

博士論文

Origins and life cycle of Venus' clouds investigated
with a high-accuracy cloud microphysics model

〔高精度雲微物理モデルを用いた金星雲の起源と生成消滅過程の研究〕

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ORIGINS AND LIFE CYCLE OF VENUS' CLOUDS INVESTIGATED WITH A HIGH-ACCURACY CLOUD MICROPHYSICS MODEL

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Résumé

Les nuages sont une composante omniprésente et essentielle des atmosphères planétaires, ayant un impact substantiel sur le transfert radiatif, la chimie et l'évolution. Ces effets dépendent de la densité numérique, de la taille, de la composition et de la distribution des nuages. Ainsi, la compréhension de la formation des nuages et de ce qui détermine ces propriétés physiques est vitale pour comprendre les processus atmosphériques planétaires. Vénus est globalement couverte d'épais nuages d'acide sulfurique et constitue un exemple frappant de planète dont le climat est fortement couplé aux processus nuageux. Reproduire avec précision la structure nuageuse observée est nécessaire pour étudier ces processus. Plus important encore, la compréhension du mécanisme de formation des nuages de Vénus nécessite de comprendre comment les noyaux de condensation sont fournis et évoluent à travers les processus microphysiques.

L'objectif principal de cette thèse est de comprendre l'origine et le cycle de vie des nuages de Vénus. Pour ce faire, nous contraignons d'abord un profil de diffusion turbulente — un paramètre critique pour le transport de matière — afin de simuler avec précision la structure nuageuse observée et la distribution des espèces condensables. Ensuite, nous développons un nouveau modèle de microphysique des nuages qui inclut des processus clés cruciaux pour l'origine des nuages de Vénus. À l'aide de ce modèle, nous testons notre hypothèse selon laquelle la poussière cosmique est la source des particules de brume inférieures. Cette brume joue un rôle central dans la formation des nuages, mais son origine reste inexpliquée. Enfin, nous étudions le bilan des noyaux de condensation sur Vénus pour en comprendre l'origine.

Nous avons contraint le profil du coefficient de diffusion turbulente en réalisant des études de cas avec un modèle de microphysique des nuages 1-D, visant à reproduire simultanément la structure nuageuse observée et les distributions de vapeurs condensables sur Vénus.¹ Ces études ont utilisé quatre profils de diffusion turbulente basés sur des estimations observationnelles antérieures. Le coefficient le mieux ajusté est d'environ $\sim 2 \text{ m}^2 \text{ s}^{-1}$ entre 60 et 70 km et d'environ $\sim 360 \text{ m}^2 \text{ s}^{-1}$ au-dessus de 85 km. Nous avons également réalisé des études de cas en utilisant différents profils de température de Pioneer Venus et de Venus Express. Cette dernière expérience a révélé qu'en raison du transport des particules et de l'évaporation qui en résulte, les abondances d'eau et d'acide sulfurique varient respectivement d'un facteur de deux et de plusieurs ordres de grandeur.

Nous avons développé un nouveau modèle de microphysique des nuages 1-D, le “Simulator of Particle Evolution, Composition, and Kinetics” (SPECK), qui intègre un schéma de bins à centre mobile pour minimiser la diffusion numérique. SPECK peut gérer n'importe quel nombre de dis-

tributions de taille avec de multiples composants, permettant des applications polyvalentes pour des systèmes nuageux complexes. Les simulations 0D ont démontré que ce cadre modélise de manière fiable la microphysique des nuages dans les conditions vénusiennes, en particulier les processus de condensation et d'évaporation. Nous avons en outre validé SPECK par rapport aux modèles microphysiques récents de Vénus dans des simulations 1-D. Le schéma à centre mobile présente moins de diffusion numérique que les modèles existants basés sur des schémas de bins entièrement stationnaires, permettant des calculs microphysiques plus précis. De plus, SPECK reproduit la structure nuageuse observée par la Grande Sonde de Pioneer Venus en utilisant les mêmes paramètres de calcul que la dernière étude de modèle microphysique.

En incorporant l'afflux de poussière cosmique dans SPECK, nous montrons qu'il est suffisant pour maintenir la couche de brume inférieure avec la distribution de taille des particules observée par les sondes d'entrée. Ces particules de brume cosmique agissent comme des noyaux de condensation efficaces, favorisant la formation de nuages dans la couche nuageuse principale, loin de leur source. De plus, ces particules fournissent une source plausible pour l'absorbant ultraviolet de la planète, longtemps non identifié. Collectivement, nos découvertes établissent la poussière cosmique comme une composante essentielle du climat de Vénus. Ces perspectives sont également cruciales pour comprendre les climats d'autres planètes à atmosphère épaisse, telles que les géants gazeux et les exoplanètes, où l'accumulation de poussière cosmique pourrait affecter de manière similaire la structure et la composition des nuages.

Pour comprendre l'origine des nuages de Vénus, nous simulons les processus de nucléation en incorporant trois sources principales de noyaux dans SPECK : la vapeur d'allotropes de soufre, la vapeur d'acide sulfurique et les particules de brume inférieures. Les taux de production de noyaux, généralement prédéfinis dans les modèles existants, sont maintenant calculés sur la base de la théorie de la nucléation homogène. Les simulations montrent que des noyaux suffisants se forment par ce processus pour reproduire la structure nuageuse observée. Nous avons constaté qu'environ $\sim 80\%$ des nuages simulés se forment via une nucléation hétérogène sur des particules d'allotropes de soufre, que la brume inférieure contribue à environ $\sim 20\%$, et que les noyaux d'acide sulfurique apportent une contribution minimale. Sans allotropes de soufre, la quantité de nuages diminue de moitié et la structure simulée s'écarte considérablement des observations, soulignant son importance primordiale dans la formation des nuages. De plus, nos résultats suggèrent que des particules d'allotropes de soufre peuvent se former dans la couche nuageuse, expliquant potentiellement les particules cristallines observées par la Grande Sonde de Pioneer Venus.

Abstract

Clouds are a fundamental component of planetary atmospheres, exerting a profound influence on radiative transfer, chemical cycles, and long-term atmospheric evolution. The physical impact of these clouds is determined by their microphysical properties—including number density, size distribution, and chemical composition—which are themselves governed by the underlying processes of nucleation, condensation, and transport. As a planet globally shrouded in thick sulfuric acid (H_2SO_4) clouds, Venus represents a prominent example for studying atmospheres where the climate is deeply coupled with cloud processes. Unlike the patchy clouds of Earth or Mars, Venus' global cloud system is sustained by continuous photochemical production, a regime common in the Solar System and beyond. Understanding how these clouds originate and evolve requires a first-principles modeling approach that accurately captures the transition from gas-phase chemistry to aerosol formation.

The primary objective of this thesis is to elucidate the origin and life cycle of the Venusian cloud system by addressing critical gaps in current microphysical models. While decades of in-situ and remote sensing observations—from Venera to Akatsuki—have characterized the complex size distribution of the clouds, existing numerical models often struggle to reconcile these structures with the observed distribution of condensable vapors due to unrealistic eddy diffusion profiles. Furthermore, while current models can reproduce the observed cloud structure by utilizing artificial parameterizations, the fundamental origin of the clouds remains poorly understood. This is largely due to the omission of elementary particle formation processes, such as the binary homogeneous nucleation of H_2SO_4 and water (H_2O) or the formation of sulfur allotrope (S_8) nuclei. Additionally, the lack of a self-consistent framework for the lower haze has forced models to rely on artificial boundary conditions, leaving the physical origin of these particles unexplained.

To address these issues, we first resolve inconsistencies in vertical transport by conducting a parameter study to constrain a realistic eddy diffusion profile, thereby reconciling the simulated distribution of cloud particles and condensable vapors (H_2SO_4 and H_2O) with recent observations. Second, we develop a novel multi-component cloud microphysics model that employs a high-accuracy condensation scheme to simulate the interactions of diverse chemical species. Using this framework, we test the hypothesis that cosmic dust ablation, rather than sulfur allotropes, serves as the primary source of the enigmatic lower haze layer. Finally, we investigate the global cloud nuclei budget to determine the relative contributions of binary H_2SO_4 - H_2O nucleation, sulfur allotrope nucleation, and cosmic dust, providing a comprehensive account of the mechanisms that initiate and sustain the clouds of Venus.

In Chapter 2, to constrain the eddy diffusion coefficient, we simultaneously simulated Venus' cloud and condensable species distributions with a 1-D cloud microphysics model. We extended the model's domain upward from the previous 70 km to the current 100 km to explicitly simulate the upper haze and condensable species for the first time. This model simulates the cloud particle size distribution across 23 discrete size bins, from submicron condensation nuclei (CN) to micron-sized $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ droplets. The CN production rate is fixed and is therefore treated in the same manner as in previous work. To constrain the eddy diffusion coefficient, we ran a series of case studies using four patterns of eddy diffusion profile compiled from various past observations. The temperature profile is set to the Venus International Reference Atmosphere (VIRA), which was established based on observations from the Pioneer Venus probe. Our simulations showed that the vertical extent of the upper haze layer is highly sensitive to the eddy diffusion coefficient above 85 km. Using a large eddy diffusion coefficient of $\sim 360 \text{ m}^2 \text{ s}^{-1}$ adapted from Mahieux et al. (2021), our model successfully reproduces the extinction coefficient of the haze layer observed by Venus Express (VEx). This demonstrates that efficient vertical mixing is required to transport particles upward from the main cloud deck to sustain this haze layer. The vertical profile of water vapor significantly depends on the eddy diffusion coefficient in the upper cloud layer. Our model replicates the observed decrease by reducing the eddy diffusion coefficient in this specific layer to $2 \text{ m}^2 \text{ s}^{-1}$. Based on the eddy diffusion case studies, we narrowed the best-fit eddy diffusion profile to $\sim 2 \text{ m}^2 \text{ s}^{-1}$ between 60 and 70 km, and $\sim 360 \text{ m}^2 \text{ s}^{-1}$ above 85 km.

We then investigated the impact of the mesospheric temperature on the cloud structure by combining our best-fit eddy diffusion profile with the temperature profile recently observed by Solar Occultation at Infrared (SOIR) onboard VEx. The SOIR temperature profile includes a recently discovered warm layer in the mesosphere, which is $\sim 50 \text{ K}$ higher than the VIRA. The warm layer above 85 km causes the evaporation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ aerosol particles transported upward by the strong eddy diffusion. This evaporation released both H_2SO_4 and H_2O back into the gas phase at high altitudes. This mechanism reproduced the observed increase in H_2O volume mixing ratio (VMR) above 85 km, as observed by the VEx. This evaporation also increased the gaseous H_2SO_4 VMR to levels consistent with observational upper limits but not enough to explain the observed SO_2 inversion layer in the mesosphere. This work demonstrates that the abundance of water in Venus's upper atmosphere, which ultimately controls the planet's hydrogen escape rate and long-term evolution, is fundamentally regulated by the interplay of cloud microphysics and dynamical transport occurring within the cloud and haze layers.

In Chapter 3, we developed a new 1-D cloud microphysics model for Venus, named Simulator of Particle Evolution, Composition, and Kinetics (SPECK). Our model simulates condensational growth by implementing the moving-center bin scheme to reduce numerical diffusion. In this approach, while the bin boundaries remain fixed, the actual particle size within each bin is tracked precisely. This allows us to calculate condensational growth as a Lagrangian advection, effectively eliminating the numerical diffusion and providing a much more accurate solution. Our model framework also includes a robust, volume-conserving scheme for coagulation and compositional mixing, allowing us to track complex interactions between multiple particle distributions. Before applying SPECK to Venus, we conducted a series of benchmark tests to rigorously validate our algorithms. First, we

ran 0-D simulations focusing purely on condensation and evaporation. We compared our moving-center results directly against the exact solution provided by a full-moving scheme. Our model shows good agreement with the exact solution, with errors less than 15% and correctly capturing the Kelvin effect, where smaller particles evaporate while larger ones grow. We then increased the complexity of 0-D tests by including both growth and coagulation. These tests confirmed that our model correctly simulates the interactions between our different particle types, such as the scavenging of small CN by larger droplets to form mixed particles.

We further validated SPECK through 1-D benchmark simulations. We first compared SPECK against a traditional full-stationary model developed in Chapter 2 across various bin resolutions. The full-stationary model produced an artificially broad size distribution due to numerical diffusion, and its results only converged on the correct solution at an extremely high, computationally expensive resolution. In contrast, SPECK’s results were accurate and consistent even at low resolutions, matching the high-resolution solution. This demonstrates that our model is both more accurate and more computationally efficient than standard methods. Finally, we performed a realistic 1-D simulation of the Venus atmosphere, using the same settings as the state-of-the-art Venus cloud microphysics model based on Community Aerosol and Radiation Model for Atmospheres (CARMA). Our model successfully reproduced the complex, multi-layered cloud structure observed at Venus, showing good agreement with both the CARMA model results and in-situ observations conducted by the Pioneer Venus Large Probe. Most significantly, we found that running SPECK at a low bin resolution (half that of the nominal case) produced nearly identical results while running six times faster. This proves that the moving-center scheme is a robust and efficient method for modeling Venus’ clouds, and it represents a very promising pathway for implementing full microphysics in future 3-D modeling.

In Chapter 4, we used SPECK to investigate the origin of the persistent lower haze on Venus. In this and the subsequent chapter (Chapter 5), the initial stage of cloud formation is simulated using nucleation theory—the most fundamental approach to study the cloud origin and a substantial advancement over previous studies. To achieve this, we extended SPECK’s capabilities to create a fully self-consistent cloud model, incorporating not only H_2SO_4 and H_2O , but also the microphysics of S_8 . Furthermore, we included an influx of meteoric smoke particles (MSPs) originating from cosmic dust ablation, which we introduced as 1 nm olivine particles at the model’s 100 km upper boundary. Our simulations revealed that the lower haze layer, observed below the main cloud deck, forms only when we include the influx of MSPs. Previous models have long assumed this haze was composed of photochemically produced sulfur or unknown involatile material, while our results show that solid sulfur is not stable in this region and does not form a haze. We propose a complete particle cycle involving MSPs. The MSPs entering at high altitudes are scavenged by sulfuric acid aerosols and sediment downward. Upon reaching the hot cloud base, the sulfuric acid evaporates, leaving behind the involatile meteoric material. This residual smoke then coagulates and accumulates, forming the persistent lower haze layer. We found that this mechanism produces the lower haze characteristics consistent with previous probe observations. The final size of the lower haze particles in our model is directly controlled by the mass influx of MSPs. When we compared our results to in-situ measurements from the Pioneer Venus and Venera probes, we found that the observed haze particle radii (0.08 to 0.21 μm) are reproduced by an MSP influx between 3 and 300 tons per day. This range encom-

passes realistic influx estimates for terrestrial planets between 5 and 60 tons per day, providing strong evidence for this new mechanism. Furthermore, we found that this haze layer acts as an efficient CN, seeding the main cloud deck and increasing its mass, which brings our model into better agreement with observations.

Our findings also have important implications for Venus' atmospheric phenomena in general. First, we have identified a viable physical source for Venus's mysterious unknown UV absorber. Iron compounds in H_2SO_4 droplets have been experimentally shown to match the Venus' UV absorption feature. The MSPs deliver metal species, including iron, to the atmosphere, which accumulates in the lower haze. These iron-bearing haze particles can be transported upward by eddy diffusion, where they are incorporated into the cloud droplets and form the UV-absorbing compounds. Second, this finding establishes that MSPs are a critical, previously overlooked component of the Venus climate system, influencing both the planet's greenhouse effect by seeding the main cloud and its energy balance by supplying the UV absorber that drives atmospheric super-rotation. The same mechanism is expected to operate in the atmospheres of outer planets, such as Jupiter, since MSPs can be deposited and accumulate in the hot lower atmosphere.

In Chapter 5, we investigated whether sufficient nuclei form through homogeneous nucleation and the contribution of each different nucleus type to cloud formation. We identified three primary sources: (1) H_2SO_4 - H_2O particles forming directly from binary homogeneous nucleation, (2) S_8 particles forming from homogeneous nucleation, and (3) pre-existing lower haze particles (which Chapter 4 showed originate from cosmic dust) being transported upward. To test these ideas, we used SPECK and self-consistently simulate all three nucleation pathways simultaneously. We established four distinct simulation cases to isolate the contribution of each nucleus source. First, we ran a simulation that included all three processes. We then ran three sensitivity tests where we systematically turned off H_2SO_4 homogeneous nucleation, S_8 vapor (and its nucleation), and the lower haze boundary source, each at a time. The nominal model accurately reproduced the complete, multi-layered Venus cloud structure, from the lower haze all the way to the upper haze. Our simulated vertical profiles for H_2SO_4 and H_2O vapor, the total cloud mass loading, the particle effective radius, and the total cloud optical thickness (our calculated value of 32.3 falls squarely within the observed range of 25-40) were all in good agreement with previous observational data. This confirmed that our model's physics were sound and capable of simulating the Venus cloud.

We estimated the relative contribution of each nuclei to cloud formation by comparing the sensitivity study results. We found that turning off H_2SO_4 homogeneous nucleation had a negligible effect, changing the total cloud optical thickness by only 1%. This process is not a significant contributor to the cloud thickness. Removing the lower haze caused the lower and middle cloud layers to lose about half their mass, resulting in a 17% decrease in the total optical thickness. The most significant contribution came from removing S_8 vapor. The total optical thickness decreased by 44%, far below any observed values. Based on these findings, we analyzed the hierarchy among different sources of cloud nuclei. We conclude that the homogeneous nucleation of S_8 vapor in the upper atmosphere is the dominant mechanism, responsible for approximately ~80% of the total cloud thickness. The lower haze particles, supplied from below, act as a secondary but important source, contributing the

remaining $\sim 20\%$. We therefore propose a complete cloud cycle starting from nucleation. First, photochemically generated S_8 vapor nucleates to form CN in the upper cloud. These particles are coated in H_2SO_4 via heterogeneous nucleation, grow, and sediment downward, forming the lower and middle cloud. In parallel, the lower haze particles are transported upward from below the cloud base and first coated by solid S_8 via condensation. This mechanism also offers a compelling explanation for the large, crystalline particles observed by the Pioneer Venus probe. Then, these solid sulfur allotrope particles undergo heterogeneous nucleation of H_2SO_4 and grow larger through H_2SO_4 condensation.

