Electronic Supplementary Material (ESI) for New Journal of Chemistry.
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Supporting information

I. Single crystal X-ray diffraction

a. 1-Ethyl-3,4,5-trimethylimidazolium hydrogen carbonate [C_2m_3im][HCO_3] (**9c**)
b. 1-Ethyl-3,4,5-trimethylimidazolium chloride [C_2m_3im][Cl] (**18c**)
c. 1-Ethyl-2,3,4,5-tetramethylimidazolium chloride [$C_2C_1m_3im$][Cl] (**18a**)

II. NMR spectroscopy: ^{13}C and ^{1}H spectroscopy

a. 1-Ethyl-3,4,5-trimethylimidazolium hydrogen carbonate [C_2m_3im][HCO_3] (**9c+12**)
b. 1-Ethyl-3,4,5-trimethylimidazolium hydrogen carbonate [C_2m_3im][HCO_3] (**9c**)
c. 1-Ethyl-3,4,5-trimethylimidazolium methyl carbonate [C_2m_3im][CH_3CO_3] (**8c**)
d. 1-Ethyl-2,3,4,5-tetramethylimidazolium hydrogen carbonate [$C_2C_1m_3im$][HCO_3] (**9a**)
e. 1-Ethyl-2-isopropyl-3,4,5-trimethylimidazolium hydrogen carbonate [$C_2C_{i3}m_3im$][HCO_3] (**9b**)
f. 1-Ethyl-3,4,5-trimethylimidazolium chloride [C_2m_3im][Cl] (**18c**)
g. 1-Ethyl-2,3,4,5-tetramethylimidazolium chloride [$C_2C_1m_3im$][Cl] (**18a**)

a) Compound **9c**

Hydrogen bonding around each $[HCO_3]^-$ anion includes interactions with four $[C_2m_3im]^+$ cations, with a total of seven hydrogen bonds: C1-H1···O3 = 2.241(10) Å; C1-H1···O2 = 2.609(8) Å; C4-H4···O2 = 2.510(11) Å; C5-H5···O2 = 2.465(12) Å; C6-H6A···O3 = 2.558(9) Å; C6-H6C···O3 = 2.480(11) Å; C7-H7A···O1 = 2.633(7) Å. Hydrogen bonding around each $[C_2m_3im]^+$ cation includes the same interactions with four $[HCO_3^-]$ anions (Figure S1, S2).

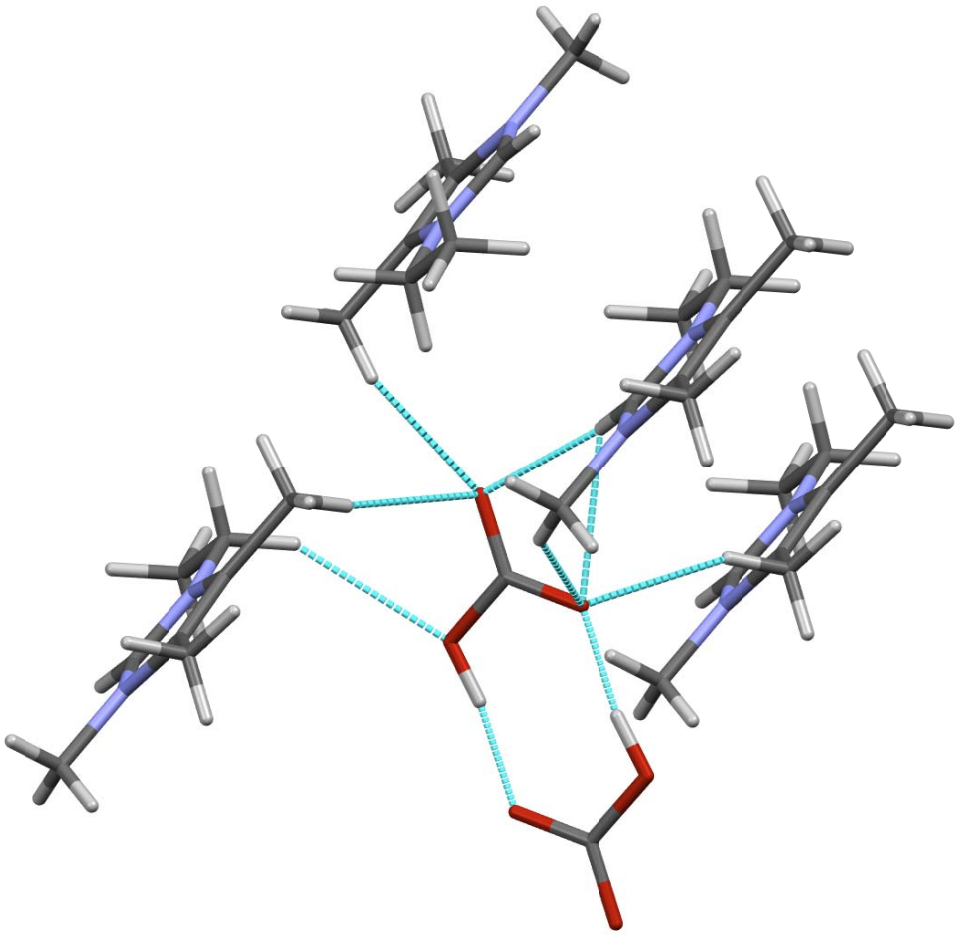


Figure S1. Hydrogen-bond interactionsbetween each $[HCO_3]^-$anion and $[C_2m_3im]^+$ cationsin the crystal structure of ***9c****.One $[HCO_3]^-$ dimer is shown.*

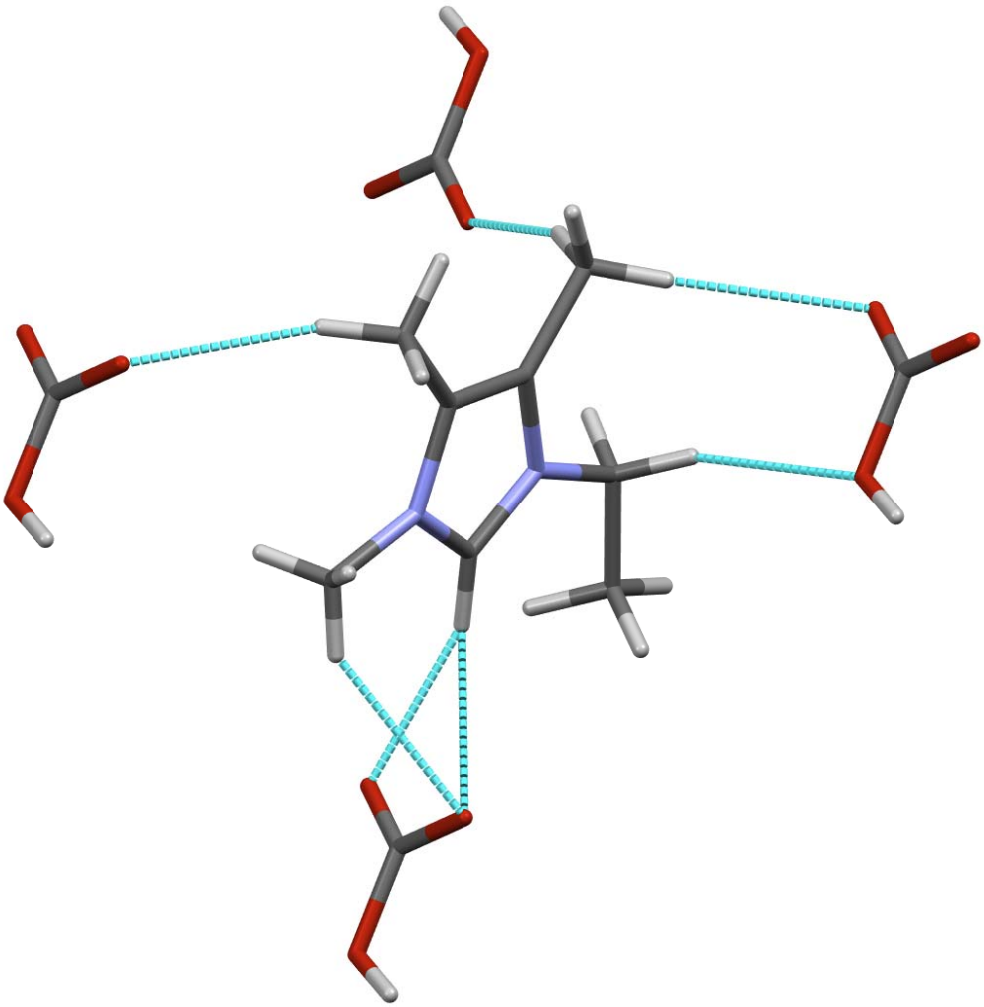


Figure S2. Hydrogen-bond interactions between each $[C_2m_3im]^+$ cationand $[HCO_3]^-$ anions in the crystal structure of ***9c****.*

In the crystal packing, short ring-interactions are observed between the $[C_2m_3im]^+$ cation imidazolium rings (ring centroid-centroid distances of 4.6055(8) Å and 5.1113(8) Å). C-H···ring interactions occur between C5-H5 and C8-H8 and the $[C_2m_3im]^+$ cation imidazolium ring (distances of 2.815(9) Å and 2.780(13) Å, respectively). Furthermore, alternating layers of $[HCO_3^-]$ anion dimers and $[C_2m_3im]^+$ cations are detected, parallel with the (010) crystallographic plane (Figure S3).

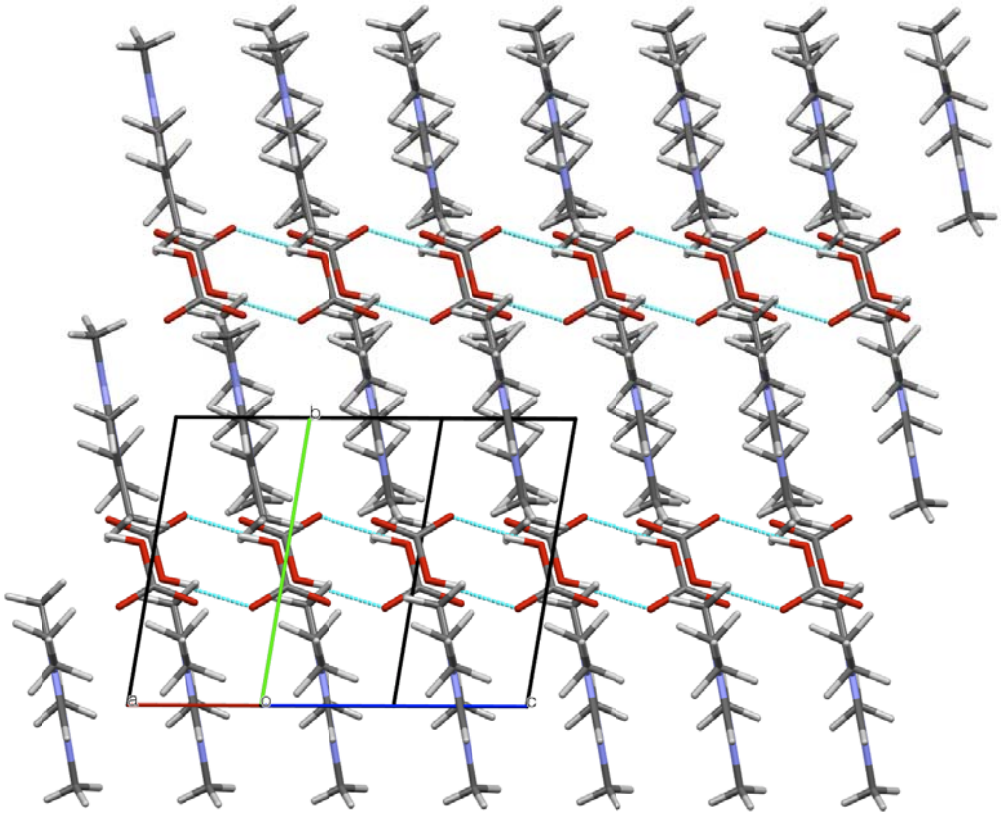


Figure S3. Packing diagram in the crystal structure of ***9c****, showing alternating layers of* $[HCO_3]^-$ *anion dimers and* $[C_2m_3im]^+$ *cations, parallel with the (010) crystallographic plane.*

b) Compound **18c**

Hydrogen bonding around each $[Cl]^-$ anion includesinteractions with five $[C_2m_3im]^+$ cations:C1-H1···Cl1 = 2.499(10) Å; C4-H4B···Cl1 = 2.794(9) Å; C4-H4C···Cl1 = 2.701(12) Å; C7-H7A···Cl1 = 2.702(9) Å; C7-H7B···Cl1 = 2.698(11) Å.Hydrogen bonding around each $[C_2m_3im]^+$ cation includes the same interactions with four $[Cl]^-$ anions (Figure S4, S5).

