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Supporting Information

Prebiotic Synthesis and Isomerization in Interstellar Analog Ice: Glycinal, Acetamide, and Their Enol Tautomers

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Experimental

The experiments were conducted in a hydrocarbon-free ultra-high vacuum (UHV) chamber evacuated to an ultimate pressure as low as 10^{-11} Torr.[1] The binary ice mixtures (Table S6) were prepared on a mirror polished silver wafer maintained at 5.0 ± 0.2 K by a cold head (Sumitomo Heavy Industries, RDK-415E). The cold head is mounted on a doubly differentially pumped rotating flange and bellows to allow both rotation and translation about or along a central axis. Acetaldehyde (acetaldehyde Sigma Aldrich, anhydrous 99.5 %; acetaldehyde-d_4, Sigma Aldrich, 99 % purity, ≥ 98 atom % D; acetaldehyde-$^{13}C_2$, Sigma Aldrich, 99 atom % ^{13}C) samples were degassed by a minimum of three freeze-pump-thaw cycles and vacuum distillation. Leak valves were used to introduce the gases through separate 10 mm diameter glass capillary arrays at a partial pressure of $2.0 \pm 0.2 \times 10^{-8}$ Torr each. The progress of the deposition was monitored by observing the effects of thin film interference on the power of a helium-neon laser reflected by the silver wafer.[2] After deposition, Fourier transform infrared (FTIR) spectra were used to determine the relative concentration of ammonia and acetaldehyde within the ice. The acetaldehyde concentration was calculated using previously reported values of absorption coefficients (A).[3] Ammonia concentration was determined by calculating the absorption coefficients from the infrared spectra of a pure ammonia ice. The integrated intensities of ν_2, ν_3, and ν_4 observed at 1077, 3394, and 1627 cm^{-1} have absorption coefficients of 2.1×10^{-17}, 2.3×10^{-17}, and 5.6×10^{-18} cm mol^{-1}, respectively. This yields ratios of acetaldehyde to ammonia of 1.6 ± 0.5 : 1. The refractive index of the ice is determined by a weighted average of the refractive indices of acetaldehyde[4] (1.303 at 15 K) and ammonia[5] (1.38 at 13 K); this yields ice thicknesses of 735 ± 50 nm.

Ices that were irradiated were exposed to an electron gun producing 5 keV electrons rastered over a 1.6 cm^2 area (Table S6). This simulates the effects of secondary electrons produced by galactic cosmic rays on interstellar ices.[6] Monte Carlo simulations (CASINO 2.42) of energy transfer by energetic electrons were used to calculate the irradiation dose per molecule and the penetration depth.[7] Rate and duration of electron irradiation was varied throughout the experiment and are detailed in Table S6. Simulations indicate that the average penetration depth of electrons in 350 ± 25 nm into the surface of the ice, less than the thickness of the ices employed for these experiments. At this depth approximately 95 % of the energy imparted by the electrons has already been absorbed. Ices are deliberately made thicker than the average penetration depth in order to

minimize any chemical interactions with the silver substrate. Selected ices were also photolyzed with 205.3 nm light using an Nd:YAG pumped dye laser operating at 615.9 nm from which the third harmonic (205.3 nm) was generated. These ices were exposed to the photolysis laser for 5 hours, either after or in lieu of irradiation with an average intensity of 10 ± 1 mW. The total photon flux was $(1.9 \pm 0.2) \times 10^{20}$ or considering the average of the densities of the ice components (0.76 g cm^{-3} for ammonia[8] and 0.787 g cm^{-3} for acetaldehyde[9]) this yields a total of 170 ± 20 photons per molecule in the ice.

Following irradiation or photolysis, ices were heated at a rate of 1 K min^{-1} from 5 K to 320 K (temperature-programmed desorption; TPD). During TPD, subliming molecules were detected through photoionization reflectron time-of-flight mass spectrometry (PI-ReToF-MS). Photoionization (PI) at 10.49 eV was accomplished by the 9th harmonic of the fundamental (1064 nm) a pulsed 30 Hz neodymium-doped yttrium aluminum garnet (Nd:YAG) laser (Spectra Physics, PRO270-30 & PRO250-30). Four-wave mixing difference frequency generation employing two-photon absorptions ($2\omega_1$) of Kr and Xe was used to produce VUV light with ω_{VUV} energy equal to $2\omega_1 - \omega_2$ for PI energies less than 10.49 eV. A pulsed dye laser (Sirah Lasertechnik, Cobra-Stretch) pumped by a Nd:YAG laser was used to produce ω_1 at 202.316 nm for Kr, and 249.628 & 222.566 nm for Xe. The 2nd or 3rd harmonic of a Nd:YAG laser, the output of a dye laser or its 2nd harmonic were employed as ω_2. These latter two arrangements were utilized while scanning ω_2 to measure photoionization efficiency (PIE) spectra. VUV light was generated in a pulsed jet of noble gas (Kr or Xe) in a differentially pumped vacuum chamber. Harmonics were separated by passing collinear ω_1, ω_2, and ω_{VUV} through an off-center biconvex lithium fluoride lens and directing the spatially separated VUV light through an aperture. The VUV light passed about 2 mm above the surface of the ice during TPD. This method of VUV production was measured to produce $4 \pm 2 \times 10^{11}$ photons s^{-1}. Photoionized molecules were extracted into the ReTOF-MS and detected with a multichannel plate after separation based on their mass-to-charge ratios. Ion counting was accomplished by amplifying the signal 100:1 before further amplification with a 100 MHz discriminator. Ion time-of-flight was recorded with a multichannel scaler which accumulated signals into 3.2 ns bins for 2 minute intervals during TPD.

Computational

All computations were carried out with Gaussian 16, Revision C.01.[10] For geometry optimizations and frequency computations, the density functional theory (DFT) B3LYP[11] functional was employed utilizing the Dunning correlation consistent split valence basis set cc-pVTZ for the ground and radical cation states of each investigated isomer.[12] Based on these geometries, the corresponding frozen-core coupled cluster[13] CCSD(T)/cc-pVDZ, CCSD(T)/cc-pVTZ, and CCSD(T)/cc-pVQZ single point energies were computed and extrapolated to complete basis set limit[14] CCSD(T)/CBS with B3LYP/cc-pVTZ zero-point vibrational energy (ZPVE) corrections. The adiabatic ionization energies were computed by taking the ZPVE corrected energy difference between the neutral and ionic species that correspond to similar conformations. As in general the difference of deuterated and non-deuterated isotopologues in the zero-point vibrational energy is marginal, we used the ZPVEs of non-deuterated isotopologues for IE calculation and assume them as the same for our experiments with heavier isotopologues. Franck-Condon factor calculations were performed with Gaussian 16. To determine the accuracy of these calculations, the experimental adiabatic ionization energies of several molecules were used to benchmark adiabatic ionization energies computed at the same level of theory (Tables S3 and S7). This comparison shows an error of ±0.05 eV. Since the adiabatic ionization energies of isotopologues typically differ by less than 0.02 eV, i.e., less than the error of the calculations employed in this study, these calculations were not repeated for isotopologues.

FTIR Analysis

The infrared spectra of the ices of the acetaldehyde–ammonia system were collected before, during, and after the radiation exposure (Table S1, Figures S1 and S2). Following the irradiation, new absorption features were detected. These can be linked to the CH bending mode of methane (CH_4, ν_4, 1311 cm^{-1}) and to the carbon monoxide (CO) stretch at 2127 cm^{-1} for the non-deuterated ices.[25] Additionally, the IR activity increases substantially in the CH and NH stretching regions covering 2800 – 3200 cm^{-1}.[26] This broad feature can be associated with amines and hydrocarbons formed during irradiation. Amides like **2** – if formed – are expected to exhibit fundamentals of the carbonyl moiety at 1650 – 1680 cm^{-1} and of the NH bending at 1620 – 1650 cm^{-1}.[26] These modes overlap with the degenerate NH bending mode of ammonia and hence could not be clearly

identified. However, the IR spectra expose radical species originating from acetaldehyde. The vinoxy radical (**A**, •CH_2CHO) was observed at 1572 cm^{-1} (acetaldehyde – ammonia) and 1510 cm^{-1} (acetaldehyde-d_4 – ammonia).[26] While the amino radical (**B**, •NH_2) – if generated and sufficiently long-lived – should be visible through its bending mode (ν_2, 1499 cm^{-1}), the isotopic shift of the peak from 1572 cm^{-1} to 1510 cm^{-1} with deuteration of acetaldehyde precludes an unambiguous assignment.[27] The newly emerged absorption at 1840 cm^{-1} with acetaldehyde and at 1847 cm^{-1} in acetaldehyde-d_4 is assigned to the acetyl radical (**C**, •$C(O)CH_3$).[28]

The confirmed presence of radical intermediates **A** and **C** for the formation of **1** and **2** is vital since the successive low-temperature chemistry of both radicals can lead to the formation of more complex molecules such as **1**, **2**, **5**, and **6**. Furthermore, the observation of these radicals after irradiation provides substantive evidence of the reaction scheme shown in Figure 2. These radicals are preserved in the ice because of the effects of these low temperatures on molecular mobility through the ice; radicals that form with no radical nearest neighbors may not be able to react. While the temperatures used in these experiments are slightly lower than may be expected in the average molecular cloud, such cold ices are able to preserve these reactive intermediates intact to provide valuable mechanistic information. With the notable exception of **A** and **C**, FTIR spectra of a complex mixture of organics can only assign *functional groups* of these newly formed molecules. This experimental technique cannot be employed identify individual species due to overlapping absorptions of these moieties (CH, NH, CO), and an alternative analytical technique is required for the firm identification of individual isomers.

Table S1. Infrared absorption features observed in ices containing acetaldehyde and ammonia.

Pristine ice, before irradiation (5 K)		
$CH_3CHO + NH_3$	$CD_3CDO + NH_3$	Assignment
1078	1076	ammonia[15] ν_2
1126	1019	acetaldehyde[2] ν_8
1348	-	acetaldehyde[2] ν_7
1425	1159	acetaldehyde[2] ν_5
1637	1635	ammonia[15] ν_4
-	1693	acetaldehyde[2] $\nu_8+\nu_9$
1716	1709	acetaldehyde[2] ν_4
1770	-	acetaldehyde[2] $2\nu_9$
-	2068	acetaldehyde[2] $2\nu_{12}$
2764	2104	acetaldehyde[2] ν_3
2862	-	acetaldehyde[2] $2\nu_6$
2912	2136	acetaldehyde[2] ν_2
2962	2181	acetaldehyde[2] ν_{11}
3001	2218	acetaldehyde[2] ν_1
-	2255	acetaldehyde[2] $2\nu_5$
3211	3211	ammonia[15] ν_1
3271	3269	ammonia[15] $2\nu_4$
3367	3368	ammonia[15] ν_3
4470	4473	ammonia[15] $\nu_1+\nu_2$
5003	5004	ammonia[15] $\nu_1+\nu_4$
Processed ice, after e^- irradiation (5 K)		
1311	-	methane (CH_4) ν_4
1572	1510	vinoxy[16] (CH_2CHO)
1840	1847	acetyl[16] (CH_3CO)
2127	-	carbon monoxide (CO) stretch
3080 – 2500	3100 – 2600	NH/CH stretch

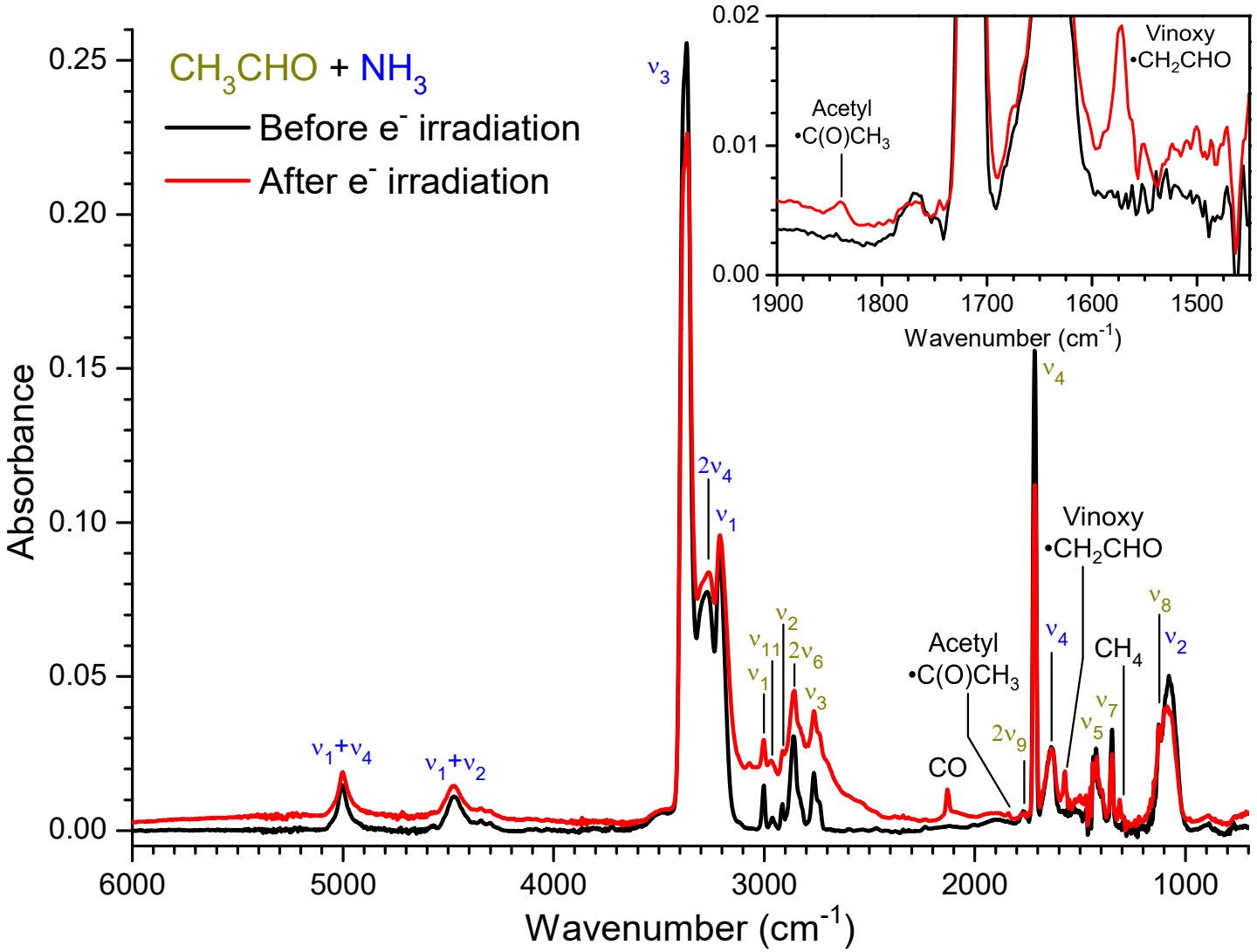


Figure S1. Infrared spectra of ice composed of acetaldehyde and ammonia before and after irradiation. A magnified view of the absorptions attributed to the radical intermediates is inset.

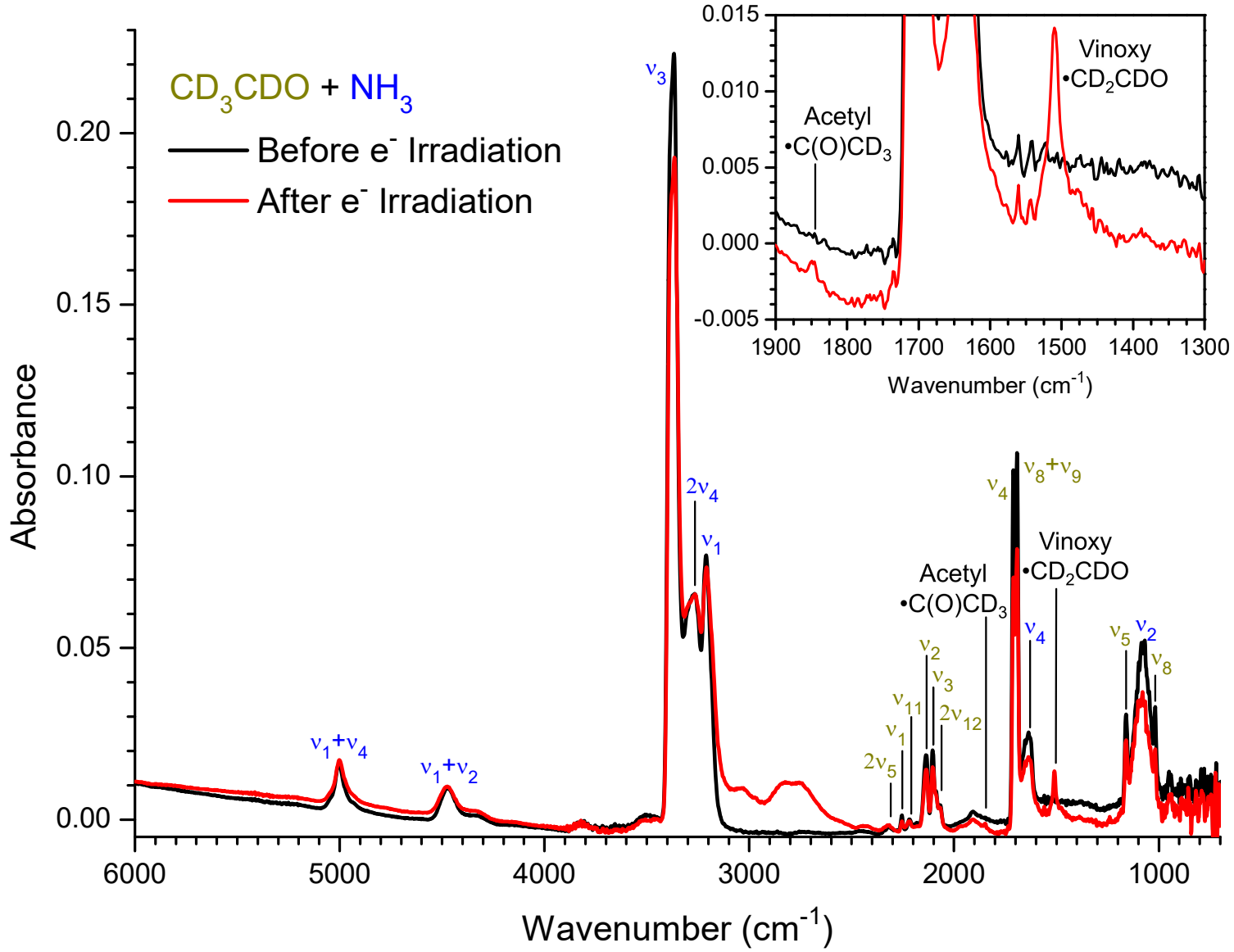


Figure S2. Infrared spectra of ice composed of acetaldehyde-d_4 and ammonia before and after irradiation. A magnified view of the absorptions attributed to the radical intermediates is inset.