The findings of this thesis and the development of the SPECK model provide a foundation for future exploration of planetary atmospheres. The inclusion of complex compositions, such as S_8 and meteoric smoke, offers a vital tool for interpreting upcoming data from the EnVision and DAVINCI missions, particularly in validating the refractive indices and abundances of cloud-forming species. Beyond Venus, the versatile architecture of SPECK can be extended to investigate water-ice clouds on Mars and photochemically-driven hydrocarbon hazes on gas giants, serving as a prototype for a universal cloud microphysics framework. Ultimately, by coupling this microphysical core with photochemical and radiative transfer models, we move closer to the goal of simulating planetary climates from first principles, enabling a deeper understanding of atmospheric evolution and climate stability on terrestrial and giant planets in the Solar System and beyond.

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List of Abbreviations

1-D	One-Dimension(al)
3-D	Three-Dimension(al)
AFES	Atmospheric GCM for the Earth Simulator
APC	Analytical Predictor of Condensation
CARMA	Community Aerosol and Radiation Model for Atmospheres
CN	Condensation Nuclei
DAVINCI	Deep Atmosphere Venus Investigation of Noble gases, Chemistry, and Imaging
EDB	Electrodynamic Balance
ESA	European Space Agency
GCM	General Circulation Model
IPSL	Institut Pierre-Simon Laplace
IR	Infrared
JAXA	Japan Aerospace Exploration Agency
K2024	Karyu et al. (2024)
LCPS	Cloud Particle Size Spectrometer
MSP	Meteoric Smoke Particle
NASA	National Aeronautics and Space Administration
OCPP	Orbitor Cloud Photopolarimeter
ODE	Ordinary Differential Equation
ppm	parts per million (10^{-6})
ppb	parts per billion (10^{-9})
ppt	parts per trillion (10^{-12})
PROTEUS	Photochemical and Radiation Transport Model for Extensive Use
PV	Pioneer Venus
SPECK	Simulator of Particle Evolution, Composition, and Kinetics
SOIR	Solar Occultation at Infrared
SPICAV	Spectroscopy for Investigation of Characteristics of the Atmosphere of Venus
SSAS	Subsolar-to-Antisolar
UV	Ultraviolet
VenSpec	Venus Spectrometer

VeRa	Venus Radio Science
VEx	Venus Express
VIRA	Venus International Reference Atmosphere
VIRTIS	Visible and Infrared Thermal Imaging Spectrometer
VMC	Venus Monitoring Camera
VMR	Volume Mixing Ratio
VMS	Venus Mass Spectrometer

Chapter 1

General Introduction

1.1 Planetary Clouds

1.1.1 Importance of clouds in planetary atmosphere

In the Solar System, clouds are a common feature on any planet with an atmosphere and thus constitute an essential component in shaping their climates (Pierrehumbert, 2010). Clouds are found on inner rocky planets (Venus, Earth, Mars) and outer planets, such as gas giants (Jupiter, Saturn) and ice giants (Uranus, Neptune) (Sánchez-Lavega et al., 2004), as shown in Figure 1.1. Furthermore, observations have found clouds on numerous exoplanets outside of our Solar System (Helling, 2019; Gao et al., 2021), highlighting their universality.

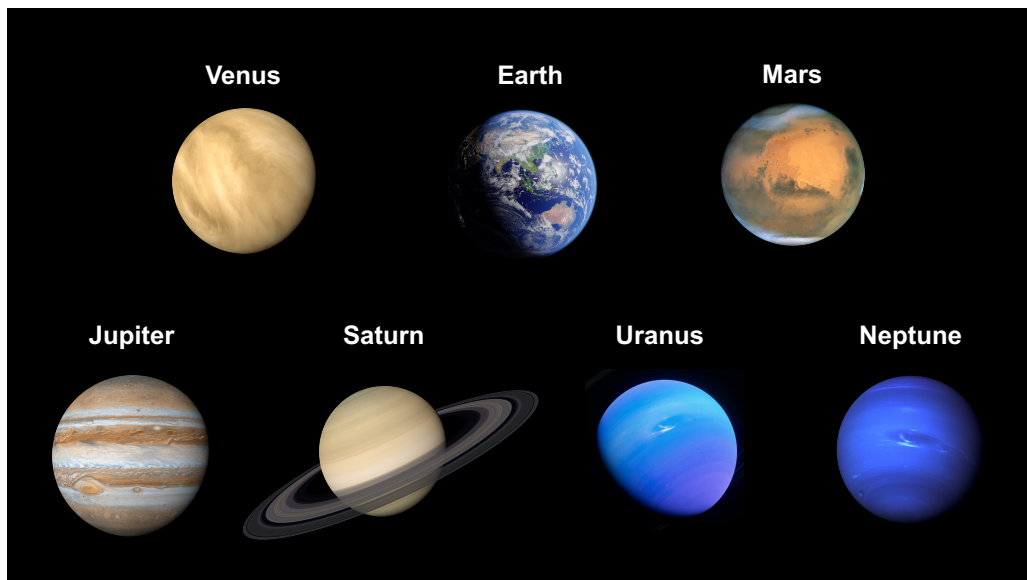


Figure 1.1: Clouds on various planets in the Solar System.

Clouds affect the planet's energy budget by reflecting the sunlight and trapping outgoing thermal radiation (e.g., Liou, 2002). Clouds function as sites of heterogeneous reactions and are also impor-

tant for understanding atmospheric chemistry (e.g., Seinfeld and Pandis, 2016). Furthermore, clouds determine the distribution of volatile substances such as water (H_2O) vapor—known as cold trap—and influence atmospheric evolution by serving as a bottleneck to atmospheric escape (e.g., Catling and Kasting, 2017). How clouds affect these atmospheric processes is determined by various factors, including their composition, quantity, particle size, and the altitude at which they form. Thus, obtaining these parameters can be considered a core part of understanding various planetary atmospheric phenomena.

1.1.2 Cloud formation mechanism

To understand how clouds form in planetary atmospheres, we first need to trace it back to the initial step. In principle, this initial step is categorized into two processes: homogeneous nucleation and heterogeneous nucleation (Pruppacher and Klett, 2010; Seinfeld and Pandis, 2016).

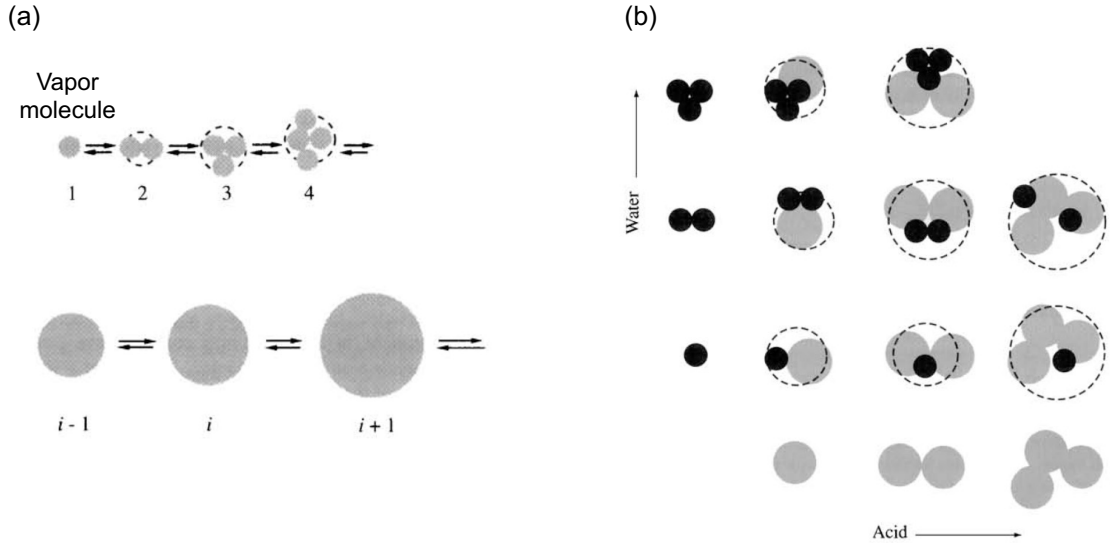


Figure 1.2: Sketch of (a) one-component homogeneous nucleation and (b) two-component binary homogeneous nucleation, adapted from Seinfeld and Pandis (2016).

In homogeneous nucleation, condensable gas molecules form clusters, generating nuclei with a typical size of ~ 1 nm, as shown in Figure 1.2a. The efficiency of homogeneous nucleation greatly depends on the system of condensing vapor. For a one-component system, classical nucleation theory is often used to describe the rate of how many nuclei form per second (homogeneous nucleation rate) as follows (Pruppacher and Klett, 2010; Seinfeld and Pandis, 2016):

$$J_{\text{hom}} = 4\pi r_c^2 Z_{\text{hom}} N_v \sqrt{\frac{k_B T}{\pi m_v}} \exp\left(-\frac{\Delta G_{\text{hom}}}{k_B T}\right), \quad (1.1)$$

$$r_c = \frac{2\sigma_v m_v}{\rho_v k_B T \ln S_v}, \quad (1.2)$$

$$\Delta G_{\text{hom}} = \frac{4\pi r_c^2 \sigma_v}{3}, \quad (1.3)$$

where, J_{hom} is the homogeneous nucleation rate ($\text{m}^{-3} \text{s}^{-1}$), k_B is the Boltzmann constant (J K^{-1}), T is the air temperature (K), N_v is the vapor concentration (m^{-3}), m_v is the molecular mass (kg) of vapor, σ_v is the surface tension (J m^{-2}) of liquid, ρ_v is the density (kg m^{-3}) of liquid, S_v is the saturation ratio of vapor, ΔG_{hom} is the Gibbs free energy for nucleus formation, and Z_{hom} is the Zeldovich nonequilibrium factor for homogeneous nucleation (Pruppacher and Klett, 2010). r_c is called the critical radius, which determines the minimum size of a particle that can exist in a condensed phase. Equation (1.2) indicates that higher supersaturation leads to smaller r_c , while Equation (1.3) indicates that smaller radius leads to smaller formation energy. Consequently, Equation (1.1) indicates that the higher supersaturation leads to larger nucleation rate with smaller critical radius. In the case of H_2O , a high degree of supersaturation ($\sim 1000\%$) is needed to produce nuclei with a size of $\sim 1 \text{ nm}$ under Earth atmospheric condition (Jacobson, 2005; Seinfeld and Pandis, 2016). This condition rarely occurs, so one-component H_2O homogeneous nucleation is not considered to be a major process on Earth atmosphere.

Addition of electrolytes (such as acid and base) to the H_2O system can greatly enhance the homogeneous nucleation rate (Figure 1.2b). These electrolytes tend to form a strong bond with H_2O molecules. Hence, they increase the stability of clusters. Sulfuric acid (H_2SO_4) is known to form a strong bond with H_2O molecules and increase the stability of molecule clusters, leading to more efficient formation of nuclei (Vehkamäki et al., 2002; Merikanto et al., 2016; Määttänen et al., 2018). This nucleation mechanism involving two components is called binary homogeneous nucleation (Seinfeld and Pandis, 2016). On Earth, particle formation rates often have a positive correlation with the abundance of gaseous H_2SO_4 (Kuang et al., 2008; Petäjä et al., 2009), highlighting the importance of the binary H_2SO_4 - H_2O homogeneous nucleation.

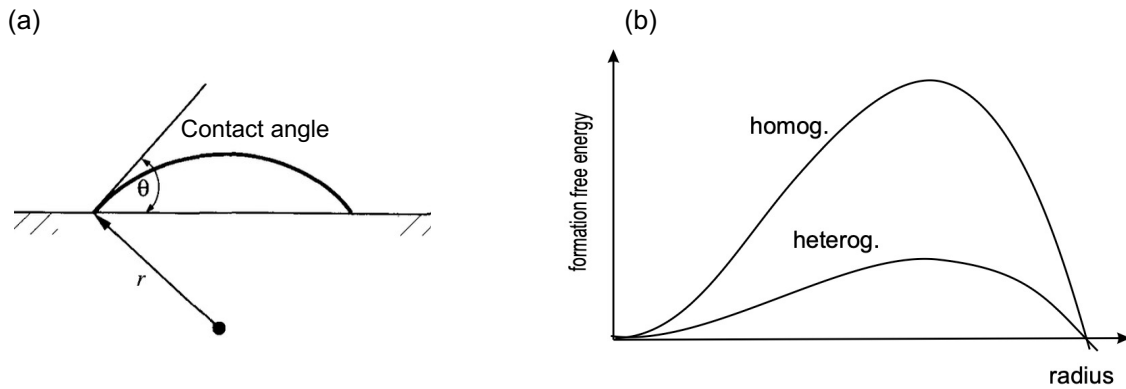


Figure 1.3: (a) Sketch of heterogeneous nucleation on a flat surface, adapted from Seinfeld and Pandis (2016). (b) Comparison of formation energy between homogeneous and heterogeneous nucleations, adapted from Vehkamäki (2006).

Heterogeneous nucleation occurs when vapors condense onto pre-existing atmospheric particles known as condensation nuclei (CN) (Pruppacher and Klett, 2010; Seinfeld and Pandis, 2016), as

illustrated in Figure 1.3. This wetting of CN is also referred to as “activation.” Since this process takes place on the CN surface, the heterogeneous nucleation rate is typically expressed in units of $\text{m}^{-2} \text{s}^{-1}$. In heterogeneous nucleation, the foreign surface provides a portion of the required interfacial area, thereby reducing the formation energy barrier. This contrasts with homogeneous nucleation, which requires building the entire spherical volume from scratch. Consequently, the energy required for nucleus formation is reduced by a factor equal to the volume ratio of the partial sphere (formed on the surface) to a perfect sphere of the same curvature radius. Accordingly, the Gibbs free energy for nucleus formation on a flat surface, ΔG^* , is expressed as follows (Seinfeld and Pandis, 2016):

$$\Delta G^* = \Delta G_{\text{hom}} f(m), \quad (1.4)$$

$$f(m) = \frac{1}{4}(2+m)(1-m)^2, \quad (1.5)$$

$$m = \cos \theta, \quad (1.6)$$

where θ is the contact angle between the condensed vapor and the surface, as illustrated in Figure 1.3a. The term $f(m)$ is known as Fletcher’s correction (Fletcher, 1958) and represents the ratio of the volume of a partial sphere to that of a perfect sphere with radius r . Since $f(m)$ ranges between 0 and 1, the energy barrier ΔG^* is lower than the homogeneous barrier ΔG_{hom} (Figure 1.3b), except when $m = -1$ (corresponding to a contact angle of $\theta = 180^\circ$). This reduction in energy means that heterogeneous nucleation is generally more efficient than homogeneous nucleation, making it a primary pathway for cloud formation.

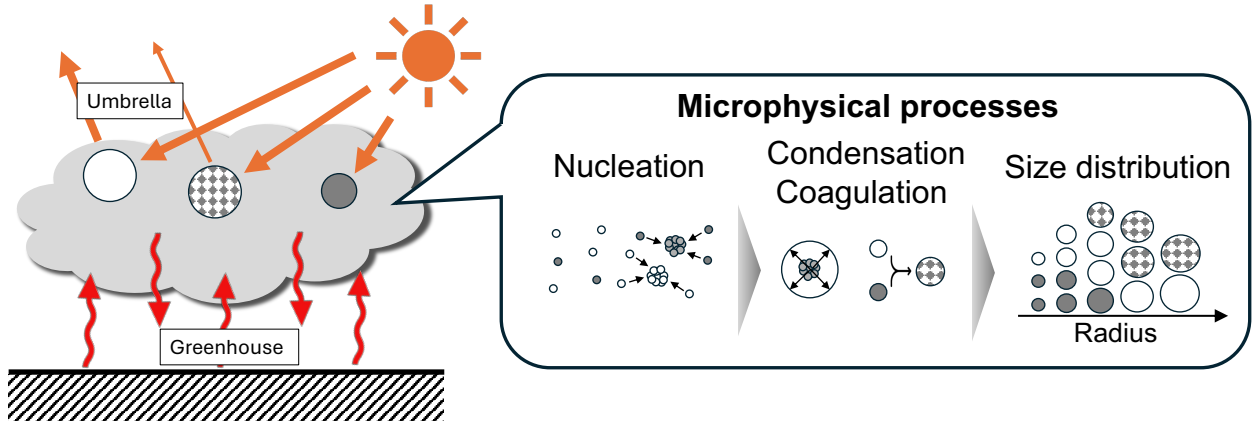


Figure 1.4: Sketch of cloud microphysics processes in planetary atmospheres and resulting climatic effects.

These nuclei further evolve through microphysical processes, including condensation and coagulation (Figure 1.4). Condensational growth occurs when a sufficient amount of vapor exists in the atmosphere. The term “sufficient amount” used here refers to the quantity derived from Equation (1.2), at which the supersaturation required for condensation to occur for a given particle radius in the atmosphere is achieved (Pruppacher and Klett, 2010). Coagulation occurs when two particles collide

with each other and make one larger particle. Since this process involves the mixing of the compositions, coagulation serves as a determining factor for the resulting particle composition (Riener et al., 2019). The combined effect of condensation and coagulation produces particles with a wide range of sizes and a variety of compositions, represented as a size distribution. The resulting diversity of clouds' microphysical properties can significantly affect the planet's climate mainly through two effects, as illustrated in Figure 1.4. First, clouds reflect incoming solar radiation—an effect often referred to as the umbrella effect. Second, clouds trap outgoing thermal radiation, thereby contributing to the greenhouse effect. Since both albedo and optical properties against thermal radiation depend on particle size and composition (Pierrehumbert, 2010), seamlessly modeling cloud microphysics from the initial nucleation stage is essential for understanding planetary climates at a fundamental level.

1.1.3 Interplanetary comparison of clouds and the niche of Venus

Since cloud formation is initiated by either homogeneous nucleation or heterogeneous nucleation on CN, quantifying the nuclei budget is essential for understanding cloud origins. The CN budget on Earth was investigated by Merikanto et al. (2009) (Figure 1.5). They found that H_2SO_4 homogeneous nucleation accounts for 45% of the total cloud nuclei budget, on a global average. The H_2SO_4 for homogeneous nucleation is predominantly provided through the photochemistry of dimethyl sulfide produced from phytoplankton (Seinfeld and Pandis, 2016). In contrast, particle emission from various sources accounts for the remaining 55% (Merikanto et al., 2009), including sea salt, carbonaceous particle emissions, and sulfate particle emissions from wildfires, biofuels, fossil fuels, and volcanoes (Stier et al., 2005). Therefore, it is evident that cloud formation on Earth is a process substantially involving both anthropogenic and biological activities.

