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Supplementary Materials for

Processing of methane and acetylene ices by galactic cosmic rays and implications to the color diversity of Kuiper Belt objects

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This PDF file includes:

Python code used to analysis the secondary ion mass spectra (SIMS)

```
import numpy as np
import matplotlib.pyplot as plt
import scipy.interpolate as inter
import scipy.optimize as opt
import pandas as pd

m_c13=13.00336 #exact mass of C-13 isotope, amu
m_h=1.00783 #exact mass of H-1 isotope, amu

path='sample_{ind}/Sample {ind}/'#path to the data file
ind=1#index of the data file
file='A22249.'#name of the data file

mspec=np.loadtxt(path.format(ind=ind)+file+'txt',skiprows=3).T #load data file
c_off=200 #intensity cut-off
plt.plot(mspec[1],c_off*np.ones(len(mspec[1])),label='Cut-off') #plot cut-off
plt.plot(mspec[1],mspec[2],label='Raw data') #plot raw data
mlist=[] #creating a list of x, y and masses for (C-13)x(H-1)y compounds, x = 1-39,
y = 0 - 2*x+3
for i in np.arange(1,500//13+1):
    for j in range(2*i+3):
        mass=m_c13*i+m_h*j
        if mass<500:
            mlist.append([i,j,mass])
mlist=np.asarray(mlist)
def findpnts(data,x,dx,NoPlot=True): #procedure to select peaks in the dx vicinity of
x
    res=np.argwhere(np.abs(data[1]-x)<dx)
    if not NoPlot:
        plt.plot(data[1,res],data[2,res])
    return res
out=np.array([[],[],[],[]]).T #creating array of processed data
for i in mlist: #populating array of processed data with number of counts within
m/5000 of mslist[i]
    res=findpnts(mspec,i[2],i[2]/5000)
    res1=np.sum(np.where(mspec[2,res]>c_off,mspec[2,res],0))
    if res1>0:
        out=np.vstack((out,np.array([i[0],i[1],i[2],res1]).T))

plotdata=np.zeros((2,3*len(out))) #creating array for stick spectrum of processed
data
f=open(path.format(ind=ind)+file+'out','a') #.out file
for i in range(len(out)): #populating stick spectrum array, saving processed data
into .out file
    plotdata[0,3*i]=out[i,2]-1e-6
```

```
    plotdata[0,3*i+1]=out[i,2]
    plotdata[0,3*i+2]=out[i,2]+1e-6
    plotdata[1,3*i+1]=out[i,3]
    f.write('C'+str(int(out[i,0]))+'H'+str(int(out[i,1]))+'    '+str(round(out[i,2],4))+'
'+str(int(out[i,3]))+'\n')
f.close()
plt.plot(plotdata[0],-plotdata[1],label='Stick spectrum') #plotting stick spectrum
gatedata=np.zeros((2,4*len(out))) #creating array for mass gates
for i in range(len(out)):
    gatedata[0,4*i]=out[i,2]*(1-1/5000)-1e-6
    gatedata[0,4*i+1]=out[i,2]*(1-1/5000)
    gatedata[1,4*i+1]=out[i,3]/10
    gatedata[0,4*i+2]=out[i,2]*(1+1/5000)
    gatedata[1,4*i+2]=out[i,3]/10
    gatedata[0,4*i+3]=out[i,2]*(1+1/5000)+1e-6
plt.plot(gatedata[0],gatedata[1],label='Gates') #plotting mass gates

plt.legend()
```

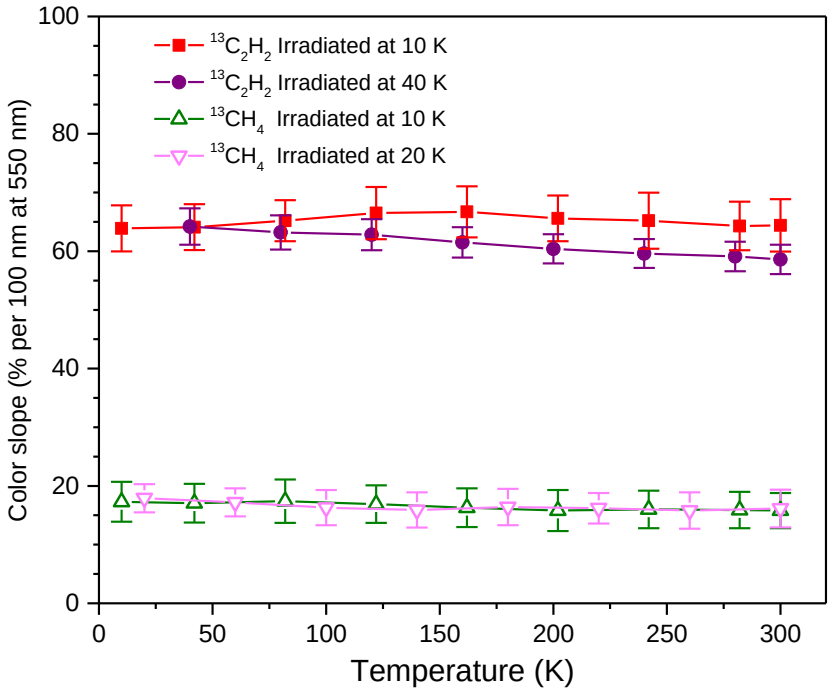


Fig. S1 The color slope evolutions of irradiated ^{13}C-acetylene ($^{13}C_2H_2$) and ^{13}C-methane ($^{13}CH_4$) during warming-up those ice mixtures up to 300 K.