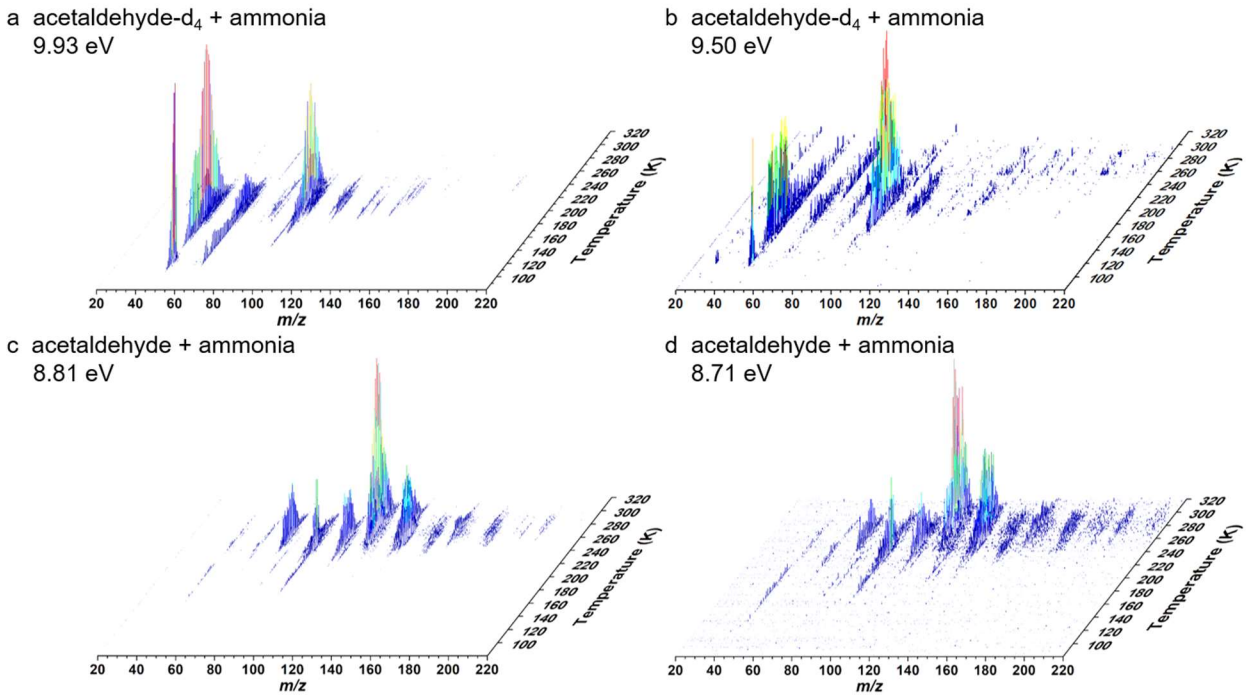


Figure S3. PI-ReToF-MS data as a function of temperature and *m/z* during the temperature program desorption (TPD) of the analog ices. Data were collected for acetaldehyde – ammonia ices with a dose of 0.41 ± 0.06 eV per molecule of acetaldehyde-d_4 and 0.15 ± 0.02 eV per molecule of ammonia at 9.93 eV (a) and 9.50 eV (b) as well as for acetaldehyde-d_4 and ammonia ices exposed to a dose of 3.9 ± 0.5 eV per molecule of acetaldehyde and 1.5 ± 0.2 eV per molecule of ammonia with photoionization at 8.81 eV (c) and 8.71 eV (d).

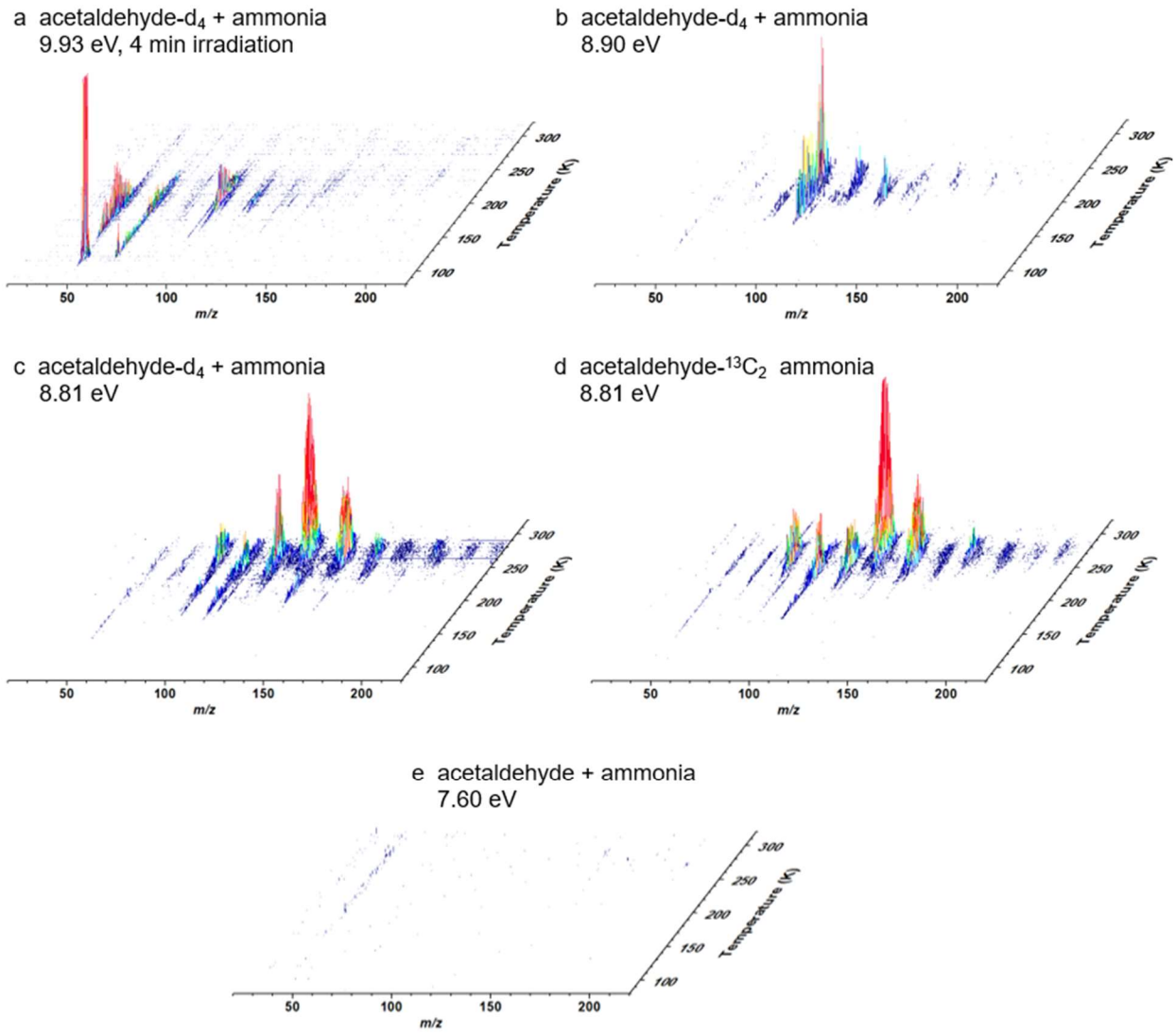


Figure S4. PI-ReToF-MS data as a function of temperature and *m/z* during the temperature program desorption (TPD) of irradiated analog ices. (a) Irradiation with 0.041 ± 0.006 eV per molecule of ammonia and 0.16 ± 0.02 eV per molecule of acetaldehyde-d_4 at 9.93 eV. (b) Irradiation with 0.15 ± 0.02 eV per molecule of ammonia and 0.42 ± 0.06 eV per molecule of acetaldehyde-d_4 at 8.90 eV. (c) Irradiation with 1.6 ± 0.2 eV per molecule of ammonia and 4.6 ± 0.6 eV per molecule of acetaldehyde-d_4 8.81 eV. (d) Irradiation with 1.5 ± 0.2 eV per molecule of ammonia and 3.9 ± 0.6 eV per molecule of acetaldehyde-$^{13}C_2$ at 8.81 eV. (e) Irradiation with 1.5 ± 0.2 eV per molecule of ammonia and 4.0 ± 0.6 eV per molecule of acetaldehyde at 7.60 eV.

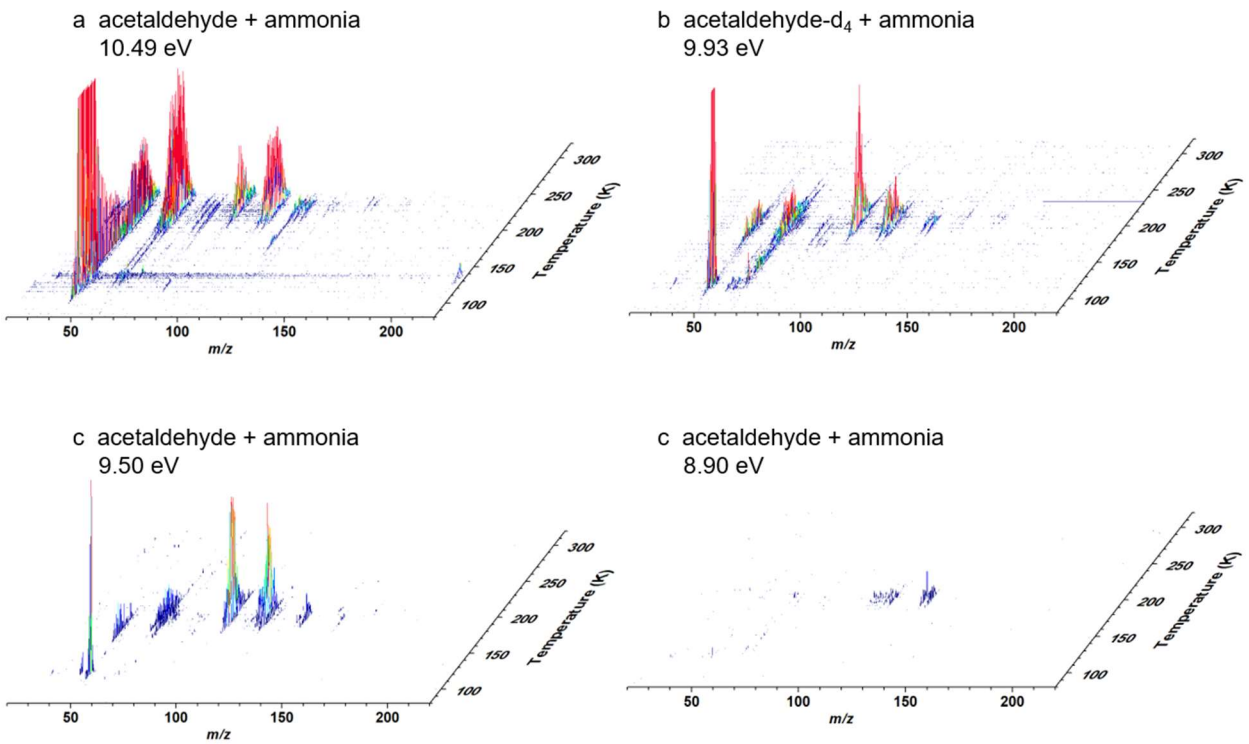
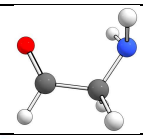
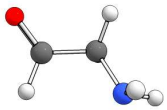
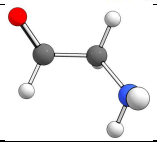
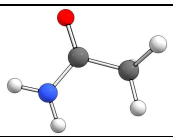
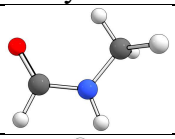
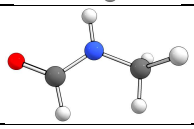
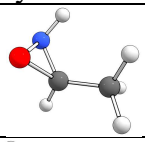
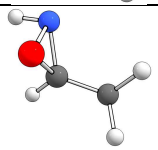
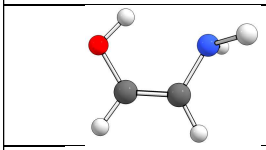
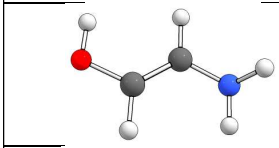
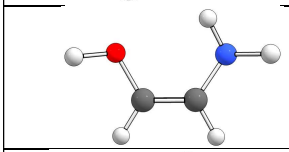
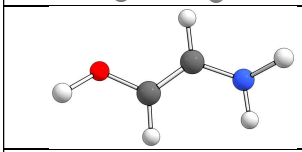
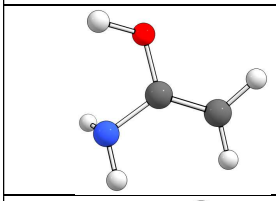
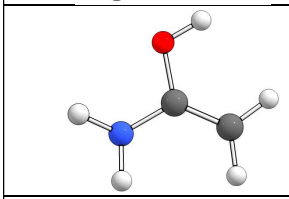
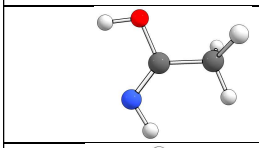
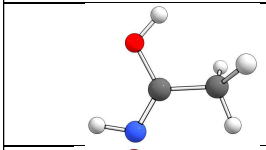
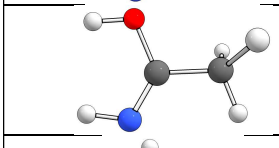
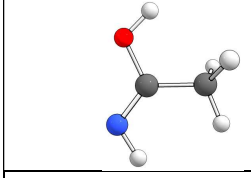
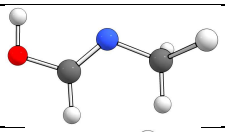
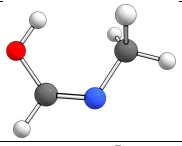
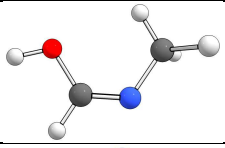
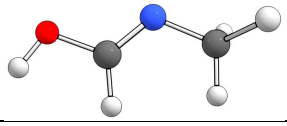
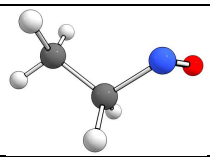
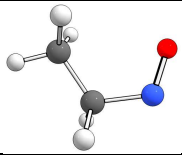
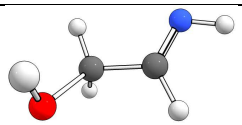
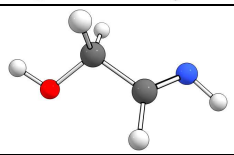
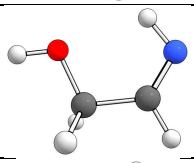
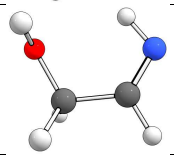
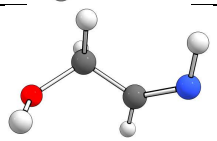


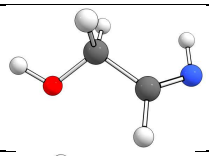
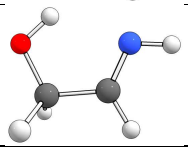
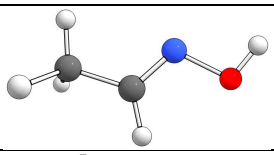
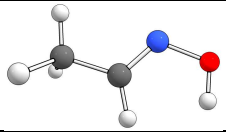
Figure S5. PI-ReToF-MS data as a function of temperature and *m/z* during the temperature program desorption (TPD) of unirradiated analog ices.