In contrast, planetary cloud formation other than Earth is expected to depend purely on physical framework, as it lacks non-physical factors like anthropogenic or biological activities (at least to date). Mars has a water cycle involving H_2O cloud formation (Määttä et al., 2024; Montmessin et al., 2024). Heterogeneous nucleation on ubiquitous dust particles is the dominant formation mechanism, while homogeneous nucleation is considered unlikely (Määttä et al., 2005). Mars' water cycle is similar to Earth's in the sense that the main source and sink of condensable species are located on the planetary surface (Haberle et al., 2017), while its CN source is more straightforward due to the lack of variety in particulate matter compared to Earth.

After Earth and Mars, Venus is the third most investigated planet, with significant attention on its global sulfuric acid clouds. Unlike on Earth and Mars, Venus' main condensable species are generated via photochemistry. This mechanism provides a continuous supply of condensable material, sustaining global cloud decks—a distinct contrast to the patchy cloud cover observed on Earth and Mars (see Figure 1.1). The fundamental difference between these two regimes lies in how easily supersaturation can be achieved. On Earth and Mars, condensable species are supplied by evaporation processes from the surface. Consequently, it is difficult for the supersaturation ratio to significantly exceed unity, regardless of the extent of evaporation. As a result, whether clouds can form is governed by factors such as the adiabatic cooling rate accompanied by vertical motion (Lohmann et al., 2016). On Venus, in contrast, condensable species are continuously supplied by photochemical processes

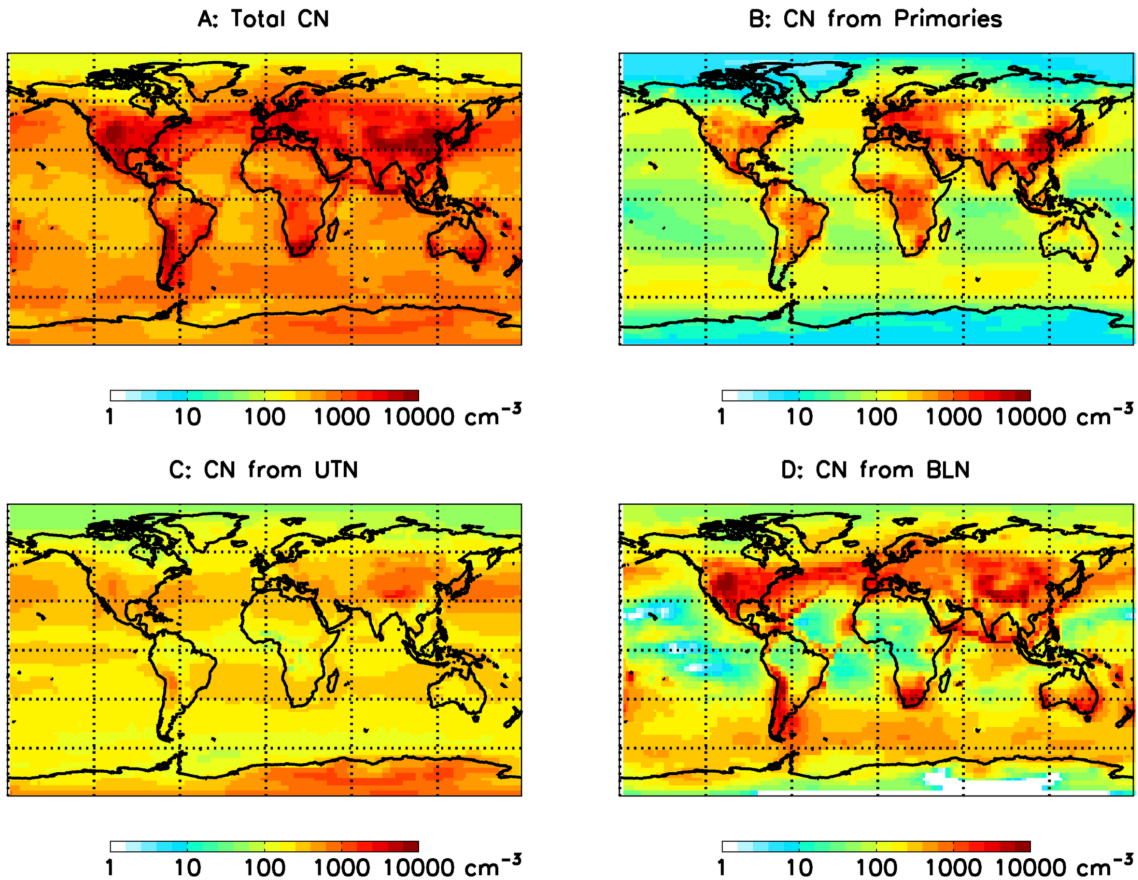


Figure 1.5: Simulated CN budget on Earth, adapted from Merikanto et al. (2009). CN from primaries represent particle emissions from various sources. CN from UTN represents the homogeneous nucleation of H_2SO_4 in the upper troposphere. CN from BLN represents homogeneous nucleation of H_2SO_4 in the boundary layer.

(Krasnopolsky, 2012; Zhang et al., 2012), allowing the supersaturation ratio to easily surpass unity. Under such conditions, one-component homogeneous nucleation, which is not a major pathway on Earth and Mars, may become the dominant process. However, the extent to which this homogeneous nucleation can supply nuclei on Venus remains unknown.

The photochemically produced clouds, analogous to those on Venus, may be the most common form of clouds, based on current observational progress both within and outside the Solar System. The atmosphere of the outer planets contains a significant amount of methane (CH_4), ammonia (NH_3), hydrogen sulfide (H_2S), and H_2O with VMR of more than hundreds of ppm. On Jupiter, CH_4 undergoes photolysis and turns into complex hydrocarbon species in the stratosphere, as depicted in Figure 1.6a. These hydrocarbon species homogeneously nucleate and condense to become micron-sized haze particles (Friedson et al., 2002; Wong et al., 2003). These haze particles are thought to serve as CN in the main cloud layer (West et al., 2004). The main condensable species are NH_3 , H_2O , and ammonium hydrosulfide (NH_4SH) produced by the chemical reaction between NH_3 and H_2S . These species condense at different altitudes due to the difference in saturation vapor pressure and produce a three-layered structure of clouds (Atreya et al., 1999, 2005), as shown in Figure 1.6b. These processes likely

operate on the other outer planets as well (Atreya and Wong, 2005), making these planets globally hazy and cloudy, as shown in Figure 1.1.

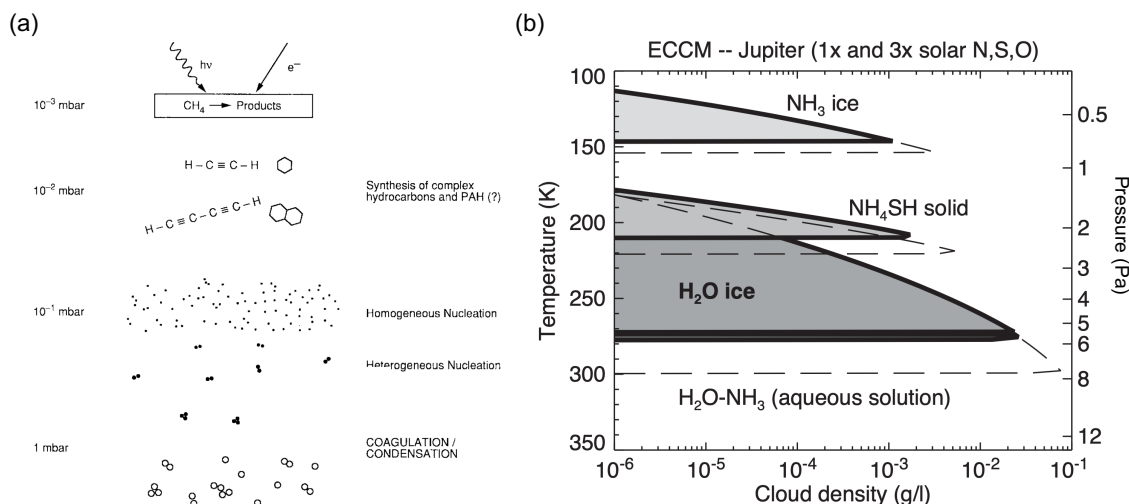


Figure 1.6: (a) Sketch of Jupiter's haze formation, adapted from Friedson et al. (2002). (b) The simulated vertical structure of the Jupiter clouds, adapted from Atreya et al. (2005).

A significant gap remains in our understanding of the Jovian cloud cycle because haze formation and subsequent cloud seeding processes are not coupled in existing microphysics models. While photochemical models calculate hydrocarbon production and haze formation, they typically omit cloud formation with NH₃, H₂O, and NH₄SH condensation (Friedson et al., 2002; Wong et al., 2003). Conversely, existing cloud microphysics models (Yair et al., 1995, 1998; Moreno, 1996) assume a constant CN production rate that is decoupled from nucleation—a limitation also prevalent in Venus' cloud modeling (see Section 1.4). Because even the most well-studied outer planets are subject to this limitation, a generalized theory of cloud formation for these worlds has yet to be established.

This lack of a fully integrated cloud cycle presents a major barrier to characterizing exoplanets, where a growing body of theoretical, experimental, and observational research suggests that hazes and clouds are ubiquitous atmospheric features (e.g., Helling, 2019; Gao et al., 2021; Nixon et al., 2024; Ohno et al., 2025) (Figure 1.7). Particulate matter on exoplanets can be composed of diverse materials, including water (Benneke et al., 2019; Kempton et al., 2023), hydrocarbons (Kawashima and Ikoma, 2018; He et al., 2018; Ohno et al., 2025), minerals (Gao et al., 2018; Dyrek et al., 2024; Inglis et al., 2024), and sulfur or sulfates (Hu et al., 2013; Tsai et al., 2021). Because cloud particles on these distant worlds are far more difficult to observe than those within our solar system, deciphering their formation mechanisms remains a profound challenge. Accordingly, the theoretical frameworks employed in exoplanetary cloud studies depend heavily on our understanding of the Solar System; thus, validating cloud formation theories using solar system analogues represents a critical step forward for the field (Gao et al., 2021). Within this broader context, the Venus cloud system serves as

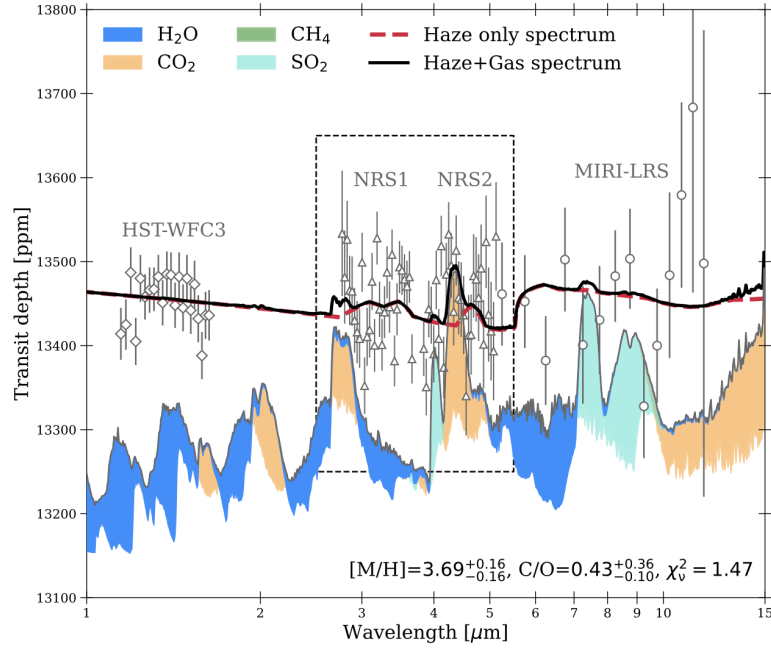


Figure 1.7: A median GJ 1214b spectrum, adapted from Ohno et al. (2025). The black solid line shows the spectrum that includes both haze and gas opacity. The gray solid and red dashed lines show the spectra including only gas and haze opacity, respectively, to illustrate their individual contributions.

an essential framework for understanding the photochemical haze and cloud formation mechanisms across a wide range of planetary environments.

On Earth, cloud formation mechanisms can be investigated in detail, as simulations can be compared to in-situ sampling of nm-sized nuclei. In contrast, such methods are not feasible in planetary atmospheric science, limiting the effective approach to theoretical studies. Thus, understanding cloud formation requires first-principle simulations—namely, nucleation theory—as these processes initiate all cloud formation (Section 1.3). However, existing Venus cloud models omit these processes, hindering comprehension of the cloud origin.

Understanding cloud formation on Venus is a critical step toward understanding planetary clouds in general for two reasons. Firstly, the cloud microphysics on Venus can be validated against abundant observational data obtained by previous spacecraft (Pioneer Venus, Venera, VeGa, Venus Express, Akatsuki, and many flybys) and ground-based telescopes. Secondly, the cloud formation theory established on Venus can be applied to many other planets, as photochemically produced clouds are the most common regime in the Solar System and possibly beyond. In the following sections, we review the previous cloud observations and modelling efforts, and then summarize the key challenges that need to be addressed to elucidate the cloud formation mechanism of Venus.

1.2 Venus and Its Clouds

1.2.1 Venus

Venus is often called Earth's twin planet, although its atmosphere is completely different from Earth's. The surface pressure is 90 times higher than that of Earth, and the atmosphere is mainly composed of carbon dioxide (CO_2). This thick CO_2 atmosphere, opaque to the thermal radiation, causes a significant greenhouse effect, resulting in the surface temperature of 740 K (Lebonnois et al., 2015), as shown in Figure 1.8. From the surface up to an altitude of ~ 60 km, the temperature profile closely follows the adiabatic lapse rate as a result of radiative-convective equilibrium, and this region is typically called the troposphere (Limaye et al., 2018). This persistent adiabatic lapse rate leads to a tropopause temperature of ~ 220 K. Above 60 km, the atmospheric structure is governed by radiative equilibrium in a region typically referred to as the mesosphere or middle atmosphere (Tellmann et al., 2009).

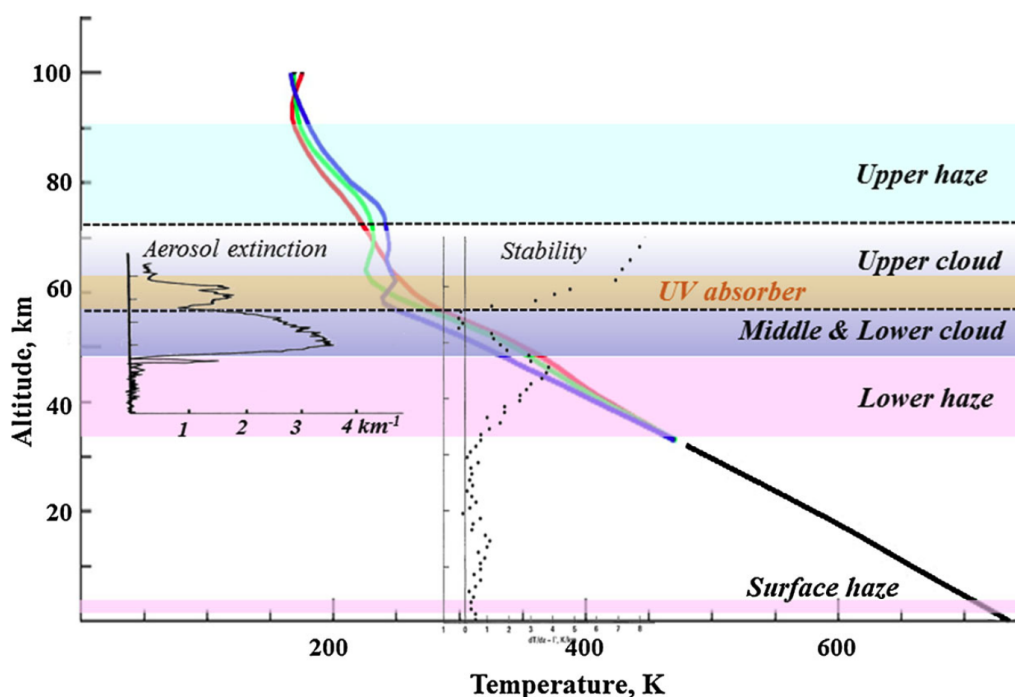


Figure 1.8: Vertical structure of the Venus clouds as derived by the Venera and Pioneer Venus descent probes, adapted from Titov et al. (2018). The color lines show mean temperature profiles at low (red), middle (blue), and high (green) latitudes with a single black line representing the temperature structure below 30 km. The typical vertical profile of aerosol extinction (Ragent and Blamont, 1980) is shown on the left. The static stability profile from the Venus international reference atmosphere model (Seiff et al., 1985) is shown in the middle.

Beyond the bulk CO_2 abundance, Venus' atmospheric composition presents another striking contrast to Earth's, as detailed in Table 1.1. Venus is severely depleted in H_2O , with concentrations reaching only ~ 30 ppm at the surface. This is attributed to hydrogen escape triggered by a hot early climate (Kasting, 1988; Turbet et al., 2021). This scarcity of water, combined with extreme surface temperatures, precludes the formation of oceans. Without a liquid ocean to sequester them, volcanic

Table 1.1: Venus' atmospheric composition

Species	VMR below cloud (~ 40 km)	VMR above cloud (~ 70 km)
CO ₂	$\sim 96\%$ ^[a]	$\sim 96\%$ ^[a]
N ₂	$\sim 4\%$ ^[a]	$\sim 4\%$ ^[a]
SO ₂	~ 130 ppm ^[b]	10-300 ppb ^[c]
H ₂ O	~ 30 ppm ^[d]	0.5-5 ppm ^[e]
CO	20-30 ppm ^[f]	~ 50 ppm ^[g]
H ₂ SO ₄	4-12 ppm ^[h]	< 3 ppb ^[i]
SO	-	10-100 ppb ^[j]
OCS	0.4-0.5 ppm ^[k]	0.3-10 ppb ^[l]
HCl	~ 0.4 ppm ^[m]	0.1-0.8 ppm ^[n]

^a Oyama et al. (1980)^b Marcq et al. (2008); Arney et al. (2014)^c Esposito et al. (1979); Belyaev et al. (2012); Jessup et al. (2015); Vandaele et al. (2017); Marcq et al. (2020); Mahieux et al. (2023a); Iwanaka et al. (2025)^d Marcq et al. (2008); Arney et al. (2014); Bézard et al. (2011); Chamberlain et al. (2013); Fedorova et al. (2015)^e Fedorova et al. (2008); Cottini et al. (2015); Chamberlain et al. (2020); Mahieux et al. (2024)^f Pollack et al. (1993); Bézard and de Bergh (2007); Satoh et al. (2021); Marcq et al. (2006)^g Krasnopolsky (2010a); Irwin et al. (2008); Iwagami et al. (2010); Vandaele et al. (2016); Mahieux et al. (2023b)^h Kolodner and Steffes (1998); Imamura et al. (2017); Oschlisniok et al. (2021)ⁱ Sandor et al. (2012)^j Sandor et al. (2010); Encrenaz et al. (2015); Belyaev et al. (2012)^k Marcq et al. (2006); Bézard and de Bergh (2007); Arney et al. (2014)^l Krasnopolsky (2010b); Mahieux et al. (2023a)^m Pollack et al. (1993); Iwagami et al. (2008); Arney et al. (2014)ⁿ Vandaele et al. (2008); Krasnopolsky (2010a); Sato and Sagawa (2023); Mahieux et al. (2023b)

gases such as CO₂ and sulfur dioxide (SO₂) have accumulated in the atmosphere (Catling and Kasting, 2017). Consequently, the abundances of SO₂ (~ 130 ppm) at the surface is significantly higher than those on Earth. This abundant SO₂ serves as a precursor for other species, including sulfur monoxide (SO) and carbonyl sulfide (OCS), driving a complex sulfur cycle within the Venus atmosphere (Mills and Allen, 2007; Zhang et al., 2012). Furthermore, SO₂ is photochemically converted into H₂SO₄ (see Section 1.3.1), which subsequently condenses to form the planet's dense, global cloud layers between 47 and 70 km, as indicated in Figure 1.8.

