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Sub-micrometer Aerosol Particles in the UT/LMS Region as Measured by CARIBIC and Modeled Using the MIT-CAM3 Global Climate Model

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In recent years, more and more aerosol microphysical processes have explicitly been incorporated in global atmospheric models. These developments make it possible to investigate in more detail the influence of different aerosol processes on clouds and climate. In particular, the influence of boundary layer particle formation on the number of available cloud condensation nuclei and related radiative forcing has been studied intensively in the last years. Deep convective clouds are considered to be the most important controllers of new particle formation in the upper troposphere/lowermost stratosphere (UT/LMS) region. However, the exact formation path and spatial and temporal extent of the formation events are not well known. Hence, model simulations of these processes are associated with large uncertainties. In the present study, we compare global distributions of sub-micrometer aerosol particle concentrations obtained from the CARIBIC project (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container) with global model simulations using the MIT-CAM3. The MIT-CAM3 is a state-of-the art global climate model with a two-moment interactive aerosol physics and chemistry module involving seven different aerosol compounds and mixtures. The main aim of the comparison is to learn more about what governs the formation as well as spatial and temporal distribution of the UT/LMS sub-micrometer aerosol. Furthermore, we investigate the performance of a state-of-the-art global climate model in terms of its representation of aerosol processes (and the interaction of aerosols with clouds) in the UT/LMS, which may indicate where more general model improvement and development is needed.

We find that the simulated global average nucleation mode particle number concentration (here particles with diameter between 4 and 12 nm, N_{4-12}) agrees fairly well with observations but the spatial correlation is poor. For the Aitken mode particles (particles with diameter larger than 12 nm, N_{12}), the model overestimates the median concentration but the geographical distribution is in rather good agreement with observations (correlation coefficients between 0.68 and 0.71). In general, the largest overestimate by the model appears to be located downwind the source regions and over South America. A clear seasonal variation of N_{12} is displayed in both the observations and in the model simulations, with overall higher concentrations in the northern hemisphere during the northern hemisphere summer. The seasonal variation in the N_{4-12} concentration is less conspicuous, but there is a tendency to higher summertime concentrations over Southeast Asia and the US.

High concentrations ($>5\times10^4$ particles/cm^{3}STP) of N_{4-12} particles are in the CARIBIC data found in a broad altitude interval between 310 and 370 K potential temperature. This is in agreement with previous studies and indicates that new particles are generated in deep convective outflows close to the tropopause and then transported into the UT/LMS region. The model simulations also display a maximum in the N_{4-12} number concentration around 340K, but the maximum value is substantially lower. The vertical extent of the maximum is also much narrower compared to the observations. This is most likely an indication of too little mixing and transport in the model around the tropopause. Comparing the observed and modeled vertical aerosol distributions, the pattern is similar for N_{12} as for N_{4-12}. A maximum in particle number concentration is found both in the model and in the observations around 340-350K, but the observations display high N_{12} values over a larger altitude range compared to the model. Again the modeled vertical gradient of the N_{12} concentration is steeper than in the observations, indicating again too weak mixing around the tropopause.