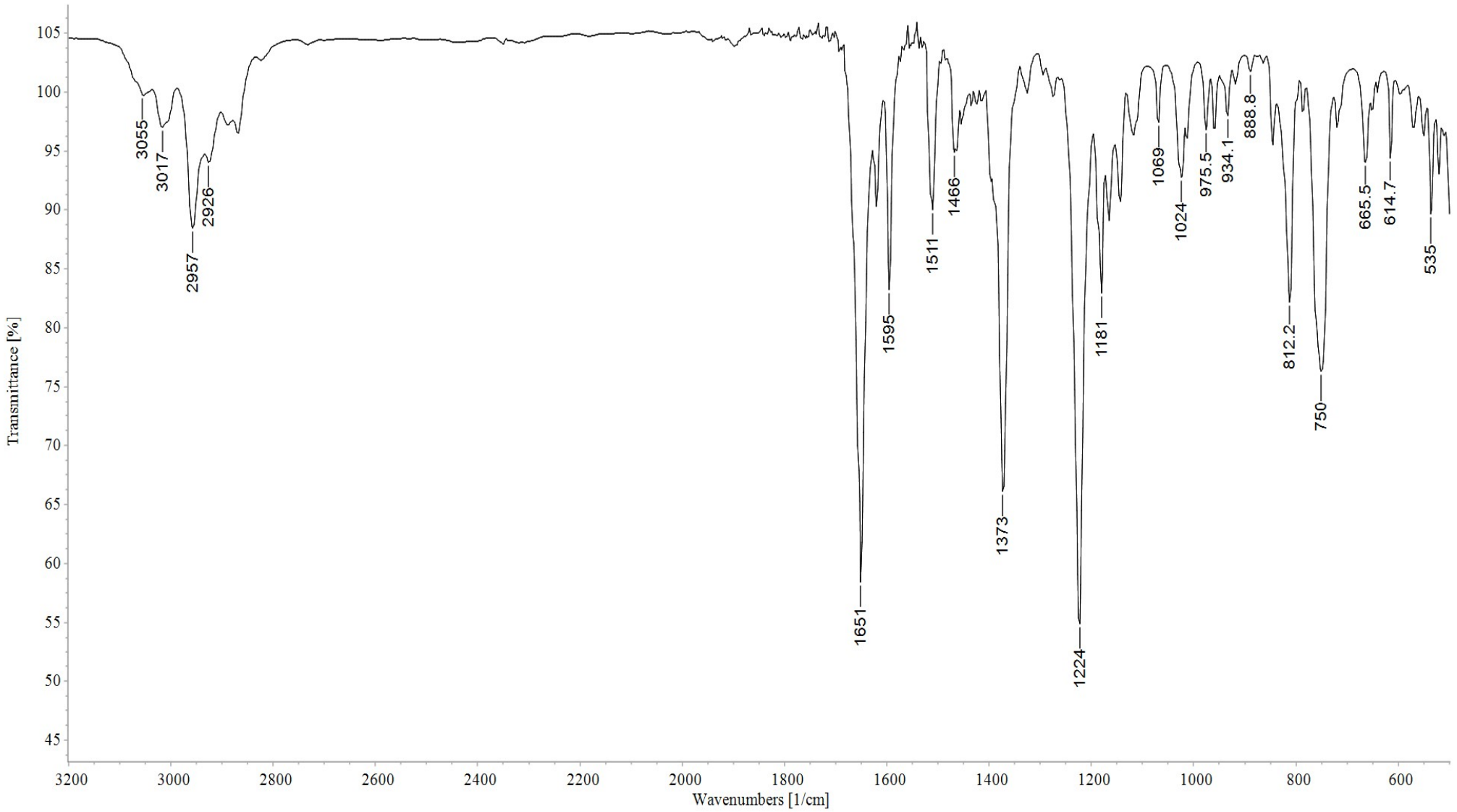

Figure S34 - FT-IR spectrum of 9,9-dimethyl-12-(4-methylphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4m), NaCl

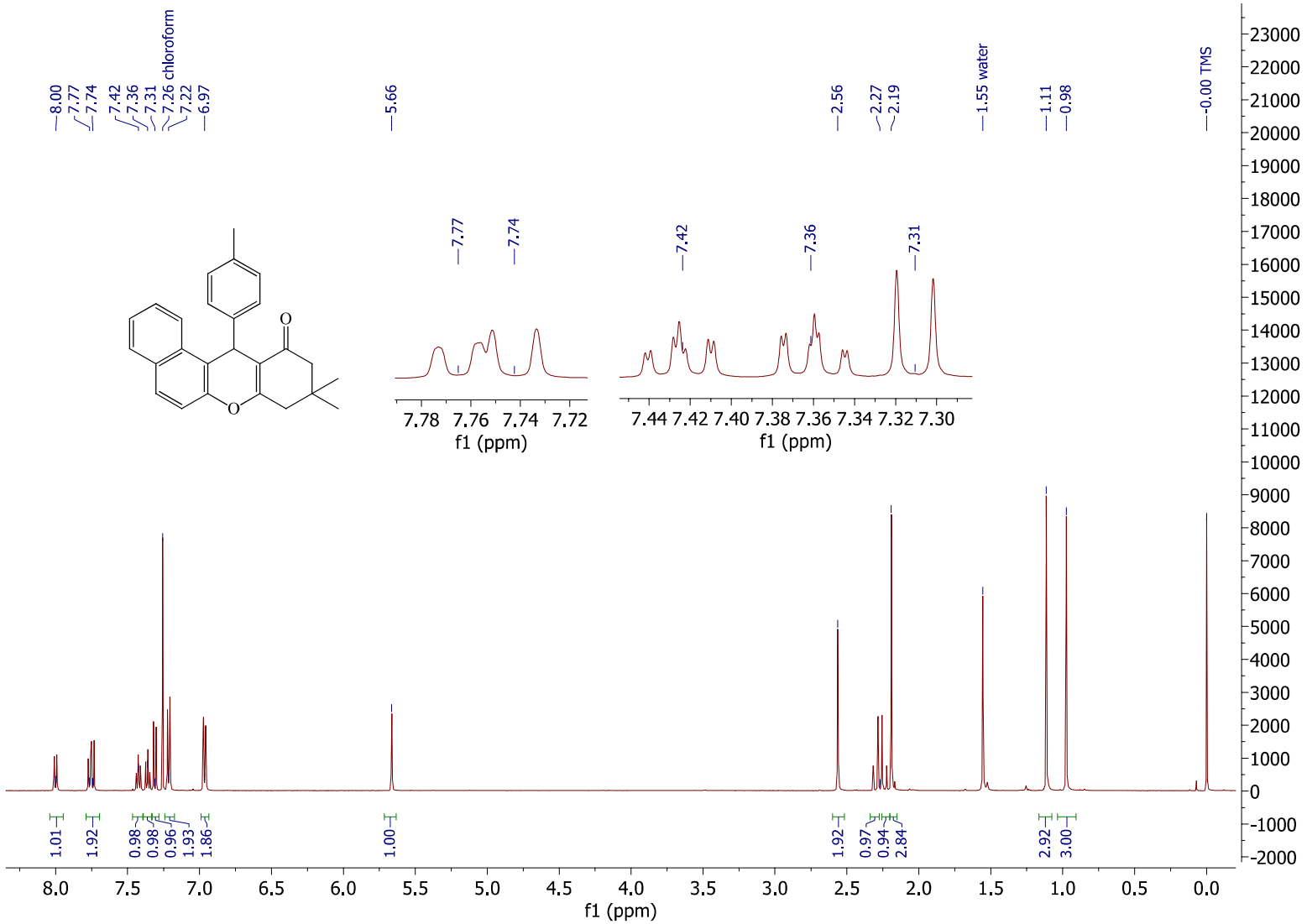

Figure S35 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-(4-methylphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4m), in chloroform-d

