Aerosol nucleation and its role for clouds and Earth's radiative forcing in the aerosol-climate model ECHAM5-HAM

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Nucleation from the gas phase is an important source of aerosol particles in the Earth's atmosphere, contributing to the number of cloud condensation nuclei, which form cloud droplets. We have implemented in the aerosol-climate model ECHAM5-HAM a new scheme for neutral and charged nucleation of sulfuric acid and water based on laboratory data, and nucleation of an organic compound and sulfuric acid using a parametrization of cluster activation based on field measurements. We give details of the implementation, compare results with observations, and investigate the role of the individual aerosol nucleation mechanisms for clouds and the Earth's radiative forcing. The results of our simulations are most consistent with observations when neutral and charged nucleation of sulfuric acid proceed throughout the troposphere and nucleation due to cluster activation is limited to the forested boundary layer. The globally averaged annual mean contributions of the individual nucleation processes to total absorbed solar short-wave radiation via the direct, semi-direct, indirect cloud-albedo and cloud-lifetime effects in our simulations are −1.15 W/m2 for charged H2SO4/H2O nucleation, −0.235 W/m2 for cluster activation, and −0.05 W/m2 for neutral H2SO4/H2O nucleation. The overall effect of nucleation is −2.55 W/m2, which exceeds the sum of the individual terms due to feedbacks and interactions in the model. Aerosol nucleation contributes over the oceans with −2.18 W/m2 to total absorbed solar short-wave radiation, compared to −0.37 W/m2 over land. We explain the higher effect of aerosol nucleation on Earth's radiative forcing over the oceans with the larger area covered by ocean clouds, due to the larger contrast in albedo between clouds and the ocean surface compared to continents, and the larger susceptibility of pristine clouds owing to the saturation of effects. The large effect of charged nucleation in our simulations is not in contradiction with small effects seen in local measurements: over southern Finland, where cluster activation proceeds efficiently, we find that charged nucleation of sulfuric acid and water contributes on average less than 10% to ultrafine aerosol concentrations, in good agreement with observations.

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