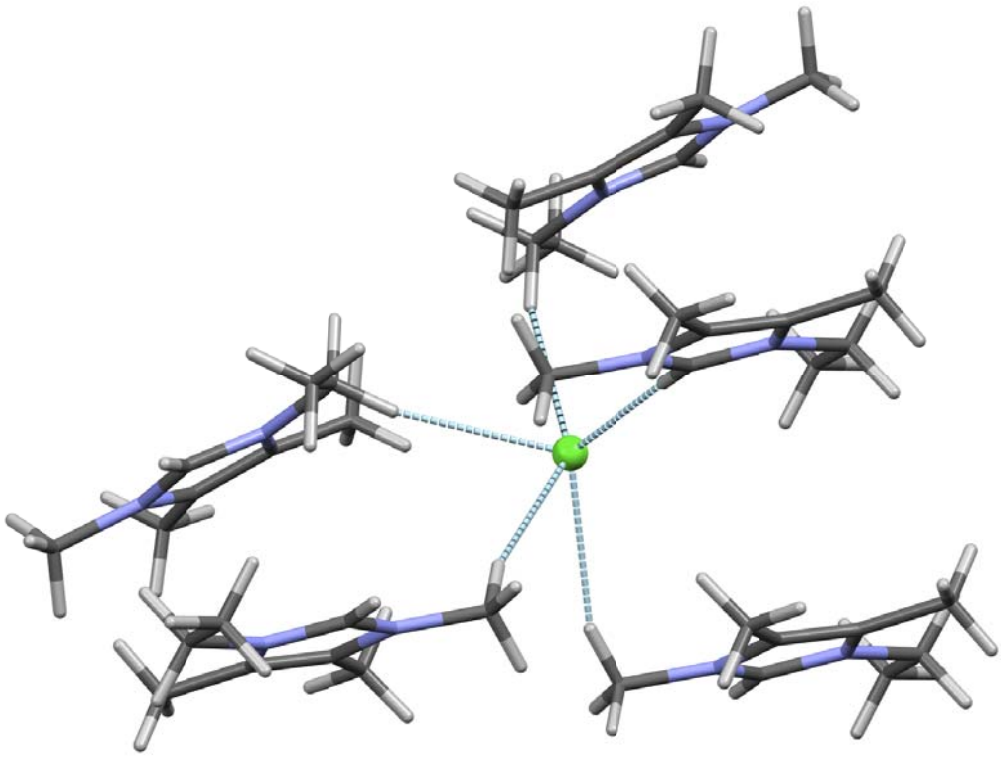


*Figure S4. Hydrogen-bond interactions between each$[Cl]^-$anion and $[C_2m_3im]^+$ cations in the crystal structure of **18c**.*

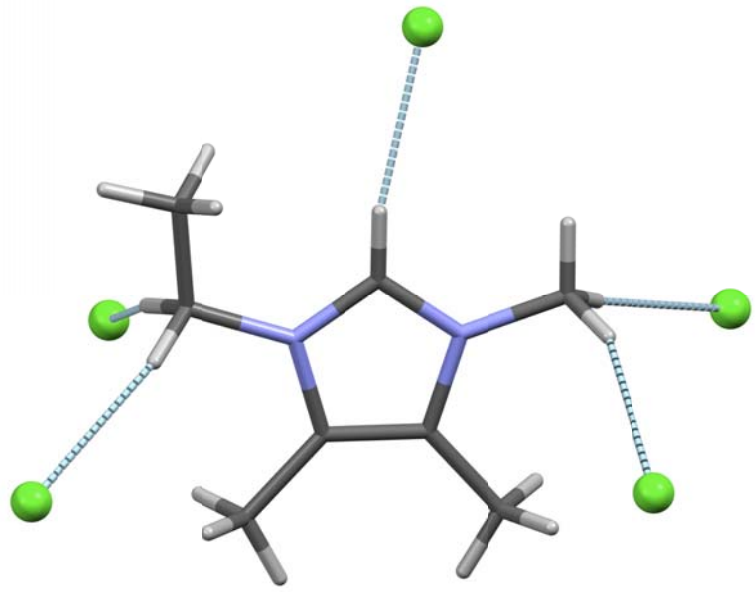


*Figure S5. Hydrogen-bond interactions between each $[C_2m_3im]^+$ cation and $[Cl]^-$anions in the crystal structure of **18c**.*

In the crystal packing, short ring-interactions are observed between the $[C_2m_3im]^+$ cation imidazolium rings (ring centroid-centroid distances of 4.9493(9) Å and 4.9492(9)Å) along the crystallographic [001] direction (Figure S6). C-H···ring interactions occur between C8-H8 and the $[C_2m_3im]^+$ cation imidazolium ring (distance of 2.981(11)Å).

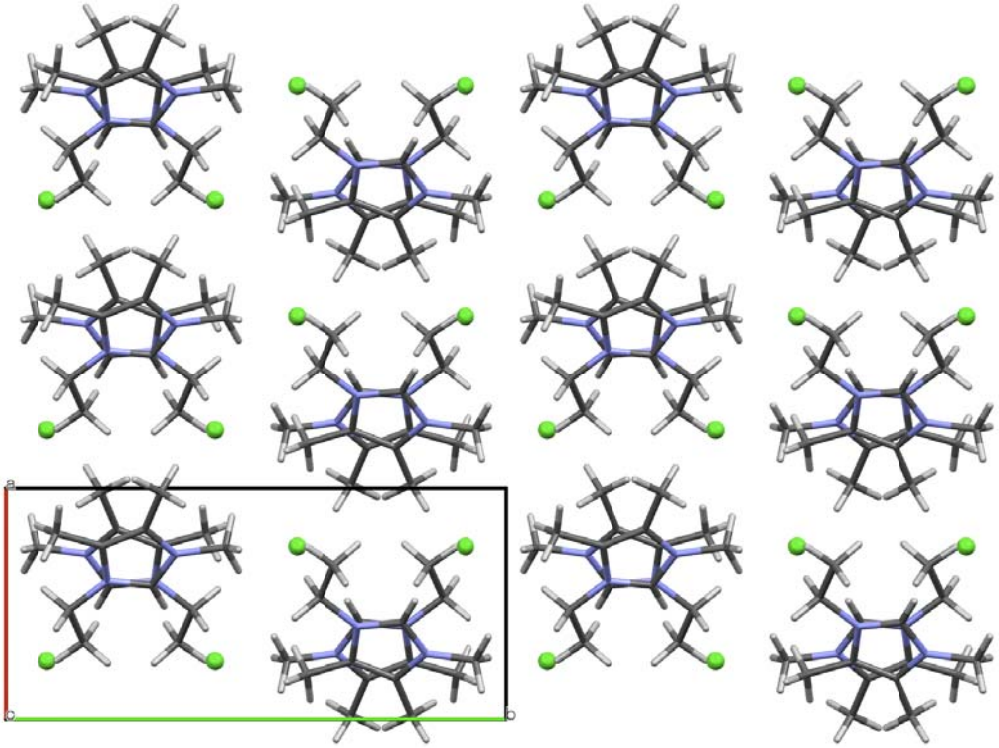


Figure S6. Packing diagram in the crystal structure of ***18c****, along the crystallographic [001] direction.*

c) Compound **18a**

Hydrogen bonding around each $[Cl]^-$ anionincludes interactions with six $[C_2C_1m_3im]^+$ cations: C4-H4B···Cl1 = 2.920(12) Å; C4-H4C···Cl1 = 2.764(16)Å; C5-H5A···Cl1 = 2.922(14) Å; C5-H5B···Cl1 = 2.746(10) Å, C5-H5C···Cl1 = 2.884(12) Å; C6-H6A···Cl1 = 2.818(12) Å; C6-H6B···Cl1 = 2.786(12) Å;C6-H6C···Cl1 = 2.913(12) Å;C7-H7A···Cl1 = 2.767(13) Å;C8-H8A···Cl1 = 2.793(11) Å;C8-H8B···Cl1 = 2.822(10) Å. Hydrogen bonding around each $[C_2C_1m_3im]^+$ cation includes the same interactions with six $[Cl]^-$anions (Figure S7, S8).

