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Cloud condensation nuclei in pristine tropical rainforest air of Amazonia:

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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 κ = 0.05-0.45. The overall median value of $\kappa \approx 0.15$ was only half of the value 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).

The CCN measurement results were fully consistent with aerosol mass spectrometry (AMS) data, which showed that the organic mass fraction ($X_{m,org}$) 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 close linear correlation with $X_{m,org}$ and extrapolation yielded the following effective hygroscopicity parameters for organic and inorganic particle components: $\kappa_{org} \approx 0.1$ which is consistent with laboratory measurements of secondary organic aerosols 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 ($\kappa_p = 0.1\ X_{m,org} + 0.6\, X_{m,inorg}$), and the CCN number concentrations predicted with κ_p were in fair agreement with the measurement results. The median CCN number concentrations at S = 0.1-0.82% ranged from $N_{CCN,0.10} \approx 30$ cm^{-3}to $N_{CCN,0.82} \approx 150$ cm^{-3}, the median concentration of aerosol particles larger than 30 nm was $N_{CN,30} \approx 180$ cm^{-3}, and the corresponding integral CCN efficiencies were in the range of $N_{CCN,0.10}/N_{CN,30} \approx 0.1$ to $N_{CCN,0.82}/N_{CN,30} \approx 0.8$.

Although the number concentrations and hygroscopicity parameters were much lower, the integral CCN efficiencies observed in pristine rainforest air were similar to those in highly polluted mega-city air. Moreover, model calculations of $N_{CCN,S}$ with a global average value of κ = 0.3 led to systematic overpredictions, but the relative deviations exceeded ~50% only at low water vapor supersaturation (0.1%) and low particle number concentrations (≤ 100 cm^{-3}). 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 in detailed process models as well as in large-scale atmospheric and climate models.

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