Land abandonment and subsequent re-vegetation have significantly modified the hydrological behavior of Mediterranean mountains. In order to assess the hydrological effect of such land-cover change, the streamflow response for a set of 26 flood events was compared in two neighboring catchments (central Spanish Pyrenees) with similar lithology and topography but different land cover: one (2.84 km$^2$) was extensively used for agriculture in the past, and the other (0.92 km$^2$) is covered by natural forest. Relative to the forested catchment, peak flows in the past agricultural catchment were always greater (566 vs. 119 l s$^{-1}$ km$^{-2}$), the response time was 2- to 3-fold faster (131 vs. 356 min), and the recession limbs were 1–2 orders of magnitude shorter (7 vs. 72 h). Storm flow differences were closely related to catchment wetness conditions and showed a marked seasonal pattern, with higher values in the past agricultural catchment under dry conditions, and usually higher values in the forested catchment under wet conditions. In the past agricultural catchment, runoff was generated during the entire hydrological year, through both surface and subsurface flow. We suggest that the forested catchment is characterized by a switching behavior controlled by soil moisture conditions, which regulates the hydrological connectivity within the forested and favor the release of large amounts of subsurface flow. Differences in soil depth and permeability, together with differences in vegetation cover, may explain the contrasting dominant runoff generation processes operating in each catchment, and consequently the differences between their hydrograph characteristics.