The global cloud cover gives Venus a planetary albedo of ~ 0.7 (Read et al., 2016; Lee et al., 2019), which is significantly higher than Earth's albedo of ~ 0.3 (Trenberth et al., 2009). These

clouds also contribute to the greenhouse effect, and removing them as a source of infrared opacity would decrease the surface temperature by 140 K (Limaye et al., 2018). However, their net effect is a significant cooling of the surface; the high albedo maintains the surface temperature at approximately 730 K. In the absence of clouds, the surface temperature could increase to 850–900 K due to the increased absorption of solar radiation (Bullock and Grinspoon, 2001; Titov et al., 2006).

Another critical component of the radiative energy budget is the UV absorber, which effectively absorbs solar radiation between 300 and 400 nm (Pollack et al., 1980; Pérez-Hoyos et al., 2018). This absorber is likely located between 57 and 70 km, overlapping with the cloud layer, as seen in Figure 1.8. The atmospheric heating generated by this absorption excites thermal tides (Kouyama et al., 2019; Fukuya et al., 2021), which serve as a major acceleration mechanism for atmospheric superrotation (Takagi et al., 2018; Horinouchi et al., 2020). Consequently, the UV absorber is vital to both the temperature structure and global atmospheric circulation. However, the exact identity of this material has remained unknown since its discovery. Recent experimental studies suggest that iron sulfate (Jiang et al., 2024) or ferric chloride (Egan et al., 2025) could satisfy the observed UV absorption features. In either case, iron appears to be a key species, although a viable physical mechanism for the supply of iron into the Venusian atmosphere has yet to be established.

1.2.2 Venus cloud observation

1.2.2.1 Before Venera, Vega, and Pioneer Venus

Before the era of spacecraft exploration, ground-based telescope measurements were the sole means of observing the Venusian clouds. During this period, researchers analyzed the reflected sunlight to investigate cloud characteristics. Polarimetry is a powerful tool to constrain the particle composition and size because the polarization signals are highly sensitive to the refractive index and the size of particles. Using a refractive index of 1.44 constrained by these polarization signals, Young (1973) proposed that the clouds are primarily composed of sulfuric acid. Shortly after, Hansen and Hovenier (1974) confirmed this composition through further polarization analysis. Additionally, by fitting theoretical polarization models to their observations, they demonstrated that cloud particles at the cloud top have a characteristic size of $\sim 1 \mu\text{m}$, as shown in Figure 1.9.

1.2.2.2 Venera, Vega, and Pioneer Venus

Venera (Keldysh, 1977), VEGA (Sagdeev et al., 1986), and Pioneer Venus (PV) (Colin, 1980) missions played a crucial role in establishing the standard structure of Venus' clouds (Titov et al., 2018), as shown in Figure 1.8. Particle counting instruments, namely nephelometers, on board Venera 9, 10, and 11 landers measured the microphysical properties of the clouds and suggested that the particle size distribution is at least bimodal between 48 and 57 km (Marov et al., 1980). The VEGA-1 balloon measured the backscattering of the clouds between 50 and 53 km and suggested that the clouds are highly variable in terms of time and altitude (Sagdeev et al., 1986). Measurement of the clouds' microphysical properties was also conducted by the Cloud Particle Size Spectrometer (LCPS) on board the PV Large probe (Knollenberg and Hunten, 1980). The in situ measurements revealed a triple-

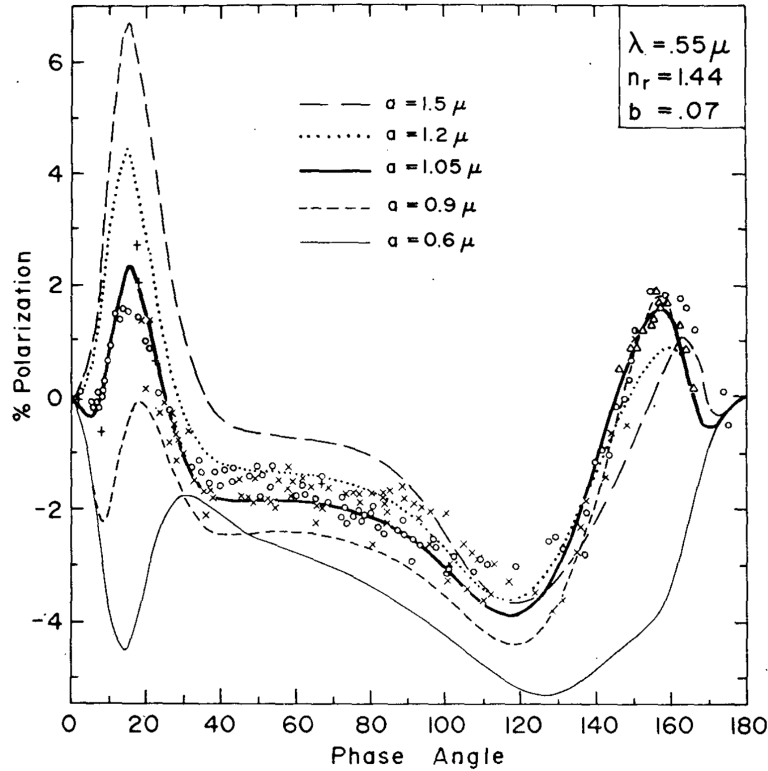


Figure 1.9: Comparison between observed polarization signals and computed signals, adapted from Hansen and Hovenier (1974).

layered cloud structure: the lower cloud spans from 47 to 51 km, the middle cloud from 51 to 57 km, and the upper cloud from 57 to 70 km. The lower and middle cloud layers, located between 47 and 57 km, exhibit a size distribution with three modal radii: Mode 1 ($\sim 0.15\text{--}0.2\ \mu\text{m}$), Mode 2 ($\sim 1\text{--}1.3\ \mu\text{m}$), and Mode 3 ($\sim 3.5\ \mu\text{m}$), as shown in Figure 1.10. The upper cloud layer, located between 57 and 70 km, displays a bimodal size distribution consisting of Mode 1 and Mode 2 particles. The parameters of Venus cloud observed by these multiple instruments are summarized in Table 1.2.

In addition to the particles in and above the cloud layer, entry probes from multiple missions—including those from the VeGa (Moshkin et al., 1986), Venera (Marov et al., 1980) missions, and PV (Knollenberg and Hunten, 1980)—have consistently detected an aerosol layer extending from the cloud base at ~ 47 km down to ~ 30 km, referred to as the lower haze layer (Titov et al., 2018). This indicates that Venus' clouds contain involatile substances other than sulfuric acid. The typical size of the lower haze particles ranges from 0.08 (Marov et al., 1980) to $0.21\ \mu\text{m}$ (Knollenberg and Hunten, 1980) and is thought to follow a lognormal distribution. These sub-cloud particles are transported upward into the main cloud deck, where they play a critical role in cloud formation by acting as condensation nuclei (James et al., 1997; Imamura and Hashimoto, 2001). The composition of these lower haze particles is unknown, but at least thought to have an involatile non-sulfuric-acid composition, given that H_2SO_4 cannot remain in the liquid phase in the intense heat of the lower atmosphere.

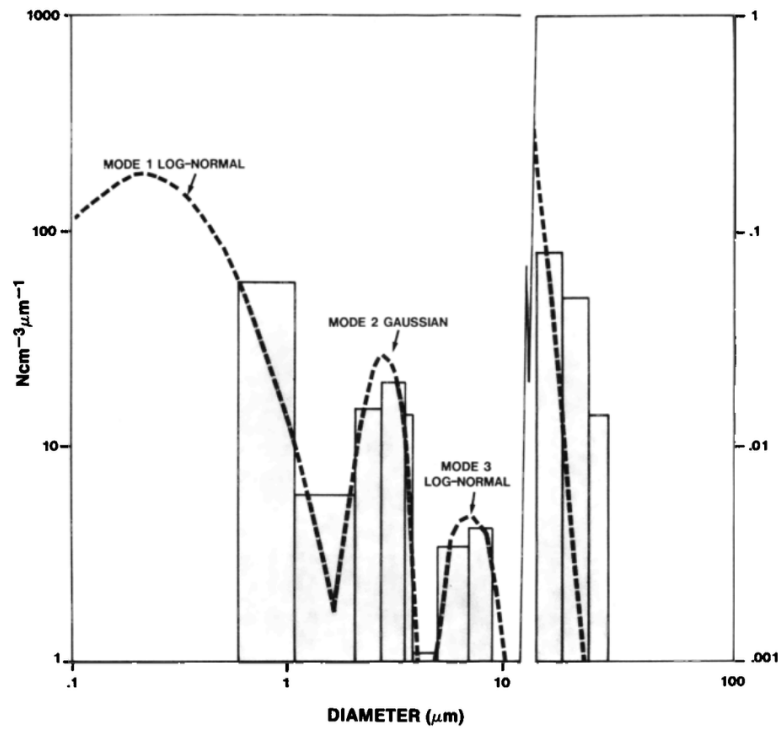


Figure 1.10: Cloud particle size distribution at 54 km observed by the PV LCPS adapted from Knollenberg and Hunten (1980)

Table 1.2: Parameters of the Venus cloud system adapted from Titov et al. (2018)

Region	Altitude range (km)	Optical thickness (at 0.63 μm)	Mean radius (μm)
Upper haze	70-90	0.2-1.0	0.2
Upper cloud	56.5-70	6.0-8.0	Mode 1: 0.2 Mode 2: 1.0
Middle cloud	50.5-56.5	8.0-10.0	Mode 1: 0.2 Mode 2: 1.3 Mode 3: 3.5
Lower cloud	47.5-50.5	6.0-12.0	Mode 1: 0.4 Mode 2: 1.0 Mode 3: 4.0

Particles have been observed in the region above the cloud top altitude of 70 km. This extended particle layer commonly referred to as the upper haze layer. Kawabata et al. (1980) characterized this layer using polarization data obtained by the Orbiter Cloud Photopolarimeter (OCP) onboard PV, reporting an effective radius of the haze particles of $\sim 0.2 \mu\text{m}$. They also reported that the obtained refractive index is consistent with that of sulfuric acid and that the optical thickness of the upper haze layer is 0.8, using 550 and 365 nm wavelengths.

1.2.2.3 Venus Express

Venus Express (VEx) (Svedhem et al., 2009) further advanced the understanding of the Venus clouds that was built up during the Pioneer Venus era. Near-infrared observations performed by the Visible and Infrared Thermal Imaging Spectrometer (VIRTIS) showed that the Venus cloud top is located at 71 km and decreases to 62 km with increasing latitude, using 1 μm wavelength (Haus et al., 2014), as shown in Figure 1.11. Haus et al. (2014) also showed that the opacity of the main cloud typically ranges from 30 to 50 near the equator and 25 to 40 in the mid-latitude region. Barstow et al. (2012) performed fitting of VIRTIS spectra to derive cloud parameters such as opacity, acidity, and cloud base altitude. They reported that the sulfuric-acid cloud acidity ranges from 80% to 100% and that cloud base is located between 44 and 46 km. In addition, the analysis of VIRTIS data supported the existence of the lower haze layer found by the previous probe measurements (Satoh et al., 2009).

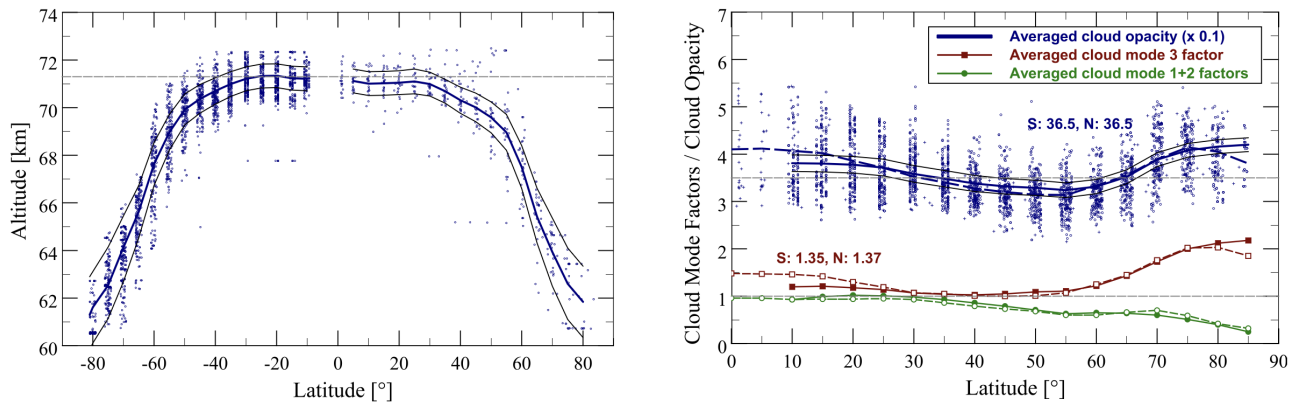


Figure 1.11: Zonal average of cloud top altitude (left panel) and total cloud opacity retrieved at 1 μm , adapted from Haus et al. (2014).

The cloud top was studied extensively using visible wavelengths from the Venus Monitoring Camera (VMC). Shalygina et al. (2015) analyzed the scattering phase functions of cloud particles, deriving a particle size of $\sim 1 \mu\text{m}$ and a refractive index of 1.44-1.47, which is slightly higher than those of a sulfuric acid solution. The VMC also captured the first image of a glory on Venus (Markiewicz et al., 2014), which was later used to further characterize cloud particles (Figure 1.12). By analyzing the phase functions obtained from these glory images, Markiewicz et al. (2018) determined the mean particle radius at the cloud top to be $\sim 1 \mu\text{m}$. They also suggested that an additional trace component with a refractive index higher than sulfuric acid, such as sulfur allotropes or ferric chloride, must be present within the cloud particles to fully match the observed scattering phase functions.

Observations by Spectroscopy for Investigation of Characteristics of the Atmosphere of Venus (SPICAV) on board VEx revealed the distribution and microphysical properties of the upper haze layer (Figure 1.13). Using the solar occultation data obtained by the Solar Occultation at Infrared (SOIR) instrument and SPICAV-UV, Wilquet et al. (2009) determined that the upper haze consists of two particle modes. Mode 1 particles have a mean radius ranging from 0.1 to 0.3 μm , while Mode 2 particles have a mean radius between 0.4 and 1.0 μm . Luginin et al. (2016) observed the upper haze layer using the dataset obtained by SPICAV-IR and obtained a mean radius of $\sim 0.5 \mu\text{m}$.

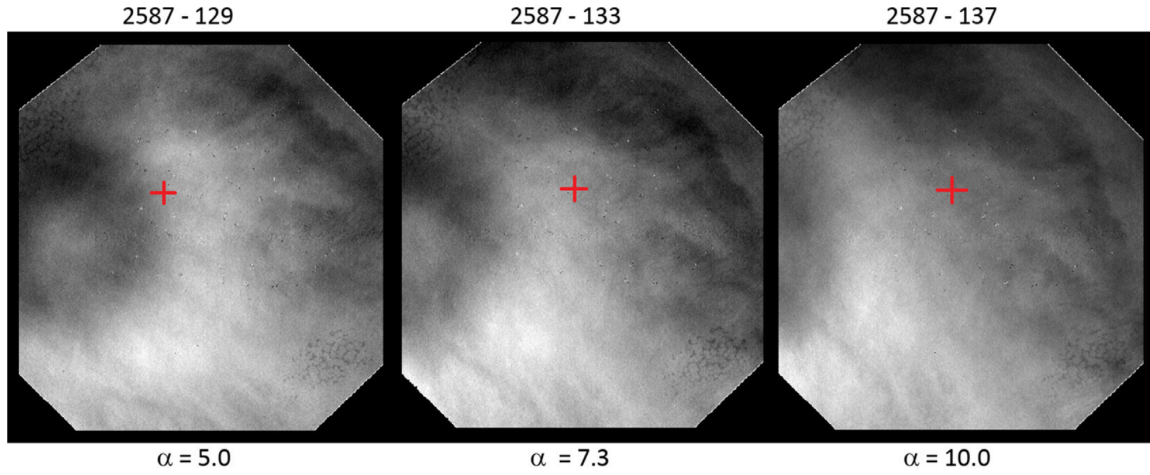


Figure 1.12: Three successive UV images taken in orbit 2587. The point marked by a cross (the coordinates are 0° in latitude and 180° in longitude) is viewed under different phase angles in the images (Markiewicz et al., 2018).

when a unimodal distribution was assumed. A follow-up study by Luginin et al. (2024) reported that a bimodal distribution is observed in more than 50% of retrieved profiles in the upper haze layer. Takagi et al. (2019) showed that haze particles are ubiquitous above 90 km using the SOIR data.

