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Ice supersaturations exceeding 100% at the cold tropical tropopause: implications for cirrus formation and dehydration

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Abstract. Recent in situ measurements at tropical tropopause temperatures as low as 187 K indicate supersaturations with respect to ice exceeding 100% with little or no ice present. In contrast, models used to simulate cloud formation near the tropopause assume a supersaturation threshold for ice nucleation of about 65% based on laboratory measurements of aqueous aerosol freezing. The high supersaturations reported here, along with cloud simulations assuming a plausible range of temperature histories in the sampled air mass, indicate that the vast majority of aerosols in the air sampled on this flight must have had supersaturation thresholds for ice nucleation exceeding 100% (i.e. near liquid water saturation at these temperatures). Possible explanations for this high threshold are that (1) the expressions used for calculating vapor pressure over supercooled water at low temperatures give values are at least 20% too low, (2) organic films on the aerosol surfaces reduce their accommodation coefficient for uptake of water, resulting in aerosols with more concentrated solutions when moderate-rapid cooling occurs and correspondingly inhibited homogeneous freezing, and (3) if surface freezing dominates, organic coatings may increase the surface energy of the ice embryo/vapor interface resulting in suppressed ice nucleation. Simulations of in situ cloud formation in the tropical tropopause layer (TTL) throughout the tropics indicate that if decreased accommodation coefficients and resulting high thresholds for ice nucleation prevailed throughout the tropics, then the calculated occurrence frequency and areal coverage of TTL cirrus would be significantly suppressed. However, the simulations also show that even if in situ TTL cirrus form only over a very small fraction of the tropics in the western Pacific, enough air passes through them due to rapid horizontal transport such that they can still effectively freeze-dry air entering the stratosphere. The TTL cirrus simulations show that even if very large supersaturations are required for

ice nucleation, these large supersaturations should occur very rarely.

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