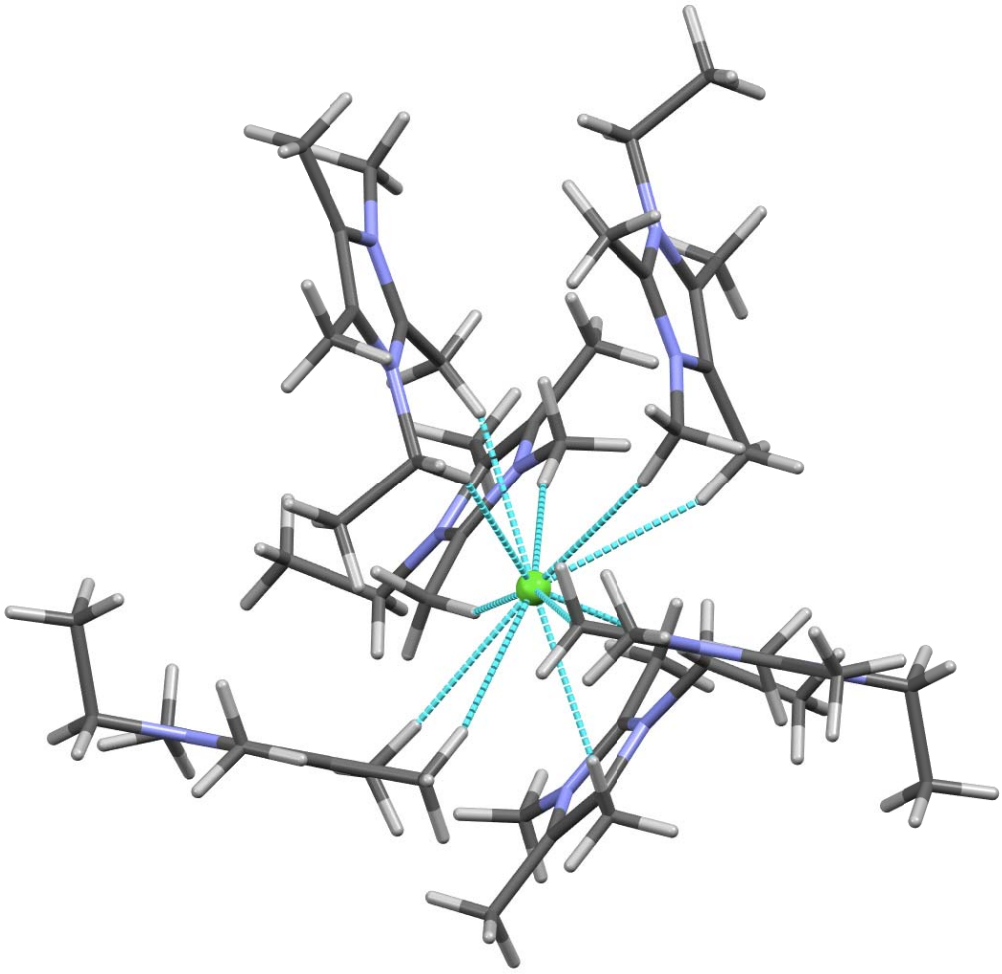


*Figure S7. Hydrogen-bond interactions between each $[Cl]^-$ anion and $[C_2C_1m_3im]^+$ cations in the crystal structure of **18a**.*

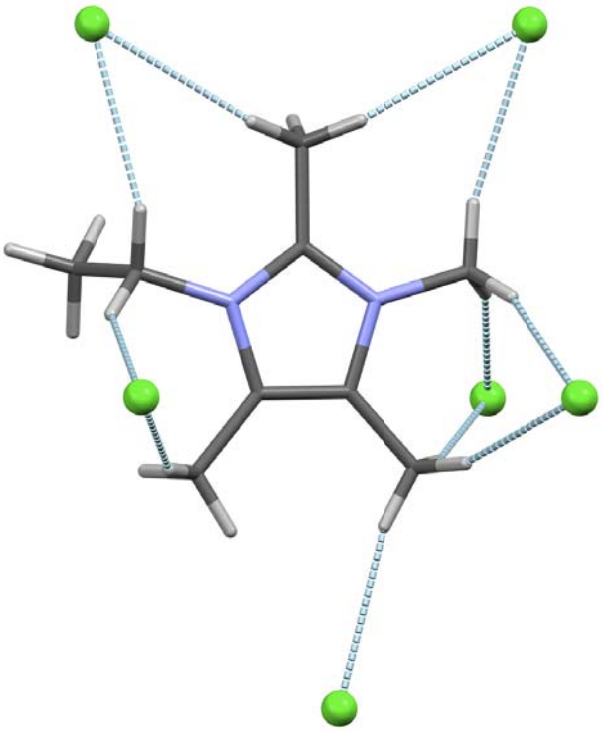


Figure S8. Hydrogen-bond interactions between each $[C_2C_1m_3im]^+$ cation and $[Cl]^-$ anions in the crystal structure of ***18a****.*

In the crystal packing, short ring-interactions are observed between two $[C_2C_1m_3im]^+$ cation imidazolium rings (ring centroid-centroid distance of 4.9241(11) Å). A typical packing pattern around the three-fold axes and inversion centers is observed along the [001] direction, clustering the $[C_2C_1m_3im]^+$ cation together in a cyclic columnar manner, surrounded by the $[Cl]^-$ anions (Figure S9).

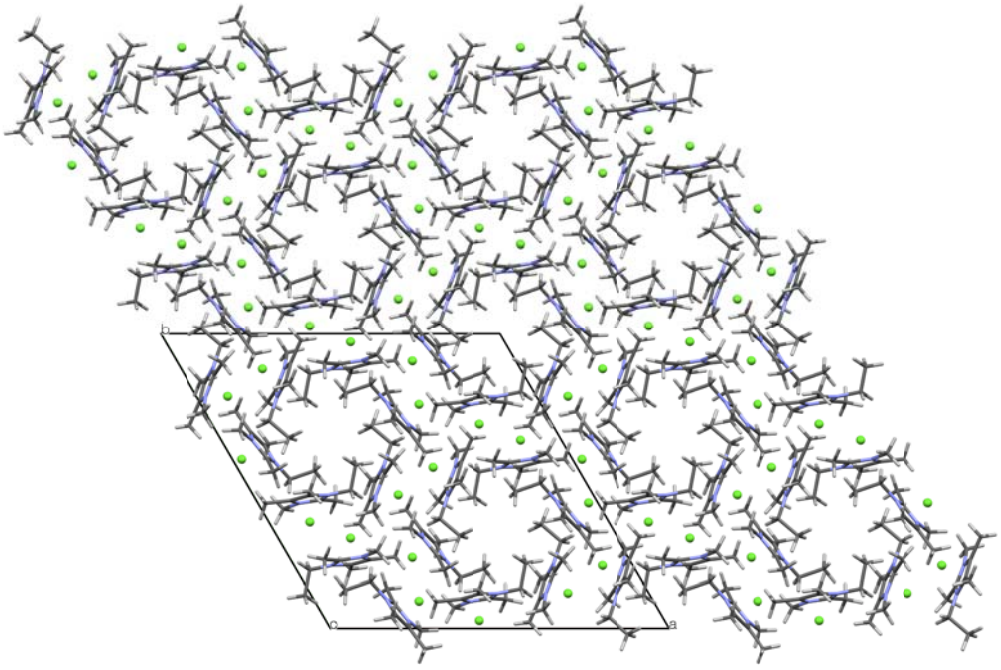
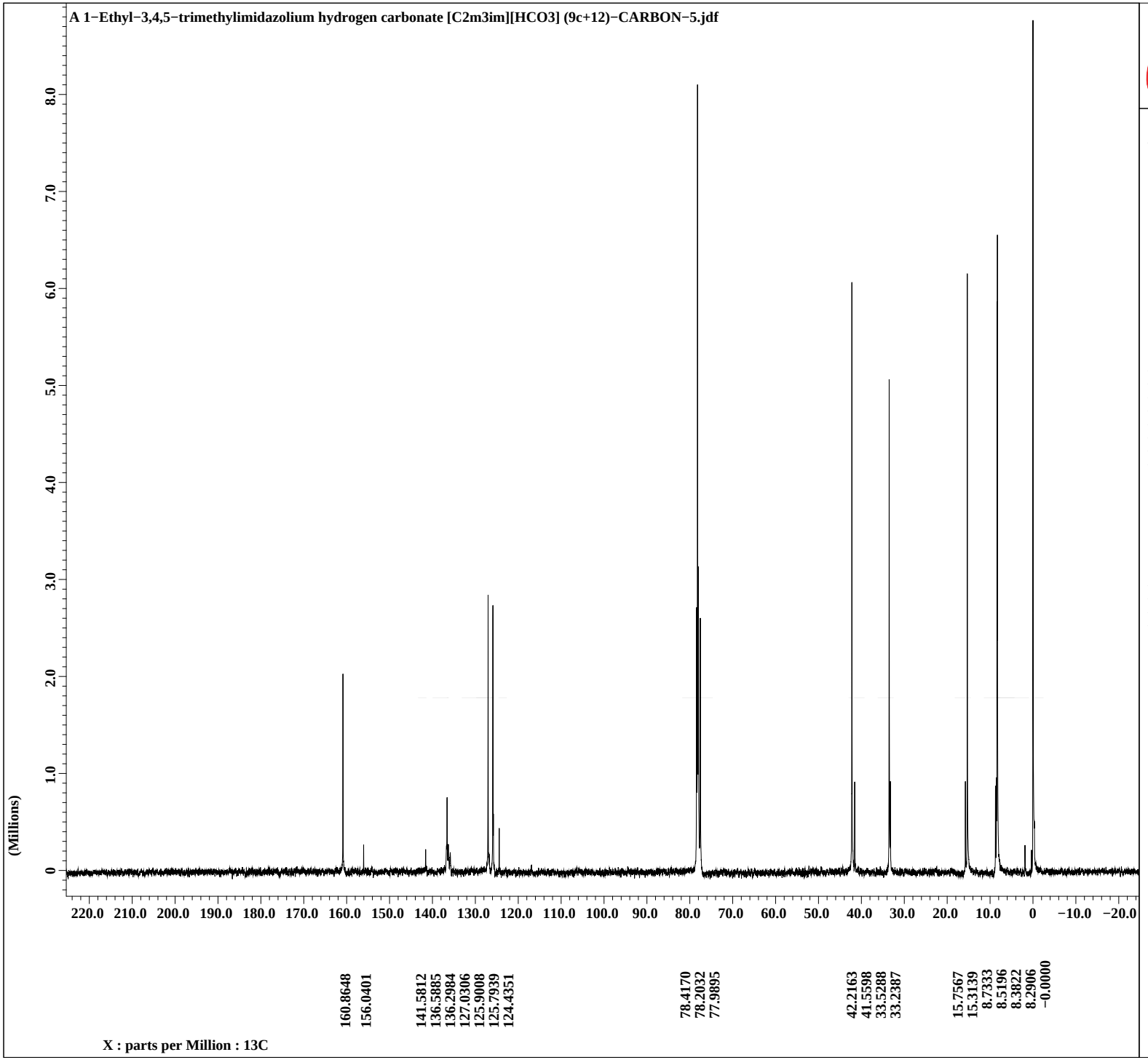


Figure S9. Packing diagram in the crystal structure of ***18a****, along the crystallographic [001] direction.*