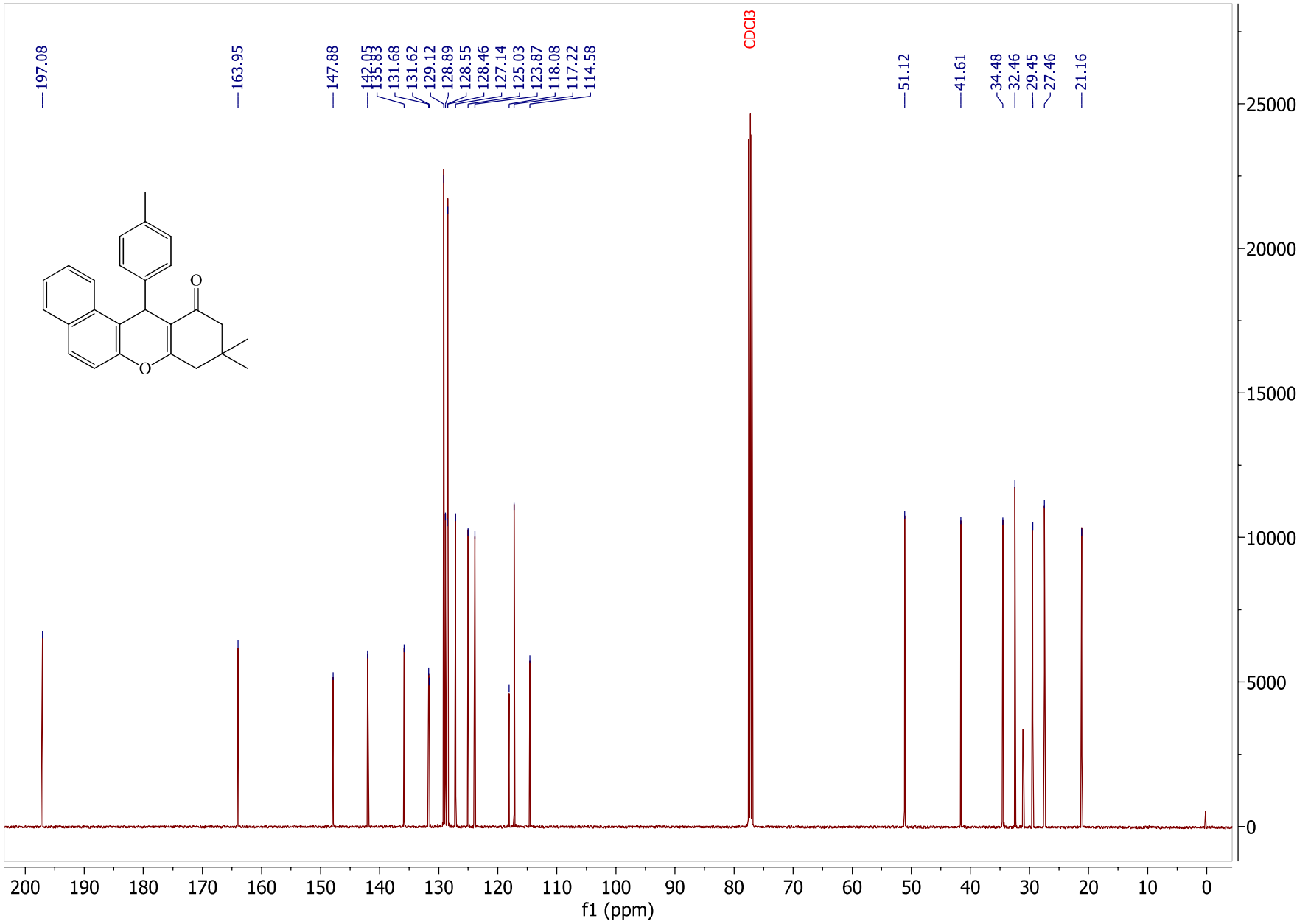

Figure S36 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-(4-methylphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4m), in chloroform-d

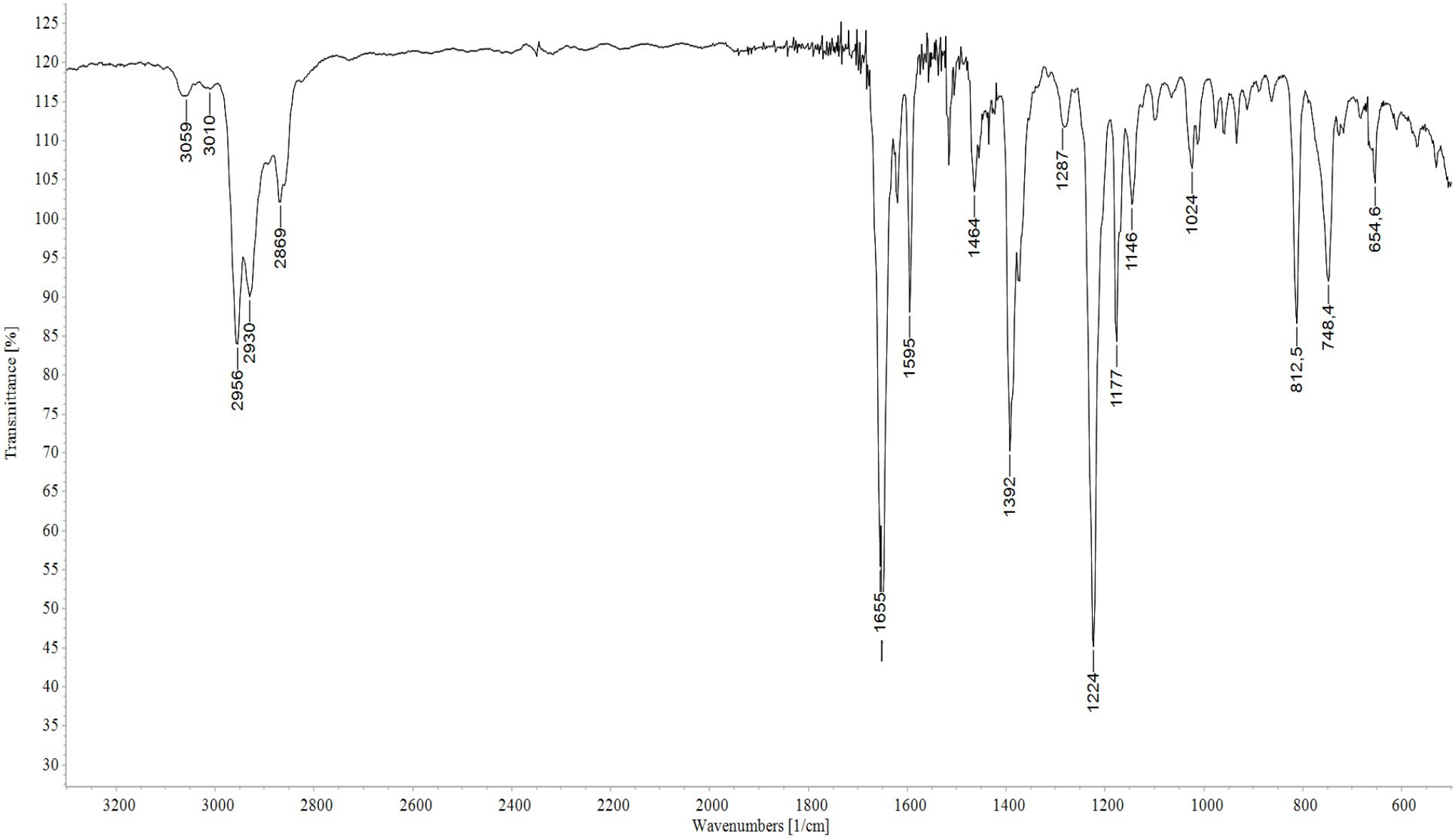

Figure S37 - FT-IR spectrum of 9,9-dimethyl-12-butyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4n), NaCl

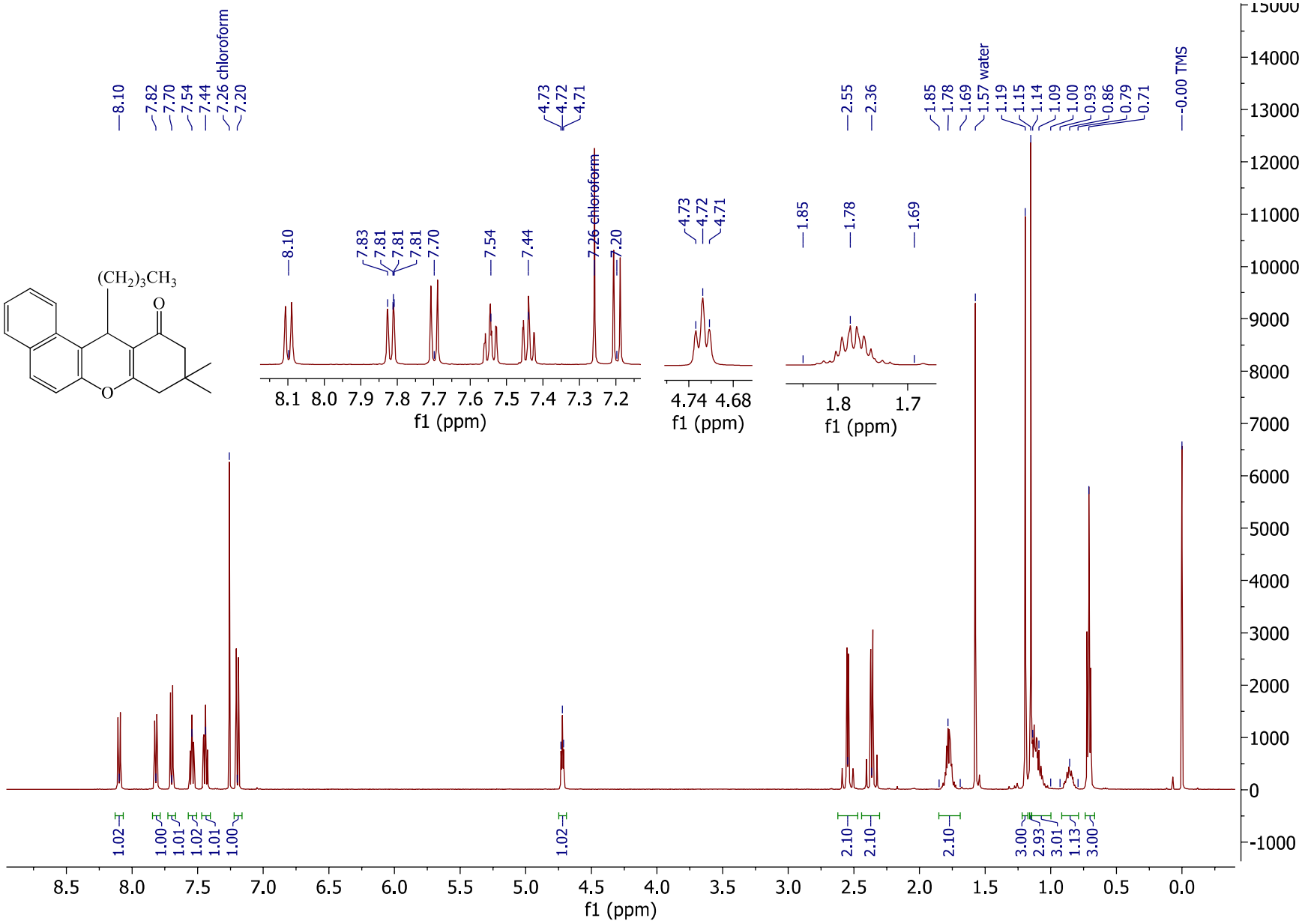

Figure S38 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-butyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4n), in chloroform-d

