Controlling the electronic properties of 2D materials down to the single-nanometer scale using conventional lithographic technologies is a very challenging task. An alternative strategy for engineering 2D potential energy landscapes at small lengthscales is to use charge-tunable molecules and atomic-scale defects. These can provide sub-nanometer structural precision and uniquely flexible, gate-tunable behavior. With this in mind we have used F4TCNQ molecules as tunable charge centers to engineer the nanoscale energy landscape of graphene. By positioning F4TCNQ molecules into atomically-precise linear arrays we have observed Coulomb-driven charging patterns at the single-molecule level via scanning tunneling microscopy (STM), as well as new multi-impurity super-critical-like extended states. In addition to molecular charge centers, we also find that charge-tunable defects in insulating substrates can be used to manipulate the energy landscape of graphene. This is accomplished by controlling local defect ionization via the electric field of an STM tip. This new technique allows the patterning of gate-tunable graphene quantum dots whose electronic wavefunctions can be directly imaged.