

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

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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 at 120 nm), we have calculated initial cloud droplet number concentrations (N_{CD}) for a wide range of updraft velocities ($w = 0.5\text{--}20\text{ m s}^{-1}$) and aerosol particle number concentrations ($N_{CN} = 10^3 - 10^5\text{ cm}^{-3}$) 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 ($> \approx 10^{-3}\text{ m s}^{-1}/(\text{cm}^{-3})$), high maximum values of water vapour supersaturation ($S_{\max} > \approx 0.5\%$), and high activated fractions of aerosol particles ($N_{CD}/N_{CN} > \approx 90\%$). 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 ($< \approx 10^{-4}\text{ m s}^{-1}/(\text{cm}^{-3})$), low maximum values of water vapour supersaturation ($S_{\max} < \approx 0.2\%$), and low activated fractions of aerosol particles ($N_{CD}/N_{CN} < \approx 20\%$). In this regime N_{CD} is directly proportional to w and practically independent of N_{CN} .

(3) An aerosol- and updraft-sensitive 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 hygroscopicity on N_{CD} . Within the range of effective hygroscopicity parameters that is characteristic for continental atmospheric aerosols ($\kappa \approx 0.05\text{--}0.6$), we found that N_{CD} depends rather weakly on the actual value of κ . Only for aerosols with very low hygroscopicity ($\kappa < 0.05$) and in the updraft-limited regime also for aerosols with higher than average hygroscopicity (κ

> 0.3) did the relative differential quotients $(\Delta N_{CD}/N_{CD})/(\Delta \kappa/\kappa)$ exceed values of ~ 0.2 , indicating that a 50% difference in κ would change N_{CD} by more than 10%. Realistic changes in the aerosol particle size distribution had practically no effect on the aerosol-limited regime and limited influence on the aerosol- and updraft sensitive regime ($\Delta N_{CD}/N_{CD} < 30\%$) but can have strong effects at low supersaturation in the updraft-limited regime ($\Delta N_{CD}/N_{CD} > 30\%$ at $S_{\max} < 0.1\%$).

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

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