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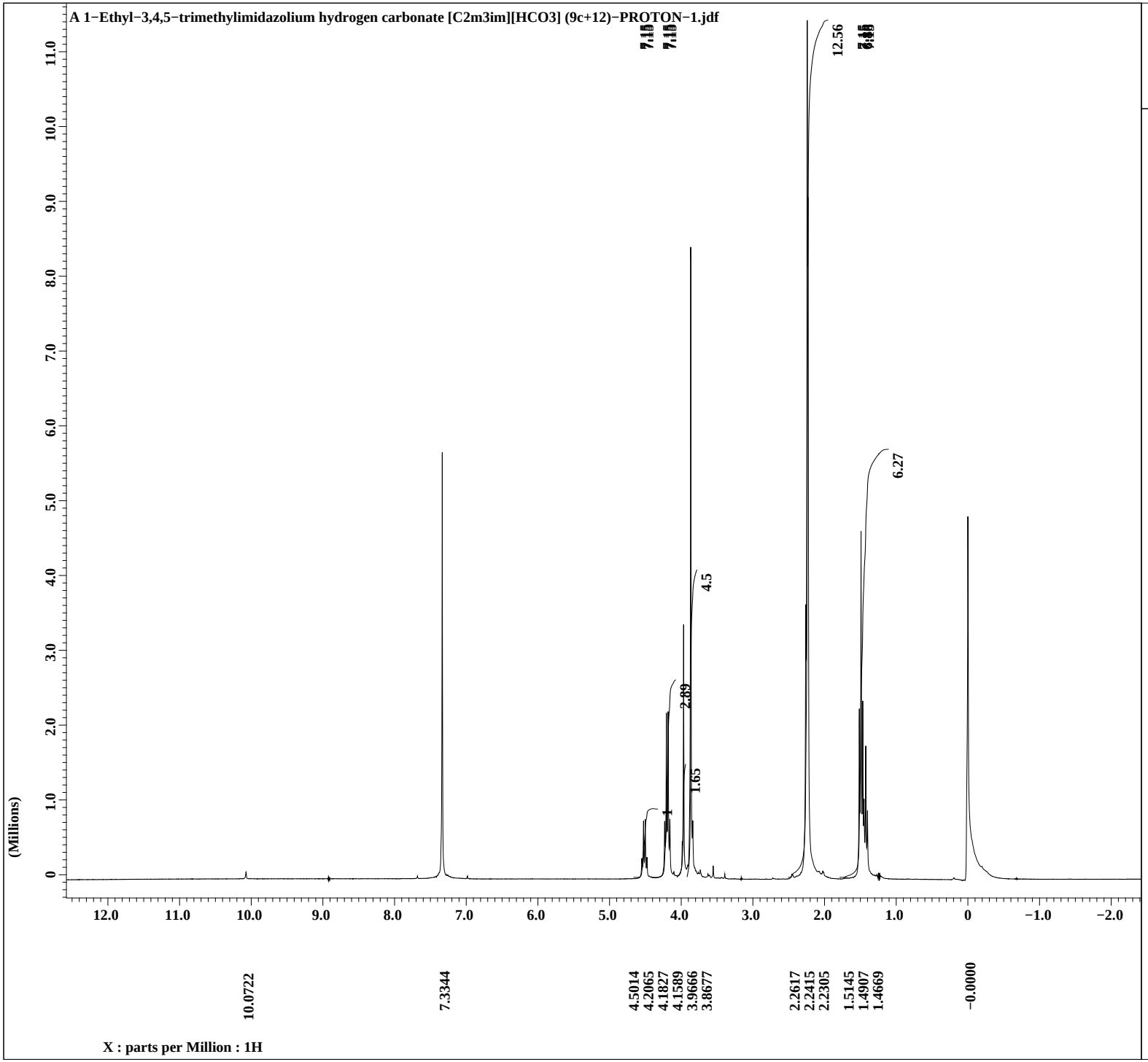


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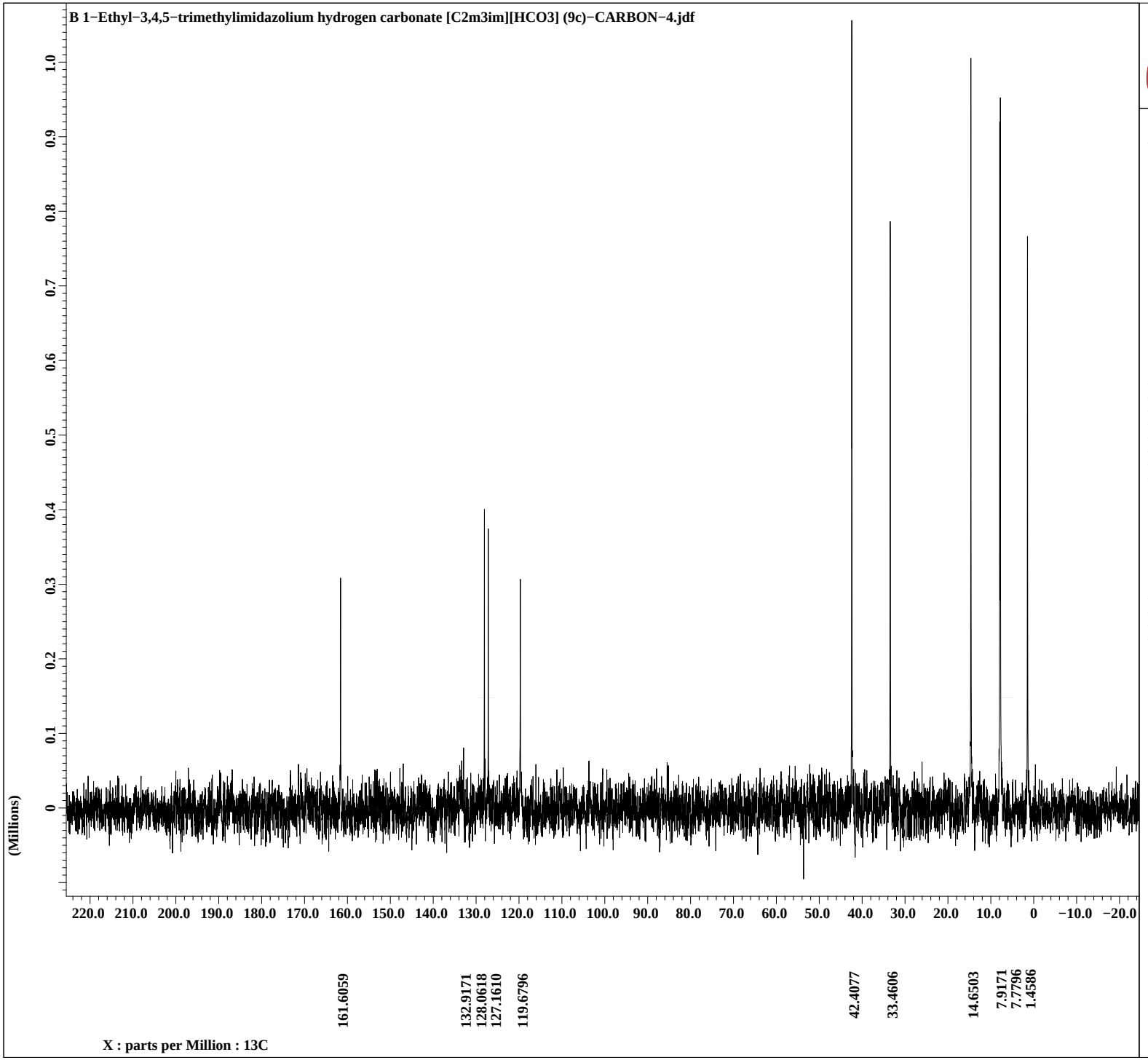

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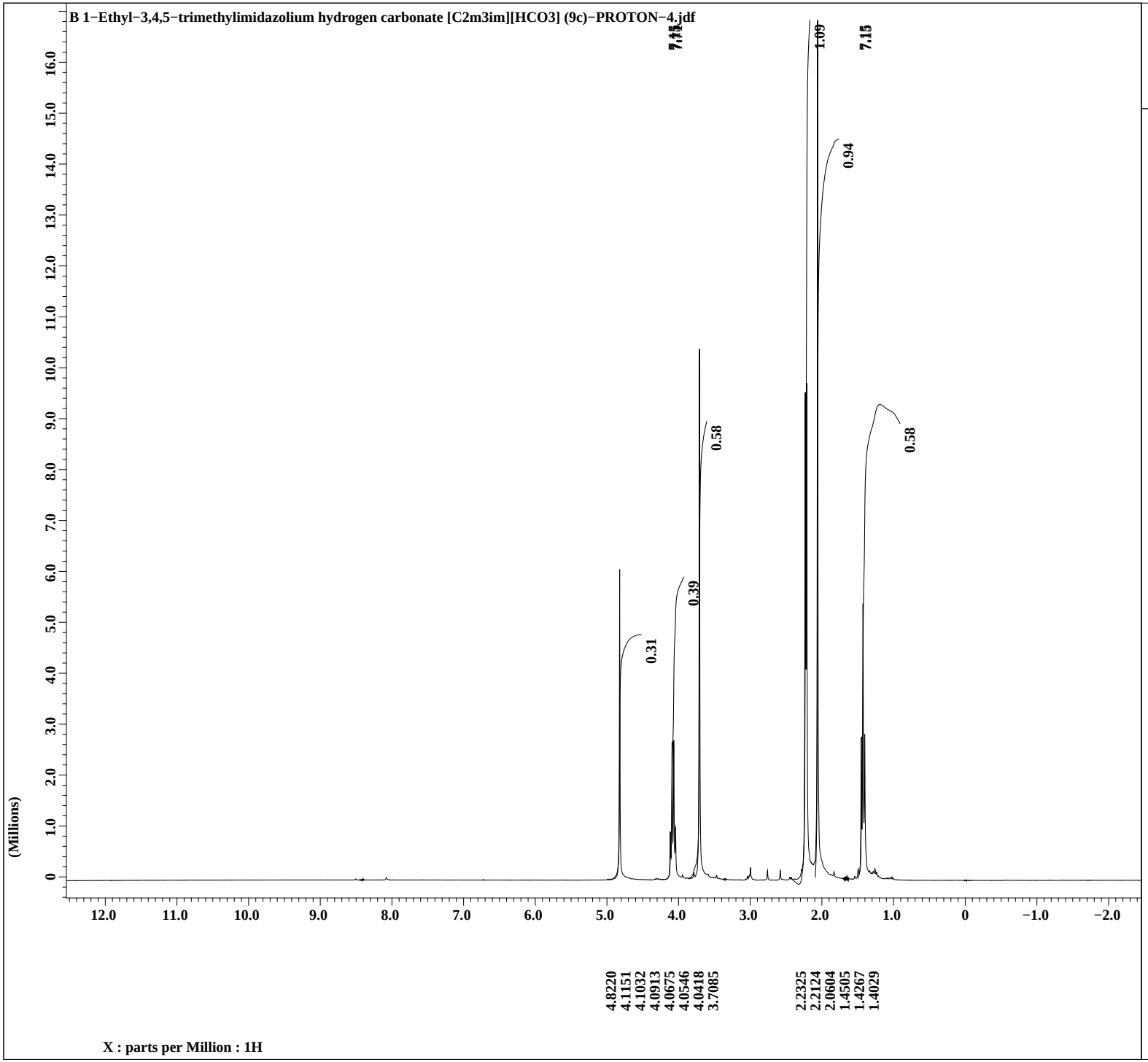