Table S2. Properties derived from electronic structure calculations at the CCSD(T)/CBS//B3LYP/cc-pVTZ level of theory, including point group and electronic state symmetries, relative energy, dipole moment, and ionization energy (IE) corrected for a 0.03 eV Stark shift resulting from the mass spectrometer electric field.[17]

		Point group	Electronic state	Rel energy (kJ mol^{-1})	Dipole moment (Debye)	IE (eV, Stark shift corrected)
1 2-aminoacetaldehyde		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 8.97 – 9.14 eV				
	**1a**	C_s	^{1}A'	103.3	1.6239	9.09 ± 0.05
	**1b**	C_1	^{1}A	110.4	3.0640	9.03 ± 0.05
	**1c**	C_1	^{1}A	111.8	2.3152	9.02 ± 0.05
2 acetamide		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.66 – 9.76 eV				
	**2a**	C_1	^{1}A	0.0	3.8180	9.71 ± 0.05
3 N-methylformamide		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.71 – 9.82 eV				
	**3a**	C_s	^{1}A'	47.6	3.9030	9.77 ± 0.05
	**3b**	C_s	^{1}A'	52.9	4.1479	9.76 ± 0.05
4 methyloxaziridine		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.10 – 9.44 eV				
	**4a**	C_1	^{1}A	296.2	2.7712	9.39 ± 0.05
	**4b**	C_1	^{1}A	320.8	2.4252	9.15 ± 0.05

5 2-aminoetheneol		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 7.31 – 7.83 eV				
	**5a**	C_s	^{1}A'	126.8	2.1908	7.78 ± 0.05
	**5b**	C_1	^{1}A	145.4	1.6015	7.48 ± 0.05
	**5c**	C_1	^{1}A	131.8	1.7020	7.40 ± 0.05
	**5d**	C_1	^{1}A	147.2	2.4121	7.36 ± 0.05
6 1-aminoethenol		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 7.79 – 7.94 eV				
	**6a**	C_1	^{1}A	106.0	2.3449	7.89 ± 0.05
	**6b**	C_1	^{1}A	101.1	1.0691	7.84 ± 0.05
7 ethanimidic acid		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.44 – 9.76 eV				
	**7a**	C_s	^{1}A'	46.7	1.5028	9.71 ± 0.05
	**7b**	C_s	^{1}A'	61.1	2.9954	9.61 ± 0.05
	**7c**	C_s	^{1}A'	58.7	1.6461	9.58 ± 0.05
	**7d**	C_s	^{1}A'	72.8	4.1319	9.49 ± 0.05
8 N-methylmethanimidic acid		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.15 – 9.47 eV				

	**8a**	C_s	^{1}A'	95.4	0.7421	9.42 ± 0.05
	**8b**	C_s	^{1}A'	109.5	1.8695	9.28 ± 0.05
	**8c**	C_s	^{1}A'	112.5	2.2560	9.23 ± 0.05
	**8d**	C_s	^{1}A'	115.5	3.1091	9.20 ± 0.05
9 nitrosoethane		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 8.84 – 8.97 eV				
	**9a**	C_1	^{1}A	281.7	2.5222	8.92 ± 0.05
	**9b**	C_s	^{1}A'	279.5	2.4709	8.89 ± 0.05
10 iminoethanol		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.04 – 9.66 eV				
	**10a**	C_1	^{1}A	14.5	1.1419	9.61 ± 0.05
	**10b**	C_1	^{1}A	16.0	1.5743	9.59 ± 0.05
	**10c**	C_s	^{1}A'	11.0	3.7469	9.53 ± 0.05
	**10d**	C_1	^{1}A	11.3	2.4292	9.53 ± 0.05
	**10e**	C_1	^{1}A	19.2	1.6053	9.49 ± 0.05

	**10f**	C_1	^{1}A	20.8	3.3750	9.47 ± 0.05
	**10g**	C_s	^{1}A'	0.0	3.0745	9.09 ± 0.05
11 acetaldoxime		IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.33 – 9.63 eV				
	**11a**	C_s	^{1}A'	216.7	0.7241	9.58 ± 0.05
	**11b**	C_s	^{1}A'	236.0	3.2056	9.38 ± 0.05

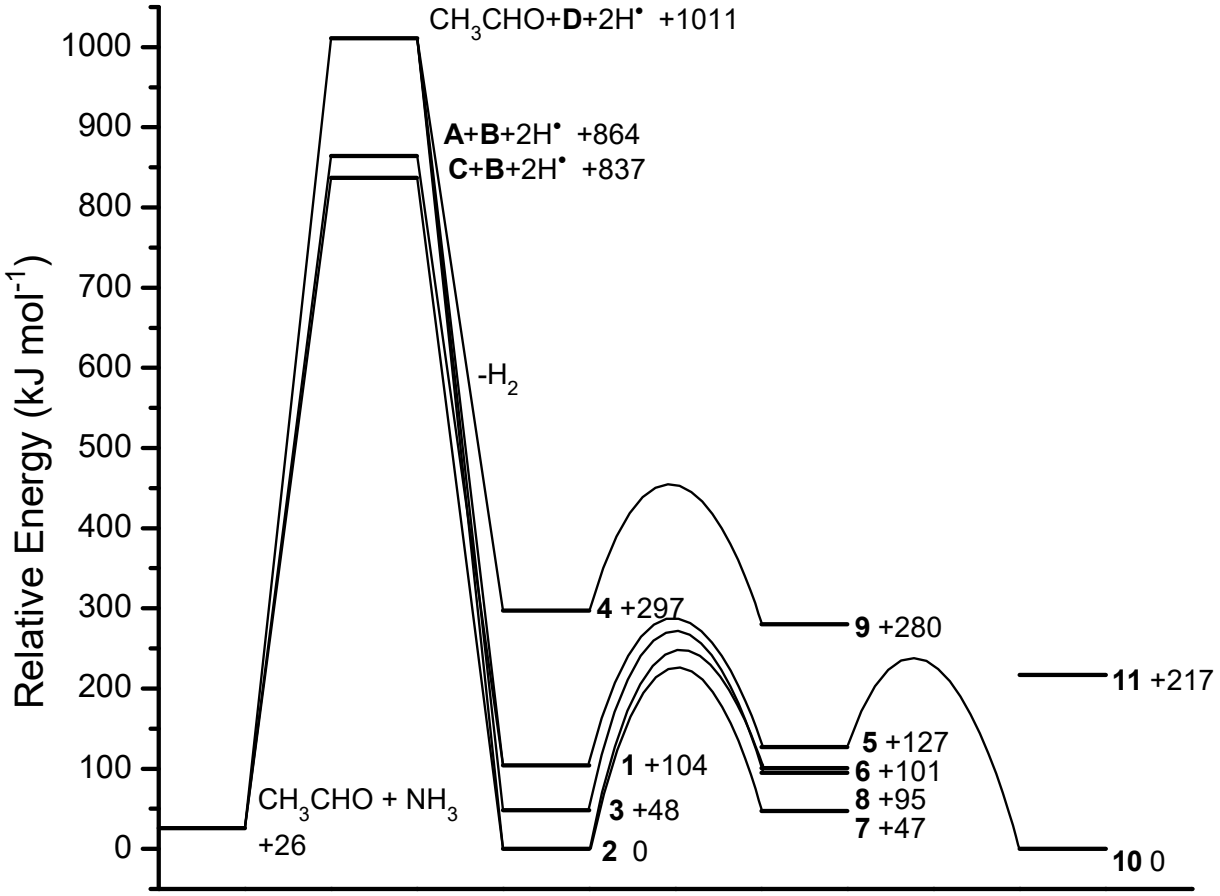


Figure S6. Potential energy surface for radical reactions of acetaldehyde (CH_3CHO) and ammonia (NH_3) via dissociation and recombination of vinoxy (**A**), acetyl (**B**), amino (**C**), and imidogen (**D**). The relative energy of **1** – **11** are those of the lowest energy conformer of each isomer presented in Table S3, and the reported $\Delta_f H°$ of acetamide (**2**) was used to determine the relative energy of the reactants and intermediates.[18] All other values are from the Active Thermochemical Tables.[18] The relative energies of **1** – **11** assume that the H radical produces in the first step recombine and their energies are no longer considered.

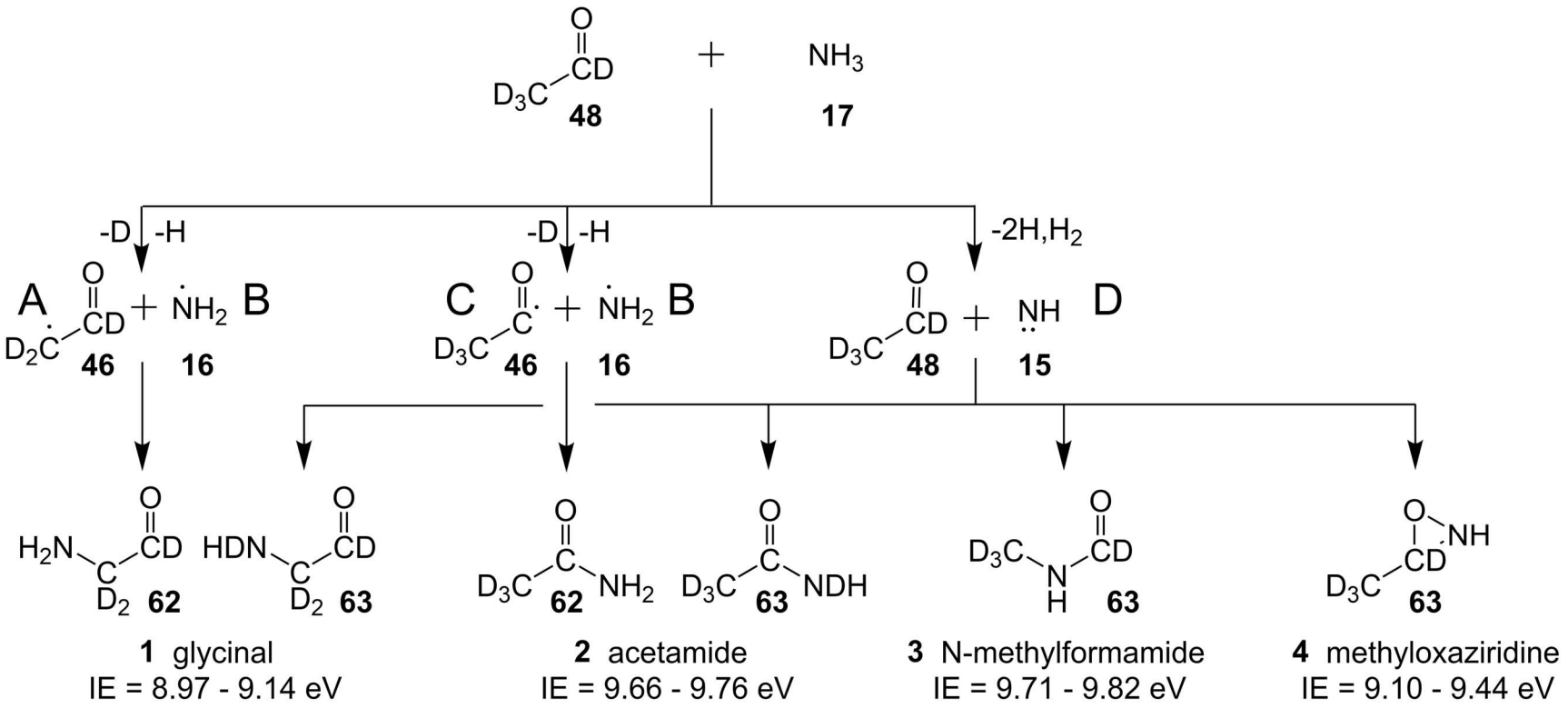


Figure S7. Radical reactions of CD_3CDO and NH_3. Energy from electron irradiation breaks reactant σ-bonds, resulting in radicals **A**, **B**, **C**, and **D**. Recombination **A** and **B** or **C** and **B** produces **1** and **2**, respectively, with *m/z* = 62. Insertion of **D** into acetaldehyde can result in **1**, **2**, **3**, or **4**, with *m/z* = 63.

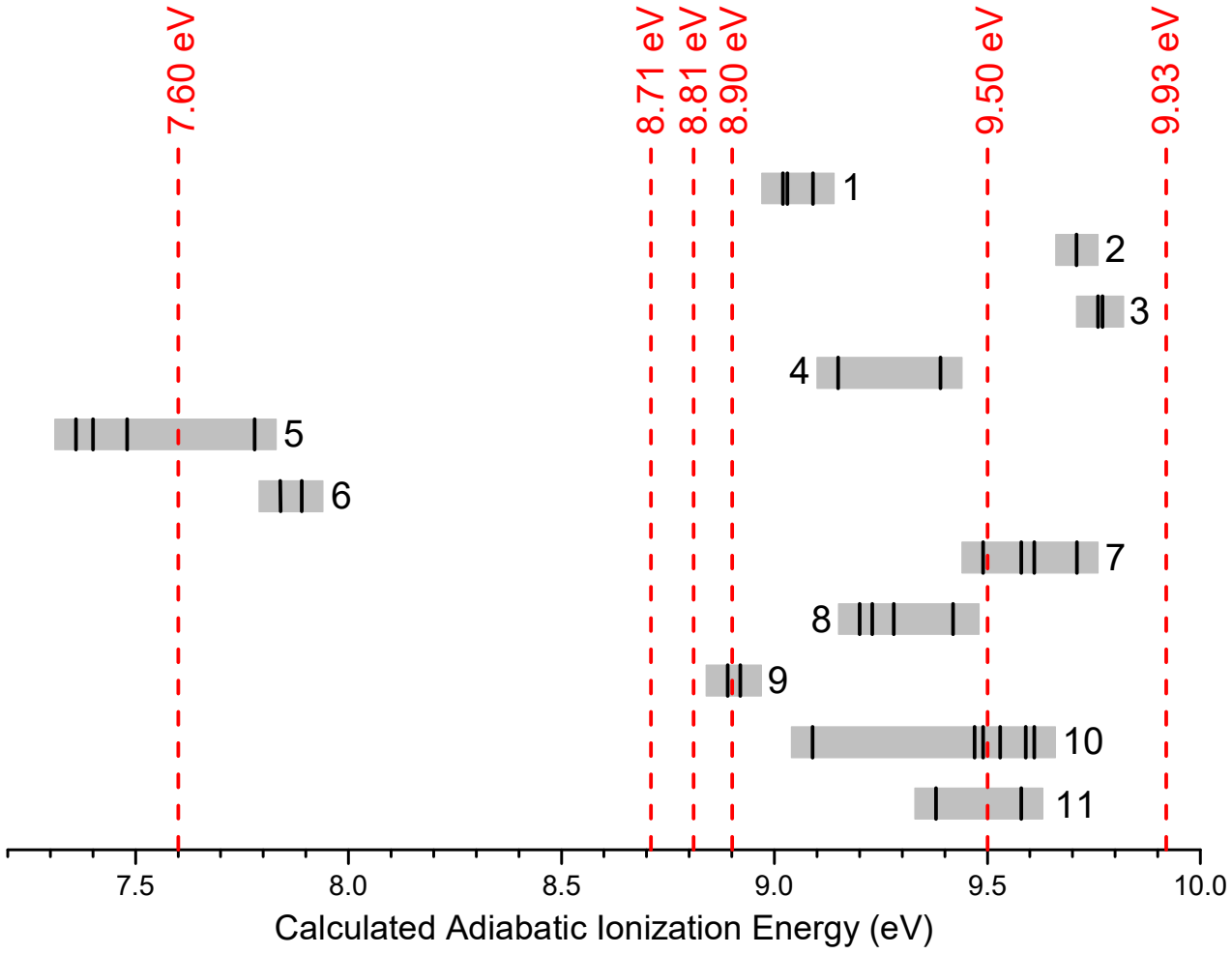


Figure S8. Calculated adiabatic ionization energy of all isomers considered are marked with black lines. The range of possible ionization energies including error bars are shown by gray rectangles. Red lines show the photon energies employed in this experiment.

Table S3. CCSD(T)/CBS//B3LYP/cc-pVTZ calculated electronic energies, zero-point vibrational energy (ZPVE), adiabatic ionization energy (IE), optimized Cartesian geometry (Å), and vibrational frequencies (cm^{-1}) with intensities (km mol^{-1}).

