β-Peptoids are peptidomimetics based on N-alkylated β-aminopropionic acid residues (or N-alkyl-β-alanines). This type of peptide mimic has previously been incorporated in biologically active ligands and has been hypothesized to be able to exhibit foldamer properties. Here we show, for the first time, that β-peptoids can be tuned to fold into stable helical structures. We provide high-resolution X-ray crystal structures of homomeric β-peptoid hexamers, which reveal right-handed helical conformations with exactly three residues per turn and a helical pitch of 9.6-9.8 Å between turns. The presence of folded conformations in solution is supported by circular dichroism spectroscopy showing length- and solvent dependency, and molecular dynamics simulations provide further support for a stabilized helical secondary structure in organic solvent. We thus outline a framework for future design of novel biomimetics that display functional groups with high accuracy in three dimensions, which has potential for development of new functional materials.