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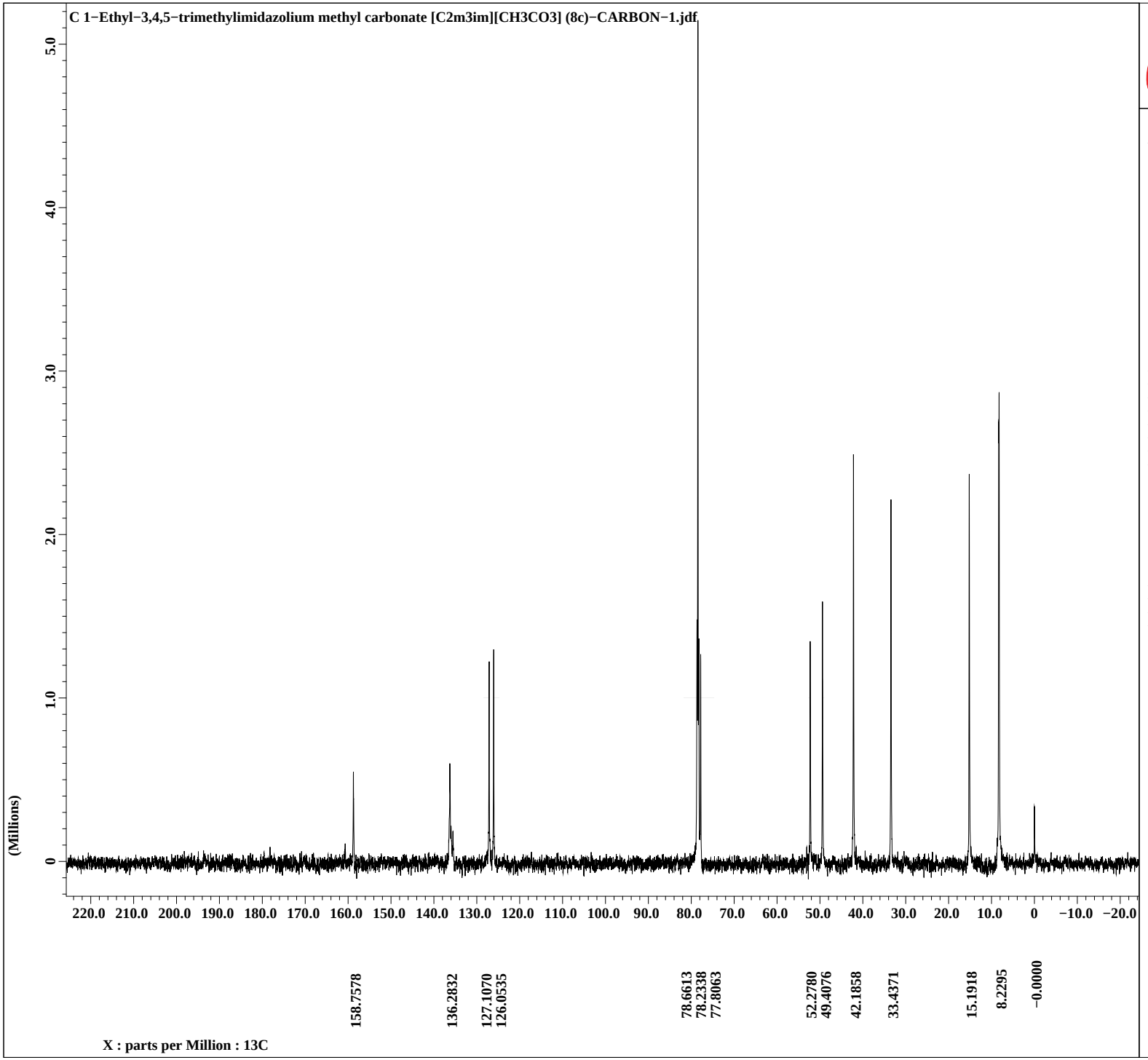

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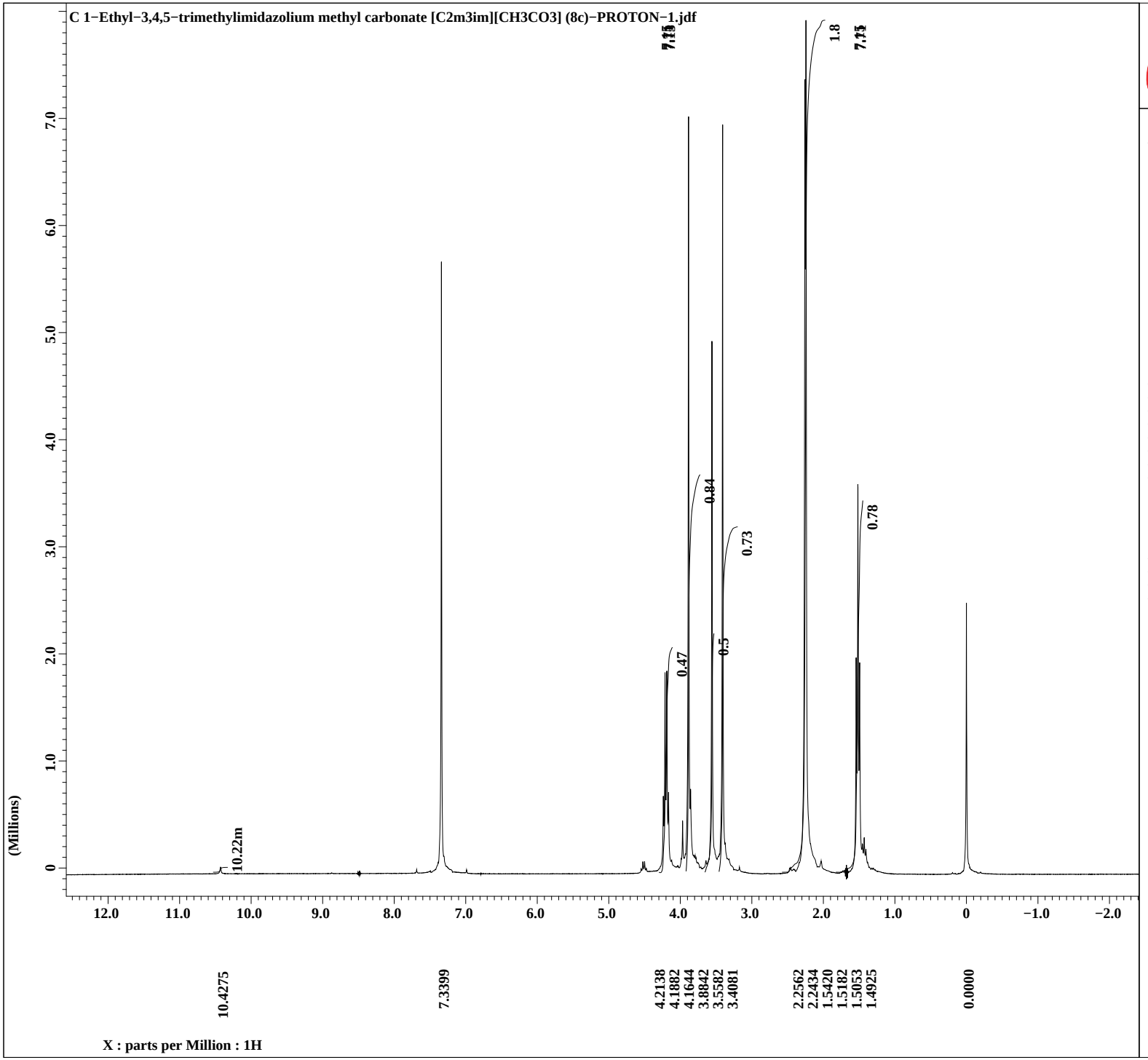


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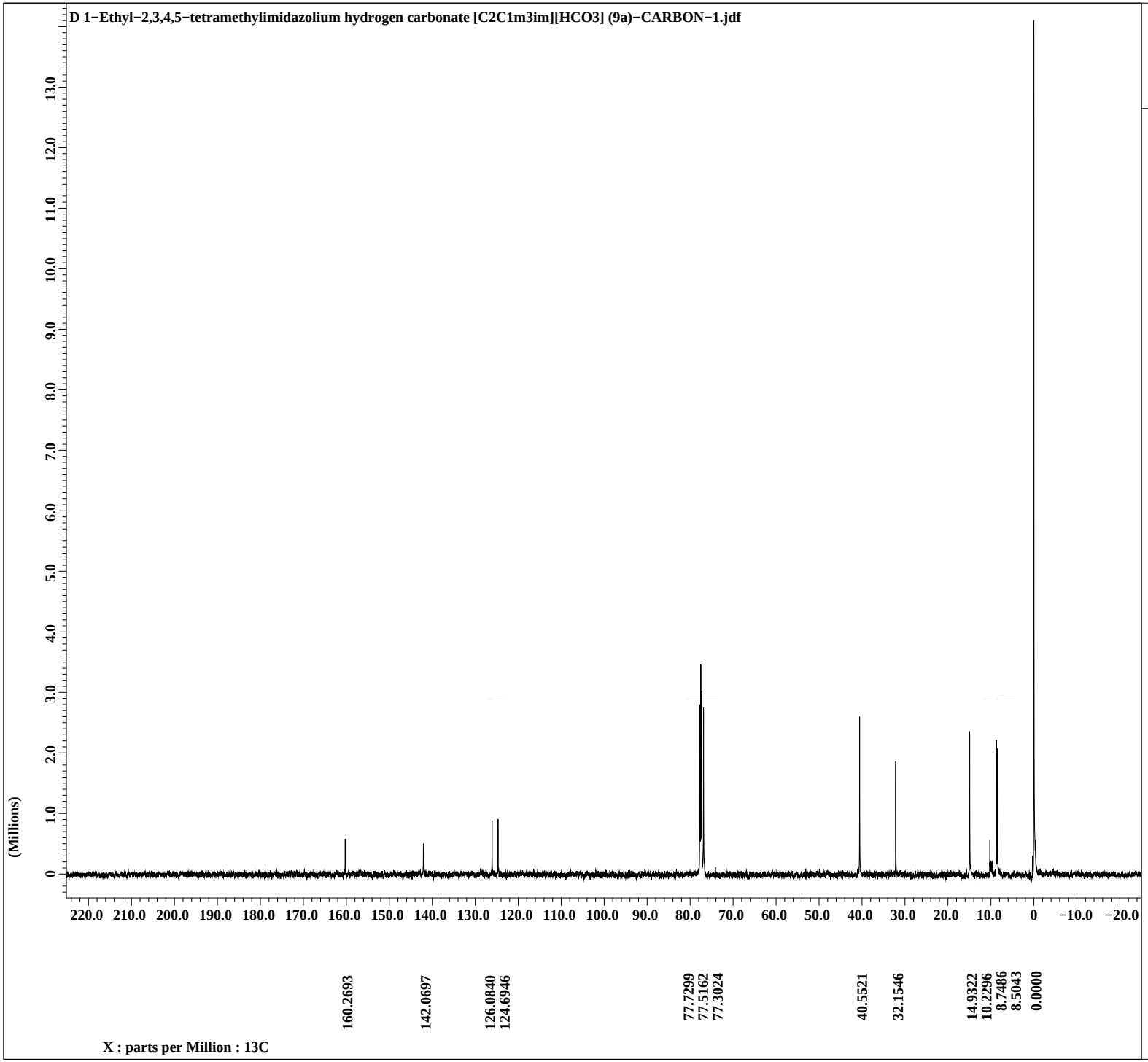

