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Ephemeral carbon dioxide ice clouds in the upper mesosphere of Venus

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Observations show that the temperature above 100 km in Venus' atmosphere intermittently falls below 100 K, when both H₂O and CO₂ become supersaturated. Profiles show temperatures can fall below 60 K at heights between 115-125 km, presumably as a result of large amplitude gravity waves. Cosmic dust particles entering the atmosphere are predicted to ablate between 110 and 125 km. This provides a source of metallic vapours (principally Mg and Fe atoms), which then form metal carbonate molecules known as meteoric smoke particles (MSPs). Because these molecules are highly polar, they are excellent nuclei for CO₂- and H₂O-ice particle formation.

In this study we examine the feasibility and kinetics of CO₂-ice cloud formation, using both classical nucleation theory (CNT) and bottom-up kinetic nucleation theory (KNT). For CNT, a dimensionless non-isothermal coefficient is included to reduce the nucleation rate of CO₂ ice particles, since the atmospheric concentration of the nucleating species (CO₂) comprises a significant fraction of the total atmosphere. For heterogeneous CNT on MSPs, a surface diffusion approach is used where molecules can diffuse on the surface to form a critical cluster for nucleation and the effect of dissipation of critical clusters is accounted for. Application of CNT shows that whereas homogeneous nucleation should be too slow for significant cloud formation, heterogeneous nucleation rates around 1 cm⁻³ s⁻¹ for CO₂ ice should be possible in the colder regions (< 80 K).

For KNT, the rate coefficients for the sequential addition of CO₂ molecules up to MgCO₃(CO₂)₄₀ were calculated explicitly with Rice Ramsperger Kassel Markus (RRKM) theory, using a solution of the Master Equation based on the inverse Laplace transform method. The rates of dissociation of the clusters i.e. MgCO₃(CO₂)_{n+1} → MgCO₃(CO₂)_n + CO₂, were calculated by detailed balance. In order to explore the evolution of the CO₂-ice clouds, a 1-dimensional model was constructed to describe the nucleation, growth, sedimentation and sublimation of the ice particles. The model is initiated with a vertical profile of atmospheric density and temperature determined using the Solar Occultation in the InfraRed (SOIR) instrument on a specified orbit of Venus Express, and then follows the fate of an MSP seed particle as it grows, sediments and finally sublimates on entering a warmer region. Two categories of cloud tend to be produced from the observed temperature profiles. The first peaks around 120 km with particles around 100-200 nm radius; and the second type persists for longer and peaks around 110 km, with particles that can exceed 2 μm in radius.

Most clouds are predicted to occur at high latitudes ($>70^\circ$). Using a probable underestimate of the MSP concentration (100 cm^{-3}), the optical extinction of these clouds at 220 nm should be readily observable by the SOIR instrument. However, the clouds are short-lived because of rapid sedimentation (typically 300 s, the longest-lived around 1200 s), so that the detection of these ephemeral “hail showers” will be challenging.