Glycinal (**1a**)

E[B3LYP/cc-pVTZ] = –209.2614427 Ha
E[CCSD(T)/CBS] = –208.9447993 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073239 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.12 eV

C	0.956581	–0.261840	0.000000
C	0.000000	0.912582	0.000000
O	0.615982	–1.416245	0.000000
H	2.033586	0.014707	0.000000
N	–1.416424	0.609271	0.000000
H	–1.645059	0.039098	0.806324
H	–1.645059	0.039098	–0.806324
H	0.252079	1.533855	–0.867852
H	0.252079	1.533855	0.867852

Frequency	Intensity
119.7893	49.8841
238.6040	9.0944
261.2268	22.5526
687.1384	1.2484
732.5360	2.4201
833.0453	75.6873
922.3828	134.0839
1006.1787	0.0127
1144.4589	13.4266
1202.8911	0.0648
1357.2394	10.0385
1381.5329	0.4872
1404.2085	11.2258
1444.9006	14.2239
1675.3578	20.1282
1812.3059	152.5074
2852.1660	145.8166
2999.2492	35.3085
3021.8936	12.8144
3491.8470	1.1112
3559.3701	2.7828

Glycinal (**1a**) radical cation

E[B3LYP/cc-pVTZ] = –208.9396675 Ha
E[CCSD(T)/CBS] = –208.6081722 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071645 Ha

C	0.981847	–0.464851	0.000000
C	0.000000	0.973799	0.000000
O	0.552531	–1.545048	0.000000
H	2.038650	–0.145241	0.000000
N	–1.341963	0.764793	0.000000
H	–1.851559	0.607051	0.859962
H	–1.851559	0.607051	–0.859962
H	0.373437	1.442145	–0.910775
H	0.373437	1.442145	0.910775

Frequency	Intensity
71.9080	14.3608
242.4451	3.0499
389.2891	70.6518
408.8161	1.4157
552.7378	12.9453
657.5094	221.1162
754.1998	13.5007
923.2961	0.7910
1105.9475	1.9895
1111.0320	39.6066
1230.5857	33.4920
1275.9587	2.9183
1352.6204	1.1912
1479.2788	22.9560
1665.8489	45.0234
1945.2438	125.8247
2945.5212	18.6180
3052.4191	10.6994
3135.0189	9.6915
3518.7133	241.2594
3630.3044	129.6709

Glycinal (**1b**)

E[B3LYP/cc-pVTZ] = –209.2586479 Ha
E[CCSD(T)/CBS] = –208.9421084 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073239 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.06 eV

C	0.758675	–0.313504	0.195568
C	–0.495059	0.531368	0.085800
O	1.846117	0.034217	–0.183062
H	0.592158	–1.300673	0.673562
N	–1.676251	–0.312757	–0.057626
H	–1.779171	–0.636804	–1.010941
H	–2.519058	0.192715	0.178145
H	–0.338651	1.281889	–0.696940
H	–0.572159	1.071250	1.035845

Frequency	Intensity
88.5238	9.1113
247.3088	41.5168
346.7216	11.8539
531.8922	4.7831
729.7550	9.4073
817.7325	147.6731
991.7192	8.3787
1061.3554	24.0086
1095.6924	3.4665
1190.5882	0.4676
1305.2299	13.1157
1371.5014	0.1735
1405.6138	8.0071
1468.6391	8.5020
1668.8506	21.5169
1813.0597	168.2549
2887.0118	95.3604
2993.1943	37.6960
3033.6963	16.8651
3510.5377	0.4486
3589.6927	4.1467

Glycinal (**1b**) radical cation

E[B3LYP/cc-pVTZ] = –208.9393122 Ha
E[CCSD(T)/CBS] = –208.6077968 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.072027 Ha

C	0.872619	–0.275902	0.347495
C	–0.625029	0.568119	0.227178
O	1.747683	0.014683	–0.365982
H	0.866352	–1.049309	1.131209
N	–1.608135	–0.265400	–0.231509
H	–1.790310	–0.363698	–1.222020
H	–2.137016	–0.864383	0.388920
H	–0.380804	1.374685	–0.456432
H	–0.768285	0.889739	1.258709

Frequency	Intensity
54.7469	9.4448
292.2303	4.4746
391.5823	1.9457
420.3513	6.3709
544.9914	95.7356
603.5878	135.6387
861.7151	13.6670
916.3032	4.9207
1078.5382	14.2206
1180.1241	8.9784
1230.5329	13.9449
1257.2884	13.4080
1342.5932	1.9397
1487.4163	22.3625
1661.1805	42.2915
1918.8373	125.0174
2980.7666	7.9168
3067.5965	10.0643
3173.0276	8.5579
3519.1355	253.4632
3633.6136	126.7770

Glycinal (**1c**)

E[B3LYP/cc-pVTZ] = –209.2583829 Ha
E[CCSD(T)/CBS] = –208.9414781 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073165 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.05 eV

C	0.746019	–0.289852	0.237418
C	–0.504871	0.564049	0.150008
O	1.815450	–0.000937	–0.231111
H	0.596954	–1.259704	0.765395
N	–1.717514	–0.178166	–0.179596
H	–1.947608	–0.857426	0.535046
H	–1.614871	–0.677189	–1.054660
H	–0.335194	1.351937	–0.584104
H	–0.647164	1.051861	1.119832

Frequency	Intensity
56.3448	7.2989
272.2417	50.1882
348.1934	7.2856
503.4403	7.8951
744.3057	0.3551
815.1215	208.0799
978.8769	1.1481
1047.4363	5.1293
1102.6011	4.4540
1191.9689	4.2448
1333.6071	3.7938
1370.7205	1.0351
1410.8633	2.9106
1461.0775	5.0465
1662.2333	21.9597
1809.4122	181.3577
2828.7725	133.6663
3017.6751	16.7022
3086.3489	13.5053
3495.9647	0.7252
3578.6690	2.2674

Glycinal (**1c**) radical cation

E[B3LYP/cc-pVTZ] = –208.9393122 Ha
E[CCSD(T)/CBS] = –208.6077971 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.072027 Ha

C	0.872586	–0.275389	0.347930
C	–0.624989	0.568426	0.226463
O	1.747569	0.014156	–0.366067
H	0.866352	–1.047650	1.132774
N	–1.608025	–0.265745	–0.231206
H	–2.136972	–0.863798	0.390049
H	–1.790257	–0.365271	–1.221575
H	–0.380703	1.374093	–0.458183
H	–0.768379	0.891371	1.257557

Frequency	Intensity
54.7107	9.4439
292.2695	4.4720
391.6542	1.9446
420.2924	6.3716
545.0248	95.9312
603.4325	135.3920
861.7430	13.6683
916.3184	4.9195
1078.5313	14.2357
1180.1527	8.9729
1230.5633	13.9366
1257.2865	13.4209
1342.5970	1.9376
1487.4052	22.3706
1661.1440	42.2916
1918.8514	125.0083
2980.7674	7.9137
3067.6194	10.0636
3173.0442	8.5586
3519.2235	253.4996
3633.7245	126.7957

Acetamide (**2**)

E[B3LYP/cc-pVTZ] = –209.3028995 Ha
E[CCSD(T)/CBS] = –208.9839054 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073001 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.74 eV

C	–1.360662	–0.338826	–0.000688
C	0.076415	0.147053	–0.007109
H	–1.937746	0.276204	–0.687463
H	–1.773322	–0.193937	0.998500
H	–1.467742	–1.387896	–0.274160
O	0.361731	1.328520	0.001050
N	1.028167	–0.830169	–0.006106
H	1.994425	–0.554702	0.032900
H	0.798851	–1.806004	0.011353

Frequency	Intensity
18.2959	4.5717
156.3451	181.3852
428.2574	4.3981
520.6508	5.5564
547.9523	13.4756
668.2511	9.0330
839.4968	2.5913
984.3811	10.6867
1056.6225	4.0732
1121.3498	0.3925
1342.0129	124.4127
1399.9600	55.5872
1473.1986	8.1542
1491.6559	8.1651
1619.4653	95.8051
1773.7886	339.9108
3044.1223	7.7374
3106.6987	14.4740
3131.0344	9.3467
3591.6496	30.4443
3728.4511	33.7646

Acetamide (**2**) radical cation

E[B3LYP/cc-pVTZ] = –208.9549320 Ha
E[CCSD(T)/CBS] = –208.6258309 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.072788 Ha

C	–1.345509	–0.539984	0.000000
C	0.061716	0.015577	0.000000
H	–2.082077	0.260495	0.000000
H	–1.461509	–1.142832	0.903128
H	–1.461509	–1.142832	–0.903128
O	0.084855	1.278562	0.000000
N	1.138176	–0.718970	0.000000
H	2.062894	–0.305634	0.000000
H	1.067180	–1.727260	0.000000

Frequency	Intensity
81.0758	0.0048
369.2073	1.2840
461.9530	5.6377
537.3497	0.5531
587.5299	156.6481
681.7905	48.4739
822.0601	1.5242
989.6209	6.9144
1012.1437	14.0946
1094.9098	14.4012
1278.8851	4.3312
1400.9900	14.3776
1428.0798	28.2863
1463.8618	18.2698
1619.5540	6.2078
1683.0808	268.7088
3027.9207	42.0931
3112.6118	15.6766
3138.9418	11.0803
3521.9748	192.3210
3636.8173	152.9924

N-Methylformamide (**3a**)

E[B3LYP/cc-pVTZ] = –209.2873181 Ha
E[CCSD(T)/CBS] = –208.9667422 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073956 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.80 eV

C	–0.950218	–0.171678	0.000000
N	–0.000000	0.796434	0.000000
C	1.424463	0.515112	0.000000
H	1.907432	0.927374	0.887482
H	1.907432	0.927374	–0.887482
H	1.545968	–0.564190	0.000000
H	–0.309825	1.752901	0.000000
O	–0.740834	–1.366464	0.000000
H	–1.969805	0.252613	0.000000

Frequency	Intensity
72.7057	0.0914
269.7749	65.3951
296.3625	13.5358
537.4395	39.0362
768.7621	0.6574
951.2977	17.0016
1022.4845	0.0029
1154.3030	0.8708
1161.2413	22.9149
1218.2766	82.5383
1422.1838	5.5746
1443.4089	20.4341
1493.4273	33.4519
1502.5451	6.4222
1555.9502	91.3101
1781.3683	325.4388
2933.9044	115.5488
3027.3600	44.5638
3074.2442	29.2024
3144.1819	0.9914
3631.8350	22.5918

N-Methylformamide (**3a**) radical cation

E[B3LYP/cc-pVTZ] = –208.9314048 Ha
E[CCSD(T)/CBS] = –208.6030861 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.07055 Ha

C	0.916155	0.443173	–0.009377
N	–0.481486	0.633619	–0.020295
C	–1.431922	–0.430141	–0.007915
H	–1.845825	–0.533450	1.011470
H	–0.957122	–1.362838	–0.298141
H	–2.278628	–0.147425	–0.642716
H	–0.821917	1.590363	0.095501
O	1.358014	–0.678170	0.007810
H	1.504385	1.365187	0.017223

Frequency	Intensity
106.6058	38.7743
264.1484	10.3665
304.5696	13.0751
510.3383	275.0540
681.6584	40.9600
732.6295	4.8092
925.2936	160.2987
1003.4493	17.4282
1027.5796	0.4273
1157.7850	11.6686
1336.2646	65.2365
1349.7519	34.8771
1400.5833	4.7951
1424.0324	10.9890
1471.8514	19.3347
1636.5641	87.4624
2912.7723	174.3003
3022.6632	26.7145
3083.3960	1.0863
3158.1984	34.5763
3457.9105	144.3722

N-Methylformamide (**3b**)

E[B3LYP/cc-pVTZ] = –209.2858119 Ha
E[CCSD(T)/CBS] = –208.9646032 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073861 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.79 eV

C	–0.285315	–0.757213	0.000000
N	0.000000	0.568907	0.000000
C	1.333944	1.131725	0.000000
H	1.512281	1.743218	0.886545
H	1.512281	1.743218	–0.886545
H	2.058477	0.318608	0.000000
H	–0.796003	1.188344	0.000000
O	–1.400928	–1.230392	0.000000
H	0.628613	–1.379674	0.000000

Frequency	Intensity
107.8608	0.3123
201.0933	0.5081
343.5659	7.8783
611.4790	13.3651
621.4576	107.1164
1008.7978	42.0387
1039.4173	1.1371
1151.9421	0.3755
1158.3605	34.5307
1301.6936	111.7254
1399.0070	10.2720
1468.6180	8.2830
1483.4393	3.9186
1485.9995	5.4608
1534.7112	15.6350
1791.0719	471.4064
2921.4046	87.7565
3020.7344	53.9073
3066.7880	28.9248
3107.7754	15.0139
3596.1528	23.4634

N-Methylformamide (**3b**) radical cation

E[B3LYP/cc-pVTZ] = –208.9316544 Ha
E[CCSD(T)/CBS] = –208.6020453 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071051 Ha

C	–0.747448	0.411937	–0.081243
N	0.391235	–0.368957	–0.129261
C	1.729703	0.105347	0.073486
H	2.386781	–0.425639	–0.625125
H	2.073867	–0.164517	1.082867
H	1.779725	1.182018	–0.061911
H	0.212927	–1.376746	–0.114342
O	–1.809071	–0.153913	0.102576
H	–0.612908	1.495177	–0.150721

Frequency	Intensity
149.3385	15.4447
185.6238	16.1296
326.7904	6.8349
550.0556	100.0602
750.0948	510.4543
786.0694	100.2642
953.8122	126.6686
998.9936	42.8008
1043.2521	22.2724
1177.2987	12.5104
1335.7554	15.5428
1369.9743	82.9513
1406.7567	36.2127
1439.6358	12.5545
1499.2754	12.2807
1554.0007	74.2672
2960.7428	200.3929
3019.6798	22.5588
3086.2733	3.4677
3154.0135	14.3827
3440.4244	155.2602

Methyloxaziridine (**4a**)

E[B3LYP/cc-pVTZ] = –209.1877753 Ha
E[CCSD(T)/CBS] = –208.8715385 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073451 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.42 eV

C	1.493418	–0.027047	–0.154369
C	0.109585	–0.019657	0.424503
H	1.452156	0.070585	–1.239641
H	2.012796	–0.952217	0.102877
H	2.075337	0.805203	0.244916
O	–0.833916	0.834319	–0.159186
N	–0.996418	–0.654037	–0.234903
H	–0.697376	–0.865368	–1.190436
H	0.054459	–0.077492	1.509801

Frequency	Intensity
219.4732	0.1627
383.3260	3.9731
442.7542	6.8825
734.8159	5.8273
858.1337	18.4385
922.8784	2.3117
1005.2916	17.9306
1073.7182	20.6503
1203.0785	6.5655
1228.1728	18.5719
1287.2238	28.3711
1353.0239	25.9121
1407.3558	6.4722
1445.7670	52.2224
1479.3198	6.3150
1496.9474	5.7339
3028.4735	9.9901
3086.0786	8.6579
3090.3389	14.3607
3109.7502	39.5101
3385.1352	2.0862

Methyloxaziridine (**4a**) radical cation

E[B3LYP/cc-pVTZ] = –208.8498374 Ha
E[CCSD(T)/CBS] = –208.5241017 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.07209 Ha

C	–1.534255	0.019074	–0.151370
C	–0.209975	–0.019695	0.491843
H	–1.474602	0.180130	–1.225692
H	–2.114840	0.824705	0.308986
H	–2.060056	–0.914526	0.058940
O	0.937164	–0.691769	–0.297614
N	0.938946	0.599179	–0.067226
H	1.172062	1.268281	–0.810944
H	–0.097748	–0.224697	1.549072

Frequency	Intensity
176.4614	0.9383
353.1011	19.2759
416.2732	3.9709
518.0997	47.6941
728.4471	134.7734
866.3124	6.5945
984.3562	11.2957
1102.8960	62.7957
1135.0081	12.6727
1148.2590	11.7595
1258.3795	16.6424
1340.7635	22.3365
1408.8727	12.8647
1419.9697	57.6196
1458.6764	22.0685
1486.5825	20.5780
3029.0754	23.8359
3088.8344	7.1884
3138.2441	2.1660
3192.7659	8.6396
3392.6547	191.3280

Methyloxaziridine (**4b**)

E[B3LYP/cc-pVTZ] = –209.1887237 Ha
E[CCSD(T)/CBS] = –208.8621268 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073407 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.18 eV

C	–4.392595	0.084825	0.054216
C	–3.029319	0.666944	–0.158134
H	–4.335774	–1.001633	0.031375
H	–5.075988	0.424448	–0.726256
H	–4.793248	0.402946	1.017911
O	–1.949204	0.061989	0.496773
N	–2.108867	–0.063151	–0.986681
H	–1.420279	0.599738	–1.350005
H	–2.977106	1.754982	–0.189449

Frequency	Intensity
207.2069	0.5092
385.6886	3.3657
438.0335	15.6817
742.9362	2.5550
854.4802	21.3082
922.7565	5.0411
1011.1175	18.7170
1065.0075	8.2626
1177.3300	3.6851
1222.4771	8.9744
1285.9269	51.9325
1344.8247	30.9060
1404.8893	3.8543
1459.9109	30.7211
1483.7370	6.7134
1494.1132	12.4325
3034.8587	10.7789
3069.4368	35.4553
3093.7320	16.1019
3127.8937	14.9272
3395.5518	1.6737

Methyloxaziridine (**4b**) radical cation

E[B3LYP/cc-pVTZ] = –208.8491705 Ha
E[CCSD(T)/CBS] = –208.5234673 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.072075 Ha

C	–4.414475	0.067600	0.049721
C	–3.126289	0.754228	–0.149892
H	–4.318040	–1.014021	–0.008058
H	–5.102266	0.414115	–0.728038
H	–4.833671	0.362083	1.012365
O	–1.853476	0.094841	0.445129
N	–2.070367	0.109968	–0.845353
H	–1.320483	0.310107	–1.518097
H	–3.043313	1.832169	–0.068026

Frequency	Intensity
195.3025	0.4503
360.1657	22.8795
429.7737	10.6479
508.0468	52.9570
691.6701	92.9604
865.2439	18.9393
993.0034	10.8330
1065.6500	63.8640
1122.3195	11.2502
1171.5631	15.1403
1265.6904	18.9311
1332.1583	26.6031
1413.1835	6.8489
1425.5567	49.0619
1460.4148	19.8341
1487.6955	18.2140
3033.1055	17.5177
3100.1560	5.2684
3146.5381	1.6068
3177.1343	6.0961
3392.8695	219.8727

2-Aminoethenol (**5a**)

E[B3LYP/cc-pVTZ] = –209.2541509 Ha
E[CCSD(T)/CBS] = –208.9371598 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0745640 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 7.81 eV