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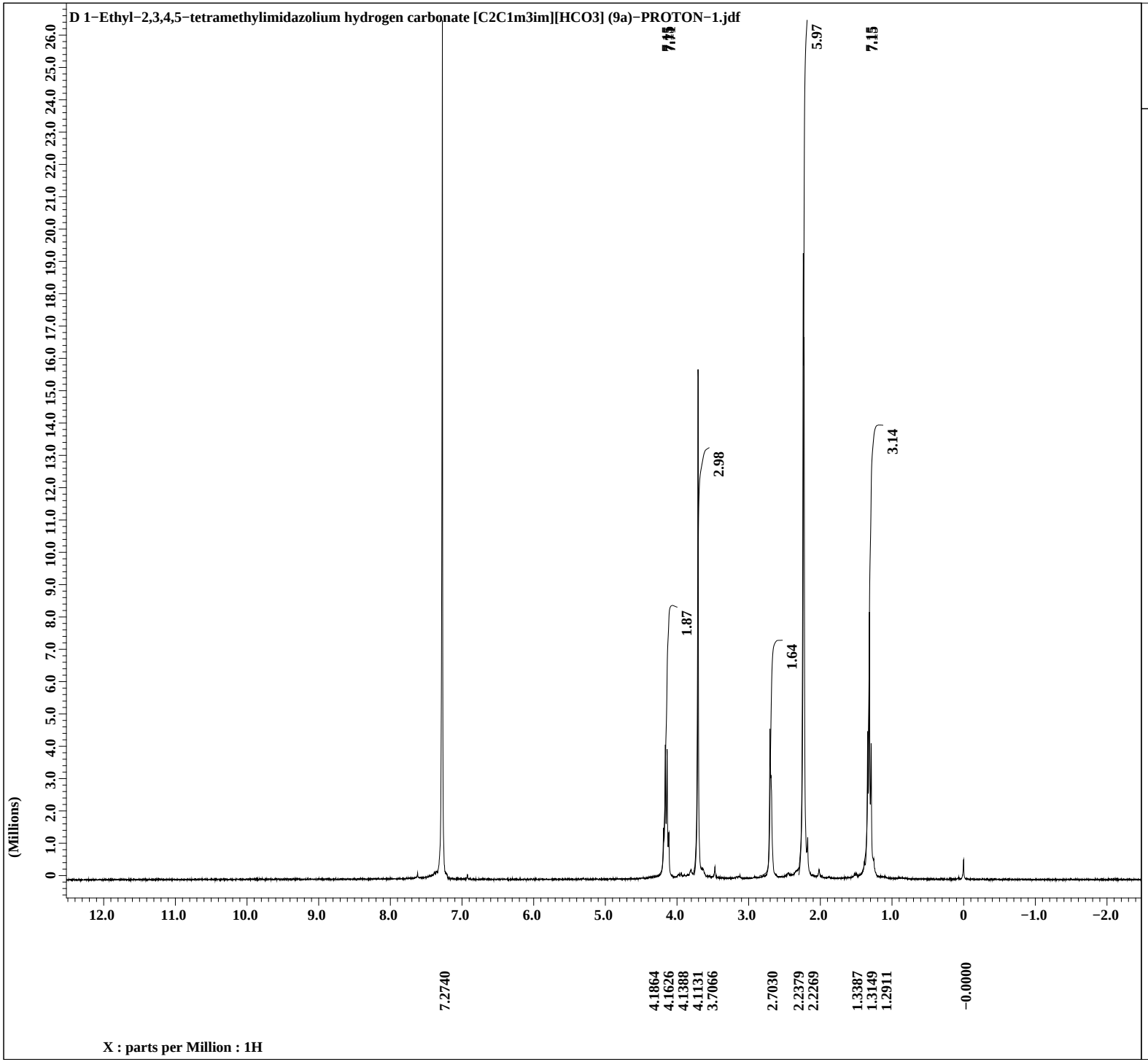

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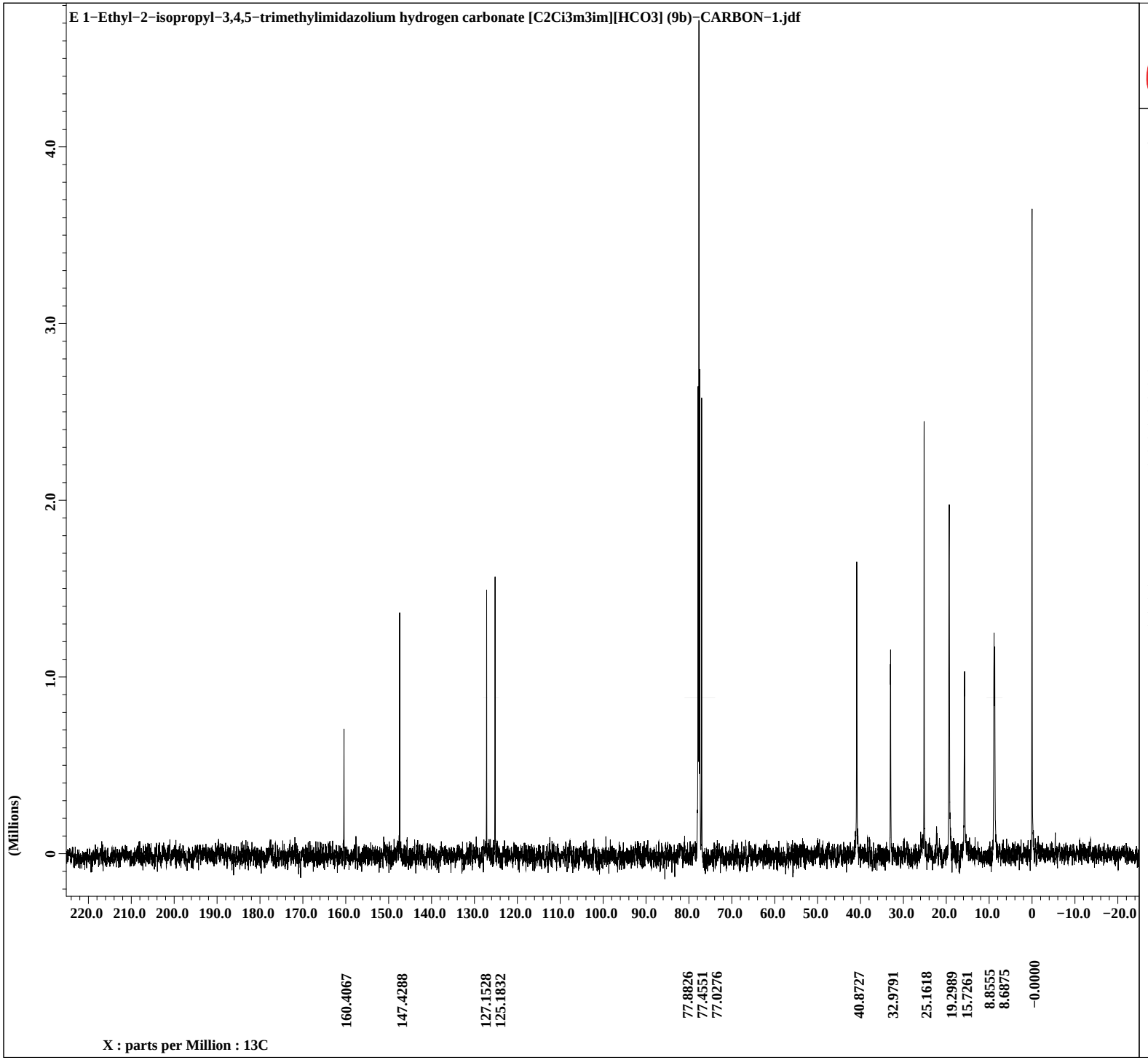
Relaxation_delay	= 5[s]
Temp_get	= 20.8[dC]
Unblank_time	= 2[us]

E 1−Ethyl−2−isopropyl−3,4,5−trimethylimidazolium hydrogen carbonate [C2Ci3m3im][HCO3] (9b)−CARBON−1.jdf

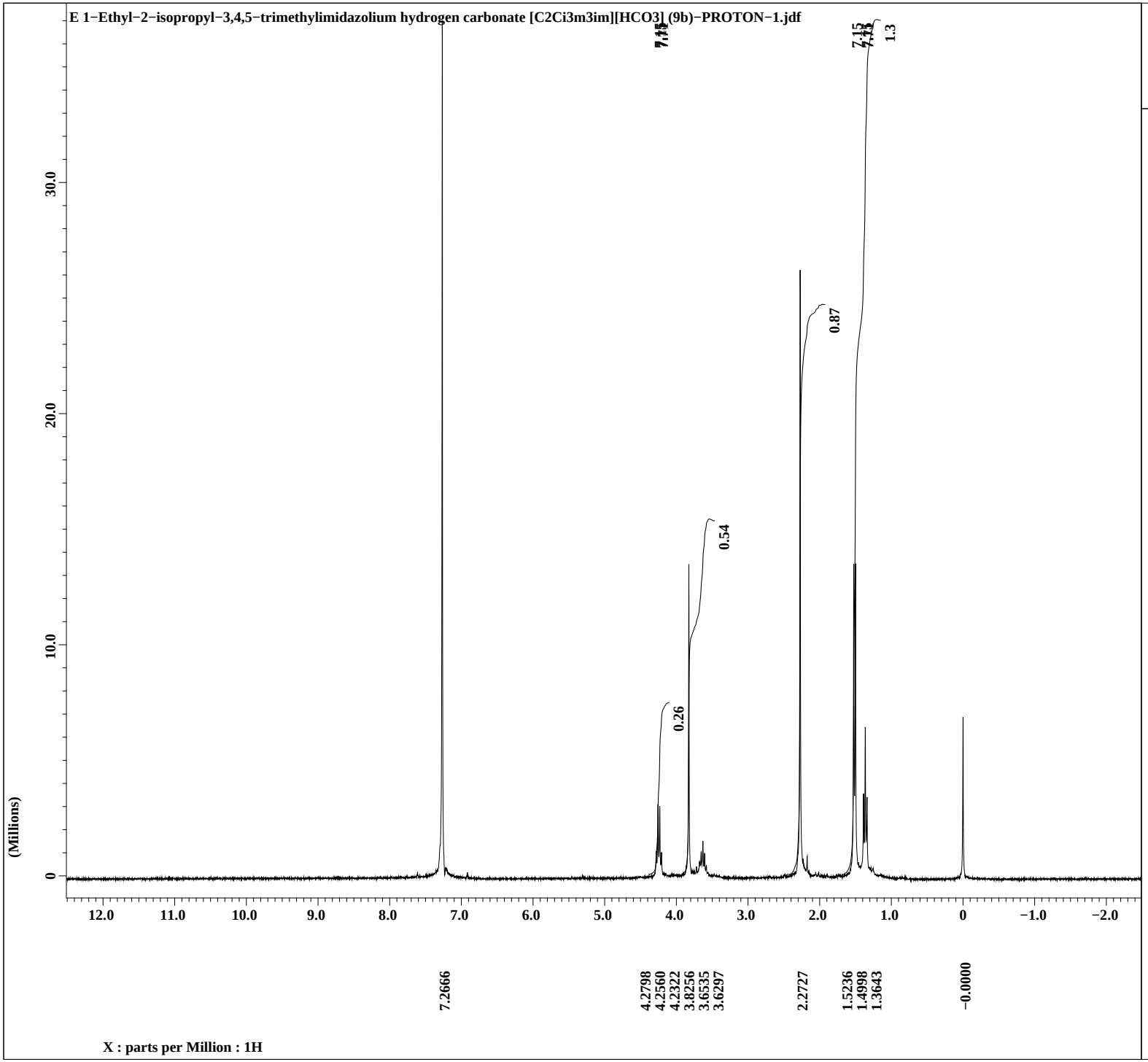


Filename = E 1−Ethyl−2−isopropyl
Author = synthese
Experiment = single_pulse_dec
Sample_id = CM/CMCIL2.47
Solvent = CHLOROFORM−D
Creation_time = 19−SEP−2012 08:44:43
Revision_time = 16−MAY−2013 15:02:25
Current_time = 31−OCT−2013 15:09:41

Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 300
Spectrometer = DELTA_NMR

Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 0.8667136[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 1.15378367[Hz]
X_sweep = 18.90359168[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 4
Scans = 300
Total_scans = 300

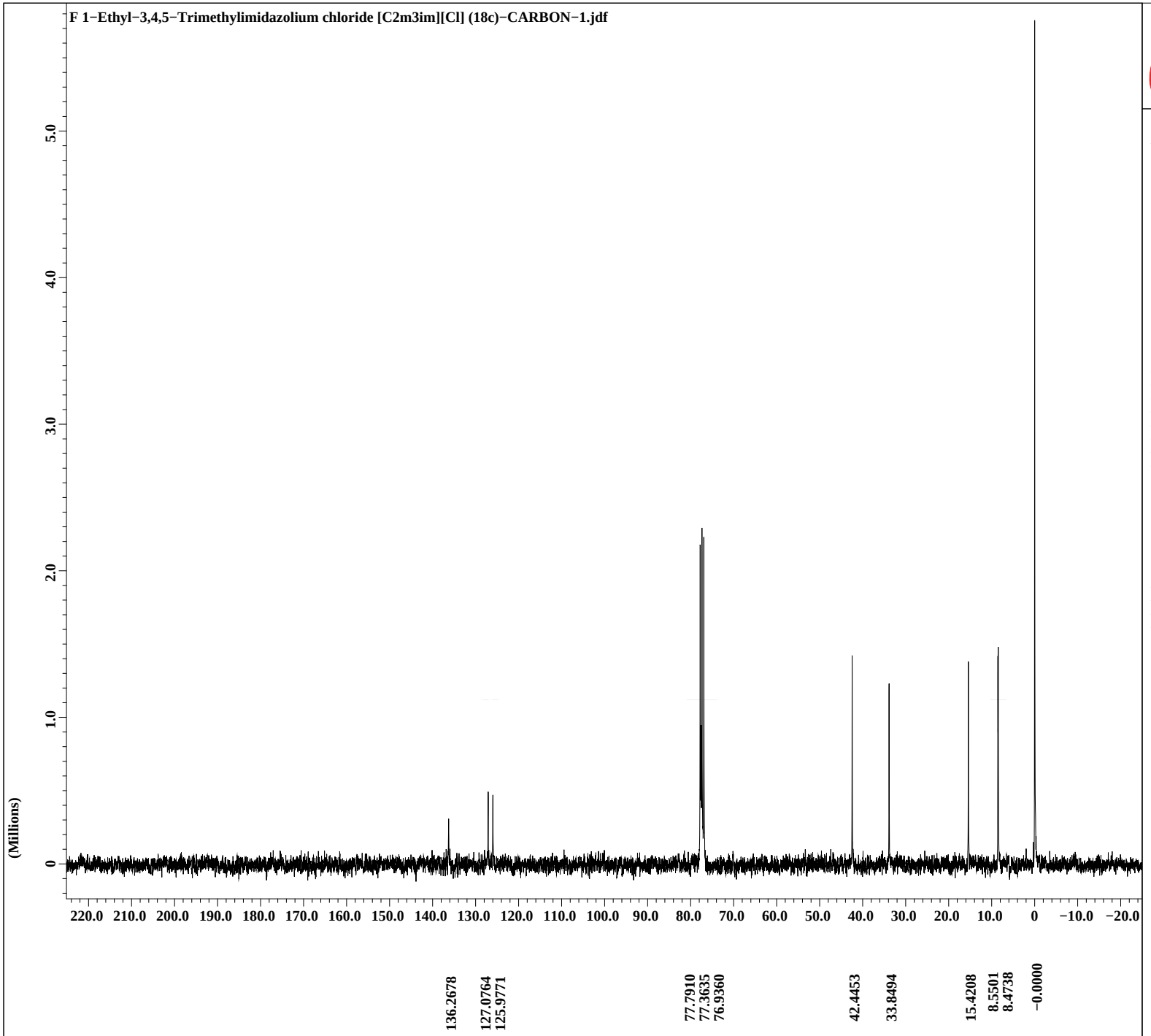
X_90_width = 10[us]
X_acq_time = 0.8667136[s]
X_angle = 30[deg]
X_pulse = 3.33333333[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 28
Relaxation_delay = 1[s]
Temp_get = 19.1[dC]
Unblank_time = 2[us]



JEOL

Filename	= E 1−Ethyl−2−isopropyl
Author	= synthese
Experiment	= single_pulse.exp
Sample_id	= CM/CMCIL2.56−lyoc
Solvent	= CHLOROFORM−D
Creation_time	= 16−MAY−2013 02:15:03
Revision_time	= 16−JUN−2013 11:01:34
Current_time	= 31−OCT−2013 15:10:05
Data_format	= 1D COMPLEX
Dim_size	= 8192
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 300
Spectrometer	= DELTA_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 1.8169856[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 8192
X_prescans	= 0
X_resolution	= 0.55036209[Hz]
X_sweep	= 4.50856628[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 8
Total_scans	= 8
X_90_width	= 14.5[us]
X_acq_time	= 1.8169856[s]
X_angle	= 90[deg]
X_pulse	= 14.5[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 22
Relaxation_delay	= 5[s]
Temp_get	= 21[dC]
Unblank_time	= 2[us]

F 1−Ethyl−3,4,5−Trimethylimidazolium chloride [C2m3im][Cl] (18c)−CARBON−1.jdf



X : parts per Million : 13C

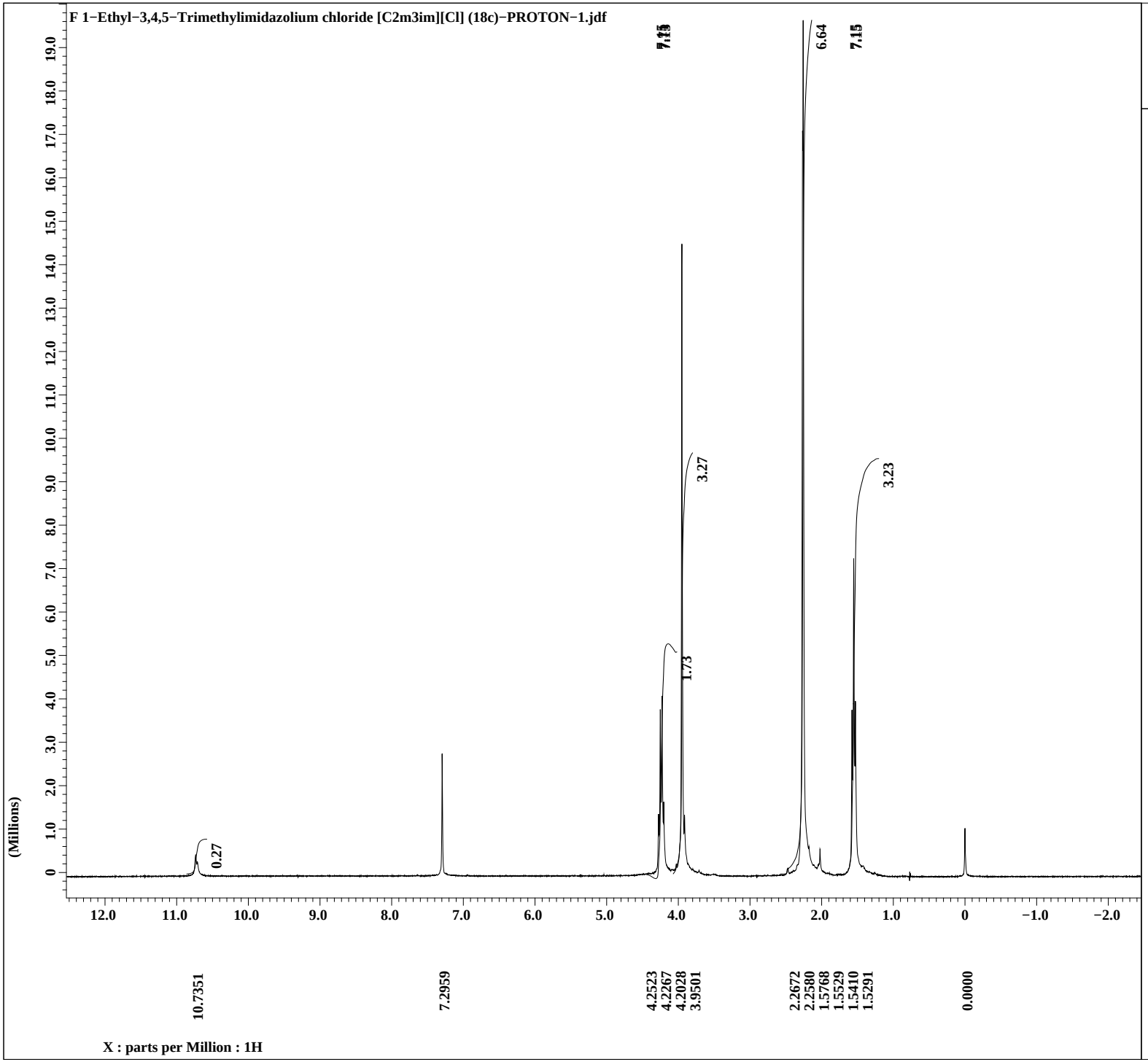
JEOL

```
Filename          = F 1−Ethyl−3,4,5−Trime
Author            = synthese
Experiment        = single_pulse_dec
Sample_id         = CM/CMCIL6.20−ow−air−c
Solvent           = CHLOROFORM−D
Creation_time     = 28−APR−2013 00:34:48
Revision_time     =  9−JUL−2013 19:40:52
Current_time      = 31−OCT−2013 15:10:45

Data_format       = 1D COMPLEX
Dim_size          = 16384
Dim_title         = 13C
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 300
Spectrometer      = DELTA_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 0.8667136[s]
X_domain          = 13C
X_freq            = 75.56823426[MHz]
X_offset          = 100[ppm]
X_points          = 16384
X_prescans        = 0
X_resolution      = 1.15378367[Hz]
X_sweep           = 18.90359168[kHz]
Irr_domain        = 1H
Irr_freq          = 300.52965592[MHz]
Irr_offset        = 5[ppm]
Clipped           = FALSE
Mod_return        = 4
Scans             = 300
Total_scans       = 300

X_90_width        = 10[us]
X_acq_time        = 0.8667136[s]
X_angle           = 30[deg]
X_pulse           = 3.33333333[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 28
Relaxation_delay  = 1[s]
Temp_get          = 21.5[dC]
Unblank_time      = 2[us]
```



