The prolate asymmetric top carbon chain molecules vinyladiacetylene (hex-1-ene-3,5-diyne), vinyltriacetylene (oct-1-ene-3,5,7-triyne) and vinylcyanodiacetylene (1-cyanohex-5-ene-1,3-diyne) have been produced in situ through discharges of selected precursor gases and have been investigated by Fourier transform microwave spectroscopy of molecular beams in the centimeter wave range. Initial searches were guided by results obtained from high-level quantum chemical calculations. Because the molecules are similar in structure and composition to known astronomical molecules and because of their significant polarity, these species are plausible candidates for radioastronomical detection.