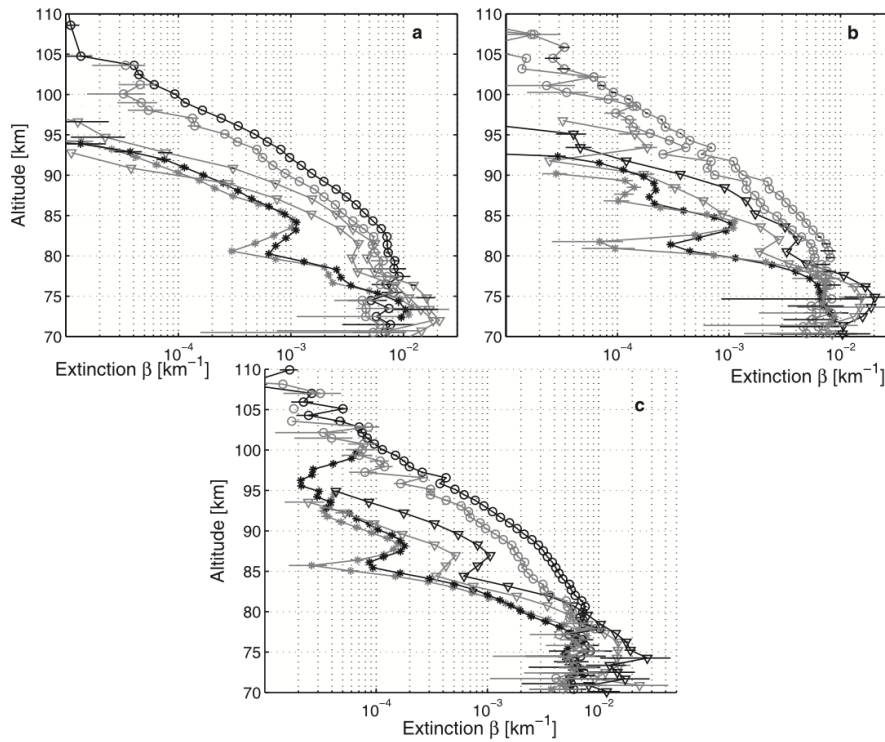


Figure 1.13: Vertical profiles of the extinction coefficient obtained with the three channels of the SPICAV instrument for the orbits (a) 485, (b) 486, and (c) 487, adapted from Wilquet et al. (2009).

1.2.2.4 Akatsuki

Following the Venus Express (VEx) era, JAXA's Akatsuki spacecraft (Nakamura et al., 2016) has provided further insights into the characteristics of Venusian clouds. Akatsuki's near-infrared cameras, IR1 and IR2, were designed to observe cloud silhouettes by detecting thermal infrared radiation emanating from the lower atmosphere. Using IR2 data (Figure 1.14), Satoh et al. (2021) investigated cloud microphysical parameters and optical thickness, demonstrating that the fraction of Mode 3 particles is higher at lower latitudes and that total cloud opacity ranges from 19.3 to 42.1. Additionally, Sato et al. (2020) utilized dayside IR2 observations to study the cloud top, finding that its altitude decreases from 71 km at the equator to 61 km at the poles. They also confirmed an average particle radius of $\sim 1 \mu\text{m}$. Collectively, these Akatsuki observations have reinforced the cloud characteristics identified by previous missions (e.g., Knollenberg and Hunten, 1980; Kawabata et al., 1980; Haus et al., 2014; Markiewicz et al., 2018).

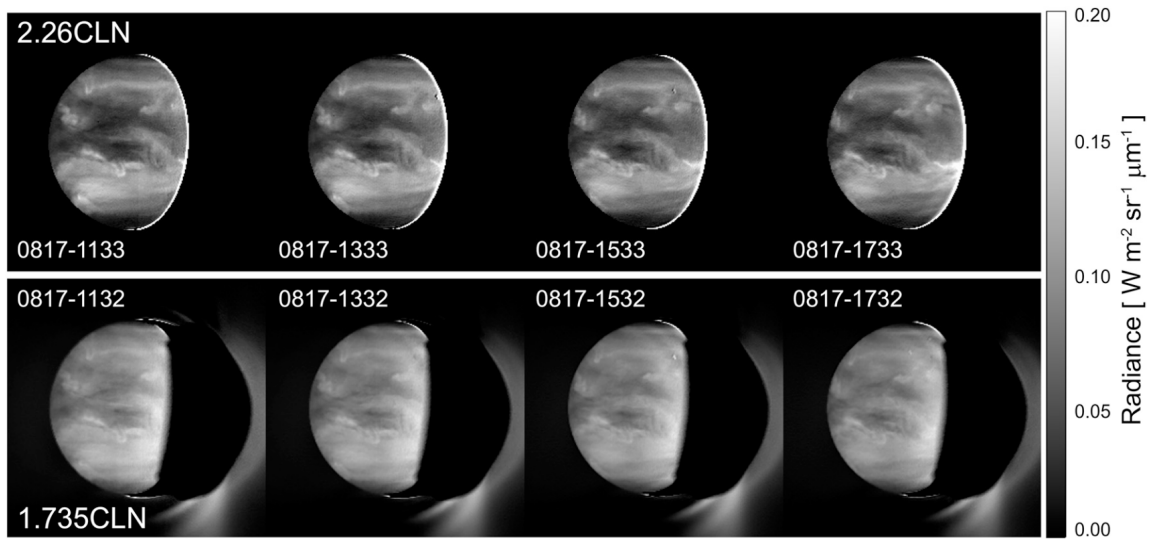


Figure 1.14: Images of cloud silhouettes taken by IR2 camera ($1.72 \mu\text{m}$ wavelength) onboard Akatsuki (Satoh et al., 2021).

1.3 Venus' Cloud Formation and Distribution

1.3.1 Production of condensable species

Condensable gases are necessary to form clouds. On Venus, the most abundant condensable species are H_2SO_4 and H_2O . The former one is provided through photochemistry. The series of chemical reactions starts from the SO_2 oxidation reaction with oxygen atom (O) provided by the photolysis of CO_2 or SO_2 as follows (Yung and DeMore, 1982; Krasnopolsky, 2012; Zhang et al., 2012):



Then, the sulfur trioxide (SO_3) readily reacts with H_2O to form H_2SO_4 as follows:

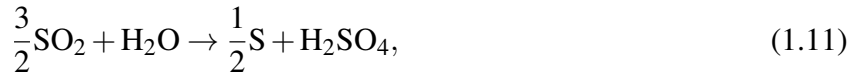


The second most important condensable species is sulfur allotropes composed of multiple sulfur atoms (S) bonded together. The sulfur atoms are generated through photolysis of SO_2 as follows:



These sulfur atoms are more energetically stable when they form molecules. Hence, they bond with each other to become various forms of molecules, whose length typically ranges from S to S_8 .

At the thermal equilibrium under room temperature, S_8 is by far the most abundant form of sulfur allotrope molecule since its molecular structure has the lowest energy state (Steudel et al., 2003; Lyons, 2008). The formation of the condensable species (H_2SO_4 and sulfur allotropes) occurs between 60 and 70 km, where SO_2 molecules effectively absorb the UV photons (Yung and DeMore, 1982; Krasnopolsky, 2012; Zhang et al., 2012). The net reaction of Equation (1.7)-(1.10) is summarized as follows (Imamura and Hashimoto, 2001):



which demonstrates that the formation of one H_2SO_4 molecule results in the loss of one H_2O molecule, and the stoichiometric ratio between the S atom and H_2SO_4 molecules is 1:2. The sulfur cycle involving the production of H_2SO_4 and sulfur allotropes are summarized in Figure 1.15

Finally, we briefly address the differences in sulfur chemistry between Earth and Venus. The primary distinction lies in the efficiency of SO_2 photolysis. On Earth, the ozone layer absorbs the short-wavelength UV radiation required to photolyze SO_2 , preventing the molecules from breaking down. Conversely, Venus lacks sufficient UV absorbers at these wavelengths, allowing SO_2 to photolyze and eventually form elemental sulfur. The second distinction is the abundance of oxidizing species. Earth's atmosphere is rich in water vapor and oxygen, leading to the high production of hydroxyl radicals (OH). These radicals rapidly oxidize SO_2 , and subsequent reactions with O_2 produce SO_3 and H_2SO_4 as the final products (Seinfeld and Pandis, 2016):



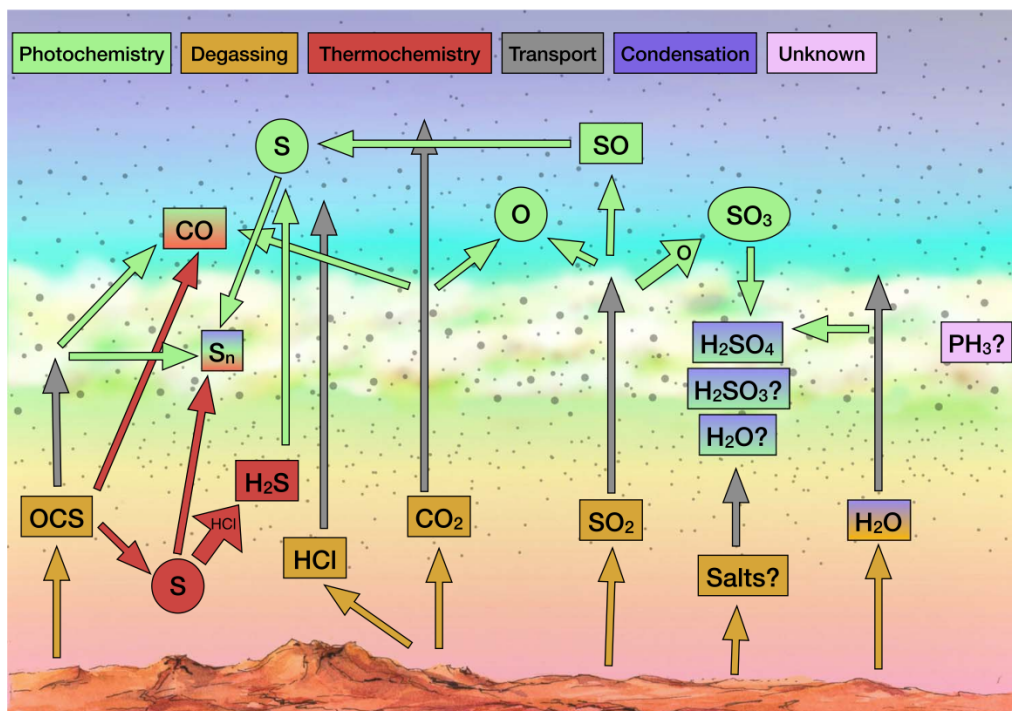


Figure 1.15: A sketch of the sulfur cycle on Venus adapted from Rimmer et al. (2021). S_n represents sulfur allotropes consisting of different numbers of sulfur atoms ($S-S_8$).

Because these oxidative pathways dominate on Earth—due to both the shielding of SO_2 from photolysis and the abundance of oxidants—photochemical pathways leading to elemental sulfur (such as disproportionation) do not occur. Consequently, sulfuric acid is the sole sulfur end product in Earth's atmosphere.

1.3.2 Formation mechanism of Venus' clouds

Based on the current understanding of Venus' atmosphere, three types of nuclei are hypothesized to exist. The first type consists of the lower haze particles, which serve as externally provided nuclei for the main cloud layer (Knollenberg and Hunten, 1980). The second is sulfur allotrope particles formed via homogeneous nucleation from sulfur allotrope vapor produced by photochemistry or thermochemistry (Krasnopolsky, 2007, 2012; Zhang et al., 2012). The first and second types of particles serve as condensation sites for H_2SO_4 vapor and become droplets via heterogeneous nucleation. Therefore, these non-sulfuric-acid particles are referred to as CN, and we use this terminology following previous studies (Imamura and Hashimoto, 2001; McGouldrick and Toon, 2007; McGouldrick, 2017; McGouldrick and Barth, 2023). The third type is H_2SO_4 - H_2O particles formed through binary homogeneous nucleation (Määttä et al., 2018).

As the initial step of cloud formation, the photochemically produced sulfur allotropes are hypothesized to form CN around 70 km and serve as condensation sites for H_2SO_4 vapor (Imamura and Hashimoto, 2001), as shown in Figure 1.16. These CN turn into cloud droplets through hetero-

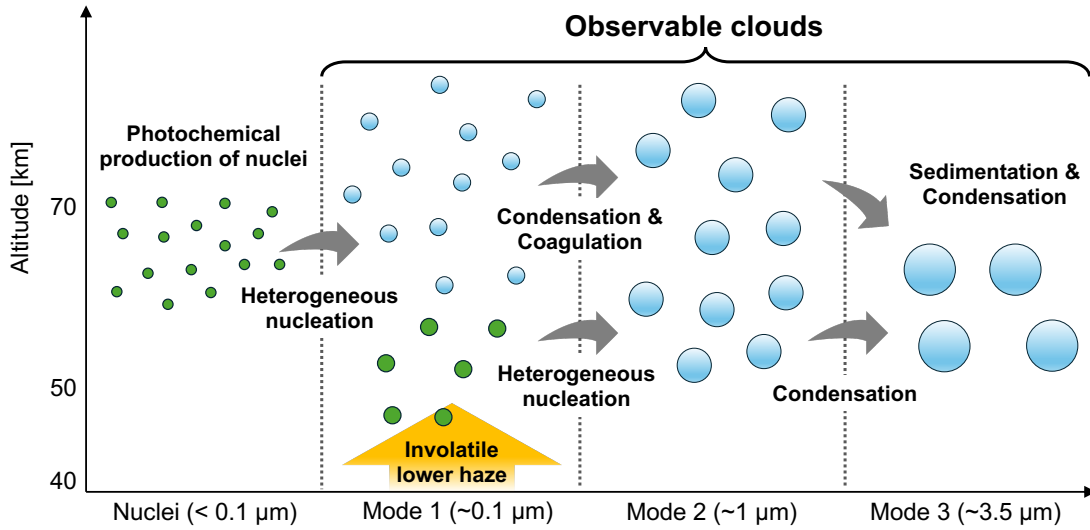


Figure 1.16: A sketch of the current understanding of cloud formation mechanism on Venus. The green and blue circles indicate CN and H₂SO₄ droplets, respectively.

geneous nucleation. This process initially forms submicron particles composed of sulfur allotrope core with H₂SO₄ coating in the upper cloud region, corresponding to Mode 1 particles (Imamura and Hashimoto, 2001; McGouldrick, 2017; McGouldrick and Barth, 2023). These particles further grow to the size of $\sim 1 \mu\text{m}$ via H₂SO₄ condensation and coagulation, corresponding to Mode 2 particles.

The Mode 2 particles in the upper cloud layer start sedimenting out since their sedimentation time scale is shorter than the condensation time scale (James et al., 1997) as shown in Figure 1.16. These particles participate in the lower and middle cloud layers and grow larger to $\sim 3 \mu\text{m}$ while the sedimentation process (Imamura and Hashimoto, 2001; McGouldrick, 2017), which corresponds to Mode 3 particles. In addition, the lower and middle cloud layers are also maintained by the lower haze particles and H₂SO₄ vapor sourced from below the cloud base (James et al., 1997; Imamura and Hashimoto, 2001; McGouldrick and Toon, 2007). The involatile lower haze particles with the size of $\sim 0.1 \mu\text{m}$ are first transported upward by strong convective eddy diffusion and undergo heterogeneous nucleation to turn into droplets. These droplets grow via H₂SO₄ condensation until they reach the size of $\sim 3 \mu\text{m}$. In essence, the lower and middle cloud layers are maintained by particle sources from the upper cloud layer and below the cloud base.

While this scenario successfully explains the observed Venus cloud structure (Knollenberg and Hunten, 1980; Titov et al., 2018), it relies on several untenable assumptions regarding the initial stages of cloud formation. The first assumption is that sulfur allotrope CN can form immediately upon the production of sulfur allotrope molecules; in other words, this scenario neglects the homogeneous nucleation of sulfur allotropes. The second assumption is that binary H₂SO₄–H₂O homogeneous nucleation does not play a significant role in cloud formation. This effect should be investigated in more detail, as a recent study (Dai et al., 2022) showed that H₂SO₄ can be highly supersaturated in the mesosphere and hosts a favorable condition for homogeneous nucleation. Hence, given the uncer-

tainties regarding the nucleation mechanism on Venus, we still do not understand whether sufficient CN can form in Venus’s atmosphere from first principles. The unknown formation mechanism of the lower haze is also problematic, since the lower haze particles seed a significant portion of the lower and middle clouds. These issues are deeply related to the advances of cloud microphysics modeling, which is discussed in Section 1.4.

1.3.3 Vertical transport of clouds and condensable species

Vertical transport is crucial for determining both the cloud structure and the condensable species distribution. In the idealized atmospheric column focused on in this study, vertical transport is often represented by an eddy diffusion coefficient. This coefficient is not an observable quantity but rather a conceptual value representing various scales of atmospheric dynamical phenomena, such as macroscopic eddies or global circulation (Catling and Kasting, 2017).

Table 1.3: Summary of observational estimates of eddy diffusion

Altitude (km)	Eddy diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)	Method	Reference
70 <	$1.4 \times 10^9 [M]^{-0.5}$	Estimated from trace gas distribution	von Zahn et al. (1980)
85 <	~ 360	Estimated from trace gas distribution	Mahieux et al. (2021)
~ 60	$4 >$	Radio scintillation	Woo and Ishimaru (1981)
~ 45	$0.2 >$	Radio scintillation	Woo et al. (1982)

On Venus, eddy diffusion coefficients are estimated at various altitudes, as summarized in Table 1.3. von Zahn et al. (1980) derived a numerical expression for calculating eddy diffusion coefficients above 70 km, by matching the carbon monoxide (CO) distribution and that predicted by the diffusion equation. This formula predicts $\sim 10 \text{ m}^2 \text{s}^{-1}$ in the upper haze layer. On the other hand, recent estimate using the CO distribution derived from SPICAV-SOIR predicts a much larger value of $\sim 360 \text{ m}^2 \text{s}^{-1}$. Hence, there is an inconsistency between classical and recent estimates in the upper haze layer.

Recent observations suggest that the transport of the upper haze particles is important to understand the material cycle in the Venus mesosphere. Mahieux et al. (2024) reported that the H_2O and HDO Volume Mixing Ratio (VMR) both increase with altitude, as shown in Figure 1.17. They argued that upward diffusive transport of H_2SO_4 - H_2O particles and their subsequent evaporation can explain such an increment. Furthermore, this region is connected to the upper atmosphere where H_2O can break down by photolysis and escape as an H atom (Figure 1.18). H_2SO_4 may evaporate as well as H_2O . In that case, sulfur species are deposited in the upper haze layer and photodissociated to SO_2 , as originally proposed by Zhang et al. (2010). Thus, the eddy diffusion transport process of particles and their phase transition in the upper haze layer may be a key to understanding atmospheric evolution and sulfur chemistry on Venus.

