This study compares the climate variability and trend on the timescales ranging from interannual to centennial in the consolidated observational "reanalysis" datasets and simulated datasets from the recent phases of the Coupled Model Intercomparison Project (CMIP3/5). The analyses focus on the dynamic climate quantity of zonal-mean zonal wind and global atmospheric angular momentum (AAM). For the observations, validation of AAM by the length-of-day (LOD) and intercomparison of AAM reveals a good agreement among reanalyses on the interannual and decadal-to-interdecadal timescales, respectively. But the most significant discrepancies among them are in the long-term mean and long-term trends. For the simulations, the CMIP5 models produce a significantly smaller bias and a narrower ensemble spread of the climatology and trend in the 20th century for AAM compared to CMIP3, while CMIP3 and CMIP5 simulations consistently produce a positive trend for the 20th and 21st century. Results show that both CMIP3 and CMIP5 models have a wide range of variability for wind component of AAM (MR) compared to observations for magnitudes of decadal and interdecadal timescales. The ensemble means of CMIP3 and CMIP5 are not statistically distinguishable for either the 20th- or 21st-century runs. We perform in-house atmospheric general circulation model (AGCM) simulations forced by CMIP5 sea surface temperature (SST) as lower boundary conditions. The results indicate that the zonal wind and MR in the CMIP5 simulations are well simulated in the AGCM simulations. This confirms that SST is an important mediator in regulating the global atmospheric changes due to GHG effect.