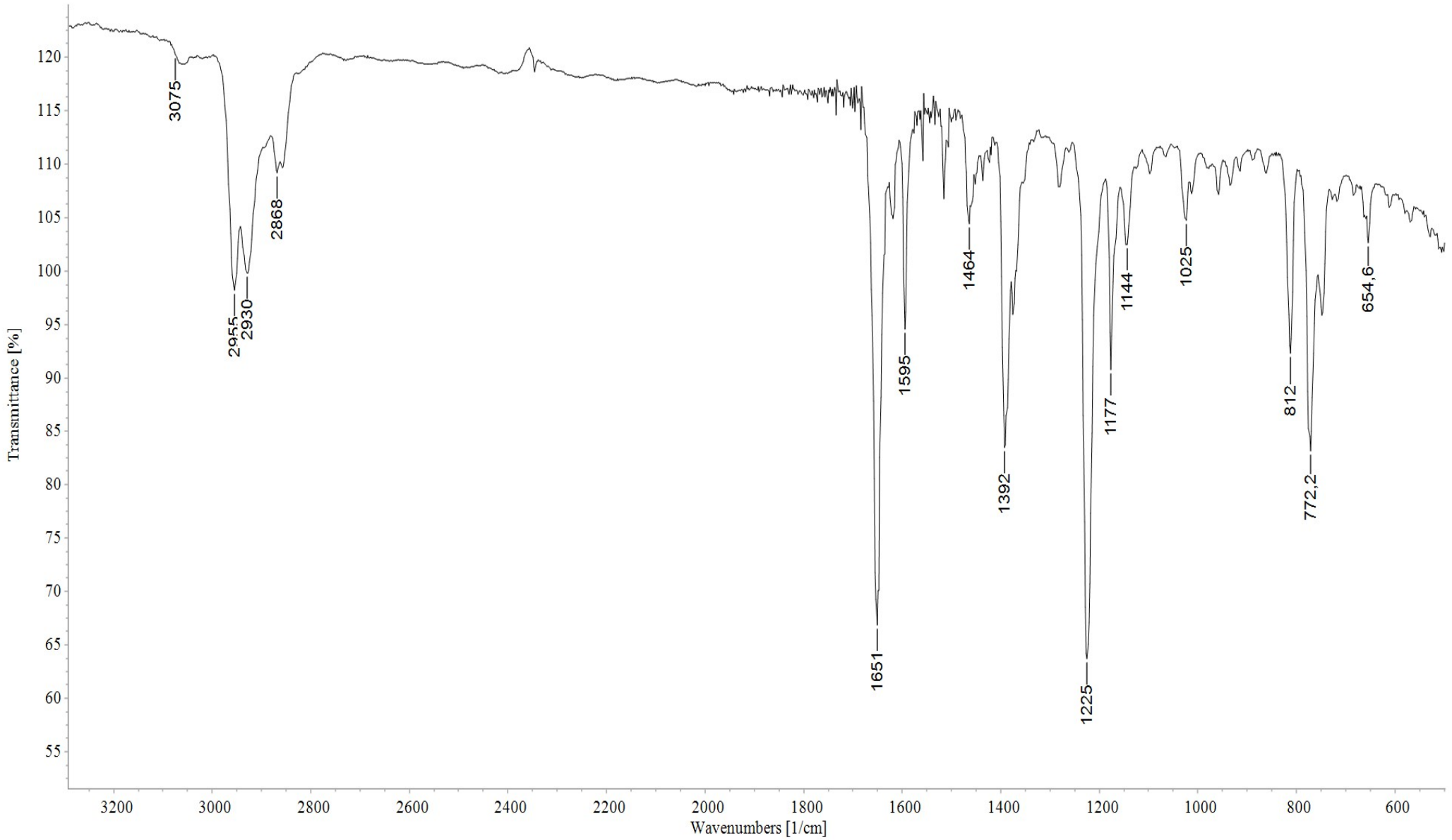

Figure S39 - FT-IR spectrum of 9,9-dimethyl-12-pentyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4o), NaCl

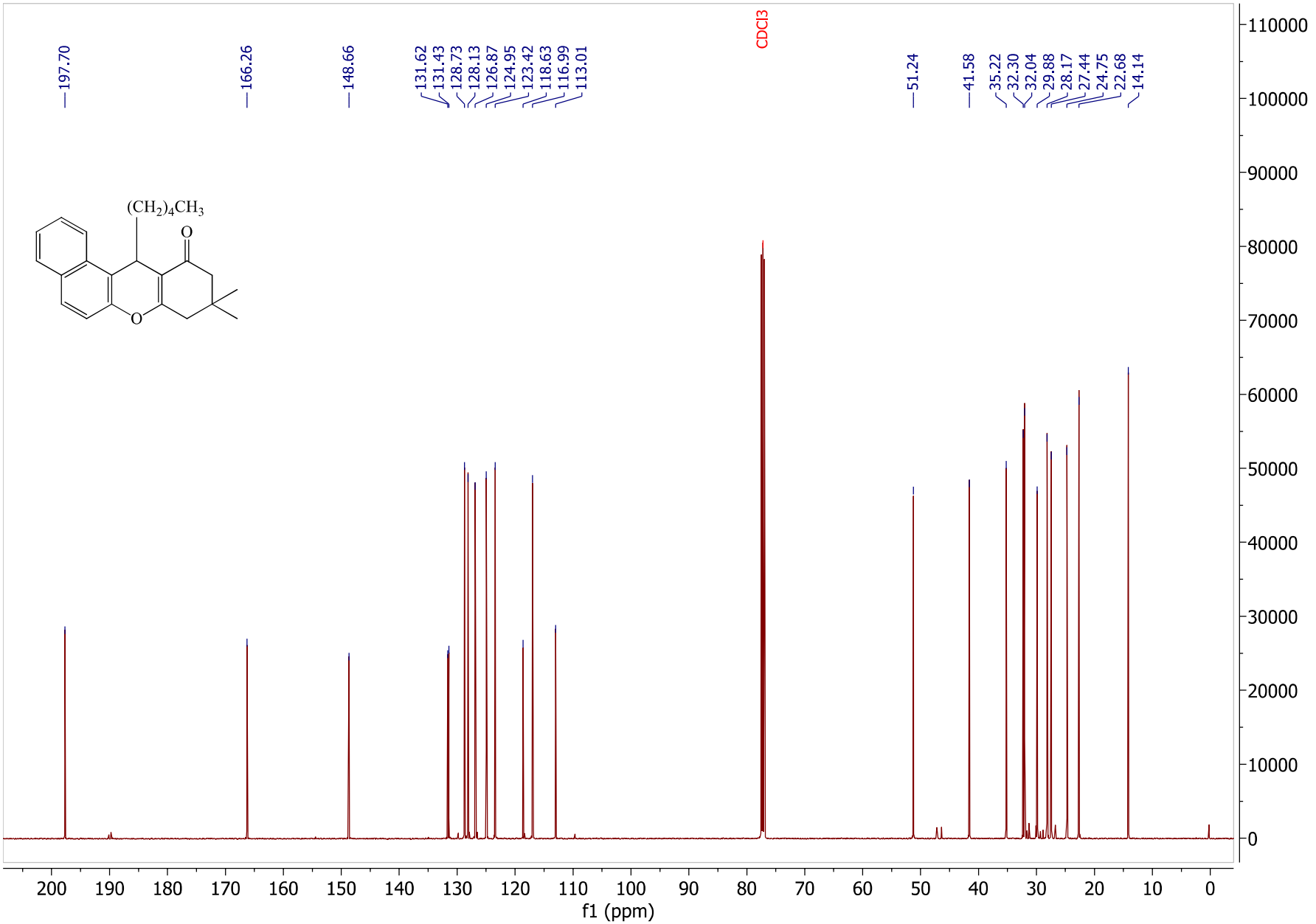

Figure S40 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-pentyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4o), in chloroform-d