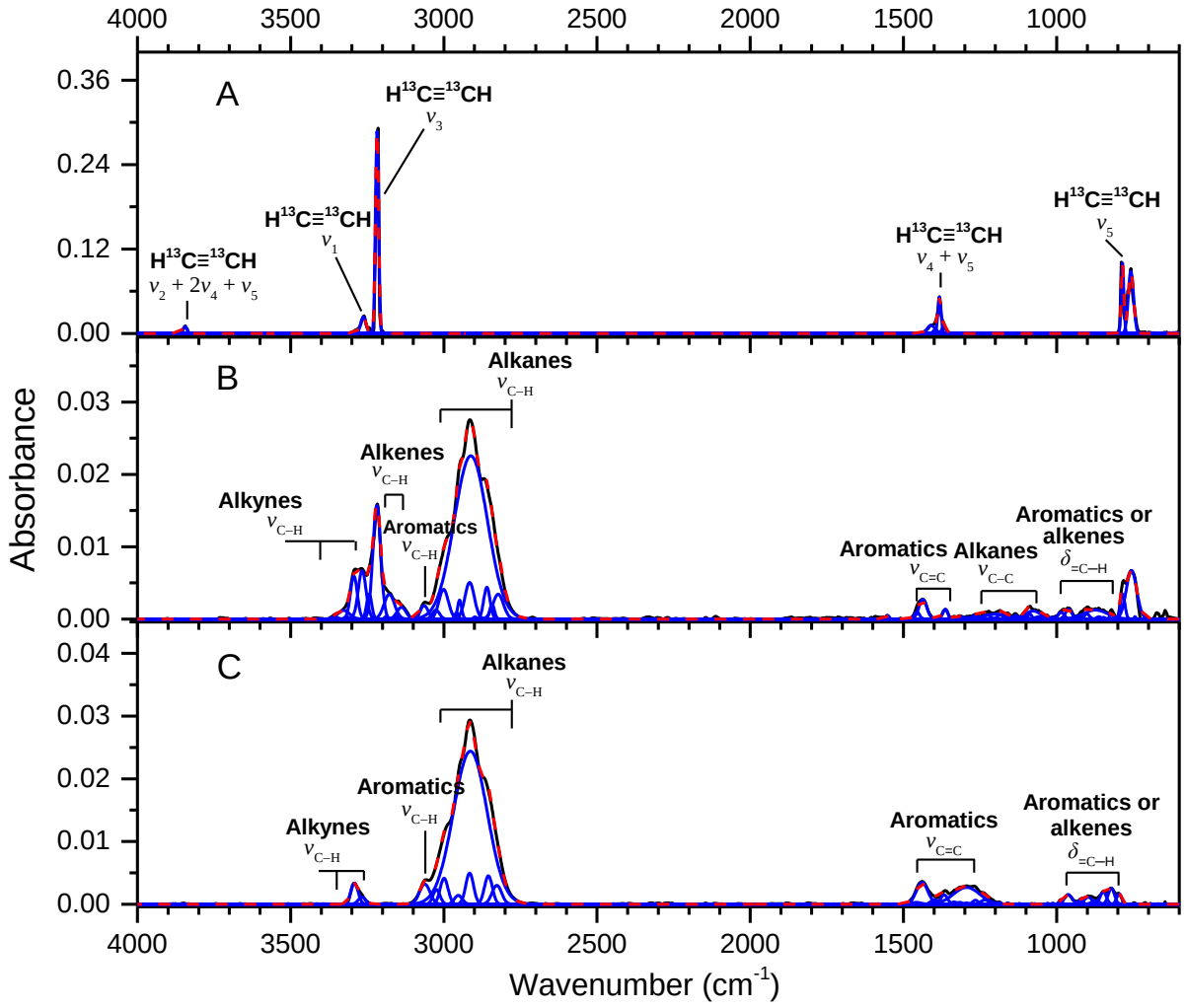


Fig. S2. Deconvoluted infrared spectra of ^{13}C-acetylene ($^{13}C_2H_2$). (**A**) Pristine ices at 40 K. **(B)** After irradiation at 40 K. **(C**) The residue at 300 K. For clarity, only significant peaks are labeled, detailed assignments can be found in Table S3.

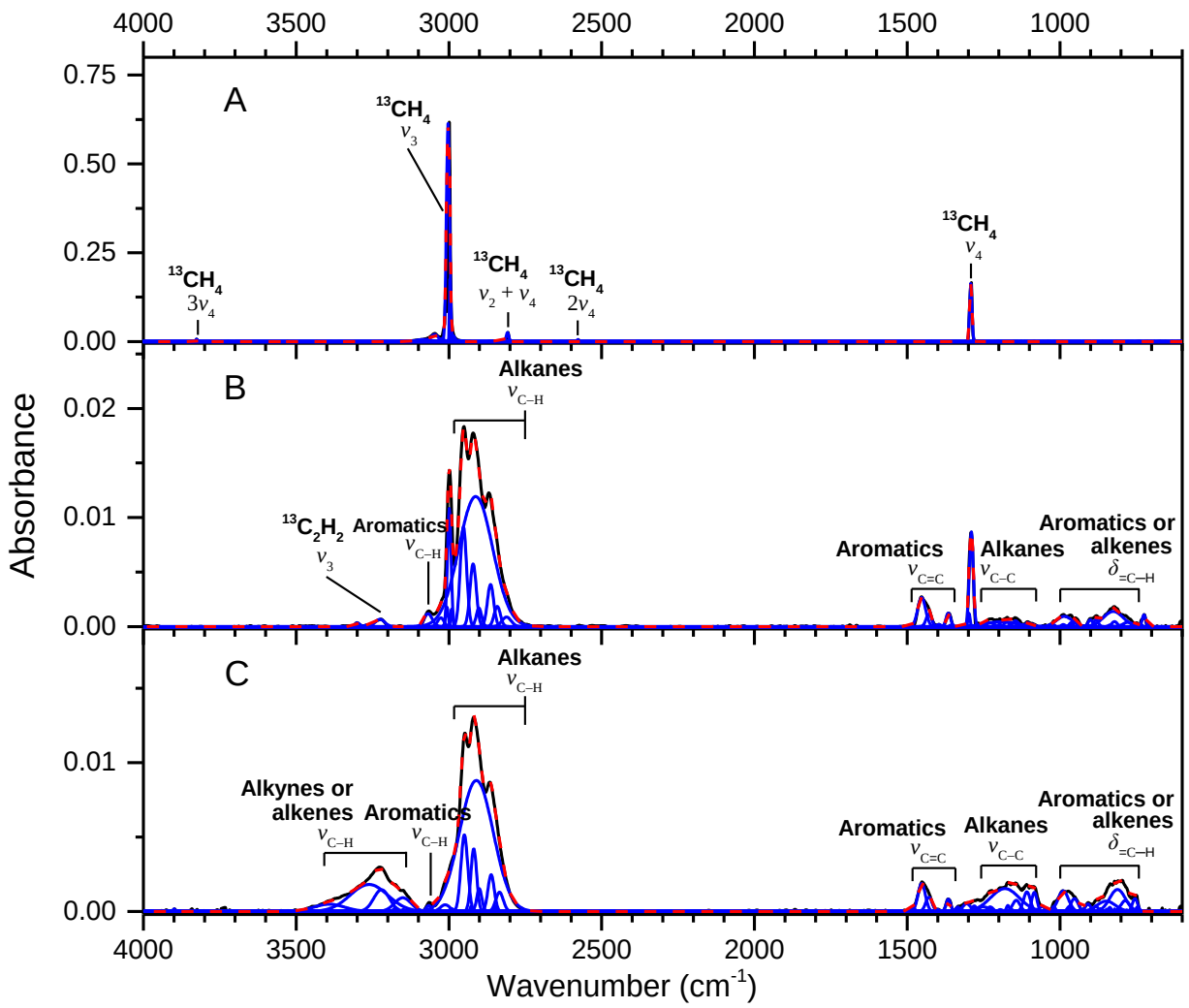


Fig. S3. Deconvoluted infrared spectra of ^{13}C-methane ($^{13}CH_4$). (**A**) Pristine ices at 10 K. **(B**) After irradiation at 10 K. **(C**) The residue at 300 K. For clarity, only significant peaks are labeled, detailed assignments can be found in Table S4.