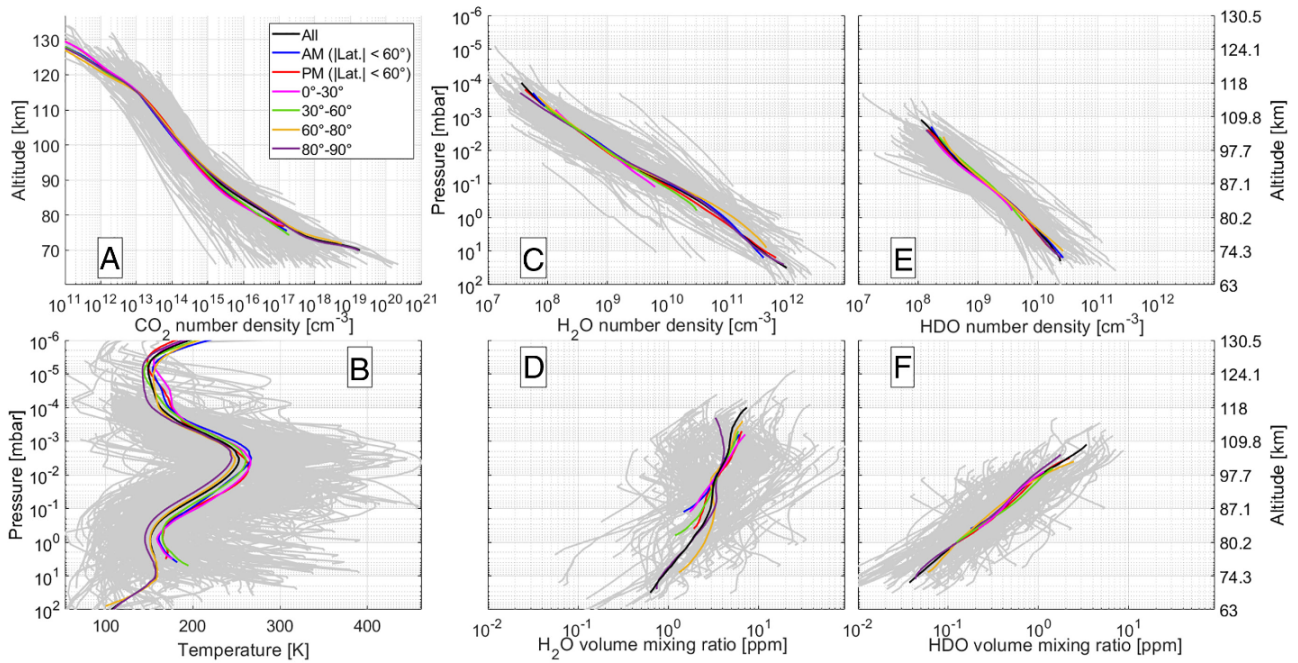


Figure 1.17: CO₂ number density (Panel A), temperature (Panel B), H₂O number density (Panel C), H₂O VMR (Panel D), HDO number density (Panel E), and HDO VMR (Panel F) vertical profiles, adapted from Mahieux et al. (2024).

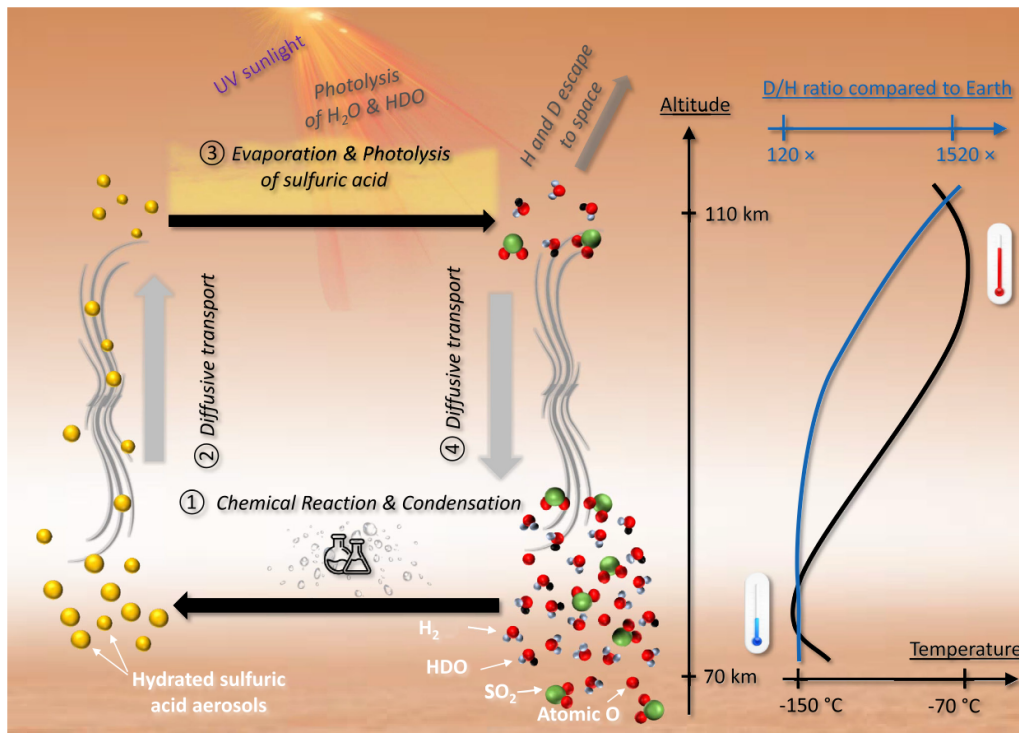


Figure 1.18: A schematic view of the mesospheric water cycle on Venus, adapted from Mahieux et al. (2024).

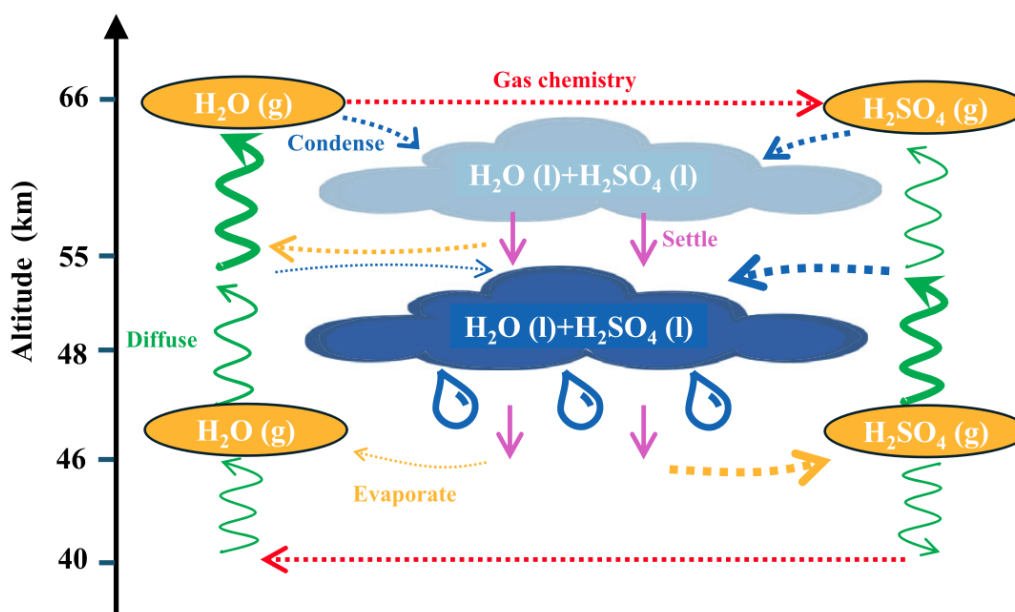


Figure 1.19: The H_2SO_4 and H_2O cycles in the Venus cloud system adapted from Dai et al. (2025).

Eddy diffusion transport is also crucial for the H_2O distribution in the cloud layers. The H_2O VMR in the Venus atmosphere decreases by more than an order of magnitude from ~ 30 ppm below the clouds (Arney et al., 2014; Bézard et al., 2011; Chamberlain et al., 2013; Fedorova et al., 2015; Marcq et al., 2008) to ~ 1 ppm above the clouds (Chamberlain et al., 2020; Cottini et al., 2015; Fedorova et al., 2008; Mahieux et al., 2023b). This profile is determined by a delicate balance among downward liquid flux, chemical loss, and upward vapor flux (Dai et al., 2025), as shown in Figure 1.19. The eddy diffusion coefficient governs the upward vapor flux, as it is the primary transport process for vapor species.

1.4 Venus Cloud Microphysics Modeling

A one-dimensional (1-D) cloud microphysics model simulates the vertical profile of clouds and condensable species based on the most fundamental processes that determine size distribution of cloud particles, such as condensation, coagulation, and nucleation. Therefore, this model has been employed to understand the observed cloud structure and particle size distribution on Venus.

Community Aerosol and Radiation Model for Atmospheres (CARMA)—originally developed for Earth atmospheric study (Turco et al., 1979; Toon et al., 1988)—has been widely used for Venus cloud studies. The first Venus cloud microphysics model was based on CARMA and used to investigate the contribution of sulfur allotropes to the unknown UV absorber (Toon et al., 1982). Later, James et al. (1997) used CARMA to investigate the formation mechanism of the lower and middle clouds and found that the heterogeneous nucleation of the lower haze particles is important to form these clouds. McGouldrick and Toon (2007) coupled CARMA with radiative transfer and showed that cloud formation and the subsequent cloud deck heating induce a positive feedback loop and further enhance

cloud formation. While the aforementioned CARMA studies covered the altitude range between 40 and 70 km, Gao et al. (2014) extended the model top to 100 km to investigate the formation mechanism of the upper haze, while fixing the H_2O profile. They showed that the upper haze exhibits a unimodal distribution at a steady state, while a bimodal structure can be induced by transient upward winds in the mesosphere. More recent studies by McGouldrick (2017) and McGouldrick and Barth (2023) focus on the dependence of the cloud structure on microphysical properties of CN. These studies showed that low coagulation efficiency between CN can result in long-term variation of cloud optical thickness while reproducing the observed cloud structure (Figure 1.20).

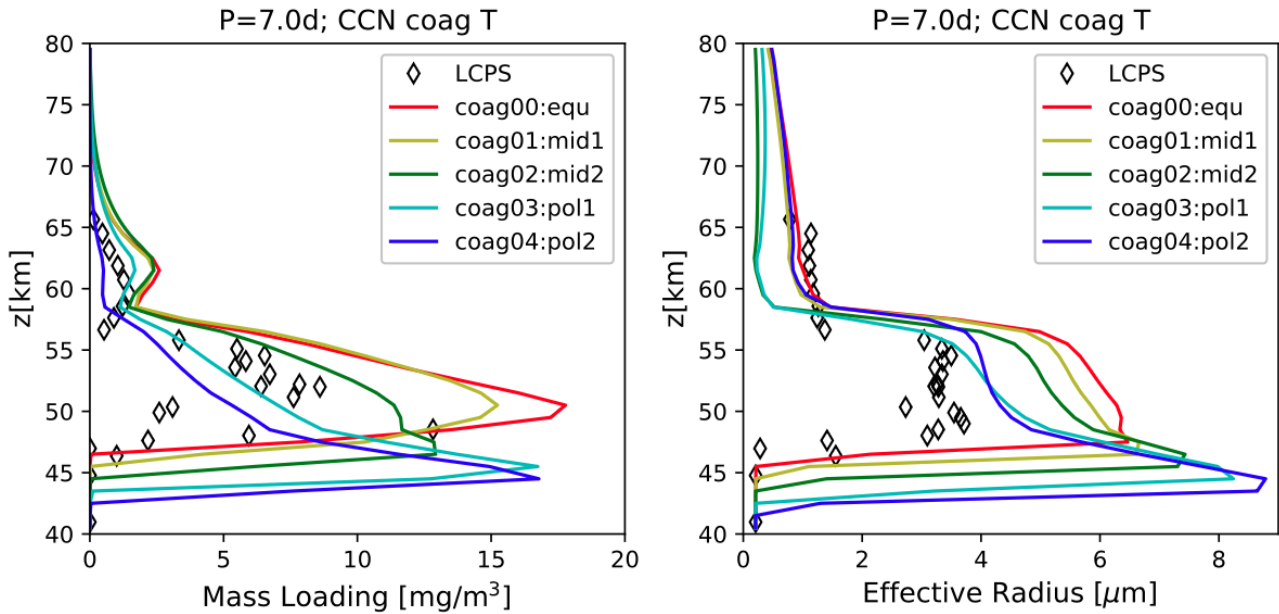


Figure 1.20: The simulated mass loading (left) and effective radius (right) for five latitudes, adapted from McGouldrick and Barth (2023). The vertical profiles are compared with the observations by LCPS (near the equator).

Besides CARMA, Imamura and Hashimoto (2001) developed their own version of a 1-D cloud microphysics model to investigate the formation mechanism of the trimodal distribution observed by the PV LCPS. They found that transient mixing induced by strong convection within the cloud deck induces rapid growth of cloud particles, resulting in a transition from a steady-state bimodal distribution to the observed trimodal distribution (Figure 1.21).

Although the aforementioned models have advanced our understanding of the Venus cloud observations, challenges remain in the simultaneous reproduction of the cloud structure and the distribution of its constituents (H_2O and H_2SO_4 vapors). In particular, previous studies (e.g., McGouldrick, 2017; McGouldrick and Barth, 2023) overestimated the H_2O VMR at the cloud top by more than an order of magnitude compared to observed values (Figure 1.22), partly because they used unrealistic eddy diffusion profiles. This discrepancy may result in inaccurate cloud structure above the upper cloud, as the mole concentration of H_2O in H_2SO_4 droplets is observed to be more than 50% and regarded as the dominant component of the clouds (Hansen and Hovenier, 1974). In addition, no cloud micro-

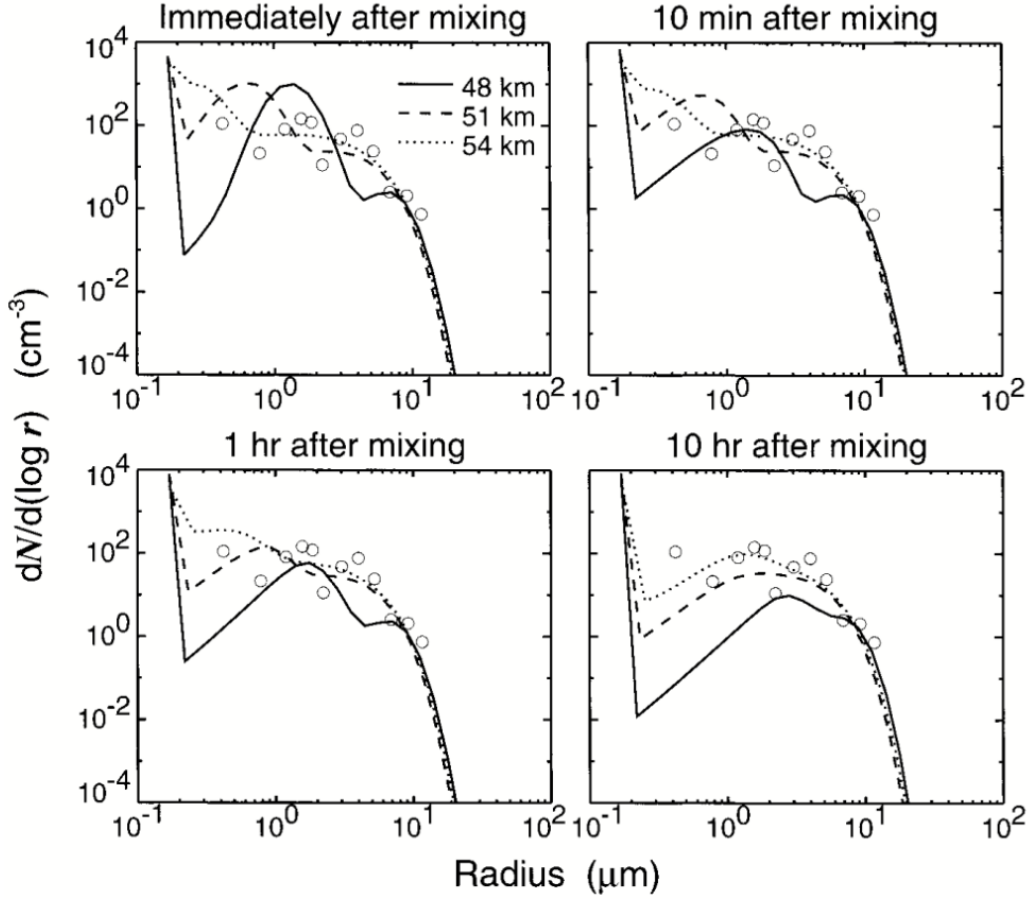


Figure 1.21: Development of particle size distribution at three altitude levels in the lower part of the cloud after mixing of an elevated column and lowered column for four times after mixing, adapted from Imamura and Hashimoto (2001).

physics model has yet simultaneously simulated cloud particles, H_2SO_4 , and H_2O up to 100 km. This represents a hurdle for understanding aerosol-mediated material transport in the mesosphere, which could affect sulfur chemistry and long-term evolution of Venus' atmosphere (Mahieux et al., 2024). Moreover, eddy diffusion coefficients for the upper haze layer are inconsistent within observations themselves, ranging from a classical value of $\sim 10 \text{ m}^2 \text{ s}^{-1}$ (von Zahn et al., 1980) to a recent value of $\sim 360 \text{ m}^2 \text{ s}^{-1}$ (Mahieux et al., 2021). Therefore, revisiting the eddy diffusion coefficient is necessary not only for reproducing the cloud structure but also for extending the model top altitude to 100 km.