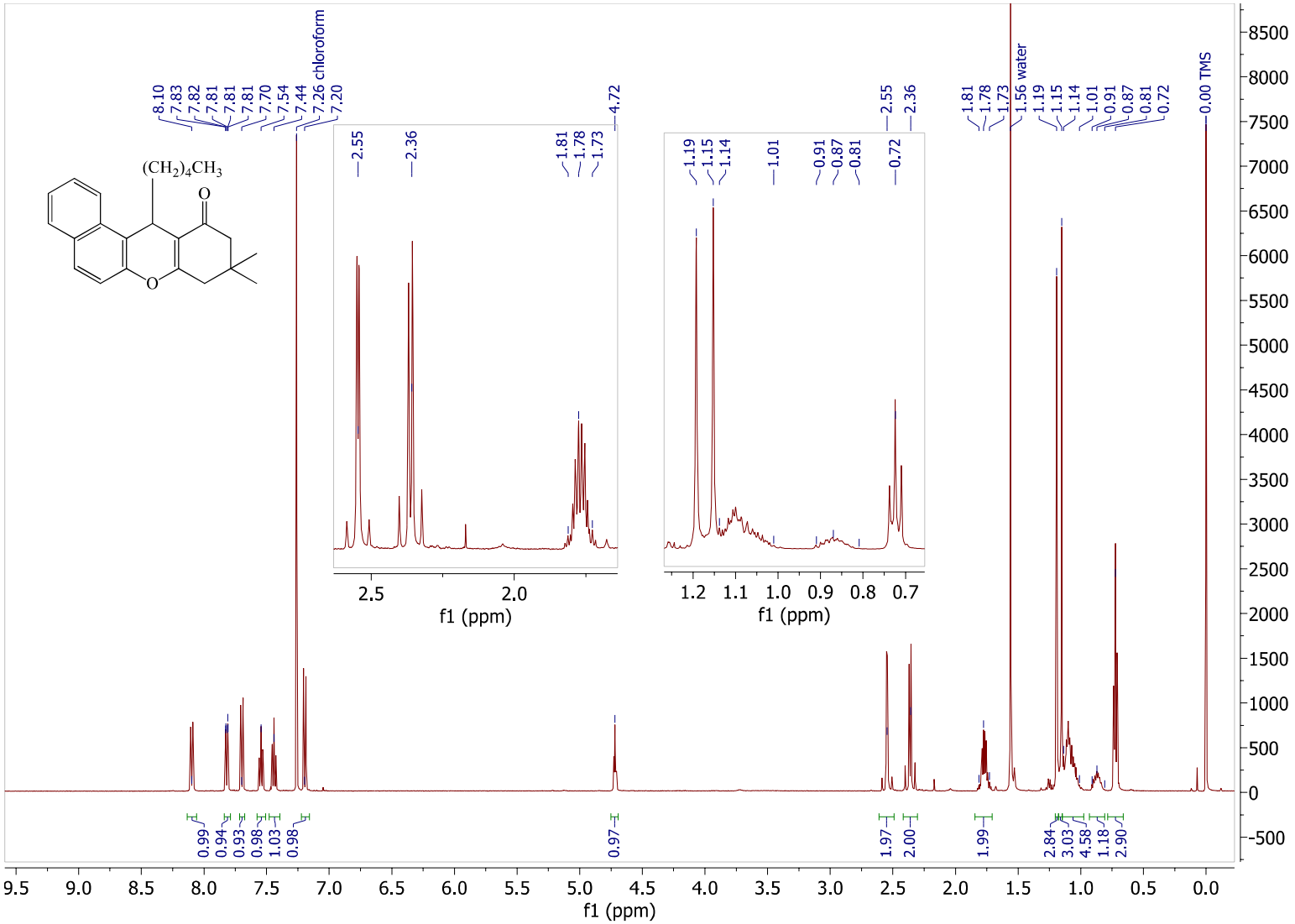

Figure S41 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-pentyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4o), in chloroform-d

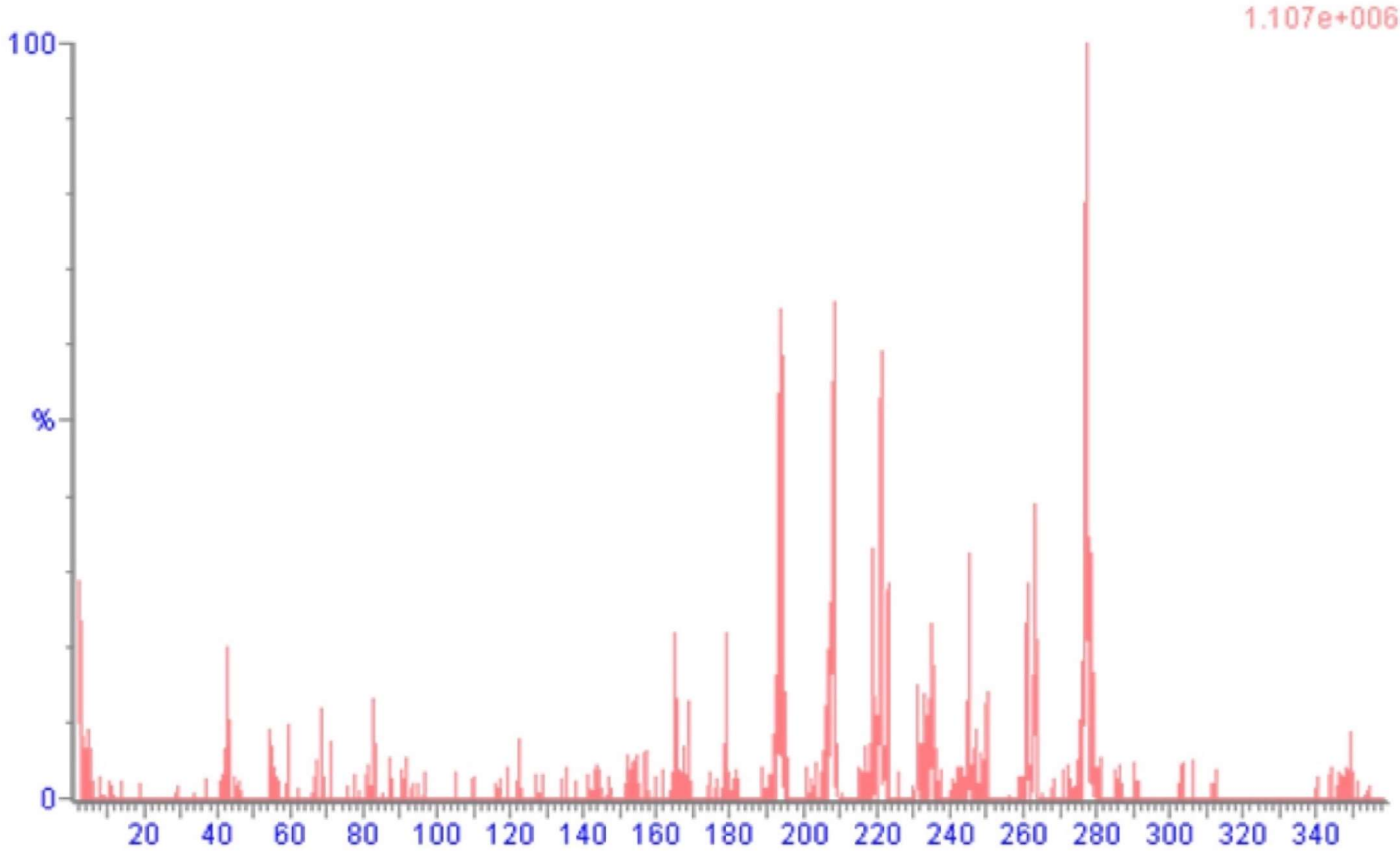

Figure S42 - MS (ES+) spectrum of 9,9-dimethyl-12-pentyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4o)

