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Aerosol- and updraft-limited regimes of cloud droplet formation: influence of particle number, size and hygroscopicity on the activation of cloud condensation nuclei (CCN)

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Abstract. We have investigated the formation of cloud droplets under pyro-convective conditions using a cloud parcel model with detailed spectral microphysics and with the κ-Köhler model approach for efficient and realistic description of the cloud condensation nucleus (CCN) activity of aerosol particles. Assuming a typical biomass burning aerosol size distribution (accumulation mode centred at 120 nm), we have calculated initial cloud droplet number concentrations (N_{CD}) for a wide range of updraft velocities (w=0.25–20 m s^{-1}) and aerosol particle number concentrations (N_{CN}=200–10^5 cm^{-3}) at the cloud base. Depending on the ratio between updraft velocity and particle number concentration (w/N_{CN}), we found three distinctly different regimes of CCN activation and cloud droplet formation:

(1) An aerosol-limited regime that is characterized by high w/N_{CN} ratios (>≈10^{-3} m s^{-1} cm^3), high maximum values of water vapour supersaturation (S_{max}>≈0.5%), and high activated fractions of aerosol particles (N_{CN}/N_{CN}>≈90%). In this regime N_{CD} is directly proportional to N_{CN} and practically independent of w.

(2) An updraft-limited regime that is characterized by low w/N_{CN} ratios (<≈10^{-4} m s^{-1} cm^3), low maximum values of water vapour supersaturation (S_{max}<≈0.2%), and low activated fractions of aerosol particles (N_{CD}/N_{CN}<≈20%). In this regime N_{CD} is directly proportional to w and practically independent of N_{CN}.

(3) An aerosol- and updraft-sensitive regime (transitional regime), which is characterized by parameter values in between the two other regimes and covers most of the conditions relevant for pyro-convection. In this regime N_{CD} depends non-linearly on both N_{CN} and w.

In sensitivity studies we have tested the influence of aerosol particle size distribution and hygroscopicity on N_{CD}. Within the range of effective hygroscopicity parameters that is characteristic for continental atmospheric

aerosols ($\kappa \approx 0.05$–0.6), we found that N_{CD} depends rather weakly on the actual value of κ. A compensation of changes in κ and S_{max} leads to an effective buffering of N_{CD}. Only for aerosols with very low hygroscopicity ($\kappa < 0.05$) and also in the updraft-limited regime for aerosols with higher than average hygroscopicity ($\kappa > 0.3$) did the relative sensitivities $\partial \ln N_{CD}/\partial \ln \kappa \approx (\Delta N_{CD}/N_{CD})/(\Delta \kappa/\kappa)$ exceed values of ~0.2, indicating that a 50% difference in κ would change N_{CD} by more than 10%.

The influence of changing size distribution parameters was stronger than that of particle hygroscopicity. Nevertheless, similar regimes of CCN activation were observed in simulations with varying types of size distributions (polluted and pristine continental and marine aerosols with different proportions of nucleation, Aitken, accumulation, and coarse mode particles). In general, the different regimes can be discriminated with regard to the relative sensitivities of N_{CD} against w and N_{CN} ($\partial \ln N_{CD}/\partial \ln w$ and $\partial \ln N_{CD}/\partial \ln N_{CN}$). We propose to separate the different regimes by relative sensitivity ratios, $(\partial \ln N_{CD}/\partial \ln w)/(\partial \ln N_{CD}/\partial \ln N_{CN})$ of 4:1 and 1:4, respectively.

The results of this and related studies suggest that the variability of initial cloud droplet number concentration in convective clouds is mostly dominated by the variability of updraft velocity and aerosol particle number concentration in the accumulation and Aitken mode. Coarse mode particles and the variability of particle composition and hygroscopicity appear to play major roles only at low supersaturation in the updraft-limited regime of CCN activation ($S_{max} < 0.2\%$).

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