C	0.953495	–0.026032	0.000000
C	0.000000	0.905778	0.000000
N	–1.367432	0.461348	0.000000
H	–1.872401	0.789090	0.814923
H	–1.872401	0.789090	–0.814923
H	0.256142	1.958472	0.000000
O	0.700251	–1.353655	0.000000
H	–0.270874	–1.423397	0.000000
H	2.008579	0.208071	0.000000

Frequency	Intensity
197.5241	16.9121
267.3091	3.6424
481.6734	0.0003
702.6247	141.2280
735.8150	68.0812
752.3165	0.2447
883.0099	74.1121
953.6491	7.3902
1018.0130	22.3723
1121.4915	127.9929
1200.3377	3.3199
1234.5187	87.2459
1372.3784	5.0671
1428.7038	43.7341
1634.3901	13.7295
1730.0837	99.6269
3144.4953	19.6096
3205.2218	16.2898
3486.0020	0.4405
3546.9110	6.6147
3633.4400	64.6162

2-Aminoethenol (**5a**) radical cation

E[B3LYP/cc-pVTZ] = –208.9800925 Ha
E[CCSD(T)/CBS] = –208.6506990 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0751630 Ha

C	0.691657	–0.573948	0.000000
C	–0.707817	–0.581133	0.000000
N	–1.511353	0.468988	0.000000
H	–2.514064	0.339825	0.000000
H	–1.196169	1.428540	0.000000
H	–1.184300	–1.551964	0.000000
O	1.498604	0.454284	0.000000
H	1.086446	1.330605	0.000000
H	1.215826	–1.519757	0.000000

Frequency	Intensity
185.5661	9.0250
275.2693	3.8221
479.3124	23.6900
584.5966	3.4590
624.5294	238.5886
679.7560	9.3593
884.4535	70.8747
976.4410	1.8132
993.6581	31.2664
1146.3454	24.8414
1210.4297	210.2636
1318.3989	20.9900
1420.5959	6.5371
1516.4557	60.0650
1644.7271	43.1176
1707.6823	79.5140
3202.4781	19.8915
3216.9013	12.6812
3539.2842	194.5879
3641.7509	106.6077
3744.1589	109.2747

2-Aminoethenol (**5b**)

E[B3LYP/cc-pVTZ] = –209.2471669 Ha
E[CCSD(T)/CBS] = –208.9291138 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0735740 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 7.51 eV

C	0.563766	0.397203	0.036963
C	–0.509954	–0.389033	0.005190
N	–1.848373	0.050471	–0.106785
H	–1.934714	1.055723	–0.037923
H	–2.456793	–0.376877	0.579635
H	–0.392795	–1.468008	–0.022991
O	1.874858	–0.014500	–0.002258
H	1.905388	–0.973847	–0.079654
H	0.495795	1.476694	0.073579

Frequency	Intensity
231.8818	49.1376
285.7285	68.2909
330.9670	18.3393
358.4473	42.1280
561.1241	0.3650
773.1011	195.6891
819.5668	53.7318
917.4509	84.5845
1046.8877	76.2552
1145.6741	177.5508
1180.2757	25.9701
1307.4008	0.1909
1340.2194	4.0842
1405.7963	46.3244
1649.7184	35.6161
1742.0752	5.4439
3131.5830	21.6192
3177.6450	17.4178
3497.5513	0.8674
3588.0207	3.4275
3804.2231	30.5941

2-Aminoethenol (**5b**) radical cation

E[B3LYP/cc-pVTZ] = –208.9849387 Ha
E[CCSD(T)/CBS] = –208.6547226 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0752850 Ha

C	–0.027102	–0.719984	0.000000
C	0.000000	0.672836	–0.000000
N	1.128408	1.365406	–0.000000
H	2.036951	0.920224	0.000000
H	1.117278	2.375697	–0.000000
H	–0.921606	1.242648	–0.000000
O	–1.113378	–1.445293	0.000000
H	–1.941767	–0.938242	–0.000000
H	0.879926	–1.312942	0.000000

Frequency	Intensity
241.9473	4.3632
318.6855	5.9552
503.6608	7.7977
564.5151	2.0896
609.0600	196.7026
633.7366	59.2895
921.6236	18.7626
971.5876	72.8909
1018.3564	19.6183
1162.7933	163.4205
1321.4180	10.9790
1349.6555	76.1062
1383.1749	17.1799
1447.7034	85.4109
1647.2164	126.6141
1702.1935	18.7929
3180.2651	3.2286
3185.8028	19.9213
3531.4574	221.1363
3642.2723	137.2709
3709.1768	171.0368

2-Aminoethenol (**5c**)

E[B3LYP/cc-pVTZ] = –209.2517350 Ha
E[CCSD(T)/CBS] = –208.9344229 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0737300 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 7.43 eV

C	0.685936	0.610849	0.010197
C	–0.645222	0.632414	0.004078
N	–1.460731	–0.500194	–0.086018
H	–0.968365	–1.356549	0.124335
H	–2.316715	–0.433532	0.442932
H	–1.152697	1.586189	–0.012588
O	1.361990	–0.598313	–0.042034
H	2.246077	–0.483174	0.313352
H	1.276617	1.515352	–0.015283

Frequency	Intensity
173.4320	129.7927
242.4395	10.8141
322.1858	37.4973
519.6571	44.7980
677.1453	220.0884
713.2852	15.7752
739.1950	95.9604
878.0498	14.2553
1014.2956	11.1724
1091.6135	98.4596
1174.9015	22.3425
1272.8209	112.9690
1313.8137	10.7192
1433.1185	24.7299
1646.9274	36.3402
1758.8018	39.6115
3179.7296	1.4893
3202.8487	18.9784
3531.9601	5.4362
3629.5359	11.9865
3848.0581	69.1193

2-Aminoethenol (**5c**) radical cation

E[B3LYP/cc-pVTZ] = –208.9923940 Ha
E[CCSD(T)/CBS] = –208.6634709 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0756540 Ha

C	–0.921690	–0.136177	0.000000
C	0.000000	0.914122	0.000000
N	1.302804	0.720047	0.000000
H	1.697484	–0.212245	0.000000
H	1.947183	1.498146	0.000000
H	–0.364738	1.931306	0.000000
O	–0.464770	–1.367722	0.000000
H	–1.164723	–2.037915	0.000000
H	–1.986529	0.054484	0.000000

Frequency	Intensity
263.5358	6.4527
333.8765	29.2076
522.1739	152.7515
618.6033	5.9533
680.7572	154.0672
700.0918	12.9131
833.5973	10.2949
984.4165	15.1578
1022.2203	9.6778
1141.3337	67.3908
1236.0995	36.6509
1309.5284	144.6278
1376.8575	31.9826
1515.6626	30.5889
1643.1185	48.5021
1691.9135	65.1948
3200.1664	18.2276
3220.6570	10.1350
3526.5925	186.0205
3641.5819	151.3515
3745.5053	322.0208

2-Aminoethenol (**5d**)

E[B3LYP/cc-pVTZ] = –209.2463307 Ha
E[CCSD(T)/CBS] = –208.9281997 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0733550 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 7.39 eV

C	0.563898	0.380229	0.028531
C	–0.524590	–0.384087	0.028423
N	–1.847156	0.070935	–0.103335
H	–1.946549	1.069403	0.016863
H	–2.495773	–0.412884	0.501055
H	–0.409100	–1.460941	0.034090
O	1.824445	–0.181190	–0.081880
H	2.436775	0.314992	0.468112
H	0.513330	1.465556	0.016533

Frequency	Intensity
219.1413	150.8265
257.7881	4.4444
313.7460	21.2114
361.4286	33.2527
563.9969	10.4695
732.1508	250.1946
803.7708	46.3369
932.4157	52.3040
1045.9986	5.6911
1152.4424	246.7430
1184.9360	29.6682
1271.3619	13.1098
1336.1494	11.4391
1386.2206	3.3038
1650.4800	32.3067
1756.8777	3.4369
3117.2143	34.8195
3168.7576	9.8754
3516.2091	2.8289
3608.3267	4.6488
3819.6427	55.2603

2-Aminoethenol (**5d**) radical cation

E[B3LYP/cc-pVTZ] = –208.9889286 Ha
E[CCSD(T)/CBS] = –208.6586899 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0752820 Ha

C	–0.022802	–0.717428	0.000000
C	–0.000000	0.675437	–0.000000
N	1.114010	1.384535	–0.000000
H	2.031502	0.958907	0.000000
H	1.082281	2.394339	–0.000000
H	–0.939357	1.213835	–0.000000
O	–1.196735	–1.302225	0.000000
H	–1.141490	–2.269923	0.000000
H	0.879685	–1.319156	0.000000

Frequency	Intensity
247.7296	0.2411
317.3452	3.6407
505.6672	131.5913
567.5872	9.0257
600.6669	2.5646
641.8034	190.4978
873.7695	5.0301
992.1575	28.5147
1019.8782	5.3676
1170.6027	67.2054
1311.0596	97.1140
1319.5006	177.5917
1364.7998	16.4623
1463.5174	32.8812
1656.2006	77.5502
1709.4961	29.9459
3165.2626	10.0660
3196.1027	16.2293
3533.6668	222.8053
3643.5517	137.3070
3744.7046	332.2238

1-Aminoethenol (**6a**)

E[B3LYP/cc-pVTZ] = –209.2616865 Ha
E[CCSD(T)/CBS] = –208.9443292 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073372 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 7.87 eV

C	–1.401357	–0.278955	0.007953
C	–0.098395	–0.000775	0.003975
H	–2.125483	0.514736	–0.076083
H	–1.748838	–1.296123	0.097711
N	0.949349	–0.926245	–0.081124
H	1.626337	–0.829223	0.665717
H	0.630093	–1.881158	–0.147579
O	0.347844	1.290601	0.047682
H	1.188203	1.329049	–0.424922

Frequency	Intensity
321.9863	22.9032
450.3706	106.9818
457.1074	21.0911
502.8328	26.2074
584.1901	72.3927
681.2500	42.2394
782.7156	76.9428
799.4673	146.2743
920.9180	28.0023
981.6332	22.8814
1150.2443	18.4520
1242.3473	61.4693
1366.9010	182.9068
1439.9986	3.2058
1635.4169	45.7170
1747.1100	216.0762
3173.0714	1.1172
3263.7755	5.3840
3506.2584	3.6580
3617.0617	10.1553
3774.1800	37.6909

1-Aminoethenol (**6a**) radical cation

E[B3LYP/cc-pVTZ] = –208.9792075 Ha
E[CCSD(T)/CBS] = –208.6532486 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073715 Ha

C	–0.961943	1.065141	–0.000000
C	0.000000	0.020208	–0.000000
H	–0.618525	2.088087	–0.000000
H	–2.018364	0.844250	–0.000000
N	–0.358019	–1.250656	0.000000
H	0.310609	–2.009012	0.000000
H	–1.335135	–1.506938	–0.000000
O	1.254077	0.393716	0.000000
H	1.906587	–0.323623	0.000000

Frequency	Intensity
376.1031	9.6486
437.3000	3.8296
448.8563	39.5494
532.5870	8.3306
554.1772	127.4502
592.8911	148.8289
662.3174	6.1869
824.1059	44.0692
973.0005	5.1519
1003.9459	14.9748
1074.1175	15.5427
1184.7131	157.5118
1456.6178	3.3692
1561.6060	119.1701
1564.8598	75.5528
1689.4262	213.9301
3171.7828	25.7441
3291.4232	13.5637
3537.7317	193.5141
3647.1797	119.4676
3733.7465	194.1789

1-Aminoethenol (**6b**)

E[B3LYP/cc-pVTZ] = –209.2635552 Ha
E[CCSD(T)/CBS] = –208.9457670 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.07381 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 7.92 eV

C	1.166433	–0.795088	0.016690
C	0.057505	–0.045105	–0.006282
H	2.144987	–0.341325	–0.022565
H	1.107742	–1.871606	0.037986
N	–1.244581	–0.506960	–0.086321
H	–1.916429	0.089524	0.372020
H	–1.358389	–1.478355	0.150575
O	0.053543	1.320435	–0.007360
H	0.962176	1.628159	0.062666

Frequency	Intensity
214.4576	39.8297
407.3125	65.0777
461.9952	20.5174
513.1642	14.6540
574.7872	221.0572
673.6356	33.6426
674.4076	0.7599
719.5864	148.4191
933.2830	30.6268
982.6938	5.0718
1119.3755	71.6457
1221.8607	110.3804
1421.1122	56.6422
1459.0693	5.3705
1637.6221	55.4196
1720.1038	353.1840
3164.6928	0.4609
3249.0076	10.1801
3559.4908	17.9777
3667.0595	19.0626
3831.7402	50.4556

1-Aminoethenol (**6b**) radical cation

E[B3LYP/cc-pVTZ] = –208.9826529 Ha
E[CCSD(T)/CBS] = –208.6569127 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073627 Ha

C	–1.046725	0.972779	–0.000000
C	0.000000	0.011646	–0.000000
H	–0.820487	2.028984	–0.000000
H	–2.079847	0.658908	–0.000000
N	–0.248010	–1.280551	0.000000
H	0.515434	–1.946013	0.000000
H	–1.192441	–1.635649	–0.000000
O	1.270387	0.332919	0.000000
H	1.430672	1.287718	0.000000

Frequency	Intensity
336.7003	1.6990
436.9507	0.1632
463.7960	58.9246
524.9559	11.2107
571.1887	1.7913
619.8999	261.5779
699.0794	0.2461
804.3888	52.2371
962.0423	10.4998
1005.9521	1.7738
1085.0280	30.0810
1202.9680	130.7509
1466.7407	34.3156
1527.9322	192.8146
1568.8682	3.8658
1691.7456	187.3317
3163.7309	21.4569
3276.8145	8.2084
3531.7278	231.3963
3656.3618	147.9559
3760.4458	218.5253

Ethanimidic acid (**7a**)

E[B3LYP/cc-pVTZ] = –209.2835655 Ha
E[CCSD(T)/CBS] = –208.9671490 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073628 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.74 eV

C	0.822620	–1.079168	0.000000
C	0.010645	0.182124	0.000000
H	0.578359	–1.678408	0.877863
H	1.887013	–0.859210	0.000000
H	0.578359	–1.678408	–0.877863
O	–1.317265	–0.089385	0.000000
H	–1.769083	0.766794	0.000000
N	0.399473	1.386194	0.000000
H	1.411718	1.452016	0.000000

Frequency	Intensity
131.8985	0.5804
426.1319	1.8867
525.6219	24.0163
552.3277	37.7189
631.8706	112.3456
844.4184	25.0694
868.6848	1.5728
1016.5672	54.0816
1072.6347	6.9955
1102.7064	172.0937
1261.5124	89.8035
1391.0875	3.0959
1437.8905	60.1693
1474.6847	8.0941
1486.3564	24.8171
1737.7333	229.8404
3043.9406	6.2963
3099.0324	6.8700
3134.8230	12.3927
3520.3860	3.6156
3742.4916	47.1683

Ethanimidic acid (**7a**) radical cation

E[B3LYP/cc-pVTZ] = –208.9335423 Ha
E[CCSD(T)/CBS] = –208.6060918 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.07082 Ha

C	0.565963	–1.321135	0.064180
C	0.024703	0.060233	–0.025904
H	0.530314	–1.748027	–0.943711
H	–0.090995	–1.905349	0.711781
H	1.584619	–1.343388	0.441757
O	–1.210480	0.188931	–0.382846
H	–1.527390	1.110842	–0.414497
N	0.739928	1.078788	0.367869
H	1.645446	1.377738	0.003310

Frequency	Intensity
98.6723	2.5407
361.4959	26.6309
446.6288	17.6037
524.3190	85.0929
542.8967	74.4166
636.6383	162.0555
716.8630	193.6729
879.4987	9.0420
1013.9701	6.8472
1030.7197	21.3414
1197.4807	140.0877
1345.9921	61.5940
1408.1869	6.1760
1436.0103	44.9732
1459.6541	32.0142
1569.6028	141.6833
3021.5095	49.9165
3093.7077	15.8551
3150.3957	5.1326
3491.2482	238.8769
3661.0041	221.1777

Ethanimidic acid (**7b**)

E[B3LYP/cc-pVTZ] = –209.2777323 Ha
E[CCSD(T)/CBS] = –208.9611886 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073551 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.64 eV

C	0.827433	–1.066502	0.000000
C	–0.038639	0.158795	0.000000
H	0.627317	–1.678675	0.882683
H	1.872587	–0.774846	0.000000
H	0.627317	–1.678675	–0.882683
O	–1.391513	–0.072745	0.000000
H	–1.569452	–1.019073	0.000000
N	0.436476	1.325660	0.000000
H	–0.310770	2.019318	0.000000

Frequency	Intensity
166.5593	0.1377
374.7953	107.4787
427.1966	3.0152
542.2862	26.6042
543.8252	11.6108
853.4617	40.9150
915.9725	64.4969
1006.9511	5.7495
1077.5054	0.7505
1116.1044	44.2335
1229.3305	50.2561
1354.6473	266.0067
1415.3125	69.7370
1478.4207	1.9478
1488.5359	7.8669
1772.4226	165.4199
3026.1102	13.1401
3074.0910	13.5456
3152.7563	3.8665
3463.4146	2.7936
3805.5744	40.1602

Ethanimidic acid (**7b**) radical cation

E[B3LYP/cc-pVTZ] = –208.9311552 Ha
E[CCSD(T)/CBS] = –208.6044307 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.070913 Ha