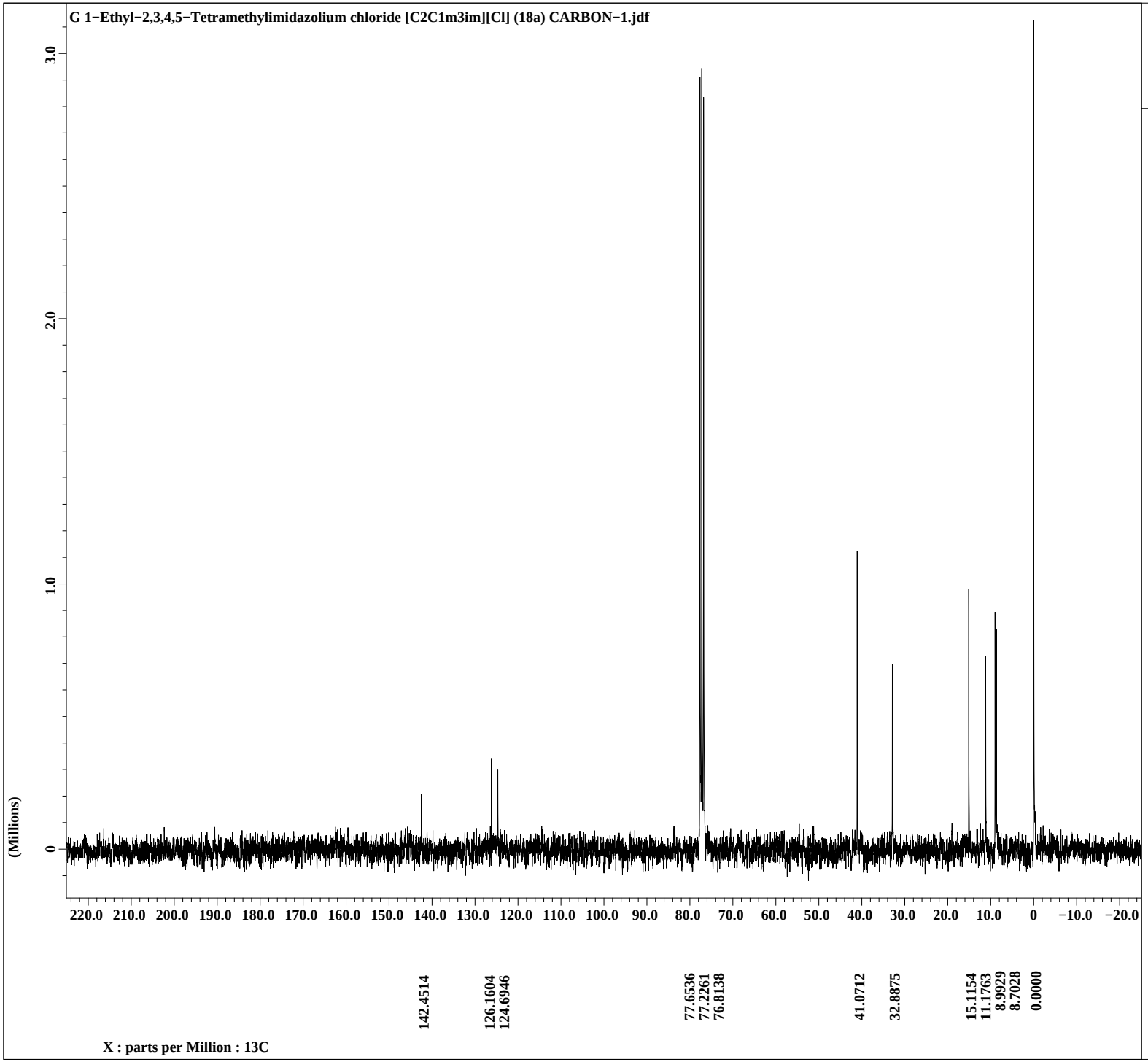
JEOL

```
Filename          = F 1−Ethyl−3,4,5−Trime
Author            = synthese
Experiment        = single_pulse.exp
Sample_id         = CM/CMCIL6.20−oh
Solvent           = CHLOROFORM−D
Creation_time     = 28−APR−2013 04:02:47
Revision_time     =  9−JUL−2013 19:43:27
Current_time      = 31−OCT−2013 15:11:25

Comment           = spectr v ow: [OH]− pr
Data_format       = 1D COMPLEX
Dim_size          = 8192
Dim_title         = 1H
Dim_units         = [ppm]
Dimensions        = X
Site              = Eclipse+ 300
Spectrometer      = DELTA_NMR

Field_strength    = 7.0586013[T] (300[MHz
X_acq_duration    = 1.8169856[s]
X_domain          = 1H
X_freq            = 300.52965592[MHz]
X_offset          = 5[ppm]
X_points          = 8192
X_prescans        = 0
X_resolution      = 0.55036209[Hz]
X_sweep           = 4.50856628[kHz]
Clipped           = FALSE
Mod_return        = 1
Scans             = 8
Total_scans       = 8

X_90_width        = 14.5[us]
X_acq_time        = 1.8169856[s]
X_angle           = 90[deg]
X_pulse           = 14.5[us]
Initial_wait      = 1[s]
Phase_preset      = 3[us]
Recvr_gain        = 16
Relaxation_delay  = 5[s]
Temp_get          = 20.8[dC]
Unblank_time      = 2[us]
```

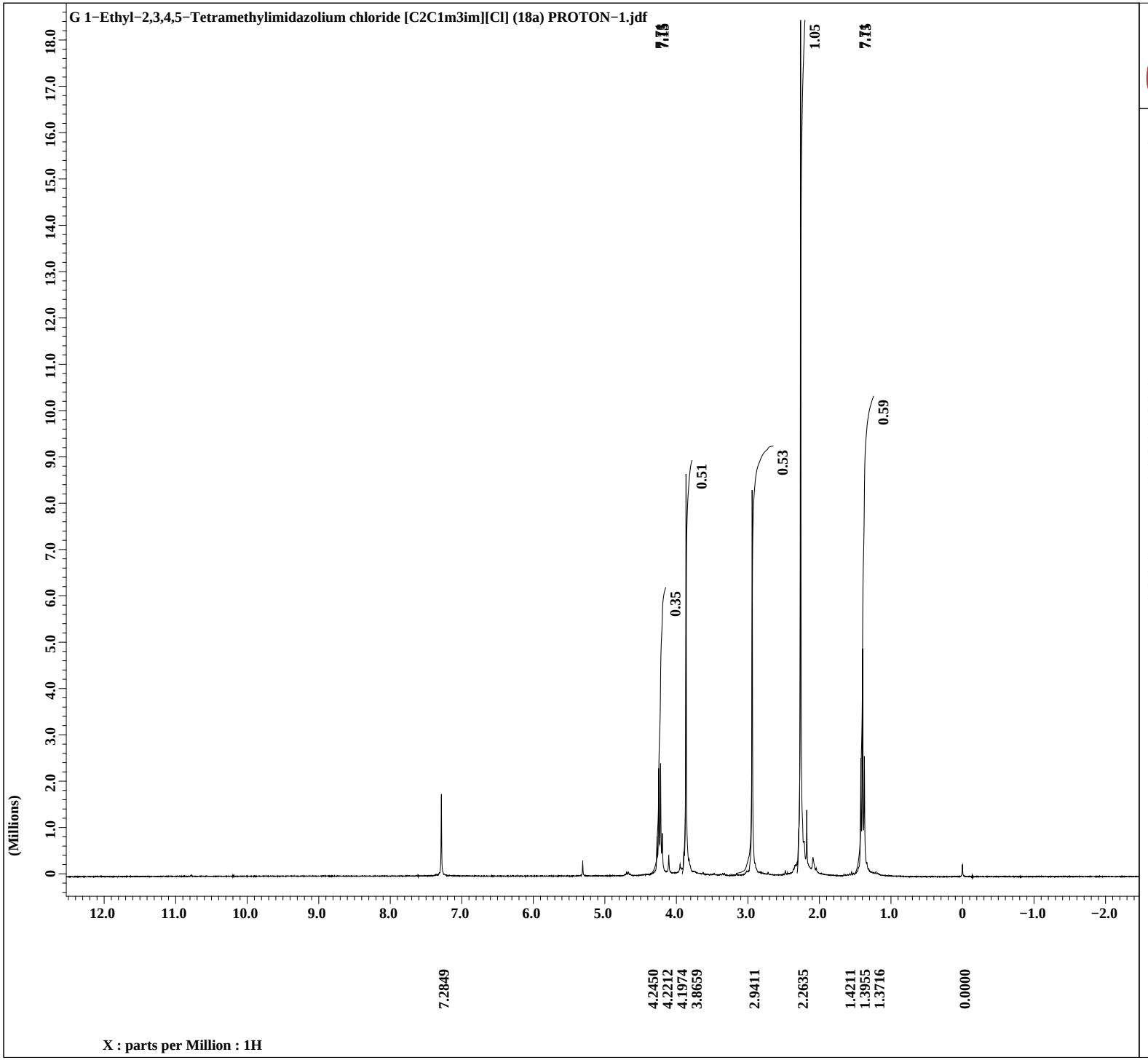


Filename = G 1−Ethyl−2,3,4,5−Tet
Author = synthese
Experiment = single_pulse_dec
Sample_id = CM/CMCIL5.9−cl−carbon
Solvent = CHLOROFORM−D
Creation_time = 9−APR−2013 22:39:25
Revision_time = 9−MAY−2013 18:07:25
Current_time = 31−OCT−2013 15:11:59

Data_format = 1D COMPLEX
Dim_size = 16384
Dim_title = 13C
Dim_units = [ppm]
Dimensions = X
Site = Eclipse+ 300
Spectrometer = DELTA_NMR

Field_strength = 7.0586013[T] (300[MHz
X_acq_duration = 0.8667136[s]
X_domain = 13C
X_freq = 75.56823426[MHz]
X_offset = 100[ppm]
X_points = 16384
X_prescans = 0
X_resolution = 1.15378367[Hz]
X_sweep = 18.90359168[kHz]
Irr_domain = 1H
Irr_freq = 300.52965592[MHz]
Irr_offset = 5[ppm]
Clipped = FALSE
Mod_return = 4
Scans = 320
Total_scans = 320

X_90_width = 10[us]
X_acq_time = 0.8667136[s]
X_angle = 30[deg]
X_pulse = 3.33333333[us]
Initial_wait = 1[s]
Phase_preset = 3[us]
Recvr_gain = 28
Relaxation_delay = 1[s]
Temp_get = 23.5[dC]
Unblank_time = 2[us]



Filename	= G 1−Ethyl−2,3,4,5−Tet
Author	= synthese
Experiment	= single_pulse.exp
Sample_id	= CM/CMCIL5.9
Solvent	= CHLOROFORM−D
Creation_time	= 9−APR−2013 08:50:41
Revision_time	= 9−MAY−2013 19:06:36
Current_time	= 31−OCT−2013 15:12:34
Comment	= precip et20 uit warme
Data_format	= 1D COMPLEX
Dim_size	= 8192
Dim_title	= 1H
Dim_units	= [ppm]
Dimensions	= X
Site	= Eclipse+ 300
Spectrometer	= DELTA_NMR
Field_strength	= 7.0586013[T] (300[MHz
X_acq_duration	= 1.8169856[s]
X_domain	= 1H
X_freq	= 300.52965592[MHz]
X_offset	= 5[ppm]
X_points	= 8192
X_prescans	= 0
X_resolution	= 0.55036209[Hz]
X_sweep	= 4.50856628[kHz]
Clipped	= FALSE
Mod_return	= 1
Scans	= 8
Total_scans	= 8
X_90_width	= 14.5[us]
X_acq_time	= 1.8169856[s]
X_angle	= 90[deg]
X_pulse	= 14.5[us]
Initial_wait	= 1[s]
Phase_preset	= 3[us]
Recvr_gain	= 16
Relaxation_delay	= 5[s]
Temp_get	= 20.4[dC]
Unblank_time	= 2[us]