Fully understanding the evolutionary state of a galaxy requires that we characterize its gas reservoir, of which the molecular component represents the mass directly available for star formation. I will discuss radio observations of molecular gas in two populations of star-forming galaxies at high redshift (selected based on their rest-frame ultraviolet and far-infrared emission) and what we can learn from them. I will highlight recent work with the "Zpectrometer" (a new, ultrawide bandwidth spectrometer for the 100m Green Bank Telescope) and prospects for future work with ALMA (Atacama Large Millimeter/submillimeter Array).