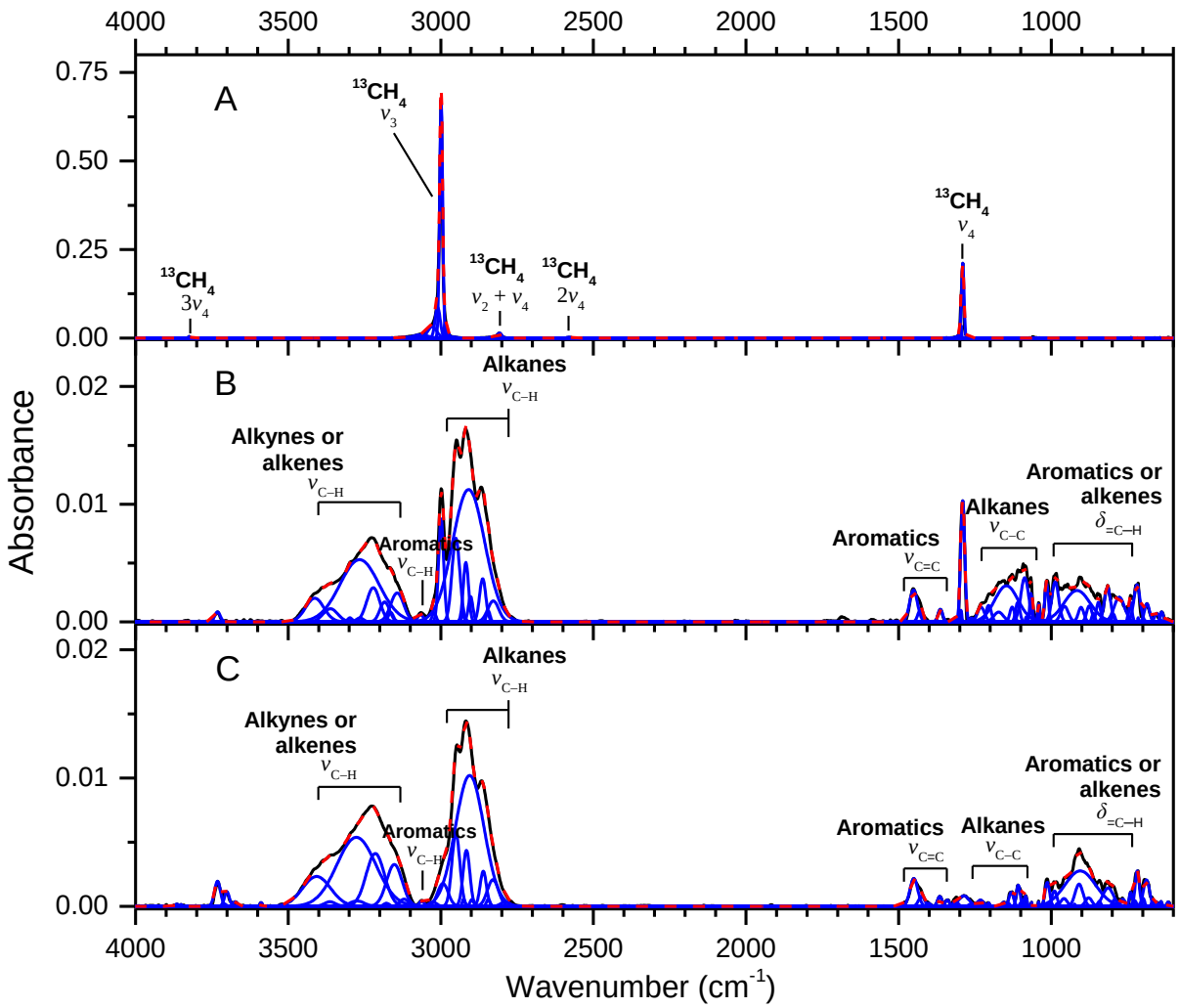


Fig. S4. Deconvoluted infrared spectra of ^{13}C-methane ($^{13}CH_4$). (**A**) Pristine ices at 20 K. **(B**) After irradiation at 20 K. **(C**) The residue at 300 K. For clarity, only significant peaks are labeled, detailed assignments can be found in Table S5.