The formation mechanism of the lower haze still remains unknown. All previous cloud microphysics models have imposed the lower haze particle population as a boundary condition in order to reproduce the observed cloud profile (James et al., 1997; Imamura and Hashimoto, 2001; McGouldrick and Toon, 2007; Gao et al., 2014; McGouldrick, 2017; McGouldrick and Barth, 2023). For simplicity, it has also been assumed that these particles are composed of either an unknown involatile material or sulfur allotropes. However, whether sulfur allotropes can remain in the solid phase below the cloud base is uncertain, because their condensation and evaporation are not included in the current

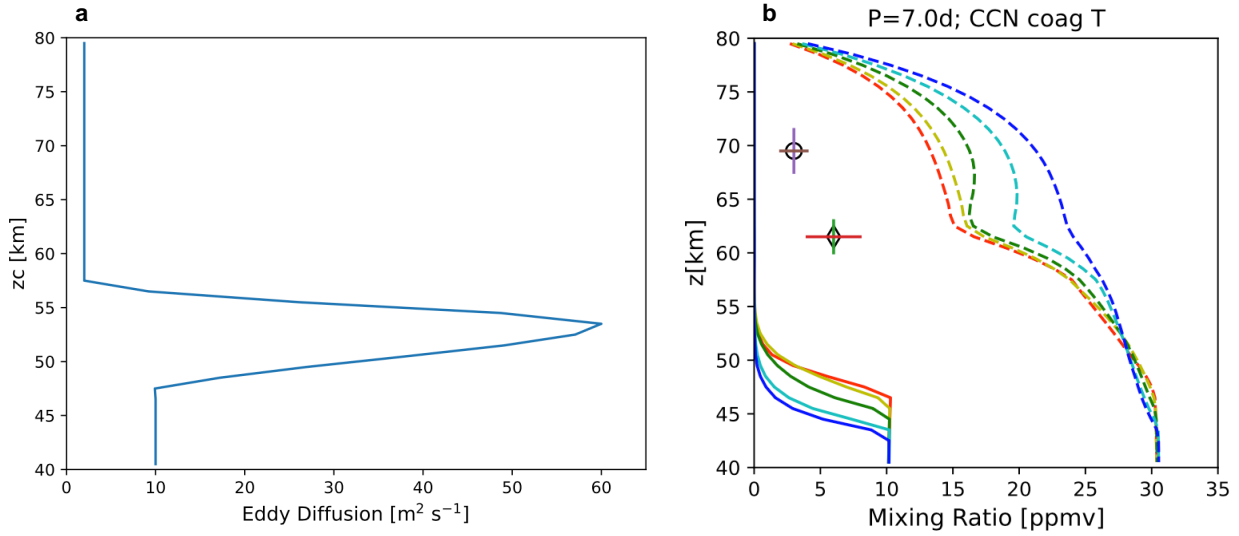


Figure 1.22: (a) Eddy diffusion profile used in CARMA and (b) the simulated H_2O VMR profile, modified from McGouldrick and Barth (2023).

cloud microphysics models (Gao et al., 2014; McGouldrick and Barth, 2023). As a result, no self-consistent model—one that simulates both the haze’s formation from its source and its interaction with cloud droplets—has yet to be developed, leaving its origin unexplained.

Another key issue in cloud microphysics modeling is the oversimplification of the sulfur allotrope CN production rate. Existing cloud microphysics models typically neglect the homogeneous nucleation of sulfur allotropes. Instead, the most widely used approach is a parameterization developed by Imamura and Hashimoto (2001), which scales the CN production rate to the chemical production rate of sulfur allotropes, as follows:

$$P_{\text{CN}}(z) = \frac{M_{\text{S}} P_{\text{S}}(z)}{\frac{4}{3} \pi \rho_{\text{CN}} r_{\text{CN}}^3}, \quad (1.15)$$

where $P_{\text{S}}(z)$ is the H_2SO_4 vapor production rate ($\text{m}^{-3} \text{s}^{-1}$), r_{CN} and ρ_{CN} are respectively the radius (m) and mass density (kg m^{-3}) of condensation nuclei, and M_{S} is the molecular mass (kg) of elemental sulfur. Because this parameterization is straightforward and easily implemented, it has been widely used in subsequent studies (e.g., Gao et al., 2014; McGouldrick, 2017; McGouldrick and Barth, 2023). Although this parameterization successfully reproduces the observed cloud structure, its oversimplification introduces several critical limitations. First, the parameterization requires that r_{CN} be artificially determined. Previous studies have set r_{CN} to 10 nm or 100 nm (Imamura and Hashimoto, 2001; Gao et al., 2014; McGouldrick, 2017; McGouldrick and Barth, 2023)—values more than an order of magnitude larger than the typical ~ 1 nm radius of nuclei formed via homogeneous nucleation. Given that the nucleation rate is inversely proportional to the radius cubed, this artificial constraint is unsuitable for accurately modeling the cloud formation process. Second, the parameterization assumes that CN forms immediately upon the production of sulfur allotropes, thereby

neglecting the thermodynamic requirements of homogeneous nucleation. This omission means that the parameterization may not reflect a realistic CN production rate. As a result of these oversimplifications, it remains unknown whether sufficient sulfur allotrope CN can form to effectively seed Venus's clouds.

The binary homogeneous nucleation of H_2SO_4 and H_2O molecules has not been extensively studied in the Venus atmosphere. Since both H_2SO_4 and H_2O are primary condensable gases on Venus, this nucleation mechanism may have an important role in cloud formation. McGouldrick (2017) investigated the effect of H_2SO_4 homogeneous nucleation on cloud structure but used a predefined production rate for sulfur allotrope CN. However, the parameterization in that study was developed for terrestrial conditions (Vehkamäki et al., 2002) and is not fully applicable to Venus' atmospheric conditions. On the other hand, more recent parameterization (Määttä et al., 2018) covers a wide range of temperature and relative humidity conditions applicable to Venus. However, this parameterization has never been included in Venus cloud microphysics models, and the effect of the binary homogeneous nucleation is yet to be assessed.

Cloud formation begins with the supply of nuclei and the various microphysical processes that follow. However, on Venus, the supply mechanism of the nuclei—the formation of the lower haze, homogeneous nucleation of sulfur allotropes and H_2SO_4 nuclei—has not been investigated based on first principles but rather artificially provided, as discussed above. Therefore, we still do not understand how and where Venus' clouds originate at a fundamental process level, and thus, their origins.

1.5 Thesis Outline and Objectives

The main objectives of this thesis are to investigate the origins of Venus' clouds. This is done by posing the following questions:

- 1. What is the origin of the lower haze?**
- 2. Can a sufficient amount of nuclei form by homogeneous nucleation?**
- 3. What is the relative contribution of each type of nucleus?**

However, several obstacles must be addressed before answering these questions. Firstly, accurately simulating the cloud distribution and condensable vapors requires a realistic eddy diffusion profile. We therefore conduct a parameter study in Chapter 2 to constrain this profile, using a 1-D cloud microphysics model based on (Imamura and Hashimoto, 2001). We also investigated how the upper haze layer affects the condensable species distribution. We found that the upward eddy diffusion transport and subsequent evaporation significantly affect both H_2SO_4 and H_2O abundances in the mesosphere.

Secondly, the existing microphysics framework is limited to H_2SO_4 and H_2O as condensable species, but simulating the lower haze and sulfur allotrope microphysics requires a more versatile,

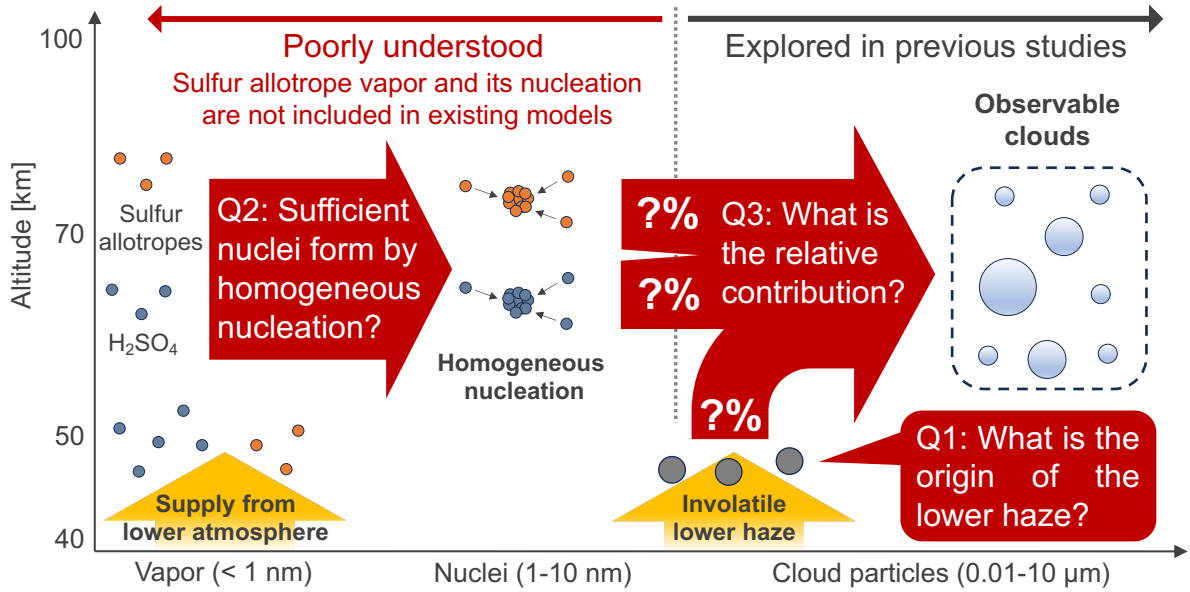


Figure 1.23: Objectives of this thesis.

multi-component model. In addition, existing models use a numerical scheme prone to numerical diffusion during condensation. This can produce an erroneous size distribution, given the vigorous condensational growth in Venus's lower and middle clouds. Therefore, in Chapter 3, we develop a new multi-component cloud microphysics model with a high-accuracy condensation scheme to address Questions 1-3.

In Chapter 4, we address **Question 1** by simulating lower haze formation. We test two non-sulfuric acid substances as cloud components: sulfur allotrope (S_8) and olivine derived from cosmic dust ablation. We found that cosmic dust ablation is a viable source for the lower haze, whereas S_8 completely evaporates below the cloud deck. Furthermore, we checked the robustness of our conclusion by carrying out parameter studies against factors that might influence the characteristics of the lower haze.

In Chapter 5, we address **Question 2** by simulating cloud formation using both binary H_2SO_4 - H_2O and S_8 homogeneous nucleation. We found that sufficient CN can form through this process to reproduce the observed cloud structure. Furthermore, we conducted case studies with various sets of nuclei sources to answer **Question 3**. We found that S_8 homogeneous nucleation accounts for 80% of the observed clouds, while the remaining 20% is attributed to the lower haze.

Chapter 2

Dependence of Venus' Cloud Structure on Eddy Diffusion and Temperature Profiles

This chapter is based on the following paper, published in the Planetary Science Journal:

Hiroki Karyu, Takeshi Kuroda, Takeshi Imamura, Naoki Terada, Ann Carine Vandaele, Arnaud Mahieux, and Sébastien Viscardy (2024). One-dimensional microphysics model of Venusian clouds from 40 to 100 km: Impact of the middle-atmosphere eddy transport and SOIR temperature profile on the cloud structure. *The Planetary Science Journal*, 5 (3): 57. 10.3847/PSJ/ad25f3

2.1 Model Description

2.1.1 Overview

The model equations used in this study are based on a 1-D cloud microphysics model developed by Imamura and Hashimoto (2001). Note, however, that the purpose of using a 1-D model is different from Imamura and Hashimoto (2001), who reproduced the equatorial atmosphere by incorporating the upward branch of the Hadley circulation into the 1-D model. Here we focus on the globally-averaged vertical structure by representing all vertical transport processes, including large-scale advection such as the Hadley circulation, by eddy diffusion, and thus the advection by vertical winds is absent in the equations. The cloud size distribution is a function of the particle radius r , the altitude z , and the time t . It is calculated based on the 1-D continuity equation defined without the advection term (modified after Imamura & Hashimoto 2001):

$$\begin{aligned}
\frac{\partial C(r, z, t)}{\partial t} = & \frac{\partial}{\partial z} \left[K_{\text{eddy}}(z) \rho(z) \frac{\partial}{\partial z} \left(\frac{C(r, z, t)}{\rho(z)} \right) \right] - \frac{\partial}{\partial z} [w_{\text{sed}}(r, z) C(r, z, t)] \\
& - \frac{\partial}{\partial r} [G(r, z, t) C(r, z, t)] \\
& + \frac{1}{2} \int_{r_{\text{CN}}}^r K_{\text{coag}}(r', r - r') C(r', z, t) C(r - r', z, t) dr' \\
& - C(r, z, t) \int_{r_{\text{CN}}}^{r_{\text{max}}} K_{\text{coag}}(r', r) C(r', z, t) dr',
\end{aligned} \tag{2.1}$$

where $C(r, z, t)$ is the cloud particle number density (m^{-3}), $\rho(z)$ is the atmospheric density (kg m^{-3}), $K_{\text{eddy}}(z)$ is the vertical eddy diffusion coefficient ($\text{m}^2 \text{s}^{-1}$) of air, $w_{\text{sed}}(r, z)$ is the sedimentation velocity (m s^{-1}) of cloud particles, $G(r, z, t)$ is the condensational radial growth rate (m s^{-1}), $K_{\text{coag}}(r, r')$ is the coagulation kernel ($\text{m}^3 \text{s}^{-1}$) between cloud particles with radius r and r' , r_{CN} and r_{max} are the radii (m) of condensational nucleus and particle with maximum size, respectively. The first term on the right-hand side is vertical transport by eddy diffusion. The second term accounts for vertical transport due to particle sedimentation. The third term represents the growth through condensation, whose numerical expression is introduced in Appendix B.1. The fourth and fifth terms represent particle size evolution resulting from coagulation. The model simulates dynamical and cloud microphysical processes in a 1-D column of CO_2 atmosphere from 40 to 100 km. As a major update from the previous work by Imamura and Hashimoto (2001), we extended the model top altitude from 70 km to 100 km to include the upper haze layer. CN and cloud droplets are modelled with discrete size bins. The size bins are divided into 23 bins between 0.17 and $\sim 30 \mu\text{m}$ in which the volume doubles from one bin to the next. The smallest size bin represents Mode 1 particles, which are assumed to be insoluble CN. Size bins larger than the smallest bin represent droplets, whose liquid composition consists of a binary solution of H_2SO_4 and H_2O .

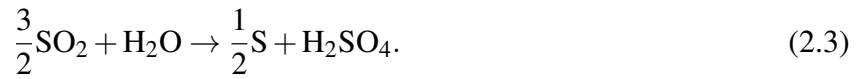
The coagulation process includes Brownian diffusion whose numerical expression is introduced in Appendix A.1. The CN is treated the same way as in the earlier work (Imamura and Hashimoto, 2001). Particles in the smallest size bin act as the CN. These CN are assumed not to coagulate with each other for simplicity because their composition and microphysical property are not known. Thermal equilibrium is assumed when calculating the H_2SO_4 mass fraction in the cloud droplets, considering that equilibrium adjustment is achieved by the exchange of H_2O molecules between the droplet surface and the surrounding environment.

Regarding the sedimentation process, while the previous work by Imamura and Hashimoto (2001) used the Stokes velocity, we implemented the Cunningham slip-flow correction as follows (Jacobson, 2005):

$$w_{\text{sed}}(r, z) = \frac{2r^2 \rho_p g}{9\eta} \left[1 + \text{Kn}(z) \left\{ A + B \exp \left(-\frac{C}{\text{Kn}(z)} \right) \right\} \right], \tag{2.2}$$

where ρ_p is the density (kg m^{-3}) of the particle, $g = 8.7 \text{ m s}^{-2}$ is the gravitational acceleration, $\eta = 1.5 \times 10^{-5} \text{ kg m}^{-1} \text{ s}^{-1}$ is the viscosity of CO_2 gas, A , B , and C are experimental parameters set to be 1.249, 0.42, and 0.87 following Kasten (1968), $\text{Kn}(z)$ is the Knudsen number for CO_2 atmosphere. The inclusion of the Cunningham slip-flow correction is crucial for accurately estimating sedimentation velocity in thin air due to strong dependency of Kn on altitude. This effect has limited impact on particle size distribution between 40 and 70 km, and it was not included in earlier work by Imamura and Hashimoto (2001).

We also consider the chemical production of H_2SO_4 vapor and loss of H_2O vapor through the following net reaction, as introduced as Equation (1.11) but reintroduced for the sake of convenience, as follows:



It demonstrates that the loss of one H_2O molecule results in the production of one H_2SO_4 molecule. We updated the production rate of H_2SO_4 using the photochemical model results from Krasnopolsky (2012). The updated production rate is a globally averaged value taken from 45° latitude, consistent with the other settings in the present study. The column integrated production rate is about $5.7 \times 10^{11} \text{ cm}^{-2} \text{ s}^{-1}$, which is approximately half of the rate used in the study by Imamura and Hashimoto (2001). In addition, the peak altitude of the production rate is located at $\sim 66 \text{ km}$ following Krasnopolsky (2012), while it was situated at 61 km in the previous work. We also adopted the CN production rate introduced by Imamura and Hashimoto (2001), who assumed that the CN are made of elemental sulfur produced by Reaction 2.3 and have the same radius as particles in Mode 1. The production rate of condensation nuclei P_{CN} is expressed in the same way as Equation (1.15) but reintroduced for convenience, as follows:

$$P_{\text{CN}}(z) = \frac{M_{\text{S}} P_{\text{S}}(z)}{\frac{4}{3} \pi \rho_{\text{CN}} r_{\text{CN}}^3}, \quad (2.4)$$

where $P_{\text{S}}(z)$ is the H_2SO_4 vapor production rate ($\text{m}^{-3} \text{ s}^{-1}$), r_{CN} and ρ_{CN} are respectively the radius (m) and mass density (kg m^{-3}) of condensation nuclei, and M_{S} is the molecular mass (kg) of elemental sulfur. The photochemical production rate profiles of H_2SO_4 vapor and the CN are shown in Figure 2.1. It should be noted that if the production rate of CN matches that of Reaction (2.3), it implies the unavailability of elemental sulfur for the chemistry of this cycle. This is a simplistic assumption, and not corroborated by current findings on the net sulfur production from photochemical models. These models also fail to account for the potential loss of sulfur into droplets, where sulfur primarily reacts with O_2 . However, interaction between the sulfur chemical cycle and CN production is out of the scope of this chapter. Selecting only one CN size bin may overlook the impact of CN on microphysical processes. Knollenberg and Hunten (1980) found a wide size distribution of CN that includes significant numbers of particles both larger and smaller than $0.17 \mu\text{m}$. Larger CN might represent a non-negligible sink of H_2SO_4 and H_2O , while smaller CN might be less relevant due to the

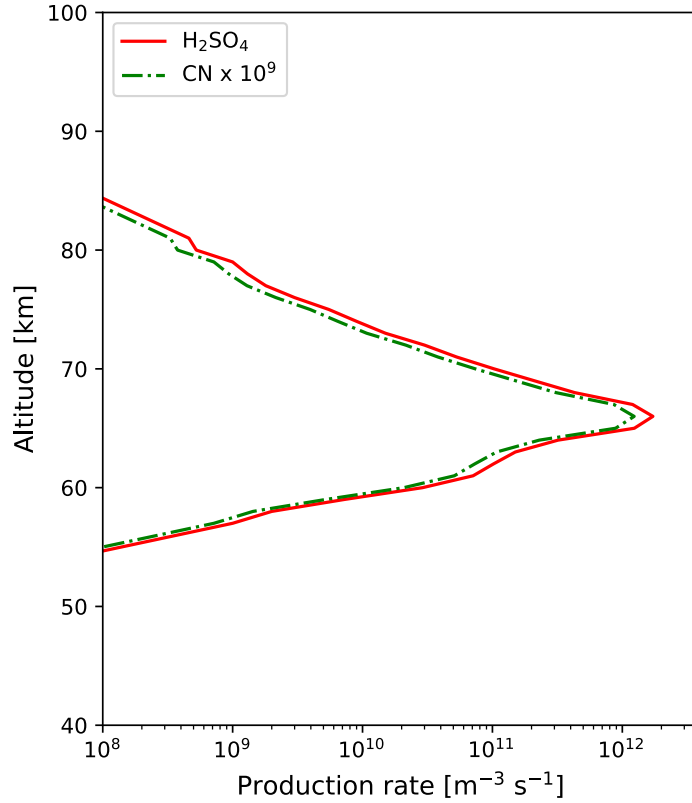


Figure 2.1: Vertical profiles of H_2SO_4 vapor (red solid line) and CN (green dashed dotted line) production rates used in the model. The H_2SO_4 vapor production rate is adapted from Krasnopolsky (2012).

