Atmospheric aerosols exert a significant impact on Earth’s climate through their direct influences on radiative energy flow (direct effect or aerosol-radiation interaction) and modulations of cloud and precipitation characteristics (indirect effect or aerosol-cloud interaction). Overall, aerosols are considered to have a cooling effect on climate that can partially offset the greenhouse-gas-induced warming, but its magnitude is still largely uncertain in current climate models, particularly due to uncertainty of the indirect effect. Cloud microphysics, which primarily shapes the aerosol-cloud interaction, is among the uncertain processes in climate models and thus a primary source of uncertainty in estimates of the aerosol climate forcing. In an attempt to overcome this difficulty, the presenter and collaborators have developed a novel methodology for observationally diagnosing the cloud microphysical processes, exploiting recent progress in satellite measurement. The methodology is to combine observables obtained from multiple satellite sensors to construct the particular statistics that “fingerprint” the process. The statistics also serve as a diagnostic tool for evaluating global models in their representations of the process. The methodology is applied to multiple state-of-the-art global models, including traditional climate models and a global cloud-resolving model, to identify a key common bias of the “too fast, too frequent” rain formation in the models, which is further traced to uncertainty in parameterizations of water conversion process. Satellite-based information in the form of the statistics constructed provides a process-level constraint on the uncertainty that have been regarded as “tunable knobs” in climate models. The models thus constrained for the process representation, however, turned out to result in the aerosol indirect forcing that is too large negative to offset the greenhouse-gas-induced warming. This is caused by pronounced perturbations to cloud water budget, which appear to be amplified due to mutual interplay between aerosols and clouds. The apparent “dichotomy” identified between the process-based model constraint and the energy-balance requirement implies a fundamental gap in our understanding of linkage of the key processes to the global climate system. This talk highlights these recent findings to discuss possible ways for mitigating the dichotomy exposed in state-of-the-art global models.