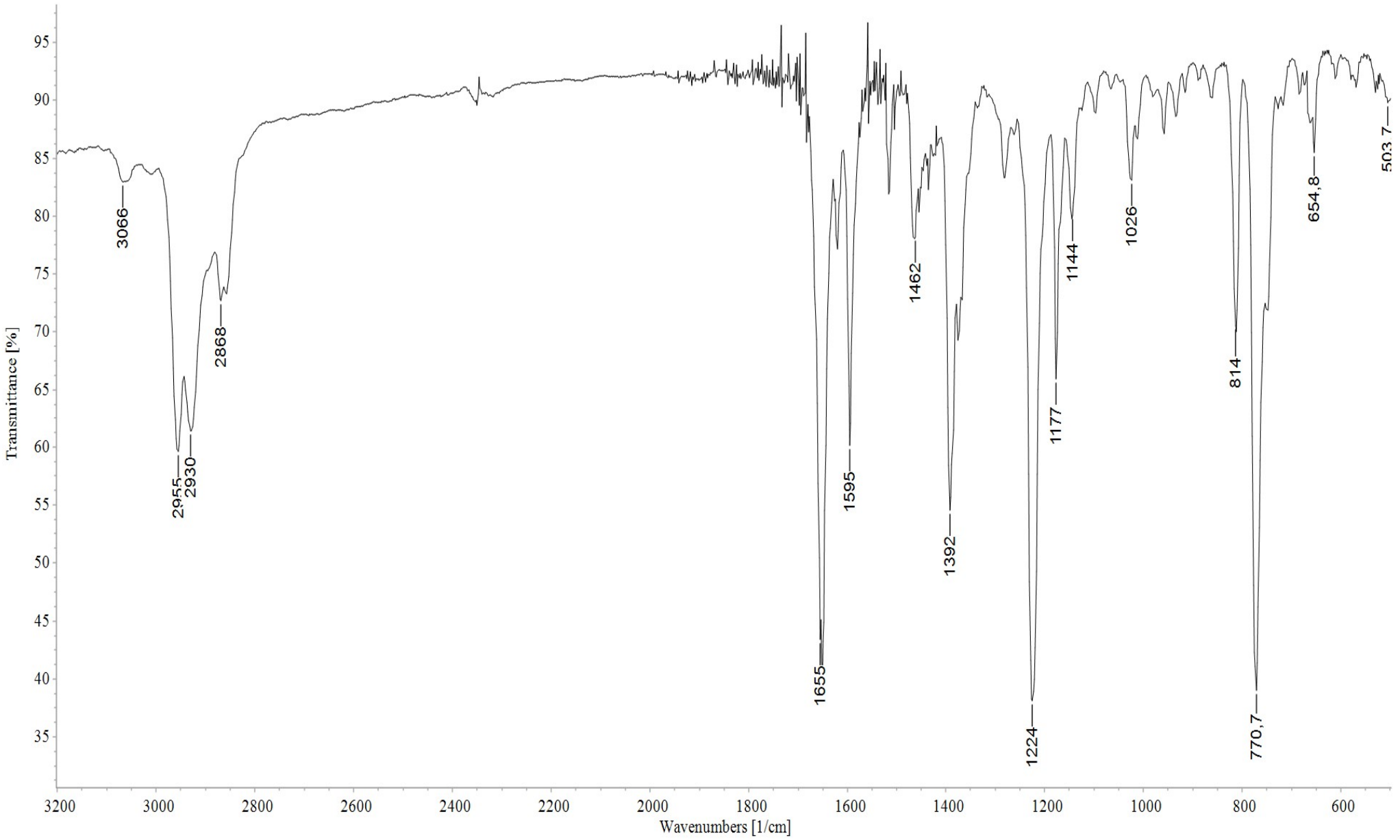

Figure S43 - FT-IR spectrum of 9,9-dimethyl-12-hexyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4p), NaCl

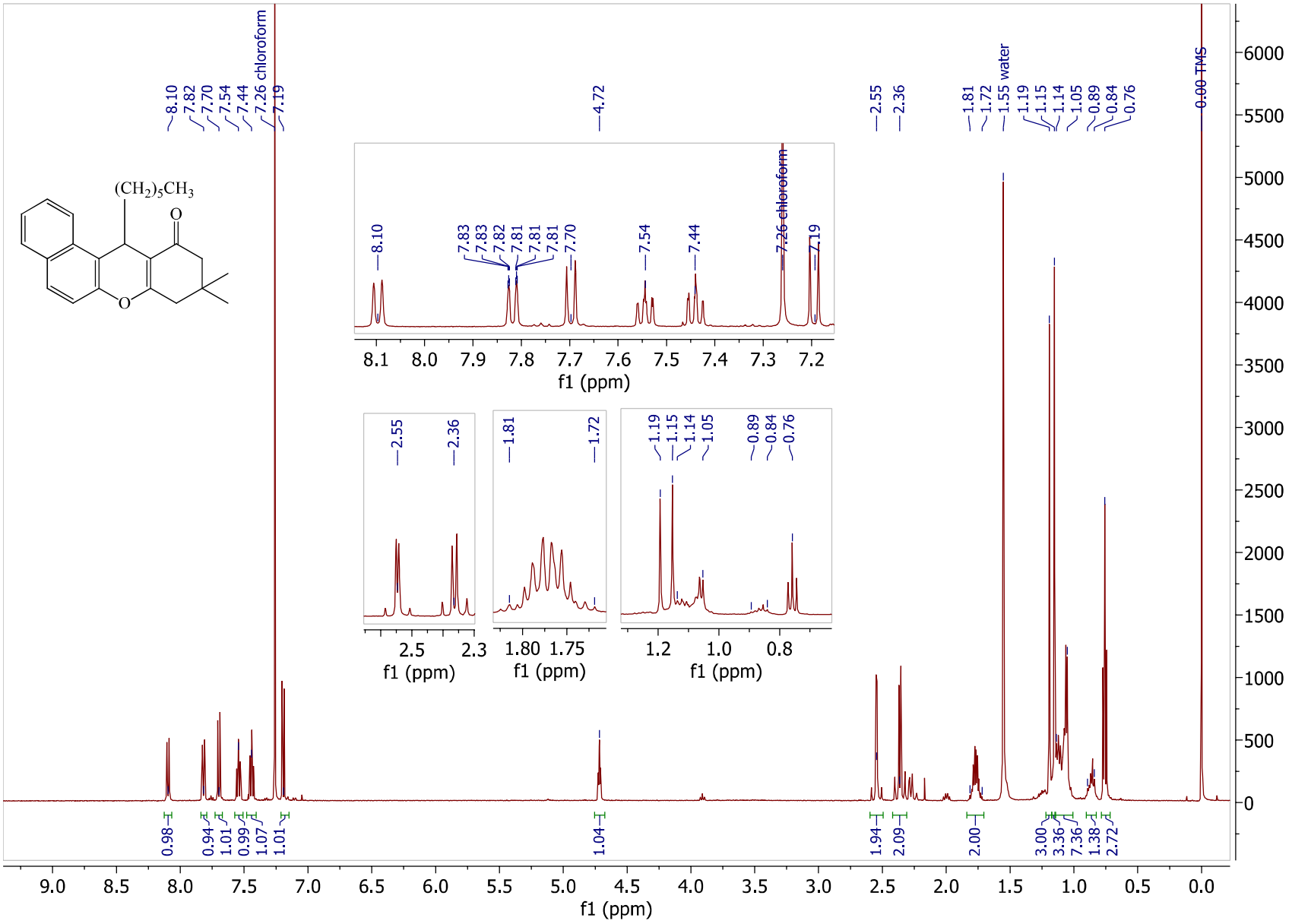

Figure S44 - ^{1}H-NMR spectrum of 9,9-dimethyl-12-hexyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4p), in chloroform-d

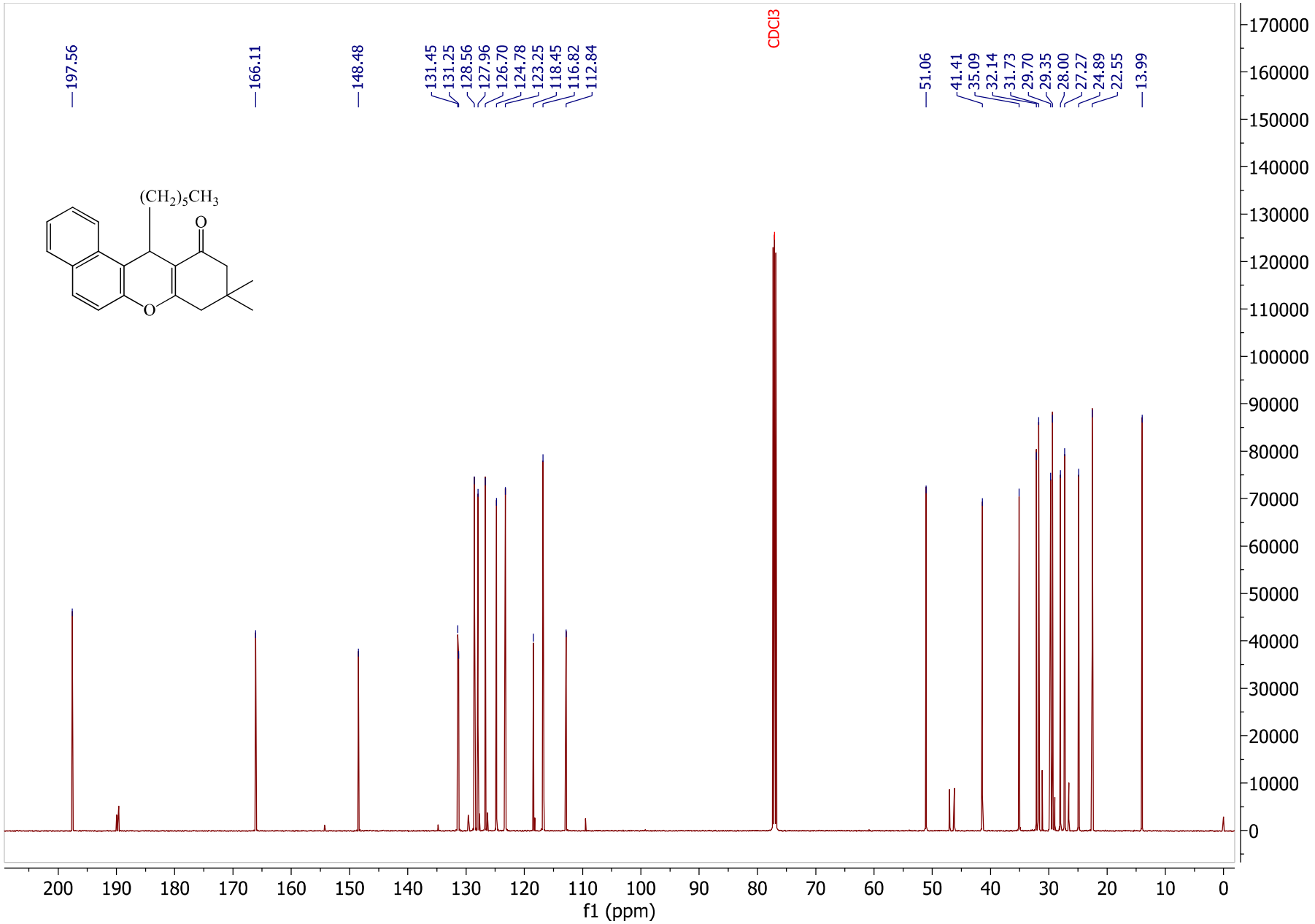

Figure S45 - ^{13}C-NMR spectrum of 9,9-dimethyl-12-hexyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4p), in chloroform-d