C	0.806076	–1.005996	–0.090346
C	–0.191437	0.097746	0.045379
H	1.774424	–0.644231	–0.426178
H	0.414751	–1.726450	–0.815330
H	0.922591	–1.496346	0.879324
O	–1.393798	–0.093489	0.474822
H	–1.576824	–1.017176	0.717269
N	0.188538	1.326161	–0.161816
H	0.552820	1.754144	–1.012403

Frequency	Intensity
119.3891	0.8318
329.7291	67.7485
423.3871	45.3936
528.4259	20.9876
547.3862	32.3161
597.5348	258.0701
687.0005	147.4197
876.2969	8.1106
1017.1329	1.6633
1036.2138	21.3997
1192.7601	106.8517
1356.4911	116.8485
1403.7439	87.0783
1441.2551	56.1732
1464.5776	18.9138
1633.7161	96.3617
3021.6677	32.1205
3091.2995	11.4302
3144.7837	4.8045
3510.2882	274.6233
3703.9147	224.8669

Ethanimidic acid (**7c**)

E[B3LYP/cc-pVTZ] = –209.2789050 Ha
E[CCSD(T)/CBS] = –208.9621928 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073628 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.61 eV

C	0.811208	–1.052907	0.000000
C	–0.029866	0.184755	0.000000
H	0.584637	–1.658393	0.878467
H	1.861641	–0.781932	0.000000
H	0.584637	–1.658393	–0.878467
O	–1.360139	–0.136906	0.000000
H	–1.883870	0.673494	0.000000
N	0.468757	1.346510	0.000000
H	–0.238924	2.081329	0.000000

Frequency	Intensity
146.1983	0.7905
426.9812	7.2979
473.9207	83.9950
551.9831	0.0346
559.1818	36.1098
863.8543	16.9546
866.8340	94.9533
1012.5382	55.2693
1078.8382	0.1588
1113.7390	48.2435
1194.1675	195.3705
1374.4536	45.5252
1426.8118	52.6590
1477.8655	8.9037
1480.9603	8.4312
1746.8922	216.1421
3046.0727	7.9051
3096.9437	9.3206
3159.0742	5.3733
3441.2413	7.9394
3780.2891	28.2926

Ethanimidic acid (**7c**) radical cation

E[B3LYP/cc-pVTZ] = –208.9335417 Ha
E[CCSD(T)/CBS] = –208.6060834 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.070814 Ha

C	0.780934	–1.061680	0.022992
C	–0.140303	0.101383	–0.062083
H	0.791668	–1.561337	–0.948074
H	0.362460	–1.751822	0.763262
H	1.788386	–0.773272	0.309921
O	–1.342754	–0.126206	–0.478228
H	–1.907724	0.666838	–0.532980
N	0.299537	1.311067	0.151931
H	0.722660	1.675311	1.006571

Frequency	Intensity
100.3687	2.5526
360.3918	27.6263
445.6362	17.8076
523.4425	84.0017
542.7230	75.0428
635.1383	161.4703
715.5109	193.6233
879.8100	8.8550
1014.2185	7.0917
1030.6572	21.3620
1197.2885	141.0169
1346.0834	61.1491
1408.0062	5.5404
1435.7821	45.6264
1459.3480	32.1743
1569.9439	140.5283
3021.1306	49.9299
3092.2555	15.8988
3151.2107	5.0696
3491.9558	239.9524
3662.6911	221.1909

Ethanimidic acid (**7d**)

E[B3LYP/cc-pVTZ] = –209.2729806 Ha
E[CCSD(T)/CBS] = –208.9568304 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073642 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.52 eV

C	–1.374637	–0.470766	0.000000
C	0.013033	0.120690	0.000000
H	–1.522214	–1.098308	–0.882146
H	–2.133481	0.307491	0.000000
H	–1.522214	–1.098308	0.882146
O	1.021759	–0.797363	0.000000
H	0.657651	–1.687347	0.000000
N	0.340436	1.338555	0.000000
H	–0.483898	1.930248	0.000000

Frequency	Intensity
153.4273	0.3298
363.0856	79.6033
436.3481	5.2151
557.9520	1.0749
566.9704	1.3399
855.0753	60.8961
866.2679	17.0118
1013.7408	10.1180
1069.5554	7.1423
1094.6411	185.4626
1251.4142	102.8888
1376.6725	118.6553
1413.7252	75.8441
1483.1541	7.5768
1487.5787	9.2552
1758.0509	171.3113
3023.9470	11.3212
3075.4317	11.1969
3127.8075	11.4830
3518.3988	4.0777
3831.7894	52.4889

Ethanimidic acid (**7d**) radical cation

E[B3LYP/cc-pVTZ] = –208.9311552 Ha
E[CCSD(T)/CBS] = –208.6044302 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.070912 Ha

C	0.554785	–1.301805	–0.125553
C	0.005893	0.077380	0.041971
H	1.585955	–1.293718	–0.469030
H	–0.060729	–1.839867	–0.850810
H	0.497192	–1.807387	0.843381
O	–1.214181	0.307755	0.395434
H	–1.740549	–0.500002	0.521817
N	0.741700	1.098620	–0.292576
H	1.635953	1.403486	0.090432

Frequency	Intensity
119.5784	0.8317
329.5666	67.9737
423.3303	45.3657
528.3305	21.3669
547.3881	32.4149
597.4794	258.4505
686.5008	146.3943
876.4462	8.1167
1017.1169	1.6785
1036.2098	21.3220
1192.8481	106.9577
1356.6112	116.5461
1403.8378	87.0997
1441.2359	56.3360
1464.6414	18.9760
1633.7860	96.4571
3021.6358	32.1066
3091.1247	11.4335
3144.7559	4.8047
3510.5250	274.9908
3703.7285	224.8358

N-Methylmethanimidic acid (**8a**)

E[B3LYP/cc-pVTZ] = –209.2670449 Ha
E[CCSD(T)/CBS] = –208.9481877 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073632 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.45 eV

O	–1.380488	–1.194977	0.000000
C	–0.158217	–0.623790	0.000000
H	–2.026402	–0.473241	0.000000
N	0.012936	0.619459	0.000000
C	1.368552	1.132369	0.000000
H	1.520346	1.762979	–0.877726
H	2.135758	0.347791	0.000000
H	1.520346	1.762979	0.877726
H	0.626456	–1.383160	0.000000

Frequency	Intensity
197.3107	2.1370
285.9748	5.9184
323.2694	12.9540
588.1338	6.6240
618.3188	117.5429
996.4855	8.6921
1002.2781	106.9103
1140.2870	15.9428
1140.7490	0.2613
1186.3151	162.9356
1335.6310	6.0721
1393.3442	14.0154
1449.4087	17.5806
1482.5006	3.9930
1507.4523	8.7008
1762.2976	315.3605
2974.3777	69.6881
3056.0249	43.5871
3072.1371	23.3508
3072.1733	37.1888
3736.2186	45.0935

N-Methylmethanimidic acid (**8a**) radical cation

E[B3LYP/cc-pVTZ] = –208.9296042 Ha
E[CCSD(T)/CBS] = –208.5983685 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071178 Ha

O	1.725836	–0.376554	0.000000
C	0.648944	0.354963	0.000000
H	1.557453	–1.334682	0.000000
N	–0.544709	–0.052837	0.000000
C	–1.937136	0.142091	0.000000
H	–2.383376	–0.270700	–0.905153
H	–2.110563	1.234965	0.000000
H	–2.383376	–0.270700	0.905153
H	0.836267	1.433097	0.000000

Frequency	Intensity
82.1989	10.2218
116.7397	2.6141
181.2697	3.4838
529.8044	18.2108
602.3547	147.5003
897.0397	18.7939
944.8305	7.7925
1050.7238	0.2230
1119.7964	4.2745
1210.1291	253.5236
1346.7477	48.9217
1364.3679	6.1412
1372.0386	33.9445
1381.7816	3.1961
1477.6075	14.4158
1762.7932	223.6638
2902.6663	85.8777
3062.2901	14.4876
3065.0936	31.1771
3121.8127	9.9906
3696.7524	194.9543

N-Methylmethanimidic acid (**8b**)

E[B3LYP/cc-pVTZ] = –209.2603270 Ha
E[CCSD(T)/CBS] = –208.9425495 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073367 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.31 eV

O	–1.341745	0.686008	0.000000
C	–0.000000	0.912809	0.000000
H	–1.514656	–0.263415	0.000000
N	0.943568	0.080333	0.000000
C	0.628895	–1.341257	0.000000
H	0.058929	–1.642493	0.887710
H	1.555527	–1.910865	0.000000
H	0.058929	–1.642493	–0.887710
H	0.196886	1.979556	0.000000

Frequency	Intensity
66.7555	0.0660
274.9126	1.3047
388.4121	18.7850
567.3556	81.2169
704.4417	22.0398
948.8907	9.9760
964.5546	5.2321
1092.8657	215.4734
1117.4395	5.0102
1166.4515	33.8782
1283.7682	1.7756
1417.9993	2.9225
1434.7804	18.9410
1493.8346	0.2027
1514.6647	6.6356
1742.5554	266.7546
2967.5228	43.2635
3010.7239	41.6661
3101.7554	22.6581
3160.7890	15.3648
3784.0659	33.7292

N-Methylmethanimidic acid (**8b**) radical cation

E[B3LYP/cc-pVTZ] = –208.9291666 Ha
E[CCSD(T)/CBS] = –208.5983709 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071282 Ha

O	1.747755	0.256612	0.000000
C	0.622942	–0.398971	0.000000
H	1.645629	1.224057	0.000000
N	–0.539735	0.090165	0.000000
C	–1.942470	–0.007976	0.000000
H	–2.358848	0.434848	–0.905163
H	–2.358848	0.434848	0.905163
H	–2.191039	–1.086231	0.000000
H	0.735628	–1.487473	0.000000

Frequency	Intensity
82.4405	10.2191
116.8342	2.6060
181.3483	3.4779
529.7349	18.1977
602.4867	147.4814
896.7278	18.7824
944.9305	7.7970
1050.7229	0.2294
1119.8047	4.2672
1210.1120	253.6417
1346.6739	48.8560
1364.2742	5.9462
1372.1342	33.9288
1381.8158	3.1581
1477.5351	14.4502
1762.6804	223.5513
2902.8280	85.8451
3062.4774	16.0496
3064.8064	29.6225
3122.0327	9.9969
3696.6441	194.8418

N-Methylmethanimidic acid (**8c**)

E[B3LYP/cc-pVTZ] = –209.2596694 Ha
E[CCSD(T)/CBS] = –208.9413021 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073231 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.26 eV

O	–1.352099	0.584172	0.000000
C	–0.016244	0.886147	0.000000
H	–1.864265	1.398296	0.000000
N	0.915548	0.049443	0.000000
C	0.628954	–1.377005	0.000000
H	1.099715	–1.825931	–0.876313
H	–0.432585	–1.629822	0.000000
H	1.099715	–1.825931	0.876313
H	0.207926	1.951761	0.000000

Frequency	Intensity
30.1586	0.0006
290.3952	13.6611
351.1770	137.2677
499.8848	0.0100
703.3652	3.6576
939.9484	3.5251
942.8085	12.6347
1094.3067	90.7354
1136.9338	1.7802
1148.4667	40.1980
1265.7451	196.5768
1419.0742	1.2201
1435.9348	4.4230
1498.6354	8.2493
1498.9189	5.8087
1784.4878	199.8658
3019.1172	47.3264
3073.7046	22.4296
3084.3467	23.7280
3100.5238	31.9071
3826.7257	66.0739

N-Methylmethanimidic acid (**8c**) radical cation

E[B3LYP/cc-pVTZ] = –208.9297351 Ha
E[CCSD(T)/CBS] = –208.5990611 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0711740 Ha

O	0.543094	–1.509037	0.000000
C	0.872432	–0.240716	0.000000
H	1.319719	–2.090558	0.000000
N	0.000000	0.667347	0.000000
C	–1.306898	1.177076	0.000000
H	–1.497466	1.754104	0.905364
H	–1.982533	0.299028	0.000000
H	–1.497466	1.754104	–0.905364
H	1.919788	0.066023	0.000000

Frequency	Intensity
87.5170	0.0715
111.6347	62.7898
141.0103	13.2216
490.2726	115.0263
616.1309	2.5599
856.8607	13.8908
959.9382	6.6335
1062.3514	15.9042
1123.7562	0.1976
1238.4820	15.4281
1269.2858	296.1283
1336.7919	43.4422
1363.3175	30.5643
1410.1242	1.4591
1471.5767	8.9138
1792.9179	146.4709
2891.0119	89.8330
3062.3081	11.4469
3098.9462	27.1238
3122.5612	9.1446
3735.0200	316.2475

N-Methylmethanimidic acid (**8d**)

E[B3LYP/cc-pVTZ] = –209.2588121 Ha
E[CCSD(T)/CBS] = –208.9400508 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.073157 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.23 eV

O	1.053564	1.461924	0.000000
C	0.001486	0.603750	0.000000
H	0.724677	2.365672	0.000000
N	0.187467	–0.632853	0.000000
C	–0.973419	–1.496900	0.000000
H	–0.944195	–2.145463	–0.877351
H	–1.929933	–0.955982	0.000000
H	–0.944195	–2.145463	0.877351
H	–0.984688	1.086097	0.000000

Frequency	Intensity
204.9634	12.2656
281.6046	21.9715
328.4436	4.0845
403.4442	66.8454
605.0484	23.1389
971.7754	2.6787
1011.8623	17.2223
1138.1042	17.8194
1140.0898	0.0095
1192.4918	30.7116
1294.7825	309.6634
1405.1324	3.6667
1449.4396	13.2008
1480.7981	3.8433
1506.9258	11.3727
1792.0934	245.4243
2958.1765	62.8346
2989.2307	87.6104
3053.3611	39.0985
3071.8393	22.6024
3832.6962	74.3102

N-Methylmethanimidic acid (**8d**) radical cation

E[B3LYP/cc-pVTZ] = –208.9296042 Ha
E[CCSD(T)/CBS] = –208.5989520 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071178 Ha

O	0.736592	–1.624962	0.000000
C	–0.227595	–0.747013	0.000000
H	0.407632	–2.537121	0.000000
N	0.000000	0.491362	0.000000
C	–0.364325	1.846666	0.000000
H	–0.002513	2.337249	0.904359
H	–1.468827	1.895103	0.000000
H	–0.002513	2.337249	–0.904359
H	–1.274995	–1.070234	0.000000

Frequency	Intensity
89.6300	4.0204
111.7175	18.0324
165.7941	5.6494
518.3678	119.8944
540.8370	5.0627
895.7941	17.7888
921.1769	0.0582
1048.3615	16.7953
1117.8826	2.1731
1248.8396	36.9116
1293.1436	280.7079
1354.3237	45.9638
1380.4612	31.6993
1400.9984	5.0826
1475.4511	11.1338
1808.8173	177.4224
2911.5431	75.6198
3036.4850	26.2204
3060.8174	7.8678
3118.6055	10.3852
3744.6457	329.2428

Nitrosoethane (**9a**)

E[B3LYP/cc-pVTZ] = –209.1929417 Ha
E[CCSD(T)/CBS] = –208.8754190 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071809 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 8.95 eV

C	–1.453766	0.872625	0.085126
C	0.046208	0.711724	–0.141820
H	–1.755740	0.412924	1.026169
H	–2.019324	0.406915	–0.721397
H	–1.714884	1.929572	0.123167
N	0.352894	–0.730212	–0.345368
H	0.650953	1.100738	0.679029
H	0.336566	1.192237	–1.080754
O	1.162423	–1.172389	0.423013

Frequency	Intensity
79.8261	0.0181
205.4826	0.4154
374.2508	4.4254
563.3802	1.5158
776.0588	20.5956
850.6407	9.5215
989.8902	2.4156
1040.2919	5.7795
1172.5039	12.3573
1254.8004	5.5702
1327.2934	3.0046
1409.3456	2.1210
1465.3411	4.1655
1493.8779	8.6402
1504.9699	7.2053
1655.6773	93.3357
3017.6426	11.6618
3040.8631	17.5183
3077.2046	8.9598
3108.4360	18.6184
3112.7698	20.4221

Nitrosoethane (**9a**) radical cation

E[B3LYP/cc-pVTZ] = –208.8706775 Ha
E[CCSD(T)/CBS] = –208.5461634 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071297 Ha

C	1.603207	–0.348145	–0.083893
C	0.478149	0.685904	–0.012993
H	1.421865	–1.088949	–0.858675
H	1.778200	–0.828487	0.875631
H	2.493734	0.227096	–0.349027
N	–0.793128	–0.061935	0.396692
H	0.226505	1.162648	–0.960741
H	0.615679	1.396748	0.801162
O	–1.664464	–0.394787	–0.241694

Frequency	Intensity
81.3264	0.7401
224.9208	0.0111
345.3362	1.9619
423.5225	6.8056
485.9686	1.1257
821.8158	2.9819
903.9539	2.2805
1035.7715	25.7531
1125.7589	8.8020
1241.8775	7.9979
1292.9447	1.0343
1404.2575	1.6821
1446.7400	18.7583
1457.3774	22.8984
1493.2341	10.8907
1991.7562	90.9671
3041.0379	17.0657
3058.9701	11.6379
3120.0287	13.6726
3138.5094	11.1496
3160.4514	3.3490

Nitrosoethane (**9b**)

E[B3LYP/cc-pVTZ] = –209.1934683 Ha
E[CCSD(T)/CBS] = –208.8760418 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071612 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 8.92 eV

