

Electronic Supplementary Information:

Table S1: Infrared absorption bands and assignments for laboratory acetonitrile aerosols compared to thin film studies at 95 K.

Mode	Assignment	Peak Position – Aerosol (95 K) / cm ⁻¹	Peak Position – Thin Film (95 K) ^a / cm ⁻¹
$\nu_2 + \nu_4$	combination	3163	3161 ^b
ν_5	CH ₃ asym. stretch	3000 (2989 sh)	3000
ν_1	CH ₃ sym. stretch	2940	2939
	-	2737	na
$\nu_2 + \nu_8$	combination	2633	2628 ^c
$\nu_6 + \nu_7$	combination	2483	2486 ^c
	-	2446	na
$\nu_3 + \nu_7$	combination	2411 (2421 sh)	2414 ^b
	CO ₂	2344	-
$\nu_3 + \nu_4$	combination	2294	2293 ^b
ν_2	C≡N stretch	2251	2251 (2247 sh)
	-	2223	na
	-	2200	
	-	2073	
$2\nu_4$	overtone	1830	1834 ^c
	HCOOH	1710	-
		1454	1454
$\nu_7 + \nu_8$	combination	1451 (1448 sh)	1451 (1448 sh)
		1443	1443
		1420 (1427 sh)	1420
ν_6	CH ₃ deform.	1416	-
		1408	1408
		1378	1378
ν_3	CH ₃ deform.	1372 (1368 sh)	1372 (1368 sh)
		1048	1049
ν_7	CH ₃ rock	1040	1040
		1036 (1032 sh)	1036 (1032 sh)
		918 (921 sh)	915 (918 sh)
ν_4	C-C stretch	903	
$2\nu_8$	overtone	774	774
		394 (392 sh)	393
ν_8	C-C≡N bend	386	387
		131 (159 sh)	123
	Lattice	93	91

(a) Dello Russo & Khanna (1996); (b) Pace & Noe (1968) – β -phase thin film at 223 K; (c) Parker, Nielson & Fletcher (1957) – room temperature gas; na: Not Assigned.

Table S2: Infrared absorption bands and assignments for laboratory propionitrile aerosols compared to thin film studies at 95 K.

Mode	Assignment	Peak Position – Aerosol (95 K) / cm ⁻¹	Peak Position – Thin Film (95 K) ^a / cm ⁻¹
$\nu_{1, \nu_{14}}$	CH ₃ asym. stretch	2998	2990
ν_{15}	CH ₂ asym. stretch	2971	2969
ν_2	CH ₃ sym. stretch	2954	2949
			2945
ν_3	CH ₂ sym. stretch	2886	2914
			2893
			2882
			2877
	CO ₂	2342	-
ν_4	C≡N stretch	2245	2261
			2245
			2218
	HCOOH	1711	-
ν_5, ν_{16}	CH ₃ asym. deform.	1462	1466
			1460
			1456
ν_6	CH ₂ bend	1427	1435
			1430
ν_7	CH ₃ deform.	1375	1383
			1380
ν_8	CH ₂ wag	1316	1310
ν_{17}	CH ₂ twist	Not observed	1269
ν_9	C-CN stretch	1077	1078
			1074
ν_{18}	CH ₂ rock	1039	-
ν_{10}	C-C stretch	1004	1015
			1009
	CH ₃ CN	918	-
ν_{11}	CH ₃ rock	Not observed	836
ν_{19}	CH ₂ rock	784	780
			764
ν_{12}	C-C-C deform.	547	546
			544
ν_{20}	C-C≡N bend	387	387
$\nu_{13, \nu_{21}}$	C-C≡N bend / CH ₃ torsion	226	225
	Lattice	115	141, 117
			106, 102
			94, 85