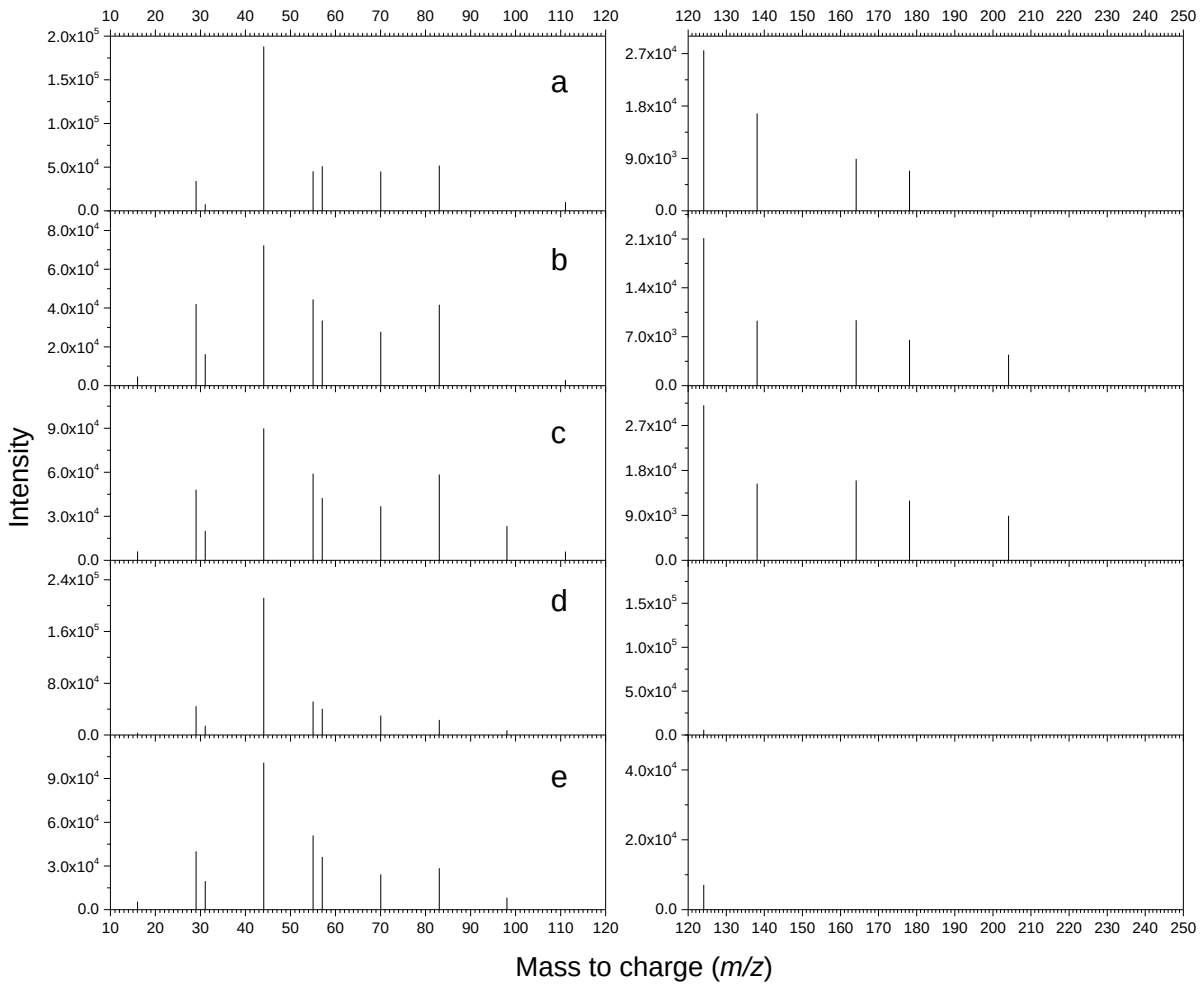


Fig. S5. Positive ion SIMS data from residues of irradiated ^{13}C–acetylene ($^{13}C_2H_2$). The data were recorded in the mass to charge from 10 to 120 (left) and mass to charge from 120 to 250 (right) correlated with (a) 0.5 eV amu^{-1}, (b) 4.6 eV amu^{-1}, (c) 8.2 eV amu^{-1}, (d) 46.4 eV amu^{-1}, and (e) 82.1 eV amu^{-1}.

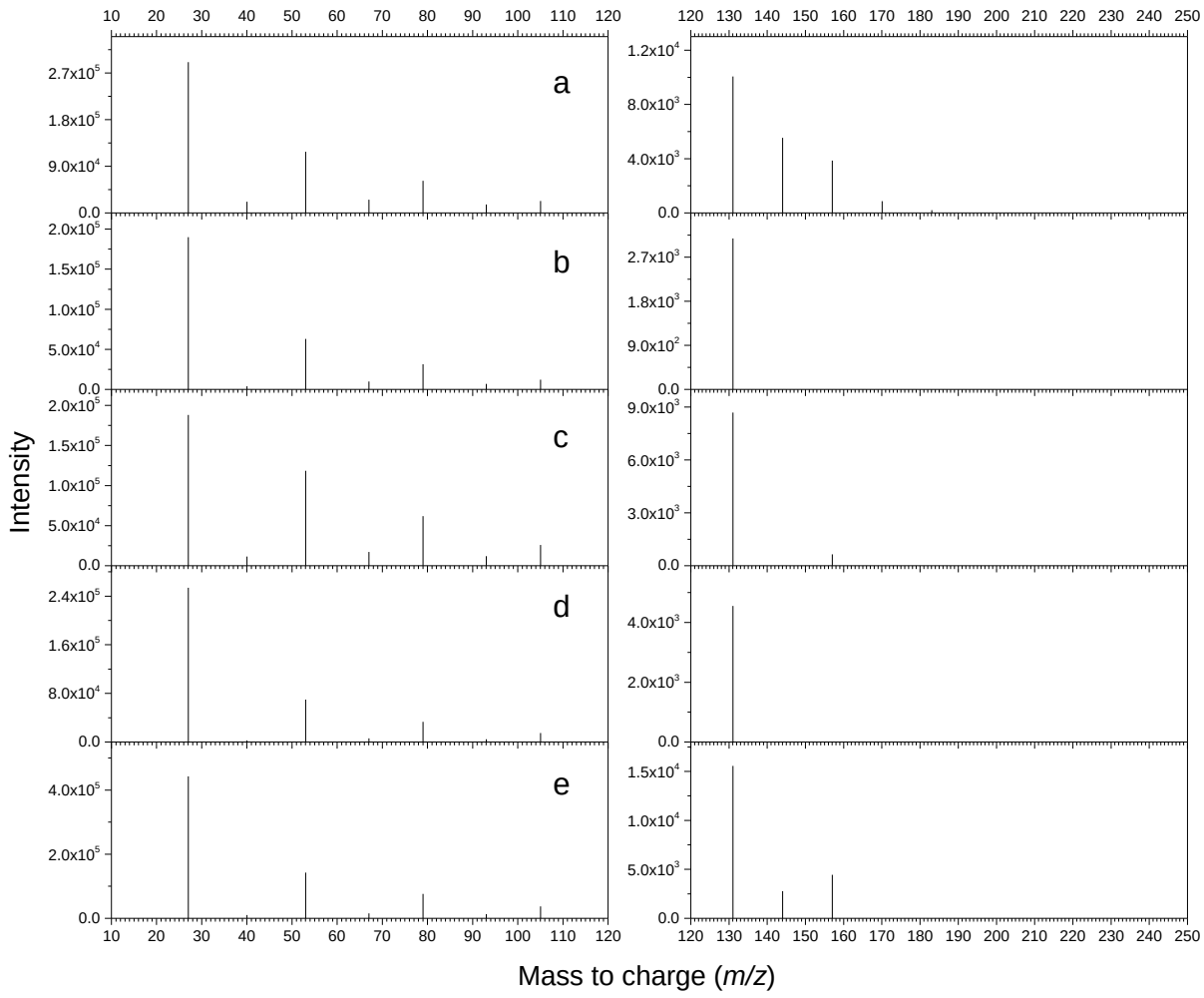


Fig. S6. Negative ions SIMS data from residues of irradiated ^{13}C-acetylene ($^{13}C_2H_2$). The data were recorded in the mass to charge from 10 to 120 (left) and mass to charge from 120 to 250 (right) correlated with (a) 0.5 eV amu^{-1}, (b) 4.6 eV amu^{-1}, (c) 8.2 eV amu^{-1}, (d) 46.4 eV amu^{-1}, and (e) 82.1 eV amu^{-1}.

Table S1. Data applied to calculate the average irradiation dose per molecule.

Initial kinetic energy of the electrons, E_{init} (keV)		5	
Ice		$^{13}C_2H_2$	$^{13}CH_4$
Irradiation current, I (nA)		1500 ± 115	1180 ± 70
Total number of electrons		3.37×10^{17}	2.65×10^{17}
Average penetration depth, l (nm)[a]		320 ± 30	330 ± 30
Average kinetic energy of backscattered electrons, E_{bs} (keV)[a]		3230 ± 320	3000 ± 300
Fraction of backscattered electrons, f_{bs}[a]		0.33 ± 0.03	0.26 ± 0.03
Average kinetic energy of transmitted electrons, E_{trans} (keV)[a]		0	0
Fraction of transmitted electrons, f_{trans}[a]		0	0
Irradiated area, A (cm^2)		1 ± 0.05	1 ± 0.05
Dose (eV $molecule^{-1}$)	$^{13}C_2H_2$	2300 ± 320	
	$^{13}CH_4$	1400 ± 190	
Dose (eV amu^{-1})	$^{13}C_2H_2$	82.1 ± 10.0	
	$^{13}CH_4$	82.1 ± 10.0	

Note.

[a] Parameters obtained from CASINO software v2.4.