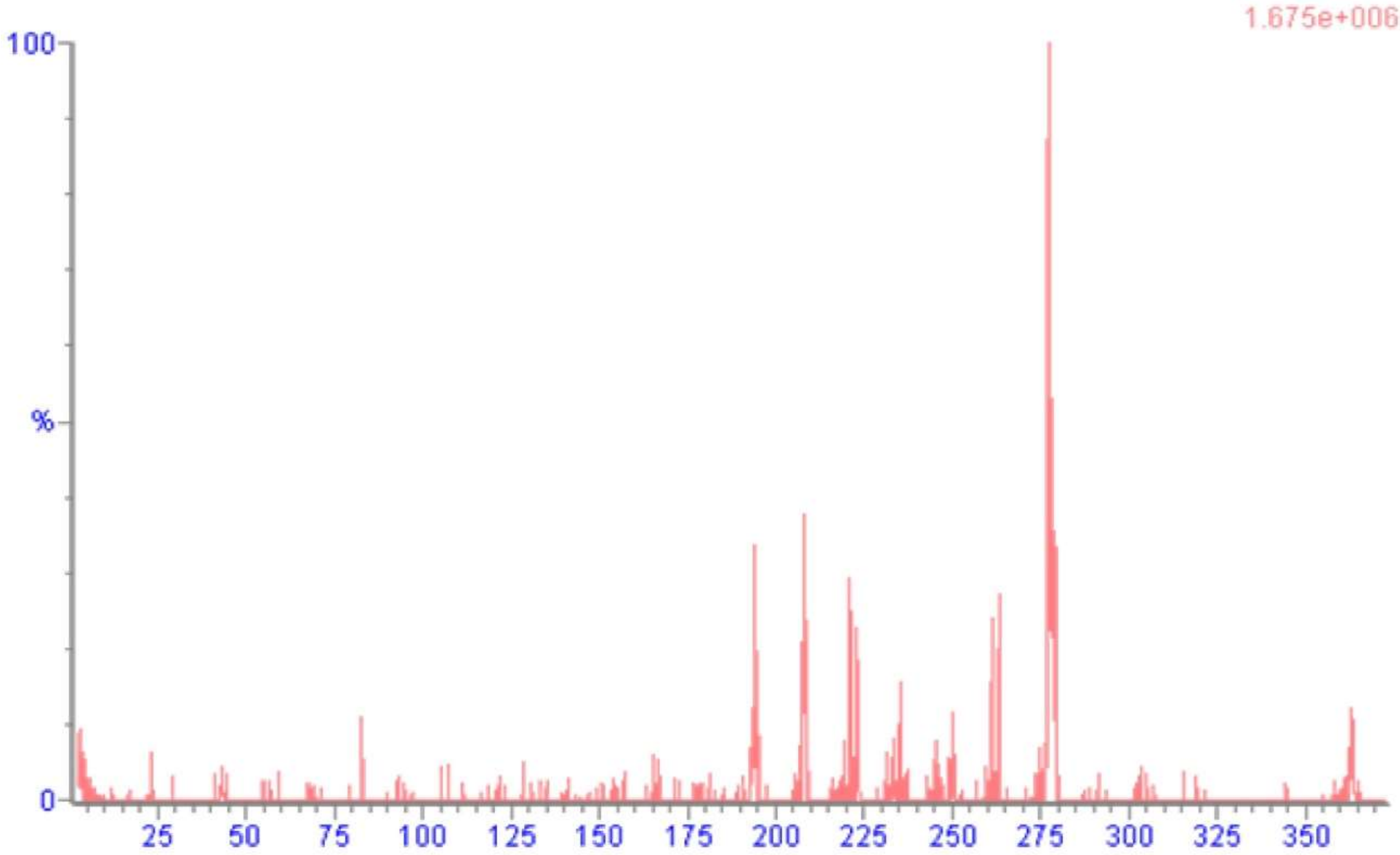

Figure S46 - MS (ES+) spectrum of 9,9-dimethyl-12-hexyl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one (4p)

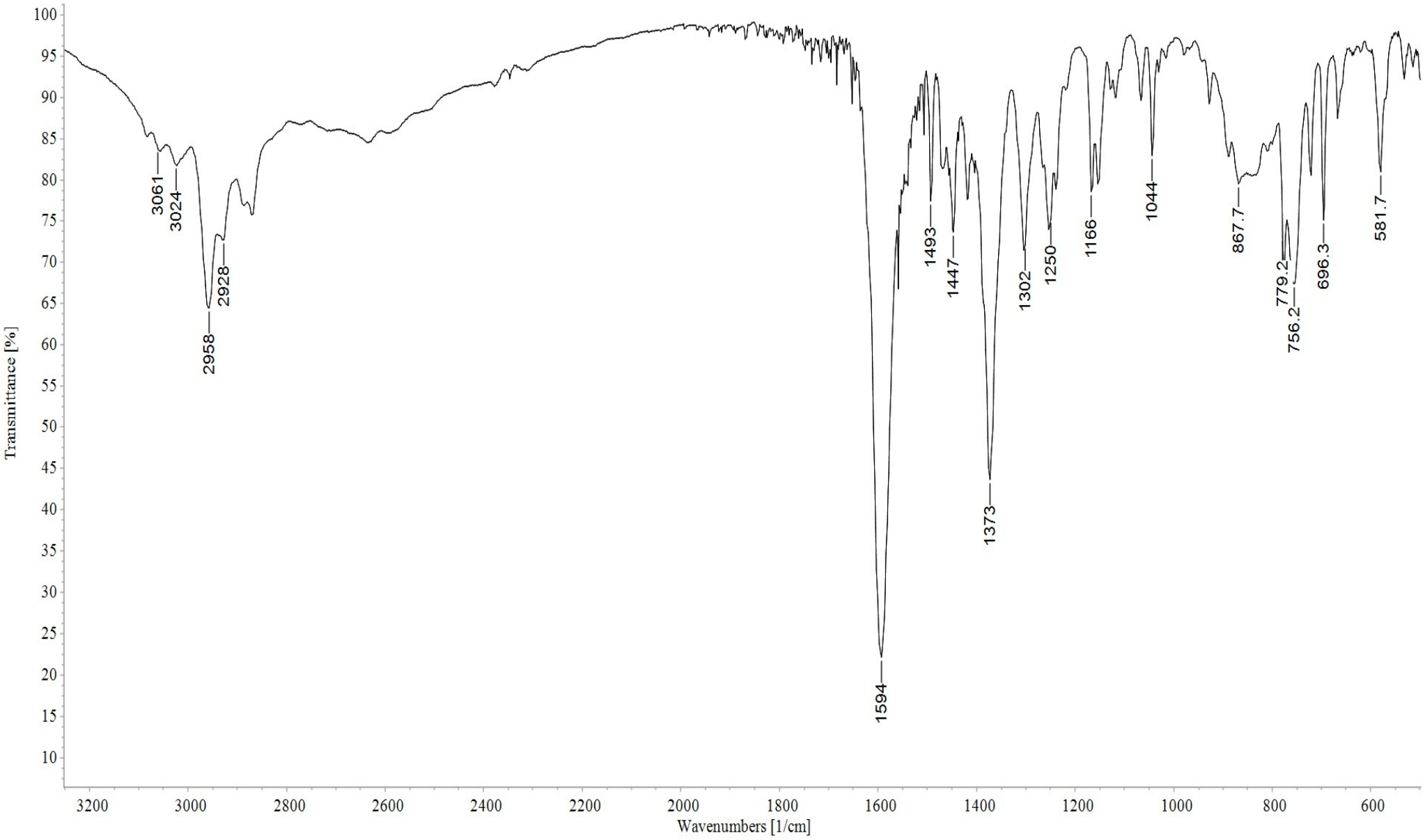

Figure S47 - FT-IR spectrum of 10,10-dimethyl-7-phenyl-10,11-dihydro-7H-benzo[c]xanthen-8(9H)-one (4q), NaCl

