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Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity

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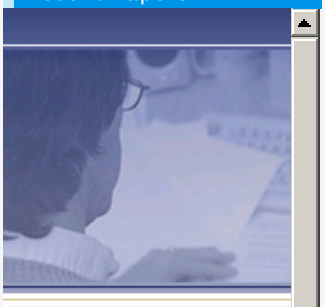
Abstract. Atmospheric aerosol particles serving as cloud condensation nuclei (CCN) are key elements of the hydrological cycle and climate. We have measured and characterized CCN at water vapor supersaturations in the range of S=0.10–0.82% in pristine tropical rainforest air during the AMAZE-08 campaign in central Amazonia.

The effective hygroscopicity parameters describing the influence of chemical composition on the CCN activity of aerosol particles varied in the range of $\kappa \approx 0.1$–0.4 (0.16 ± 0.06 arithmetic mean and standard deviation). The overall median value of $\kappa \approx 0.15$ was by a factor of two lower than the values typically observed for continental aerosols in other regions of the world. Aitken mode particles were less hygroscopic than accumulation mode particles ($\kappa \approx 0.1$ at $D \approx 50$ nm; $\kappa \approx 0.2$ at $D \approx 200$ nm), which is in agreement with earlier hygroscopicity tandem differential mobility analyzer (H-TDMA) studies.

The CCN measurement results are consistent with aerosol mass spectrometry (AMS) data, showing that the organic mass fraction (f_{org}) was on average as high as ~90% in the Aitken mode ($D \leq 100$ nm) and decreased with increasing particle diameter in the accumulation mode (~80% at $D \approx 200$ nm). The κ values exhibited a negative linear correlation with f_{org} (R^2=0.81), and extrapolation yielded the following effective hygroscopicity parameters for organic and inorganic particle components: $\kappa_{org} \approx 0.1$ which can be regarded as the effective hygroscopicity of biogenic secondary organic aerosol (SOA) and $\kappa_{inorg} \approx 0.6$ which is characteristic for ammonium sulfate and related salts. Both the size dependence and the temporal variability of effective particle hygroscopicity could be parameterized as a function of AMS-based organic and inorganic mass fractions ($\kappa_p = \kappa_{org} \times f_{org} + \kappa_{inorg} \times f_{inorg}$). The CCN number concentrations predicted with κ_p were in fair agreement with the measurement results (~20% average deviation). The median CCN number concentrations at



S=0.1–0.82% ranged from $N_{CCN,0.10}$≈35 cm^{-3} to $N_{CCN,0.82}$≈160 cm^{-3}, the median concentration of aerosol particles larger than 30 nm was $N_{CN,30}$≈200 cm^{-3}, and the corresponding integral CCN efficiencies were in the range of $N_{CCN,0.10}/N_{CN,30}$≈0.1 to $N_{CCN,0.82}/N_{CN,30}$≈0.8.

Although the number concentrations and hygroscopicity parameters were much lower in pristine rainforest air, the integral CCN efficiencies observed were similar to those in highly polluted megacity air. Moreover, model calculations of $N_{CCN,S}$ assuming an approximate global average value of κ≈0.3 for continental aerosols led to systematic overpredictions, but the average deviations exceeded ~50% only at low water vapor supersaturation (0.1%) and low particle number concentrations (≤100 cm^{-3}). Model calculations assuming a constant aerosol size distribution led to higher average deviations at all investigated levels of supersaturation: ~60% for the campaign average distribution and ~1600% for a generic remote continental size distribution. These findings confirm earlier studies suggesting that aerosol particle number and size are the major predictors for the variability of the CCN concentration in continental boundary layer air, followed by particle composition and hygroscopicity as relatively minor modulators.

Depending on the required and applicable level of detail, the information and parameterizations presented in this paper should enable efficient description of the CCN properties of pristine tropical rainforest aerosols of Amazonia in detailed process models as well as in large-scale atmospheric and climate models.

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