C	–1.417973	0.347374	0.000000
C	–0.002559	0.883703	0.000000
H	–1.606751	–0.268043	0.878654
H	–1.606751	–0.268043	–0.878654
H	–2.126588	1.175233	0.000000
N	1.097999	–0.126132	0.000000
H	0.220404	1.510299	0.869791
H	0.220404	1.510299	–0.869791
O	0.726512	–1.267603	0.000000

Frequency	Intensity
152.7733	0.7700
222.5695	2.5441
298.1081	2.8997
675.6516	0.7543
729.5976	0.4046
789.7955	29.3565
986.1200	0.5402
1038.4405	8.2943
1118.4588	11.0261
1259.7997	1.0024
1330.0078	14.3292
1411.3627	10.1538
1444.6400	18.8454
1487.9746	7.3130
1496.3857	6.4443
1658.0756	74.8652
3020.7035	13.1770
3043.2004	4.0417
3046.2567	11.5142
3110.9493	14.6828
3113.1695	18.6655

Nitrosoethane (**9b**)

E[B3LYP/cc-pVTZ] = –208.8723184 Ha
E[CCSD(T)/CBS] = –208.5474482 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.07098 Ha

C	–1.451362	0.350691	0.000000
C	–0.048373	0.904846	0.000000
H	–1.657838	–0.236318	0.892862
H	–1.657838	–0.236318	–0.892862
H	–2.114329	1.219426	0.000000
N	1.020776	–0.173060	0.000000
H	0.246281	1.492947	0.878093
H	0.246281	1.492947	–0.878093
O	0.919357	–1.299271	0.000000

Frequency	Intensity
135.9865	1.4775
247.0541	1.1000
252.0262	0.9272
489.2177	11.1792
585.7164	2.2193
732.9950	5.1959
922.3800	10.2758
1084.7517	12.6833
1084.9323	5.5700
1227.4484	1.5813
1292.8652	2.2695
1398.5950	56.5927
1410.4168	3.9296
1456.1393	16.9375
1487.7563	5.4576
1988.4908	83.6595
3018.6933	44.8383
3040.2995	15.4805
3045.2040	23.3495
3112.2907	9.3242
3143.2261	0.5482

Iminoethanol (**10a**)

E[B3LYP/cc-pVTZ] = –209.2478773 Ha
E[CCSD(T)/CBS] = –208.9326334 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0737620 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.64 eV

H	0.552024	2.543554	–0.288668
N	0.768271	1.567805	–0.492384
C	0.043682	0.785476	0.189420
H	–0.730839	1.119618	0.889805
C	0.176444	–0.707366	0.069108
O	–1.090087	–1.332310	–0.118762
H	–1.420429	–1.087340	–0.988426
H	0.568772	–1.113390	1.004341
H	0.883878	–0.942113	–0.730161

Frequency	Intensity
94.1649	15.1252
334.6756	100.3978
351.4108	40.8856
494.8916	7.2581
729.3745	27.0717
970.6256	65.3240
1028.4526	19.1972
1053.5123	79.0365
1129.0074	7.5863
1178.9265	39.4713
1270.6661	7.8401
1357.3830	6.0554
1393.6697	12.6516
1422.8479	59.1153
1490.5056	3.4577
1722.4523	43.5604
3007.0277	44.5249
3024.1739	30.9343
3069.5405	23.9933
3440.8549	0.3491
3813.6794	29.1624

Iminoethanol (**10a**) radical cation

E[B3LYP/cc-pVTZ] = –208.9053681 Ha
E[CCSD(T)/CBS] = –208.5750024 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0702800 Ha

H	–2.764419	0.281852	0.208103
N	–1.774809	0.064040	0.144774
C	–0.733291	0.318633	–0.413758
H	–0.569696	1.093598	–1.174584
C	0.622475	–0.517283	–0.071523
O	1.643739	0.366254	0.098776
H	1.847348	0.516995	1.032582
H	0.762987	–1.088410	–0.993729
H	0.406861	–1.163035	0.776263

Frequency	Intensity
89.4035	3.7764
343.2980	6.1501
373.3434	31.6382
430.5165	89.5776
572.0626	95.8088
628.9653	119.4729
663.6864	460.6665
816.7549	56.5720
968.0165	64.0725
1105.4673	56.6135
1165.9271	117.7262
1198.3967	36.3158
1326.6934	0.5204
1377.0408	24.9532
1489.3518	3.6003
1825.4082	10.5809
2994.7546	43.9610
3032.1892	15.5500
3138.2661	3.9184
3552.7999	482.8561
3757.2061	268.7928

Iminoethanol (**10b**)

E[B3LYP/cc-pVTZ] = –209.2465331 Ha
E[CCSD(T)/CBS] = –208.9316104 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0733070 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.62 eV

H	–1.348956	–2.237680	0.154974
N	–1.336386	–1.227036	0.285540
C	–0.242048	–0.730669	–0.106663
H	0.592483	–1.311676	–0.515133
C	0.002095	0.750020	–0.062070
O	1.385014	0.957001	0.203045
H	1.572023	1.897707	0.152304
H	–0.643531	1.195322	0.699602
H	–0.279172	1.172644	–1.036529

Frequency	Intensity
91.4897	18.7753
219.7214	125.5722
334.7163	27.5591
516.3308	6.5905
698.1979	26.4227
965.9739	6.0296
1043.6119	49.4260
1070.7604	78.9470
1127.6360	5.3243
1205.0706	93.9854
1231.3603	21.3097
1283.2766	5.8587
1396.7170	25.0458
1449.3262	12.5503
1501.0517	7.2950
1731.5677	47.1475
2965.4250	42.4035
3014.9475	31.3629
3039.6203	39.1138
3452.3287	0.1218
3838.8647	37.8823

Iminoethanol (**10b**) radical cation

E[B3LYP/cc-pVTZ] = –208.9053684 Ha
E[CCSD(T)/CBS] = –208.5750139 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0702670 Ha

H	2.612368	–0.337548	–0.536287
N	1.657991	–0.079572	–0.305742
C	0.655451	–0.395335	0.291283
H	0.494316	–1.321711	0.858793
C	–0.644898	0.586955	0.317541
O	–1.755527	–0.157931	0.064145
H	–2.061681	–0.062473	–0.848737
H	–0.453900	1.404811	–0.372986
H	–0.640945	0.922562	1.358528

Frequency	Intensity
89.4209	3.8025
342.9617	5.8018
373.5373	30.6185
430.8936	90.7555
570.1742	95.2926
627.1070	124.7301
660.7687	452.6365
816.2194	56.3609
967.9215	63.8750
1105.2303	56.6645
1165.7856	118.8522
1198.2564	35.8728
1326.4758	0.5403
1377.1017	24.9833
1489.2297	3.5718
1825.4782	10.6816
2994.5749	43.7865
3032.5619	15.5685
3138.4599	3.9131
3554.3929	484.1894
3757.0314	267.9508

Iminoethanol (**10c**)

E[B3LYP/cc-pVTZ] = –209.2487151 Ha
E[CCSD(T)/CBS] = –208.9337995 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0736210 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.56 eV

H	–1.544142	–0.183918	0.000000
N	–1.275773	0.803149	0.000000
C	–0.018090	0.900925	0.000000
H	0.431466	1.896876	0.000000
C	0.973987	–0.231473	0.000000
O	0.281858	–1.464960	0.000000
H	0.917739	–2.184241	0.000000
H	1.619102	–0.124770	0.882794
H	1.619102	–0.124770	–0.882794

Frequency	Intensity
140.1496	107.9297
247.2950	20.1601
289.8065	1.1937
680.2851	11.7262
686.0581	44.7762
870.5961	4.6221
1061.8312	0.0026
1100.4068	50.9971
1150.2613	53.4942
1184.4687	33.7055
1251.9759	0.1417
1284.8945	129.4500
1425.5546	17.9411
1440.5513	10.0354
1497.1975	13.0220
1731.3438	53.6334
2963.7106	39.2768
2986.2592	36.0153
3057.8627	46.7862
3416.7052	2.2546
3848.7179	46.0352

Iminoethanol (**10c**) radical cation

E[B3LYP/cc-pVTZ] = –208.9084248 Ha
E[CCSD(T)/CBS] = –208.5795948 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0707340 Ha

H	1.352493	–1.528655	0.254483
N	1.509314	–0.537439	0.097010
C	0.882761	0.494818	–0.033673
H	1.366548	1.470368	–0.152773
C	–0.745224	0.630519	–0.007949
O	–1.402909	–0.549981	0.018639
H	–1.743987	–0.801306	–0.850823
H	–0.960999	1.278824	–0.860272
H	–0.868821	1.163063	0.941645

Frequency	Intensity
160.4236	12.2944
235.1633	6.5230
406.8073	37.1163
502.8914	183.1355
573.2109	81.0756
637.5171	73.9074
779.1475	88.6702
821.1399	408.2228
981.7851	32.8470
1103.9879	100.1109
1189.1387	65.3065
1241.3737	43.9848
1316.1656	1.2643
1384.5176	22.1540
1478.0753	1.6086
1790.7143	10.8517
3008.9724	26.4607
3043.2158	38.5799
3088.4130	4.1922
3545.5174	271.7288
3760.4432	251.1049

Iminoethanol (**10d**)

E[B3LYP/cc-pVTZ] = –209.2492253 Ha
E[CCSD(T)/CBS] = –208.9339407 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0738800 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.56 eV

H	–1.574872	0.016405	–0.414140
N	–1.256912	0.981270	–0.282177
C	–0.011717	0.989059	–0.073229
H	0.486456	1.951856	0.068333
C	0.897655	–0.216045	0.002764
O	0.214729	–1.451801	–0.090943
H	–0.231582	–1.623627	0.744001
H	1.502363	–0.150313	0.915936
H	1.598927	–0.181166	–0.835360

Frequency	Intensity
211.4102	11.2268
280.5127	53.6382
299.7783	79.5646
672.4685	5.5280
707.2385	16.3313
864.2730	9.2430
1012.5262	31.3959
1093.6080	47.8943
1155.9877	50.9158
1205.0849	22.1906
1238.7144	84.1690
1380.9412	9.0797
1390.1957	21.0784
1423.6153	25.4857
1478.4989	11.2758
1725.3565	50.3836
2986.1322	32.9992
3046.7660	13.4485
3053.9580	50.8475
3389.1725	0.1183
3813.4241	33.6962

Iminoethanol (**10d**) radical cation

E[B3LYP/cc-pVTZ] = –208.9084248 Ha
E[CCSD(T)/CBS] = –208.5795948 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0707340 Ha

H	1.352493	–1.528655	0.254483
N	1.509314	–0.537439	0.097010
C	0.882761	0.494818	–0.033673
H	1.366548	1.470368	–0.152773
C	–0.745224	0.630519	–0.007949
O	–1.402909	–0.549981	0.018639
H	–1.743987	–0.801306	–0.850823
H	–0.960999	1.278824	–0.860272
H	–0.868821	1.163063	0.941645

Frequency	Intensity
160.4236	12.2944
235.1633	6.5230
406.8073	37.1163
502.8914	183.1355
573.2109	81.0756
637.5171	73.9074
779.1475	88.6702
821.1399	408.2228
981.7851	32.8470
1103.9879	100.1109
1189.1387	65.3065
1241.3737	43.9848
1316.1656	1.2643
1384.5176	22.1540
1478.0753	1.6086
1790.7143	10.8517
3008.9724	26.4607
3043.2158	38.5799
3088.4130	4.1922
3545.5174	271.7288
3760.4432	251.1049

Iminoethanol (**10e**)

E[B3LYP/cc-pVTZ] = –209.2460527 Ha
E[CCSD(T)/CBS] = –208.9307625 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0736950 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.52 eV

H	–1.868168	–0.733384	–0.742743
N	–1.256018	–1.382217	–0.240216
C	–0.265492	–0.772477	0.257629
H	0.470494	–1.357380	0.814351
C	0.040655	0.702911	0.175743
O	1.411871	0.957195	–0.096660
H	1.626251	0.594826	–0.961988
H	–0.153136	1.162101	1.148246
H	–0.620996	1.182055	–0.556235

Frequency	Intensity
95.1131	4.1988
331.7895	81.8616
345.0612	59.8302
493.9004	5.8032
724.9433	14.2436
973.2780	6.5978
1039.8058	21.9264
1059.9177	96.3528
1143.9472	57.8321
1190.5074	1.4109
1267.6749	78.9610
1362.6390	7.1726
1398.6197	53.7410
1415.7711	3.9735
1487.9338	3.1234
1716.5543	44.3182
2985.8021	42.3200
3051.3740	11.7894
3065.8309	35.2135
3388.2930	5.3425
3809.4595	29.3975

Iminoethanol (**10e**) radical cation

E[B3LYP/cc-pVTZ] = –208.9085387 Ha
E[CCSD(T)/CBS] = –208.5784957 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0711220 Ha

H	2.198210	0.846431	0.659355
N	1.819962	0.051006	0.153576
C	0.759875	–0.351487	–0.286591
H	0.640410	–1.300289	–0.814977
C	–0.643775	0.489617	–0.167487
O	–1.641929	–0.381903	0.123820
H	–1.879431	–0.378013	1.062413
H	–0.746493	0.854893	–1.193814
H	–0.496367	1.299289	0.544112

Frequency	Intensity
97.8381	0.8414
339.1594	9.3929
363.4923	11.0910
437.3628	93.6741
595.3146	303.7070
665.9472	180.0250
800.8258	200.7107
891.8430	57.4940
986.7010	44.0288
1156.7314	3.0840
1169.3213	203.5507
1212.3032	10.2918
1318.2466	3.4974
1377.1634	22.8380
1485.5083	3.3520
1789.6736	1.4741
3027.3530	19.2102
3076.3653	41.1836
3130.9944	4.0816
3546.9560	296.1937
3749.6589	283.4340

Iminoethanol (**10f**)

E[B3LYP/cc-pVTZ] = –209.2445985 Ha
E[CCSD(T)/CBS] = –208.9297104 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0732500 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.50 eV

H	–2.004457	–0.748638	0.495670
N	–1.278327	–1.388463	0.162960
C	–0.224119	–0.761372	–0.141842
H	0.634562	–1.332975	–0.499151
C	–0.005261	0.728793	–0.104812
O	1.345131	0.972541	0.262851
H	1.548280	1.899138	0.111247
H	–0.710799	1.188023	0.597856
H	–0.218942	1.128224	–1.105725

Frequency	Intensity
80.4309	6.2696
215.5048	102.9761
335.3553	5.0521
503.8043	14.5008
708.5541	15.6375
963.5980	37.7035
1054.6724	6.3344
1073.0303	71.1436
1140.9470	51.9097
1217.2865	23.0324
1230.0237	37.8165
1295.5830	93.5795
1403.0057	22.4807
1440.8859	0.9525
1497.6121	4.9588
1724.9555	47.3971
2963.2215	41.4602
3000.5443	35.8381
3075.0104	26.6215
3393.7337	5.4477
3835.0815	37.1335

Iminoethanol (**10f**) radical cation

E[B3LYP/cc-pVTZ] = –208.9085382 Ha
E[CCSD(T)/CBS] = –208.5785053 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0711190 Ha

H	2.086170	0.852308	–0.746473
N	1.702840	0.017681	–0.312996
C	0.671615	–0.374522	0.199262
H	0.534259	–1.382777	0.596977
C	–0.657134	0.568473	0.385527
O	–1.754486	–0.178841	0.105741
H	–2.100108	–0.016960	–0.783983
H	–0.526374	1.465594	–0.215909
H	–0.604870	0.780098	1.457952

Frequency	Intensity
97.8246	0.8449
339.2412	9.4797
364.2337	11.5590
436.5520	94.2818
595.4477	303.4910
665.5358	181.8849
800.0982	198.9293
891.4671	56.3150
986.6481	44.0861
1156.6865	3.2893
1169.1555	203.0195
1212.3077	10.4100
1318.5542	3.5878
1376.8919	22.8414
1485.6800	3.3554
1789.3574	1.4685
3026.5794	19.2257
3076.6851	41.2262
3130.6667	3.9689
3547.7959	296.8566
3750.3866	283.1551

Iminoethanol (**10g**)

E[B3LYP/cc-pVTZ] = –209.2536838 Ha
E[CCSD(T)/CBS] = –208.9383305 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0739480 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.12 eV

H	–2.356716	–0.534120	0.000000
N	–1.340727	–0.572966	0.000000
C	–0.804078	0.572156	0.000000
H	–1.359883	1.514701	0.000000
C	0.692292	0.678874	0.000000
O	1.321625	–0.573149	0.000000
H	0.605067	–1.228023	0.000000
H	0.995285	1.265772	0.878665
H	0.995285	1.265772	–0.878665

Frequency	Intensity
220.0464	0.0209
307.8156	13.2904
443.3677	87.6094
645.1517	33.3142
725.3695	23.9799
882.7171	23.7481
1041.0996	22.7439
1104.5919	105.4585
1111.6363	3.1183
1177.5381	36.0421
1254.0749	3.5376
1338.9966	21.6513
1406.0886	58.2687
1460.1346	65.2590
1493.6975	13.1733
1720.6600	61.3376
2965.4142	33.5801
2976.3045	33.1311
3033.8776	61.1036
3487.8697	1.5227
3662.9008	82.8580

Iminoethanol (**10g**) radical cation

E[B3LYP/cc-pVTZ] = –208.9289472 Ha
E[CCSD(T)/CBS] = –208.6025123 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.0733020 Ha