Table S2. Infrared absorption features were recorded before the electron irradiation, after the electron irradiation (82.1 ± 10 eV amu^{-1}), and after the temperature-programmed desorption (TPD) of ^{13}C-acetylene ice ($^{13}C_2H_2$) at 10 K.

Absorption before irradiation (cm^{-1})	Absorption after irradiation (cm^{-1})	Absorption after TPD (cm^{-1})	Assignment	Reference
5101, 4052, 3845			$5\nu_4 + 3\nu_5$, $\nu_1 + \nu_5$, $\nu_2 + 2\nu_4 + \nu_5$	(*74*, *75*)
	3529, 3457, 3374	3293, 3261, 3226	C–H stretch (ν_{C-H}, alkynes, –C≡C–H)	(*74-76*)
3317			ν_1	(*74*, *75*)
3242, 3216			ν_3	(*74*, *75*)
	3170, 3128	3021, 3000	C–H stretch (ν_{C-H}, alkenes, –CH=CH$_2$)	(*76*)
	3070, 3045, 3000	3062	C–H stretch (ν_{C-H}, aromatics, =CH–)	(*76*, *77*)
	2952, 2917, 2912	2917, 2913, 2898,	asymmetric C–H stretch (ν_{C-H}, acylic alkanes, –CH$_2$–)	(*76*)
	2859, 2842, 2825	2859, 2842, 2825	symmetric C–H stretch (ν_{C-H}, acylic alkanes, –CH$_2$–)	(*76*)
2648			$\nu_2 + \nu_5$	(*74*, *75*)
	2088, 2048		C≡C stretch ($\nu_{C\equiv C}$, monosubstituted alkynes, –C≡CH)	(*75*, *76*)
1899			ν_2	(*74*, *75*)
	1581, 1562, 1550, 1546, 1531, 1521, 1500	1581, 1558, 1537, 1519	C=C stretch ($\nu_{C=C}$, alkenes or aromatics)	(*76*, *77*)
	1440, 1363, 1334, 1321, 1307, 1290, 1268,	1440, 1360, 1317, 1286, 1268	C=C stretch ($\nu_{C=C}$, aromatics)	(*76*, *77*)
1381			$\nu_4 + \nu_5$	(*74*, *75*)
	1197, 1178, 1170, 1160, 1135, 1132, 1103, 1070, 1043, 1022	1058, 1012, 962	C–C stretch (ν_{C-C}, alkanes)	(*76*)
	989, 960, 912, 894, 881, 854, 833	929, 906, 894, 873, 827, 798, 788, 777	Aromatic or alkenes =C–H out-of-plane deformation	(*76*, *77*)
782, 754, 745			ν_5	(*74*, *75*)

Table S3. Infrared absorption features were recorded before the electron irradiation, after the electron irradiation (82.1 ± 10 eV amu^{-1}), and after the temperature-programmed desorption (TPD) of ^{13}C-acetylene ice ($^{13}C_2H_2$) at 40 K.

Absorption before irradiation (cm^{-1})	Absorption after irradiation (cm^{-1})	Absorption after TPD (cm^{-1})	Assignment	Reference
5101, 4474, 4051, 3845			$5\nu_4 + 3\nu_5$, $\nu_3 + 2\nu_4$, $\nu_1 + \nu_5$, $\nu_2 + 2\nu_4 + \nu_5$	(*74*, *75*)
3399, 3364			$\nu_2 + 2\nu_5$	(*74*, *75*)
	3328, 3293, 3268	3291, 3263	C–H stretch ($\nu_{C–H}$, alkynes, –C≡C–H)	(*74-76*)
3261			ν_1	(*74*, *75*)
3215			ν_3	(*74*, *75*)
	3176, 3149, 3133		C–H stretch ($\nu_{C–H}$, alkenes, –CH=CH$_2$)	(*74-76*)
	3064, 3028,	3066, 3028	C–H stretch ($\nu_{C–H}$, aromatics, =CH–)	(*76*, *77*)
	2998, 2958, 2948, 2912	2995, 2954, 2915	asymmetric C–H stretch ($\nu_{C–H}$, acylic alkanes, –CH$_2$–)	(*74*, *76*)
	2859, 2842, 2823	2854, 2828	symmetric C–H stretch ($\nu_{C–H}$, acylic alkanes, –CH$_2$–)	(*74*, *76*)
2673			$\nu_2 + \nu_5$	(*74*, *75*)
2336			$3\nu_5$	(*74*, *75*)
	1454, 1436, 1363, 1268	1457, 1436, 1398, 1365, 1334, 1315, 1295, 1265,	C=C stretch ($\nu_{C=C}$, aromatics)	(*76*, *77*)
1383			$\nu_4 + \nu_5$	(*74*, *75*)
	1228, 1203, 1186, 1161, 1135, 1106, 1087, 1074, 1058, 1031	1236, 1186, 1157, 1029, 1014	C–C stretch ($\nu_{C–C}$, alkanes)	(*74*, *76*)
	987, 977, 960, 922, 901, 879, 862, 846, 821	987, 962, 935, 921, 910, 897, 887, 873, 848, 821, 794	Aromatic or alkenes =C–H out-of-plane deformation	(*74*, *76*, *77*)
788, 758			ν_5	(*74*, *75*)

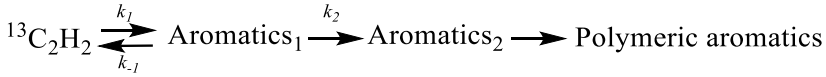
Table S4. Infrared absorption features were recorded before the electron irradiation, after the electron irradiation (82.1 ± 10 eV amu^{-1}), and after the temperature-programmed desorption (TPD) of ^{13}C-methane ice ($^{13}CH_4$) at 10 K.

