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Formation and growth of nucleated particles into cloud condensation nuclei: Comparison of a global microphysics model with observations

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Aerosol nucleation occurs frequently in the atmosphere and is an important source of particle number. Observations suggest that nucleated particles are capable of growing to sufficiently large sizes that they act as cloud condensation nuclei (CCN), but some global models have reported that CCN concentrations are only modestly sensitive to large changes in nucleation rates. The connection between nucleation and CCN represents a key uncertainty in understanding the CCN budget and its implications for Earth's radiative balance. Here we present a novel approach for evaluating, in observations and models, the impact of single-day nucleation and growth events on the regional CCN budget. We also compare model-predicted nucleation rates, diameter growth rates, condensation and coagulation sinks, and survival probabilities to observations. This work uses the TwO-Moment Aerosol Sectional algorithm (TOMAS) hosted by the global chemical transport model GEOS-Chem to simulate nucleation events predicted by ternary (with a 10^{-5} tuning factor) or activation nucleation over one year. To evaluate model performance, we compare GEOS-Chem-TOMAS output in 30-minute intervals against a full year of size distribution datasets measured at the five following locations: Pittsburgh, Hyytiälä, Atlanta, St. Louis, and Po Valley. Results show that GEOS-Chem-TOMAS does not understate the importance of nucleation to CCN concentrations, as most metrics are within a 50% error in the model-measurement agreement and tend to be biased high. Median survival probabilities to 100 nm within one day for the model and measurements range from less than 1% to 9% across the five locations we considered. The strength of the coagulation sink and the infrequency of strong growth events (greater than 10 nm h^{-1}) are mainly responsible for these relatively low single-day survival probabilies. Additionally, both observations and models suggest that single-day nucleation and growth events contribute less than 5% to annually averaged CN100 concentrations (a proxy for CCN). Growth of nucleated particles on subsequent days means that nucleation makes a larger contribution, but this is difficult to constrain with available measurements. When a 50 nm CCN size limit is considered, particle survival probabilities for single-day events range from 25-50% across the different locations, and CN50 enhancements from nucleation are at least an order magnitude larger than CN100. This detailed exploration of new particle formation and growth dynamics adds support to the use of global models as tools for assessing the contribution of microphysical processes such as nucleation to the total number and CCN budget.