H	–1.874135	1.307783	0.000000
N	–1.161795	0.585306	0.000000
C	0.086768	0.843856	0.000000
H	0.412946	1.879201	0.000000
C	1.072372	–0.288527	0.000000
O	0.490863	–1.510188	0.000000
H	–1.457051	–0.393274	0.000000
H	1.758886	–0.187028	0.862736
H	1.758886	–0.187028	–0.862736

Frequency	Intensity
232.9636	10.3338
287.0130	21.2004
567.4739	20.0353
675.7780	2.1342
790.9388	0.9013
863.8057	3.3402
907.5994	164.8490
1067.4529	17.0675
1094.2185	14.6244
1139.6394	13.2074
1181.7443	0.8075
1300.5970	59.1668
1351.0172	8.3408
1411.4036	5.4020
1561.3881	39.4195
1747.4945	109.8246
2910.7324	29.3075
2915.4264	20.3748
3176.1610	7.6553
3421.2797	121.2516
3571.7444	207.7157

Acetaldoxime (**11a**)

E[B3LYP/cc-pVTZ] = –209.2205174 Ha
E[CCSD(T)/CBS] = –208.9008213 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.072437 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.61 eV

C	1.905155	0.000503	0.000000
C	0.470582	–0.404953	0.000000
H	2.419185	–0.398709	0.877762
H	1.995561	1.084786	0.000000
H	2.419185	–0.398709	–0.877762
N	–0.441956	0.474819	0.000000
O	–1.721757	–0.110422	0.000000
H	–2.303219	0.655948	0.000000
H	0.210064	–1.463480	0.000000

Frequency	Intensity
205.8477	0.2955
286.7196	0.8283
326.5093	3.1568
404.0974	126.1751
566.1932	14.7695
905.2652	25.0674
917.7094	8.5388
1002.3700	135.6573
1083.9248	1.1017
1151.5206	3.8815
1290.0106	56.3691
1404.0323	19.4256
1440.5315	7.6090
1479.1504	7.9252
1483.5468	11.0765
1732.1489	1.4190
3022.5030	18.9635
3065.4650	15.6967
3082.5903	14.1865
3122.8920	13.8169
3823.2756	88.5292

Acetaldoxime (**11a**) radical cation

E[B3LYP/cc-pVTZ] = –208.8788445 Ha
E[CCSD(T)/CBS] = –208.5466786 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071417 Ha

C	–0.877389	1.632394	–0.000247
C	0.155316	0.586747	0.077035
H	–0.798329	2.270101	0.886127
H	–1.880125	1.227433	–0.093800
H	–0.644513	2.276139	–0.855259
N	–0.123711	–0.656032	0.120841
O	0.637967	–1.666190	–0.174017
H	0.453941	–2.386595	0.467052
H	1.213094	0.848172	0.174213

Frequency	Intensity
172.5875	5.1317
236.6588	20.7196
284.7002	2.0821
453.3717	39.5602
561.2271	69.1878
876.7059	41.6823
905.5763	6.3962
1053.1647	6.5910
1142.7471	97.2518
1156.4987	63.7467
1299.1974	89.1489
1373.5434	23.5748
1391.9087	72.8072
1442.4189	17.9164
1466.1356	19.4926
1648.4213	9.2792
3016.8300	39.6614
3061.5961	10.2051
3066.5862	27.0270
3162.3367	7.8803
3576.1697	365.8408

Acetaldoxime (**11b**)

E[B3LYP/cc-pVTZ] = –209.2128357 Ha
E[CCSD(T)/CBS] = –208.8929545 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.07194 Ha
IE[CCSD(T)/CBS//B3LYP/cc-pVTZ] = 9.41 eV

C	–1.280865	1.313228	0.000000
C	–0.003559	0.541363	0.000000
H	–1.343718	1.958977	0.878789
H	–2.129489	0.633098	0.000000
H	–1.343718	1.958977	–0.878789
N	–0.011666	–0.726693	0.000000
O	1.217830	–1.362701	0.000000
H	1.918590	–0.687566	0.000000
H	0.941612	1.096864	0.000000

Frequency	Intensity
195.6572	4.8783
280.8732	15.3696
333.3321	20.8805
442.0611	57.6493
581.7455	0.4169
893.0864	13.0063
916.0223	36.0372
1009.0973	102.3432
1075.5313	0.5544
1156.3674	3.6453
1320.9274	0.6015
1406.6085	2.7602
1449.3499	65.8724
1478.6553	8.1218
1481.6389	14.8610
1707.9265	2.6025
3004.9646	49.9241
3026.6100	16.2949
3069.7541	15.3015
3128.4499	8.5057
3619.5153	0.0028

Acetaldoxime (**11b**) radical cation

E[B3LYP/cc-pVTZ] = –208.8788447 Ha
E[CCSD(T)/CBS] = –208.5466828 Ha
ZPVE[B3LYP/cc-pVTZ] = 0.071418 Ha

C	–1.302672	1.321740	0.022762
C	0.056980	0.774603	–0.113321
H	–1.280050	2.141235	0.748387
H	–2.030909	0.571374	0.314072
H	–1.582978	1.774807	–0.934145
N	0.346576	–0.431887	0.176999
O	1.361722	–1.138221	–0.220432
H	1.664685	–1.690941	0.532331
H	0.893750	1.416078	–0.404789

Frequency	Intensity
172.6235	5.0630
236.5318	20.6821
284.7276	2.0536
453.5969	39.5744
561.0601	69.1323
876.6225	42.0926
905.3861	6.2386
1053.3798	6.3132
1142.9691	96.5361
1156.1699	64.0452
1299.2136	89.1558
1373.5435	22.8009
1391.7983	73.3880
1442.5846	17.8542
1466.0918	19.4793
1649.1650	9.1620
3017.0401	39.4811
3061.9902	10.1943
3066.7779	27.0406
3162.2898	7.8731
3575.3906	365.7610

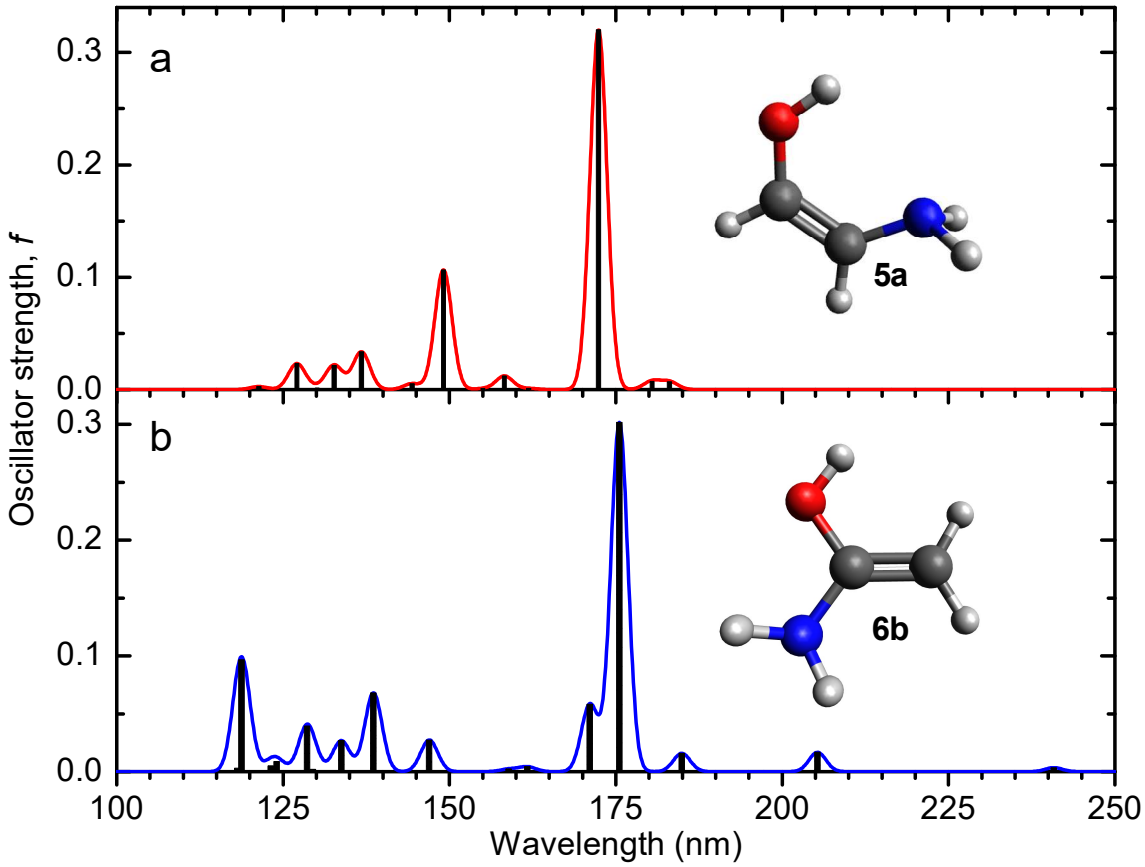


Figure S9. Calculated electronic absorption spectra of (a) **5a** and (b) **6b**. These spectra were calculated at the B3LYP/cc-pVTZ level using time-dependent density functional theory (TD-DFT). The simulated spectra include 3 nm FWHM broadening and show that if **6b** is present it can be expected to absorb at 206 nm while **5a** has no electronic transitions that lie below 185 nm.

Table S4. Calculated ultraviolet absorption bands of **5a**, **5b**, **6a**, and **6b** calculated at the B3LYP/cc-pVTZ level using time-dependent density functional theory (TD-DFT) reported with f (oscillator strength) as a function of wavelength (nm).

5a		**5b**		**6a**		**6b**	
wavelength (nm)	f	wavelength (nm)	f	wavelength (nm)	f	wavelength (nm)	f
211.53	0	248.72	0.003	229.31	0.0102	240.82	0.0035
183.06	0.0069	233.88	0.0052	192.03	0.0859	205.31	0.0167
180.45	0.0073	206.96	0.008	183.66	0.0897	184.91	0.0158
179.89	0.0002	192.50	0.3086	171.61	0.214	175.54	0.3016
172.40	0.3201	174.70	0.0123	164.61	0.014	171.10	0.0582
161.91	0.0011	169.23	0.0027	163.09	0.014	161.70	0.0043
158.26	0.0121	150.03	0.0287	152.29	0.0313	158.90	0.0022
155.04	0.0009	145.02	0.0116	149.70	0.018	146.96	0.0274
149.09	0.1066	140.00	0.0224	140.30	0.0139	138.58	0.0681
144.39	0.0053	133.09	0.0095	134.45	0.007	133.74	0.0268
136.77	0.0336	130.40	0.0051	131.06	0.0679	129.49	0.0021
134.41	0.0006	128.70	0.0497	128.28	0.0238	128.61	0.0394
132.66	0.0215	127.04	0.0145	123.61	0.0347	124.00	0.0088
130.11	0.0015	122.78	0.0168	123.45	0.0089	123.17	0.0049
127.07	0.0231	121.18	0.0112	122.72	0.0063	118.77	0.0968
121.36	0.0027	119.55	0.001	122.39	0.0637	118.09	0.0029

Table S5. Ices studied in this experiment containing acetaldehyde and ammonia or their isotopomers. The ratio of the concentration of the components, ice thickness, irradiation parameters, and photon energy used are listed.

Ice	[NH_3] : [CH_3CHO]	Thickness (nm)	Irradiation Time (min)	Irradiation Current (nA)	Dose (eV/NH_3)	Dose (eV/CH_3CHO)	PI Energy (eV)
NH_3/CH_3CHO	1.6 ± 0.8 : 1	736 ± 50	-	-	-	-	10.49
NH_3/CH_3CHO-d_4	2.5 ± 0.5 : 1	733 ± 50	-	-	-	-	9.93
NH_3/CH_3CHO-d_4	1.3 ± 0.5 : 1	735 ± 50	15 ± 0.1	9.5 ± 1	0.15 ± 0.02	0.41 ± 0.06	9.93
NH_3/CH_3CHO-d_4	1.7 ± 0.5 : 1	736 ± 50	4 ± 0.1	10.0 ± 1	0.041 ± 0.006	0.12 ± 0.02	9.93
NH_3/CH_3CHO-d_4	1.3 ± 0.6 : 1	737 ± 50	-	-	-	-	9.50
NH_3/CH_3CHO-d_4	1.4 ± 0.3 : 1	736 ± 50	15 ± 0.1	9.4 ± 1	0.15 ± 0.02	0.41 ± 0.06	9.50
NH_3/CH_3CHO-d_4	2.2 ± 0.7 : 1	734 ± 50	-	-	-	-	8.90
NH_3/CH_3CHO-d_4	1.4 ± 0.2 : 1	734 ± 50	15 ± 0.1	9.7 ± 1	0.15 ± 0.02	0.42 ± 0.06	8.90
NH_3/CH_3CHO	2.1 ± 0.9 : 1	734 ± 50	30 ± 0.1	50.2 ± 1	1.5 ± 0.2	3.9 ± 0.5	8.81
$NH_3/^{13}C_2$-CH_3CHO	1.5 ± 0.3 : 1	736 ± 50	30 ± 0.1	51.2 ± 1	1.5 ± 0.2	3.9 ± 0.6	8.81
NH_3/CH_3CHO-d_4	1.6 ± 0.8 : 1	737 ± 50	30 ± 0.1	52.5 ± 1	1.6 ± 0.2	4.6 ± 0.6	8.81
NH_3/CH_3CHO	1.5 ± 0.6 : 1	736 ± 50	30 ± 0.1	49.6 ± 1	1.5 ± 0.2	3.8 ± 0.5	8.71
NH_3/CH_3CHO*	1.6 ± 0.6 : 1	732 ± 50	30 ± 0.1	52.1 ± 1	1.5 ± 0.2	3.8 ± 0.5	8.71
NH_3/CH_3CHO*	1.6 ± 0.5 : 1	733 ± 50	-	-	-	-	8.71
NH_3/CH_3CHO	1.5 ± 0.7 : 1	736 ± 50	30 ± 0.1	52.7 ± 1	1.6 ± 0.2	4.0 ± 0.6	7.60
NH_3/CH_3CHO	1.6 ± 0.6 : 1	736 ± 50	30 ± 0.1	50.0 ± 1	1.5 ± 0.2	3.9 ± 0.5	8.29 – 8.64
NH_3/CH_3CHO	2.2 ± 0.8 : 1	736 ± 50	30 ± 0.1	48.5 ± 1	1.4 ± 0.2	3.7 ± 0.5	8.23 – 8.36
NH_3/CH_3CHO	1.4 ± 0.5 : 1	736 ± 50	30 ± 0.1	49.9 ± 1	1.5 ± 0.2	3.8 ± 0.5	7.90 – 8.00
NH_3/CH_3CHO	1.4 ± 0.5 : 1	736 ± 50	30 ± 0.1	50.3 ± 1	1.5 ± 0.2	3.9 ± 0.5	7.73 – 7.90
NH_3/CH_3CHO	1.8 ± 0.5 : 1	734 ± 50	30 ± 0.1	51.1 ± 1	1.5 ± 0.2	3.9 ± 0.5	7.73 – 7.90

*Ice was photolyzed with 10 mW of 205.3 nm laser light for 5 hours

Table S6. Parameters Used in Irradiation Dose Calculation and Resulting Doses

Irradiated area	1.6 ± 0.1 cm^2		
Initial kinetic energy of e$^-$	5.000 keV		
Irradiation current	30 nA		
Total number of e$^-$	$(5.6 \pm 0.1) \times 10^{14}$		
Total molecules irradiated	9.93×10^{17}		
	Acetaldehyde–Ammonia	Acetaldehyde-$^{13}C_2$–Ammonia	Acetaldehyde-d_4–Ammonia
Approximate density of mixed ice*	0.77_4g cm^{-1}	0.79_2 g cm^{-1}	0.80_9 g cm^{-1}
Average Penetration depth	348 ± 10 nm	337 ± 10 nm	329 ± 10 nm
Fraction of backscattered e$^-$	0.329	0.326	0.332
Average energy of backscattered e$^-$	1.06 keV	1.06 keV	1.072 keV
Fraction of Transmitted e$^-$	0.002	0.001	0.000
Average energy of transmitted e$^-$	0.002 keV	0.001 keV	0.000 keV
Dose per molecule of ammonia	1.4 ± 0.2	1.5 ± 0.02	1.5 ± 0.02
Dose per molecule of acetaldehyde	3.7 ± 0.5	3.9 ± 0.06	4.0 ± 0.06

*average density with 0.76 g cm^{-3} for ammonia[8] and 0.787 g cm^{-3} for acetaldehyde[9], with corrections made for isotopic labelling

Table S7. Error range demonstrated by ionization energies calculated with CCSD(T)/CBS//B3LYP/cc-pVTZ. The larger of the two bounds, 0.05 eV, is applied as ± 0.05 eV error for all presented ionization energies.

Name	IE calc (eV)	IE exp (eV)	IE Difference (lower limit)	IE Difference (upper limit)
Acetonitrile	12.20	12.20 ± 0.01	0.01	0.01
Methyl isocyanide	11.25	11.24 ± 0.01	0.02	0.00
Azirine	10.02	10.05 ± 0.03	0.00	0.06
Acetic acid	10.65	10.65 ± 0.02	-0.02	0.02
		Average:	0.00	0.02
		Std Dev:	0.01	0.02
		Error Bound:	-0.02	0.05

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