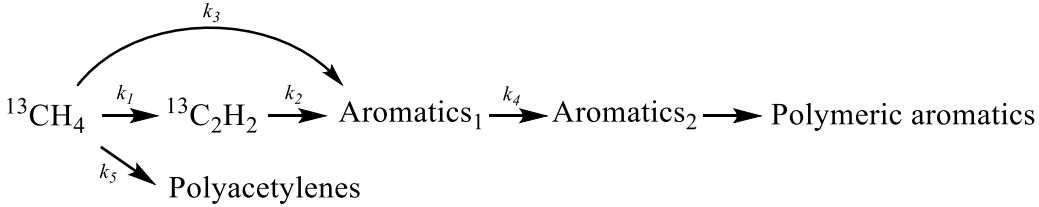
Absorption before irradiation (cm^{-1})	Absorption after irradiation (cm^{-1})	Absorption after TPD (cm^{-1})	Assignment	Reference
5968, 5782, 5540, 4518, 4283, 4192, 4098, 3825			$2\nu_3$, $\nu_1 + \nu_3$, $\nu_3 + 2\nu_4$, $\nu_2 + \nu_3$, $\nu_3 + \nu_4$, $\nu_1 + \nu_4$, $\nu_2 + 2\nu_4$, $3\nu_4$	(*78*, *80*)
	4127		$\nu_7 + \nu_{12}$ (C_2H_6)	(*74*)
	3300		$\nu_3 + \nu_4 + \nu_5$ (C_2H_2)	(*78*, *80*)
	3222		ν_3 (C_2H_2)	(*78*, *80*)
		3409, 3263, 3174, 3149,	C–H stretch ($\nu_{C–H}$, alkynes, –C≡C–H, or alkenes, –CH=CH_2)	(*74-76*)
	3065, 3025	3065, 3016	C–H stretch ($\nu_{C–H}$, aromatics, =CH–)	(*76*, *77*)
2999			ν_3	(*78*, *80*)
	2952, 2921, 2912, 2902	2952, 2919, 2910, 2900	asymmetric C–H stretch ($\nu_{C–H}$, acylic alkanes, –CH_2–)	(*74*, *76*)
	2863, 2840, 2809	2862, 2846, 2834	symmetric C–H stretch ($\nu_{C–H}$, acylic alkanes, –CH_2–)	(*74*, *76*)
2854			ν_1	(*78*, *80*)
2807			$\nu_2 + \nu_4$	(*78*, *80*)
2577			$2\nu_4$	(*78*, *80*)
	1454, 1429, 1363	1484, 1454, 1429, 1363	C=C stretch ($\nu_{C=C}$, aromatics)	(*76*, *77*)
	1230, 1199, 1172, 1145, 1106, 1087, 1020	1334, 1305, 1280, 1267, 1251, 1228, 1193, 1170, 1145, 1108, 1095, 1087, 1058, 1020	C–C stretch ($\nu_{C–C}$, alkanes)	(*74*, *76*)
1291			ν_4	(*78*, *80*)
	989, 966, 949, 903, 879, 823, 775, 748, 723	989, 966, 950, 910, 880, 868, 850, 835, 821, 811, 809, 796, 781, 759, 750	Aromatic or alkenes =C–H out-of-plane deformation	(*76*, *77*)

Table S5. Infrared absorption features were recorded before the electron irradiation, after the electron irradiation (82.1 ± 10 eV amu^{-1}), and after the temperature-programmed desorption (TPD) of ^{13}C-methane ice ($^{13}CH_4$) at 20 K.

Absorption before irradiation (cm^{-1})	Absorption after irradiation (cm^{-1})	Absorption after TPD (cm^{-1})	Assignment	Reference
5971, 5782, 5543, 4520, 4286, 4194, 4100, 3825			$2\nu_3$, $\nu_1 + \nu_3$, $\nu_3 + 2\nu_4$, $\nu_2 + \nu_3$, $\nu_3 + \nu_4$, $\nu_1 + \nu_4$, $\nu_2 + 2\nu_4$, $3\nu_4$	(*78*, *80*)
	4123		$\nu_7 + \nu_{12}$ (C_2H_6)	(*74*)
	3731	3731, 3702	$\nu_{11} + \nu_{10}$ (C_2H_4)	(*78*, *80*)
	3415, 3361, 3299, 3270, 3263, 3222, 3183, 3162, 3143	3407, 3365, 3278, 3274, 3222, 3178, 3151, 3120	C–H stretch (ν_{C-H}, alkynes, –C≡C–H, or alkenes, –CH=CH_2)	(*74-76*)
	3068, 3021	3068, 3023	C–H stretch (ν_{C-H}, aromatics, =CH–)	(*76*, *77*)
2999			ν_3	(*78*, *80*)
	2950, 2927, 2918, 2911, 2902	2994, 2950, 2925, 2915, 2906, 2898	asymmetric C–H stretch (ν_{C-H}, acylic alkanes, –CH_2–)	(*74*, *76*)
	2862, 2844, 2829	2862, 2844, 2829	symmetric C–H stretch (ν_{C-H}, acylic alkanes, –CH_2–)	(*74*, *76*)
2908			ν_1	(*78*, *80*)
2809			$\nu_2 + \nu_4$	(*78*, *80*)
2579			$2\nu_4$	(*78*, *80*)
	1450, 1429, 1363	1479, 1450, 1429, 1402, 1363, 1340	C=C stretch ($\nu_{C=C}$, aromatics)	(*76*, *77*)
	1259, 1230, 1203, 1174, 1147, 1128, 1110, 1088, 1066, 1041,1012	1311, 1286, 1230, 1207, 1155, 1133, 1108, 1083, 1012	C–C stretch (ν_{C-C}, alkanes)	(*74*, *76*)
1291			ν_4	(*78*, *80*)
	985, 956, 916, 903, 876, 862, 847, 818, 796, 777, 740, 719	985, 956, 908, 877, 842, 815, 788, 767, 738, 715, 692	Aromatic or alkenes =C–H out-of-plane deformation	(*76*, *77*)

Table S6. Rate constants calculated via the solution of the coupled differential equations for the complex reaction of irradiated methane and acetylene ices. Errors of the rate constants were derived between 20 and 30 %.

Reaction	$^{13}C_2H_2$ Ice	10 K	40 K
	k_1	0.60	0.19
	k_{-1}	0.25	0.205
	k_2	0.055	0.080

	$^{13}CH_4$ Ice	10 K	20 K
	k_1	0.0012	0.0010
	k_2	0.15	0.10
	k_3	0.13	0.13
	k_4	0.16	0.10
	k_5	0.0033	0.0015

Note. Units of k are amu eV^{-1}.

Table S7. Quantified hydrogen molecules of irradiated methane and acetylene during the irradiation.

