

Exploring the effect of deep-convective cloud systems on particle mass and number: source or sink?

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Incomplete knowledge of aerosol-cloud interactions contributes significant uncertainty to the understanding of the Earth's climate sensitivity, and thus to the large-scale theoretical models used to predict future climate properties. The total number, size, and composition of an aerosol population entrained into a cloud system can affect cloud physical features (e.g. updraft velocity or precipitation rate) while cloud properties can affect features of the aerosol population (e.g. through wet scavenging efficiency or aqueous-phase chemical processing) (Yin *et al.*, 2005; Ekman *et al.*, 2006). This tightly-coupled system exhibits complicated feedbacks that are hard to discern in a theoretical model without faithfully representing the detailed transport, microphysical and chemical processes taking place. We present a new three-dimensional cloud-resolving model, CRM-ORG, which accounts for the processing of inorganic and organic aerosol components in a fully-coupled, dynamic simulation. We apply this model to a case study of a deep convective cloud under clean conditions above the tropical Amazon, and use it to elucidate the net influence of the cloud system on aerosol mass, number, and composition throughout the atmospheric column.

Following the Volatility Basis Set approach of Donahue *et al.* (2006), the CRM-ORG model incorporates a suite of lumped species to describe the oxidation products of isoprene and monoterpene compounds across a spectrum of volatility. The mass transfer of these species to/from particles and cloud drops is treated dynamically along with explicit representation of aqueous-phase chemical reactions relevant to the production of low-volatility organic compounds that partition significantly to the particulate phase after cloud drop evaporation. The model also accounts for particle nucleation and growth due to condensation of inorganic as well as organic vapors, and this work probes the sensitivity of the model's results to both the formulation of the nucleation mechanism and the volatility of condensable organic vapors.

The detailed considerations of organic aerosol formation and growth are implemented into the three-dimensional cloud-resolving model described by Wang *et al.* (1993) and Ekman *et al.* (2006). This elastic, non-hydrostatic model solves prognostic equations for momentum, air mass density, ice-liquid potential temperature, water vapor and four hydrometeor types. The model then considers chemical species in the gas, particulate and aqueous phases in addition to species adsorbed onto solid water particles. The sulfate and

organic aerosol species are represented with three internally-mixed modes (one each in the nucleation, Aitken and accumulation size ranges). Important aerosol microphysical processes, such as coagulation and condensation, are considered in the model.

We compare the predicted aerosol and cloud properties from CRM-ORG to airborne measurements made during actual deep convective events in the Amazon. These observational constraints will give us confidence in the ability of the model to faithfully represent the complex effects of clouds on aerosol mass and number in the tropical environment. In order to gain a more general understanding of these interactions, we report the net effect of cloud processing on aerosol mass and number at various altitudes throughout the column. Of particular focus is the generation of new particles in the relatively clean background environment of the upper troposphere where substantial amounts of trace gases and particles are injected within the cloud outflow.

Several other models have explored the effects and feedbacks between organic aerosols and clouds in adiabatic parcel or two-dimensional models (Ervens *et al.*, 2005; Yin *et al.*, 2005). These studies opted to consider more detailed representations of chemistry or spectrally-resolved microphysics while sacrificing an atmospherically relevant description of transport. The configuration of CRM-ORG, on the other hand, achieves a realistic description of the spatial complexity of a deep convective system, while maintaining enough chemical and microphysical detail to explore aerosol-cloud interactions in depth. We compare our model's results and sensitivities to those of past studies to highlight the influence of model configuration.

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