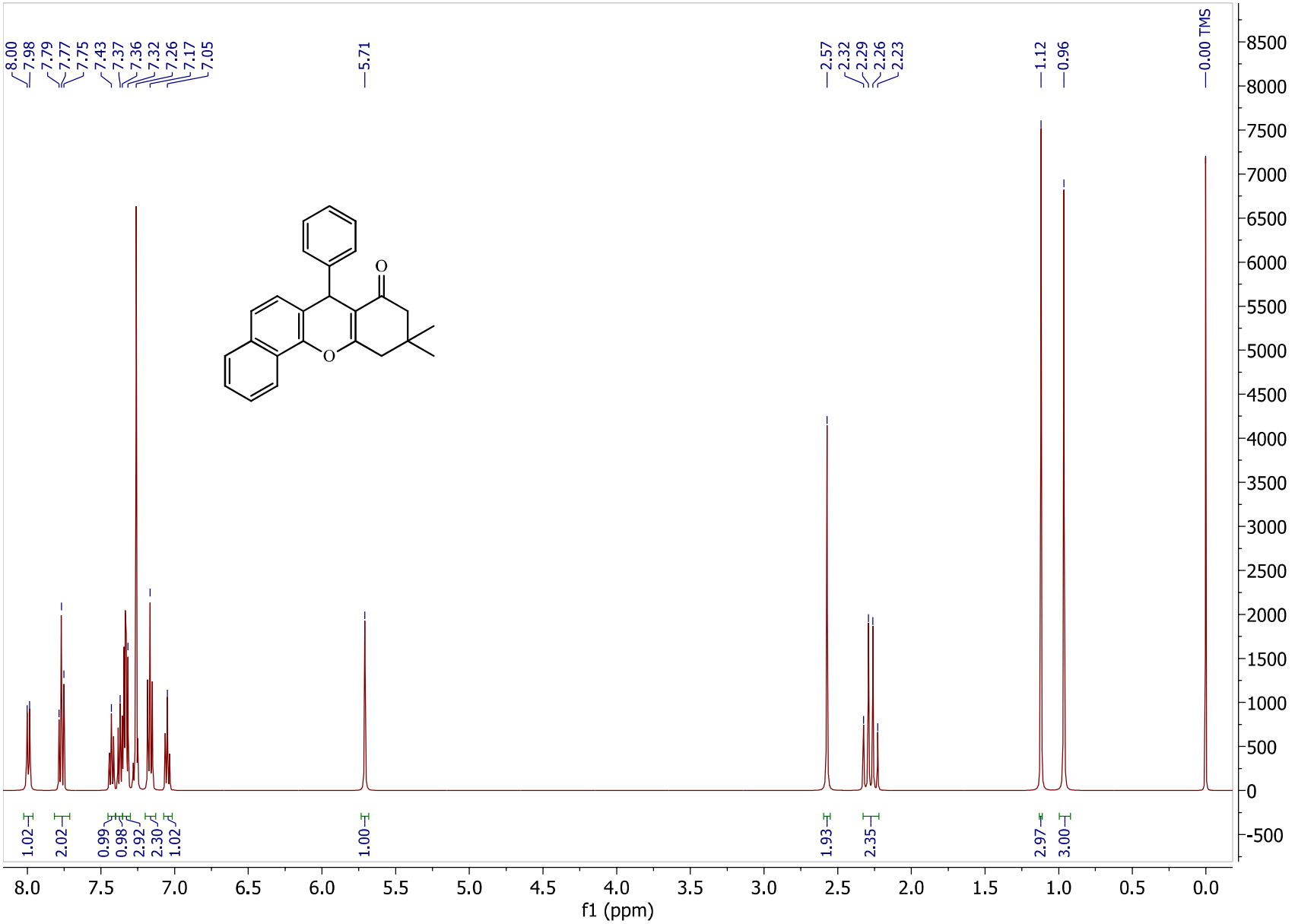

Figure S48 - ^{1}H-NMR spectrum of 10,10-dimethyl-7-phenyl-10,11-dihydro-7H-benzo[c]xanthen-8(9H)-one (4q), in chloroform-d

2. XRF analysis of the catalyst

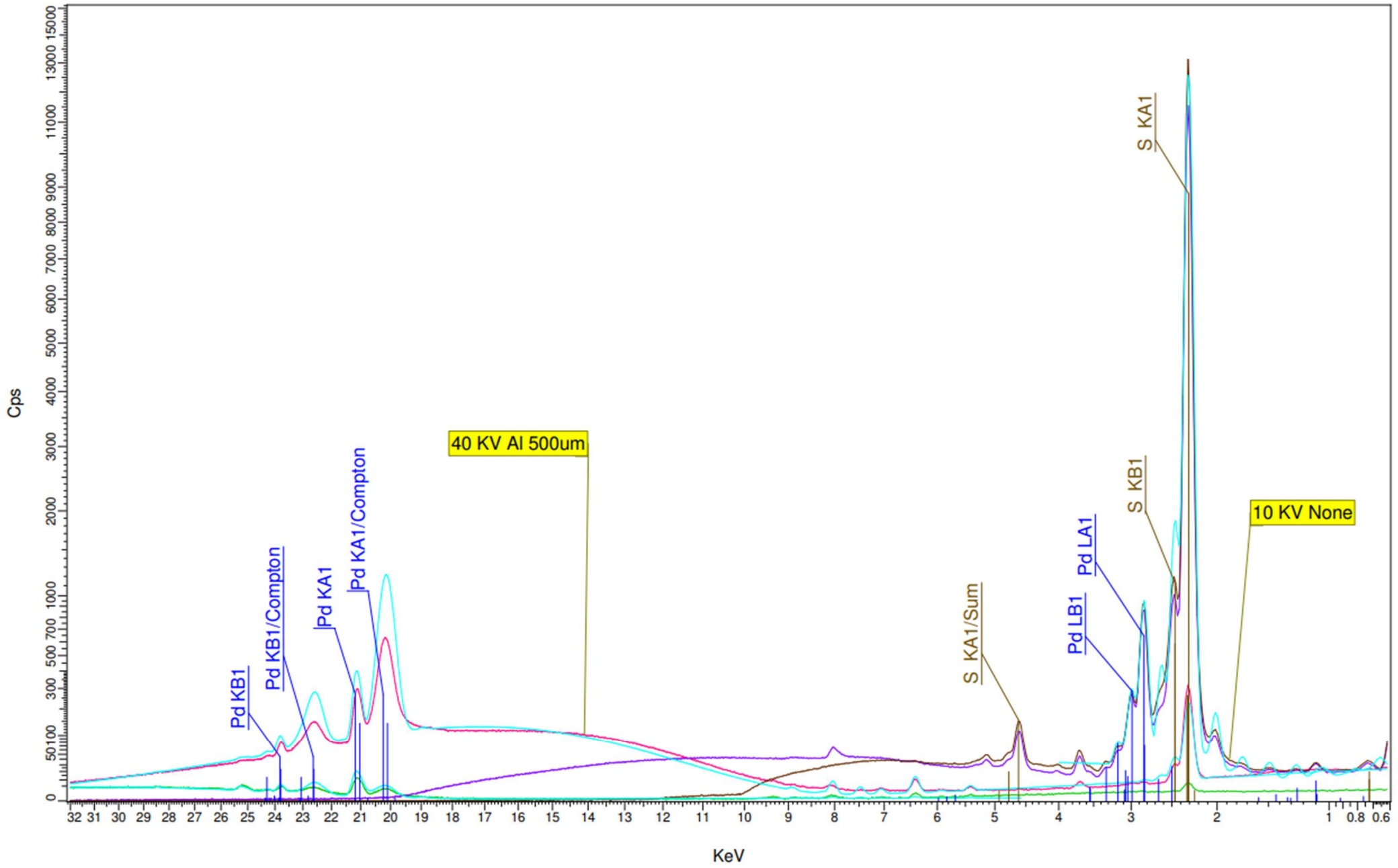

Figure S49 - XRF spectrum of Amberlyst-15

Table S1 - results from XRF analysis of Amberlyst-15

Amberlyst-15 R/R0 = 26.2 Polymers Evaluation

Formula	Z	Concentration	Status	Line 1	Net int.
CH2	6	88.8	Matrix		
S	16	6.24	Fit spectrum	S KA1/EQ10	8364
Na	11	3.79	Fit spectrum	Na KA1/EQ10	6.26
Mg	12	0.695	Fit spectrum	Mg KA1/EQ10	9.26
Al	13	0.143	Fit spectrum	Al KA1/EQ10	13.9
P	15	0.139	Fit spectrum	P KA1/EQ10	107
Cl	17	0.138	Fit spectrum	Cl KA1/EQ10	271.3
Si	14	0.0401	Fit spectrum	Si KA1/EQ10	19.3
Fe	26	0.003	Fit spectrum	Fe KA1/EQ10	28.4
Sn	50	0.0026	Fit spectrum	Sn KA1/EQ40	3.69
Cr	24	0.0016	Fit spectrum	Cr KA1/EQ10	2.02
Cu	29	0.0008	Fit spectrum	Cu KA1/EQ10	14.7
Ni	28	0.0004	Fit spectrum	Ni KA1/EQ10	8.59
Ag	47	0.0003	Fit spectrum	Ag KA1/EQ40	0.95
Sr	38	0	Fit spectrum	Sr KA1/EQ40	
Bi	83	0	Fit spectrum	Bi LA1/EQ40	
Ca	20	0	Fit spectrum	Ca KA1/EQ10	
Br	35	0	Fit spectrum	Br KA1/EQ40	
Pb	82	0	Fit spectrum	Pb LA1/EQ40	
Mo	42	0	Fit spectrum	Mo KA1/EQ40	
Nb	41	0	Fit spectrum	Nb KA1/EQ40	
Zn	30	0	Fit spectrum	Zn KA1/EQ10	
Co	27	0	Fit spectrum	Co KA1/EQ10	
Mn	25	0	Fit spectrum	Mn KA1/EQ10	
V	23	0	Fit spectrum	V KA1/EQ10	
Ti	22	0	Fit spectrum	Ti KA1/EQ10	
Ba	56	0	Fit spectrum	Ba KA1/EQ40	
Sb	51	0	Fit spectrum	Sb KA1/EQ40	
Cd	48	0	Fit spectrum	Cd KA1/EQ40	
Zr	40	0	Fit spectrum	Zr KA1/EQ40	
K	19	0	Fit spectrum	K KA1/EQ10	