Ice	Dose (eV amu^{-1})	Detected hydrogen molecules	Reactants molecules remaining	Theoretical yield hydrogen molecules	Reactants consumed (%) [a]	Detected hydrogen efficiency (%) [b]
10 K Irradiation $^{13}CH_4$	0	0	7.88×10^{17}	0	0	0
	0.7	1.42×10^{13}	7.01×10^{17}	1.30×10^{17}	11.01	0.01
	1.4	3.23×10^{13}	6.37×10^{17}	2.27×10^{17}	19.22	0.01
	2.8	6.31×10^{14}	5.24×10^{17}	3.95×10^{17}	33.46	0.16
	5.7	2.23×10^{15}	3.56×10^{17}	6.48×10^{17}	54.84	0.34
	11.4	5.22×10^{15}	1.64×10^{17}	9.36×10^{17}	79.21	0.56
	17.1	8.43×10^{15}	7.55×10^{16}	1.07×10^{18}	90.42	0.79
	22.8	1.23×10^{16}	3.47×10^{16}	1.13×10^{18}	95.59	1.09
	34.3	1.90×10^{16}	7.36×10^{15}	1.17×10^{18}	99.07	1.62
	45.7	2.72×10^{16}	1.56×10^{15}	1.18×10^{18}	99.80	2.31
	82.2	5.04×10^{16}	1.11×10^{14}	1.18×10^{18}	99.99	4.26
20 K Irradiation $^{13}CH_4$	0	0	7.88×10^{17}	0	0	0
	0.7	1.25×10^{13}	7.16×10^{17}	1.08×10^{17}	9.10	0.01
	1.4	3.68×10^{14}	6.51×10^{17}	2.05×10^{17}	17.38	0.18
	2.8	7.06×10^{14}	5.38×10^{17}	3.75×10^{17}	31.72	0.19
	5.7	2.05×10^{15}	3.68×10^{17}	6.31×10^{17}	53.36	0.33
	11.4	4.94×10^{15}	1.71×10^{17}	9.25×10^{17}	78.25	0.53
	17.1	8.16×10^{15}	7.99×10^{16}	1.06×10^{18}	89.86	0.77
	22.8	1.21×10^{16}	3.72×10^{16}	1.13×10^{18}	95.27	1.07
	34.3	1.92×10^{16}	8.10×10^{15}	1.17×10^{18}	98.97	1.65
	45.7	2.71×10^{16}	1.76×10^{15}	1.18×10^{18}	99.78	2.30
	82.2	5.11×10^{16}	1.35×10^{15}	1.18×10^{18}	99.83	4.33
10 K Irradiation $^{13}C_2H_2$	0	0	5.81×10^{17}	0	0	0
	0.7	4.31×10^{14}	3.64×10^{17}	4.35×10^{16}	37.40	0.99
	1.4	1.18×10^{15}	2.70×10^{17}	6.22×10^{16}	53.54	1.89
	2.8	2.50×10^{15}	1.86×10^{17}	7.89×10^{16}	67.91	3.16
	5.7	4.49×10^{15}	1.43×10^{17}	8.75×10^{16}	75.31	5.13
	11.4	8.00×10^{15}	1.16×10^{17}	9.31×10^{16}	80.12	8.60
	17.1	1.16×10^{16}	9.47×10^{16}	9.73×10^{16}	83.70	11.90
	22.8	1.56×10^{16}	7.77×10^{16}	1.01×10^{17}	86.62	15.47
	34.3	2.29×10^{16}	5.23×10^{16}	1.06×10^{17}	91.00	21.66
	45.7	3.01×10^{16}	3.52×10^{16}	1.09×10^{17}	93.94	27.59
	82.2	5.40×10^{16}	9.95×10^{15}	1.14×10^{17}	98.29	47.26

40 K	0	0	5.81×10^{17}	0	0	0
Irradiation	0.7	1.37×10^{13}	5.09×10^{17}	1.43×10^{16}	12.34	0.10
$^{13}C_2H_2$	1.4	2.71×10^{13}	4.49×10^{17}	2.63×10^{16}	22.64	0.10
	2.8	7.76×10^{14}	3.56×10^{17}	4.50×10^{16}	38.72	1.72
	5.7	2.85×10^{15}	2.39×10^{17}	6.85×10^{16}	58.94	4.16
	11.4	5.79×10^{15}	1.21×10^{17}	9.20×10^{16}	79.15	6.29
	17.1	9.77×10^{15}	6.53×10^{16}	1.03×10^{17}	88.77	9.47
	22.8	1.36×10^{16}	3.56×10^{16}	1.09×10^{17}	93.87	12.48
	34.3	2.10×10^{16}	1.07×10^{15}	1.14×10^{17}	98.17	18.44
	45.7	2.83×10^{16}	3.19×10^{15}	1.16×10^{17}	99.45	24.48
	82.2	5.32×10^{16}	2.82×10^{15}	1.16×10^{17}	99.51	46.05

Note: [a] The number of reactants at 0 eV amu^{-1} minus the number of reactants in the corresponding dose divided by the number of reactants at 0 eV amu^{-1} and then multiplied by 100. [b] The number of detected hydrogen molecules divided by the hydrogen produced from the decay of reactants in equation 3-5 and then multiplied by 100.

Table S8. Identified aromatic compounds detected from the room temperature residues of irradiated ^{13}C-acetylene ($^{13}C_2H_2$) ices by two-dimensional gas chromatography combined with time-of-flight mass spectrometry (GC×GC-TOF-MS) at a dose of 8.2 eV amu^{-1}. At lower doses, only indene, naphthalene, and biphenyl were identified.

	Aromatic Compound	Molecular formula	*RI*[a]	*RI*[b]	R_{t1}, R_{t2} [min:sec; sec]	*m/z*	Quantities[c] (× 10^{-12} mol)				
							0.1 eV amu^{-1}	0.5 eV amu^{-1}	0.8 eV amu^{-1}	4.6 eV amu^{-1}	8.2 eV amu^{-1}
1	2-Methylstyrene	$^{13}C_9H_{10}$	156	-	10:40; 1.88	127					23.7 [d]
2	4-Methylstyrene	$^{13}C_9H_{10}$	159	-	10:45; 1.86	127					
3	*α*-Methylstyrene	$^{13}C_9H_{10}$	151	-	10:20; 1.84	127					6.14
4	*β*-Methylstyrene	$^{13}C_9H_{10}$	166	-	11:35; 1.95	127					63.9
5	Indane	$^{13}C_9H_{10}$	169	-	11:55; 2.02	127					7.48
6	1*H*-Indene	$^{13}C_9H_8$	172	-	12:10; 2.17	125	0.32		0.40	1.12	25.6
7	1-Methylindene	$^{13}C_{10}H_{10}$	192	-	15:25; 2.23	140					7.21
8	2-Methylindene	$^{13}C_{10}H_{10}$	185	-	15:15; 2.21	140					11.5
9	3-Methylindene	$^{13}C_{10}H_{10}$	184	-	15:05; 2.36	140					3.57
10	Methylindane	$^{13}C_{10}H_{12}$	-	-	14:15; 1.99	142					7.18
11	Naphthalene	$^{13}C_{10}H_8$	200	200	16:15; 2.51	138	0.43	0.29	0.86	0.51	46.8
12	1-Methylnaphthalene	$^{13}C_{11}H_{10}$	225	224	19:50; 2.58	153					5.03
13	2-Methylnaphthalene	$^{13}C_{11}H_{10}$	220	221	19:20; 2.47	153					6.53
14	1,2,3,4-tetrahydro-naphthalene	$^{13}C_{10}H_{12}$	195	-	15:40; 2.33	142					4.01
15	9*H*-Fluorene	$^{13}C_{13}H_{10}$	271	270	26:40; 2.87	179					1.62
16	1,2-Dimethylnapthalene	$^{13}C_{12}H_{12}$	247	250	23:35; 2.63	168					0.42
17	1,4-Dimethylnapthalene	$^{13}C_{12}H_{12}$	243	247	23:10; 2.57	168					0.30
18	1-Ethylnaphthalene	$^{13}C_{12}H_{12}$	240	240	22:05; 2.53	168					23.9 [e]
19	2-Ethylnaphthalene	$^{13}C_{12}H_{12}$	239	239	22:00; 2.53	168					
20	Azulene	$^{13}C_{10}H_8$	224	223	19:40; 2.90	138					3.69
21	Biphenyl	$^{13}C_{12}H_{10}$	237	236	21:35; 2.56	166	0.24		1.02		53.1
22	2-Methylbiphenyl	$^{13}C_{13}H_{12}$	240	240	22:10; 2.42	181					0.50
23	3-Methylbiphenyl	$^{13}C_{13}H_{12}$	254	254	24:15; 2.52	181					1.16
24	4-Methylbiphenyl	$^{13}C_{13}H_{12}$	257	259	25:00; 2.81	181					0.66
25	Diphenylmethane	$^{13}C_{13}H_{12}$	242	246	23:00; 2.55	181					0.33
26	Phenalene[c]	$^{13}C_{13}H_{10}$	268	269	26:40; 2.47	179					*q.l.* [f]
27	Phenanthrene	$^{13}C_{14}H_{10}$	300	300	31:00; 3.26	192					1.14
28	Acenaphthylene	$^{13}C_{12}H_8$	247	249	23:25; 2.92	164					0.24
29	Acenaphthene	$^{13}C_{12}H_{10}$	255	258	24:45; 3.15	166					0.48