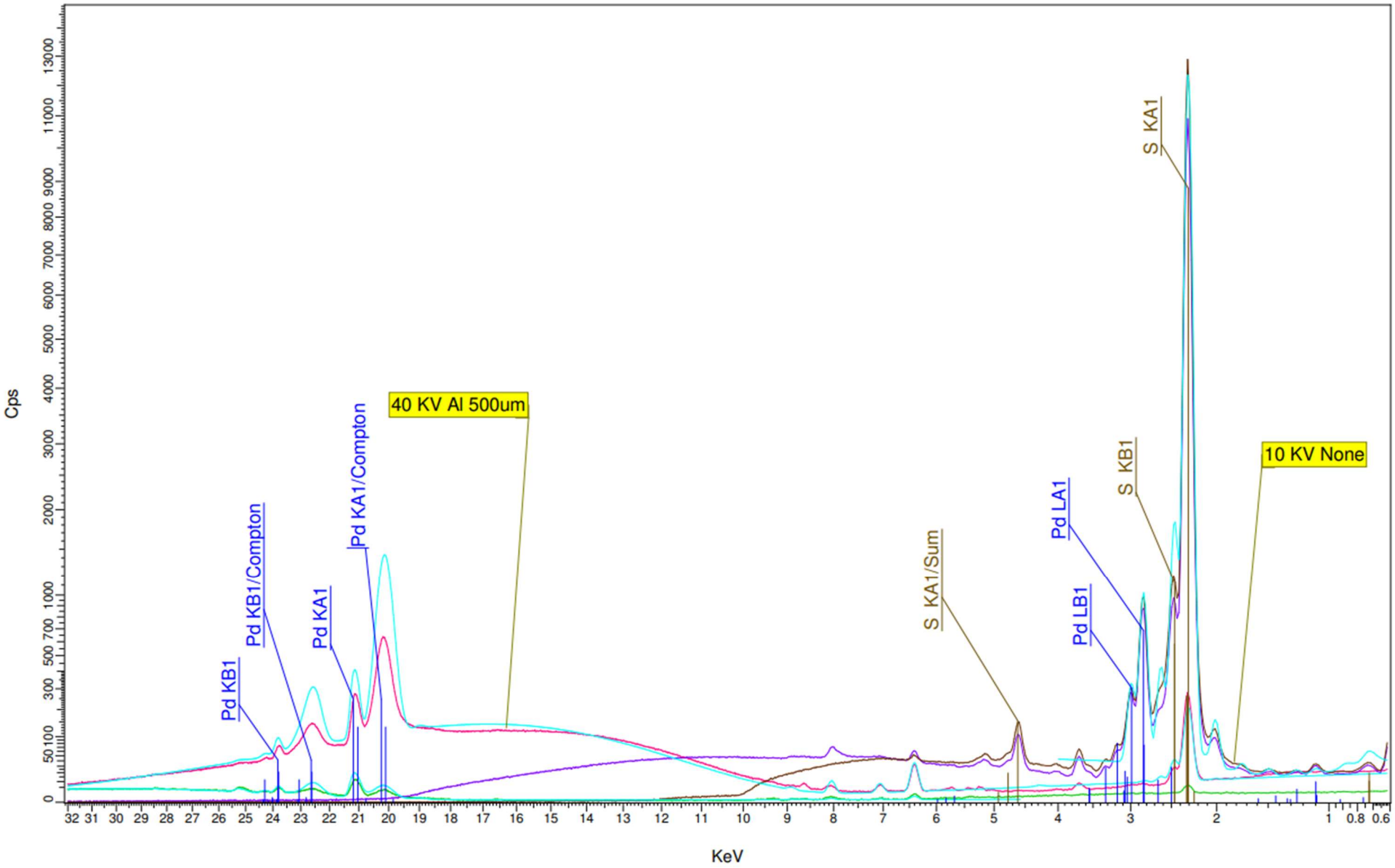

Figure S50 - XRF spectrum of DABCO / Amberlyst-15 (30 w/w%)

Table S2 - results from XRF analysis of DABCO / Amberlyst-15 (30 w/w%)

DABCO / Amberlyst-15 $R/R_0 = 31.1$ Polymers Evaluation

Formula	Z	Concentration	Status	Line 1	Net int.
CH2	6	95.1	Matrix		
S	16	4.5	Fit spectrum	S KA1/EQ10	7949
Mg	12	0.136	Fit spectrum	Mg KA1/EQ10	2.19
Cl	17	0.0942	Fit spectrum	Cl KA1/EQ10	250.3
P	15	0.0837	Fit spectrum	P KA1/EQ10	83.6
Al	13	0.0467	Fit spectrum	Al KA1/EQ10	5.69
Si	14	0.0149	Fit spectrum	Si KA1/EQ10	9.18
Fe	26	0.0062	Fit spectrum	Fe KA1/EQ10	80.5
Sn	50	0.0018	Fit spectrum	Sn KA1/EQ40	2.74
Cu	29	0.0006	Fit spectrum	Cu KA1/EQ10	16.6
Ag	47	0.0002	Fit spectrum	Ag KA1/EQ40	0.64
Sr	38	0	Fit spectrum	Sr KA1/EQ40	
Bi	83	0	Fit spectrum	Bi LA1/EQ40	
Ca	20	0	Fit spectrum	Ca KA1/EQ10	
Br	35	0	Fit spectrum	Br KA1/EQ40	
Pb	82	0	Fit spectrum	Pb LA1/EQ40	
Mo	42	0	Fit spectrum	Mo KA1/EQ40	
Nb	41	0	Fit spectrum	Nb KA1/EQ40	
Zn	30	0	Fit spectrum	Zn KA1/EQ10	
Ni	28	0	Fit spectrum	Ni KA1/EQ10	
Co	27	0	Fit spectrum	Co KA1/EQ10	
Mn	25	0	Fit spectrum	Mn KA1/EQ10	
V	23	0	Fit spectrum	V KA1/EQ10	
Cr	24	0	Fit spectrum	Cr KA1/EQ10	
Ti	22	0	Fit spectrum	Ti KA1/EQ10	
Na	11	0	Fit spectrum	Na KA1/EQ10	
Ba	56	0	Fit spectrum	Ba KA1/EQ40	
Sb	51	0	Fit spectrum	Sb KA1/EQ40	
Cd	48	0	Fit spectrum	Cd KA1/EQ40	
Zr	40	0	Fit spectrum	Zr KA1/EQ40	
K	19	0	Fit spectrum	K KA1/EQ10	

3. *E*-factor

Table S3 - Calculation of the E-factor for the synthesised products

Entry	Yield (%)	Total mass of starting materials (mg)	Total mass of product (mg)	Total mass of waste (mg)	E-factor
Model Reaction (4a)	92	Mass of dimedone + mass of benzaldehyde + mass of 2-naphthol Dimedone (364.5 mg) + benzaldehyde (212.2 mg) + 2-naphthol (288.3 mg) = 865 mg	Mass of product 4a 655 mg	Total mass of starting materials – mass of product 865 mg – 655 mg = 210 mg	$\frac{\text{Total mass of waste}}{\text{Total mass of product}}$ = 0.32
4b	88	792.08	574.45	217.63	0.38
4c	80	901.00	590.00	311.00	0.53
4d	79	933.94	614.45	319.49	0.52
4e	78	1,002.82	657.00	345.82	0.53
4f	76	933.94	591.11	342.83	0.58
4g	92	955.04	736.00	219.04	0.30
4h	43	897.05	316.00	581.05	1.84
4i	40	897.05	296.30	600.75	2.03
4j	43	925.30	330.64	594.66	1.80
4k	51	844.969	351.29	493.68	1.41
4l	90	915.06	680.30	234.76	0.35
4m	57	893.10	411.00	482.10	1.17
4n	79	825.07	528.44	296.63	0.56
4o	61	853.12	422.00	431.12	1.02
4p	44	941.18	319.00	622.18	1.95
4q	84	865.04	595.47	269.57	0.45

4. Atom Economy

Table S4 - Calculation of the AE % for the synthesized products

Entry	$\sum$ RMM $_{\text{starting materials}}$ (g/mol)	RMM $_{\text{product}}$ (g/mol)	AE %
	RMM of dimedone + RMM of benzaldehyde + RMM of 2-naphthol	RMM of product 4a	$\frac{\text{RMM of product}}{\sum \text{RMM starting materials}} \times 100$
Model Reaction (4a)	Dimedone (140.180 g/mol) + benzaldehyde (106.121 g/mol) + 2-naphthol (144.170 g/mol) = 390.471 g/mol	354.448 g/mol	$\frac{354.448}{390.471} \times 100 = 90.77\%$
4b	362.404	326.395	91.18
4c	408.460	372.439	91.52
4d	424.920	388.894	92.16
4e	459.360	423.339	91.52
4f	424.920	388.894	91.73
4g	435.470	399.446	91.14
4h	406.473	370.448	93.44
4i	396.470	370.448	91.43
4j	420.500	384.475	91.35
4k	380.430	344.400	91.44
4l	415.480	379.916	91.09
4m	404.498	368.475	90.28
4n	370.484	334.458	90.63
4o	384.510	348.485	90.96
4p	398.540	362.512	90.77
4q	390.471	354.448	91.14