[a] Lee retention index, NIST Chemistry WebBook, *National Institute of Standards and Technology*; [b] Lee retention index, this study. [c] Absolute quantities based on external calibration curves. [d] Sum of 2-, 4- and potentially 3-methylstyrene. [e] Sum of 1- and 2-ethylnaphthalene. [f] Quantification limit.

Table S9. Prominent positive ion SIMS data from residues of irradiated ^{13}C-acetylene.

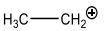
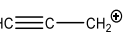
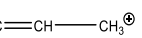
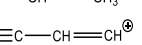
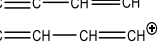
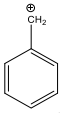
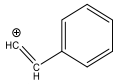
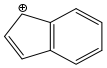
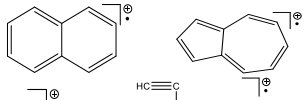
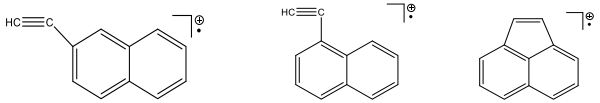
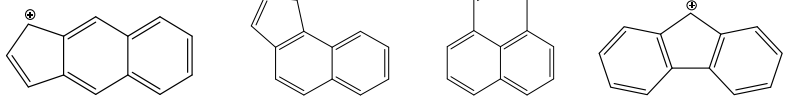
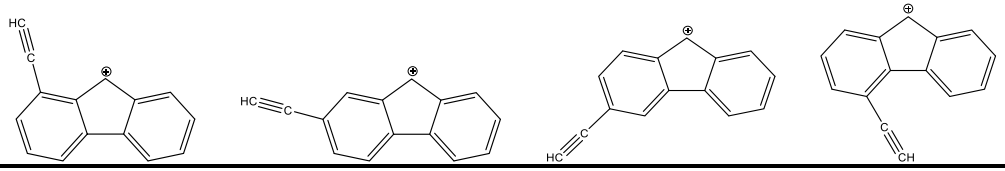
Formula	*m/z*	Representative structures (all carbon atoms are ^{13}C)
$^{13}CH_3$	16.0268	
$^{13}C_2H_3$	29.0302	
$^{13}C_2H_5$	31.0459	
$^{13}C_3H_3$	44.0492	
$^{13}C_3H_5$	44.0492	
$^{13}C_4H_3$	55.0369	
$^{13}C_4H_5$	57.0526	
$^{13}C_5H_5$	70.056	
$^{13}C_6H_5$	83.0593	
$^{13}C_7H_7$	98.0783	
$^{13}C_8H_7$	111.0817	
$^{13}C_9H_7$	124.085	
$^{13}C_{10}H_8$	138.0962	
$^{13}C_{12}H_8$	164.103	
$^{13}C_{13}H_9$	178.1142	
$^{13}C_{15}H_9$	204.1209	

Table S10. Prominent negative ion SIMS data from residues of irradiated ^{13}C-acetylene.

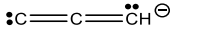
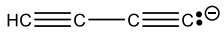
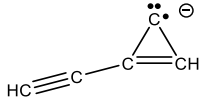
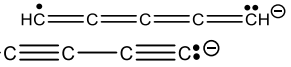
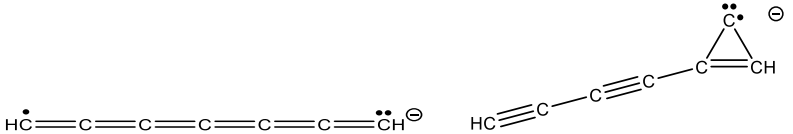
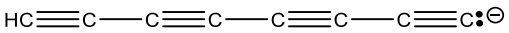
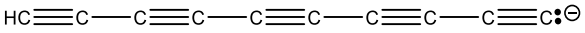
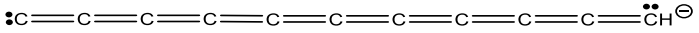
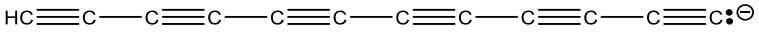
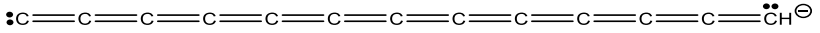
Formula	*m/z*	Representative structures (all carbon atoms are ^{13}C)
$^{13}C_2H$	27.0146	
$^{13}C_3H$	40.0179	
$^{13}C_4H$	53.0213	
$^{13}C_5H_2$	67.0325	
$^{13}C_6H$	79.028	
$^{13}C_7H_2$	93.0392	
$^{13}C_8H$	105.0347	
$^{13}C_{10}H$	131.0414	
$^{13}C_{11}H$	144.0448	
$^{13}C_{12}H$	157.0482	
$^{13}C_{13}H$	170.0728	
$^{13}C_{14}H$	183.0307	

Table S11. The temperature, composition, and distance to Sun of Kuiper belt object whose color can match irradiated methane and acetylene ices.

Name	$B-V$ color	$V-R$ color	Temperature/K	Distance to Sun/AU
275809-2001QY297	0.947	0.479		43.697
469362-2001KB77	0.89	0.457		39.755
136472-Makemake	0.84	0.48	40 K	45.43
15788-1993SB	0.802	0.475		39.294
59358-1999CL158	0.8	0.39		32.894
181855-1998WT31	0.774	0.453		46.264
1998WV24	0.77	0.502		40.656
24952-1997QJ4	0.763	0.431		39.252
86047-1999OY3	0.726	0.345		43.708
2002XV93	0.72	0.375		39.416
120216-2004EW95	0.693	0.375	35 K	39.78
307261-2002MS4	0.69	0.38		41.73
55636-2002TX300	0.679	0.359	<41 K	43.12
19308-1996TO66	0.671	0.389		43.16
90482-Orcus	0.67	0.376	< 44 K	39.174
120347-Salacia	0.664	0.403		44.8
416400-2003UZ117	0.663	0.354		44.05
120178-2003OP32	0.662	0.375	~42 K	43.52
24835-1995SM55	0.652	0.357		41.628