Kelvin effect. Moreover, it is improbable that CN with radii as large as 0.17 microns originate directly from photochemical processes because the initial products are considerably smaller and contain far fewer molecules. These particles grow to sizes around 0.17 microns over time, leading to a broad size distribution. Thus, the simplified treatment of CN in this chapter may overestimate the number of photochemically produced CN. These effects are investigated by incorporating CN size distribution in Chapter 5. The detailed settings for cloud microphysical processes and underlying chemistry are adapted from Imamura and Hashimoto (2001); we refer the reader to this paper for more details. In the following subsections, we outline the parameters used in case studies and describe the calculation settings.

2.1.2 Eddy diffusion profiles

The nominal eddy diffusion profile is established based on the past estimations (see Figure 2.2). Eddy diffusion is not an observable quantity; rather, it serves as a parameterization representing atmospheric dynamics across a range of scales. The past radio scintillation measurements estimated the contribu-

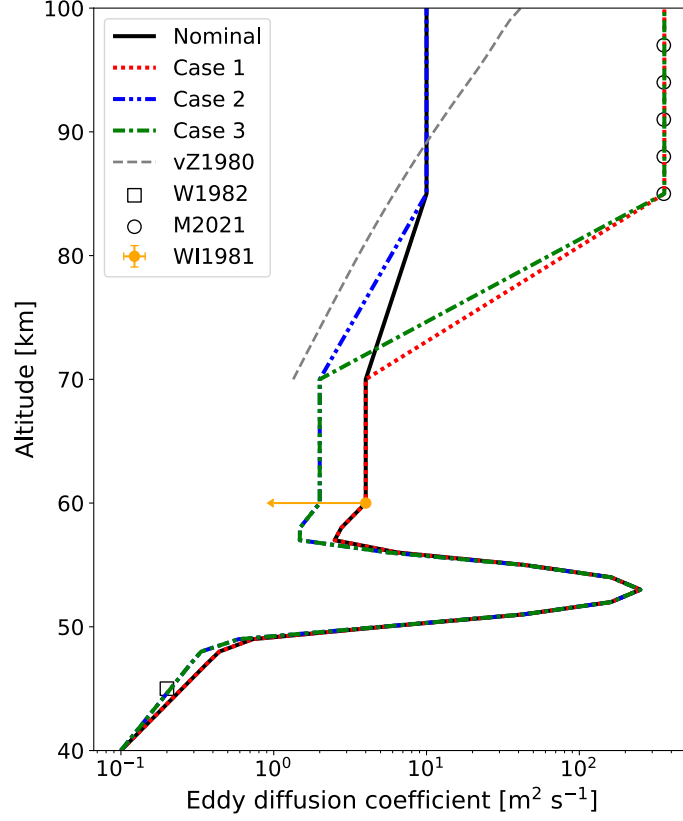


Figure 2.2: Vertical profiles of eddy diffusion coefficient for the nominal case (black solid line), Case 1 (red dotted line), Case 2 (blue dashed double dotted line), and Case 3 (green dashed dotted line) with estimated values by von Zahn et al. (1980), Woo et al. (1982), Mahieux et al. (2021), and Woo and Ishimaru (1981), labeled as vZ1980, W1982, M2021, and WI1981, respectively. The arrow of WI1981 indicates that the value is the upper limit.

tion of small-scale turbulence to the diffusion coefficient to be $\sim 0.2 \text{ m}^2 \text{ s}^{-1}$ at 45 km (Woo et al., 1982) and $\sim 4 \text{ m}^2 \text{ s}^{-1}$ at 60 km (Woo and Ishimaru, 1981). To match the estimated eddy diffusion profile, we interpolate the eddy diffusion coefficient from $0.1 \text{ m}^2 \text{ s}^{-1}$ at the bottom boundary to $4 \text{ m}^2 \text{ s}^{-1}$ at 60 km, resulting in an eddy diffusion of $\sim 0.2 \text{ m}^2 \text{ s}^{-1}$ at 45 km, consistent with past observations. To account for convective mixing in the cloud due to infrared heating of the cloud base, which is indicated by the neutral static stability observed by entry probes (e.g., Seiff et al., 1980) and the PV, VEx, and Akatsuki radio occultation (Kliore and Patel, 1982; Tellmann et al., 2009; Imamura et al., 2017), we used an enhanced eddy diffusion coefficient of $250 \text{ m}^2 \text{ s}^{-1}$ centered at 53 km, following the earlier work by Imamura and Hashimoto (2001). It is important to note that earlier theoretical research by Baker et al. (1999, 2000a,b) indicated that downwelling plumes within the convective layer may extend into the sub-cloud region, potentially causing substantial vertical mixing. Such convective mixing can alter the size distribution, as reported in the studies by Imamura and Hashimoto

Table 2.1: List of case studies and the corresponding eddy diffusion and temperature profiles

Case name	Eddy diffusion coefficient at 60-70 km ($\text{m}^2 \text{s}^{-1}$)	Eddy diffusion coefficient at 85-100 km ($\text{m}^2 \text{s}^{-1}$)	Temperature profile
Nominal	4 ^[a]	10 ^[b]	VIRA ^[d]
Case 1	4 ^[a]	360 ^[c]	VIRA ^[d]
Case 2	2	10 ^[b]	VIRA ^[d]
Case 3	2	360 ^[c]	VIRA ^[d]
Case 4	4 ^[a]	10 ^[b]	VEx/SOIR ^[e]
Case 5	2	360 ^[c]	VEx/SOIR ^[e]

^a Woo and Ishimaru (1981)

^b von Zahn et al. (1980)

^c Mahieux et al. (2021)

^d Seiff et al. (1985)

^e Mahieux et al. (2015)

(2001) and McGouldrick and Toon (2007). While this effect is not extensively examined in the current study, its possible impacts on cloud microphysics will be addressed in Section 2.2.1. Between 60 and 70 km, the eddy diffusion profile is held constant at the 60 km value for the convenience of parameter studies. The eddy diffusion above 85 km is determined based on past estimations by von Zahn et al. (1980). The eddy diffusion coefficient was derived by comparing observed gas species profiles with those calculated numerically using a 1-D diffusion equation. The formulated coefficient was $\sim 1.4 \times 10^{13} [M]^{-0.5} \text{ cm}^2 \text{ s}^{-1}$, yielding a value of $\sim 10 \text{ m}^2 \text{ s}^{-1}$ at 90 km. Thus, we use this value as a typical eddy diffusion coefficient above 85 km. The eddy diffusion coefficient between 70 and 85 km is derived by interpolating the two values.

2.1.3 Background atmosphere

The background temperature and pressure profiles are taken from the Venus International Reference Atmosphere (VIRA) (Seiff et al., 1985) for the nominal case and eddy diffusion case studies. We chose the mid-latitude profiles to represent the global average of the background atmosphere, as shown in Figure 2.3a. The temperature and pressure profiles are held constant in the calculation.

In addition, we defined case studies using a different temperature profile obtained by the VEx observations. Bertaux et al. (2007) found a warm layer in the Venus mesosphere using the SOIR dataset, indicating that the temperature above 85 km could be up to 50 K higher than previously reported. Subsequent studies also confirmed the existence of this warm layer during the course of VEx observations (Mahieux et al., 2012, 2015, 2023b). Mahieux et al. (2023b) proposed that the observed significant temperature differences between the VIRA and SOIR data can be attributed to the variations in temperature distribution with respect to local time. The temperature difference between the VIRA and SOIR could impact particle and gas distributions by changing the saturation vapor

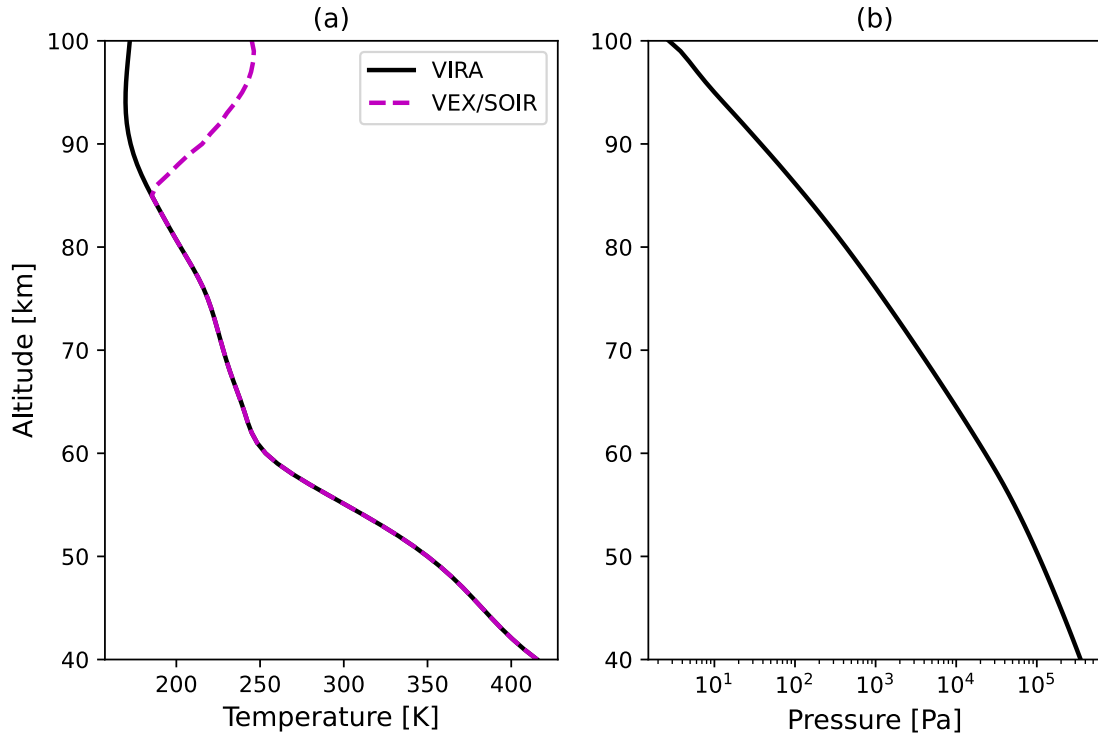


Figure 2.3: (a) Vertical profiles of temperature used in the model. The black solid line represents the temperature profile adapted from Seiff et al. (1985) and is used for the nominal case as well as for Case 1, Case 2, and Case 3. The magenta dashed line represents the temperature profile above 85 km adapted from Mahieux et al. (2015) and is used for Case 4 and Case 5. (b) Vertical profile of pressure Seiff et al. (1985) used in the model.

pressure. Therefore, we considered the mid-latitude temperature profile from Mahieux et al. (2015) to test the sensitivity towards temperature (Figure 2.3a) in Case 4 and Case 5. The SOIR temperature profiles are confined to altitudes above 80 km. To establish temperature distributions for all modeled altitudes, we connected these profiles at the point where the distributions from VIRa and SOIR intersect. We use the same eddy diffusion coefficient for the nominal case for Case 4 as a control experiment with the SOIR temperature profile. On the other hand, Case 5 uses the same eddy diffusion coefficient as in Case 3, as it well replicates the observed particle and gas distributions. The name of each case (nominal, Case 1, Case 2, Case 3, Case 4, and Case 5) along with their corresponding parameters is listed in Table 2.1. It is important to note that the SOIR temperature profiles exhibit inter-profile variability. This variation can impact condensation and evaporation processes, potentially resulting in changes to aerosol size and acidity. However, the study of the microphysical response to such variation is beyond the scope of our current research.

2.1.4 Boundary and initial conditions

Table 2.2 shows the initial and boundary conditions in our simulations, which are determined based on past observations. The H_2O vapor VMR is reported to be ~ 30 ppm below 40 km (Arney et al., 2014;

Table 2.2: Boundary conditions

Altitude	CN number density	Droplet number density	H ₂ SO ₄ VMR	H ₂ O VMR
40 km	40 cm ⁻³	0 cm ⁻³	4 ppm	30 ppm
100 km	$\frac{\partial f_{\text{CN}}}{\partial z} = 0$	$\frac{\partial f_{\text{droplet}}}{\partial z} = 0$	$\frac{\partial f_{\text{H}_2\text{SO}_4}}{\partial z} = 0$	$\frac{\partial f_{\text{H}_2\text{O}}}{\partial z} = 0$

Bézard et al., 2011; Chamberlain et al., 2013; Fedorova et al., 2015; Marcq et al., 2008). We take this value as the bottom boundary condition for the H₂O vapor. The bottom boundary for the H₂SO₄ vapor VMR is set at 4 ppm, based on previous observations from both Magellan (Kolodner and Steffes, 1998) and VEx (Oschlisniok et al., 2021). The in-situ observations reported by Knollenberg and Hunten (1980) suggested that the Mode 1 particles extended below the cloud layer. These particles are assumed to be involatile since H₂SO₄ vapor cannot exist in the liquid phase within the sub-cloud region. Therefore, the particle number density for the smallest radius is set at 40 cm⁻³, based on the number density of the Mode 1 particles from in-situ observations. The mixing ratio gradient of cloud particles, H₂SO₄ vapor, and H₂O vapor are set to be zero at the top boundary. The initial conditions for H₂O, H₂SO₄ VMR, and the smallest radius bin are set to match the bottom boundary values across all vertical grids. The aerosol and gas species profiles reach a steady state within ~ 50 Venus years (11250 Earth days) starting from these initial conditions. This time scale is longer than the time scales of the physical processes incorporated within the model such as eddy diffusion, sedimentation, condensation, and coagulation. We performed the simulation for 100 Venus years (22500 Earth days) and present the steady state results in the next section.

2.2 Model Results

2.2.1 Sensitivity to eddy diffusion profiles

Figures 2.4 and 2.5 show the simulated size distribution in the nominal case and Case 1. The particle size distribution can be divided into three regions by its formation processes, being consistent with the previous microphysics studies (e.g., Gao et al., 2014; McGouldrick and Barth, 2023): the lower and middle cloud (47-57 km), the upper cloud (57-70 km), and the upper haze (>70 km) regions. The upper cloud particles are formed through the condensation of photochemically produced H₂SO₄ vapor onto CN, which are also produced photochemically within the same altitude range. The calculated mode radius for the upper cloud layer is ~ 0.7 μm , which corresponds to Mode 2 particles but is somewhat underestimated. The lower and middle clouds exhibit a bimodal size distribution, with peaks at the smallest radius bin (0.17 μm) and ~ 3 μm , corresponding to Mode 1 and Mode 3, respectively (Figures 2.5a and 2.5c). Between 51 and 53 km, the Kelvin effect inhibits the condensational growth of sub-micron particles, allowing larger particles to grow. The evaporation of the sub-micron particles and the upward diffusion transport of CN from the bottom boundary contributes to the formation of the peak at the smallest bin of 0.17 μm . Conversely, the larger peak forms as a result of condensational growth of the photochemically produced particles descending from the upper cloud

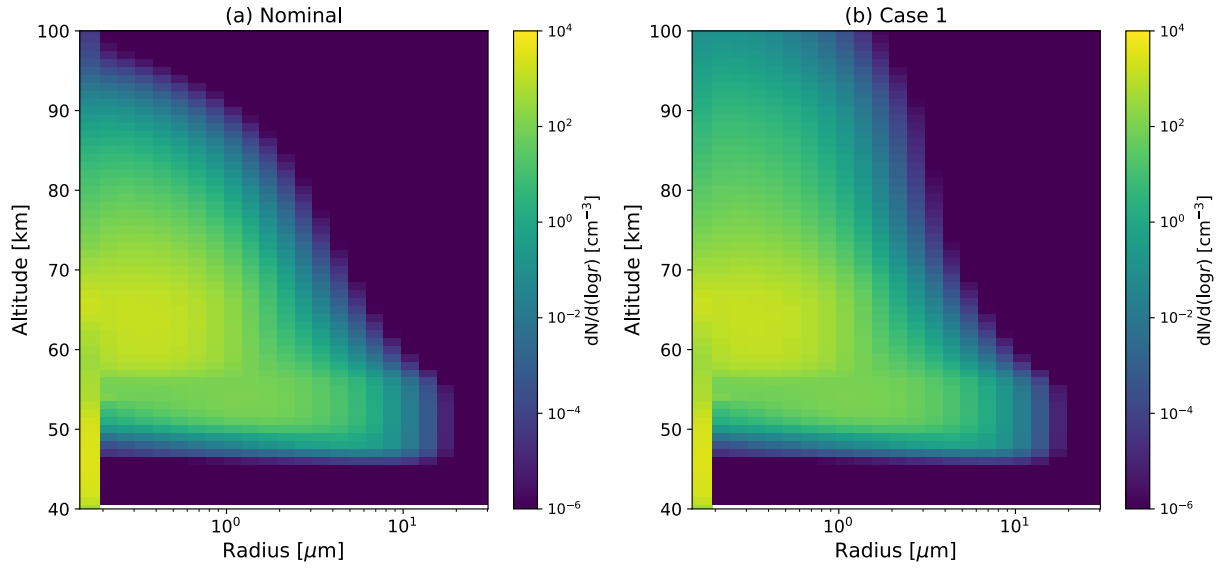


Figure 2.4: The simulated particle size distribution as a function of radius and altitude in (a) the nominal case and (b) Case 1.

region. The condensation process is driven by H_2SO_4 -rich air ascending from lower altitudes in the convective layer, where strong eddy diffusion is assumed (see Figure 2.2).

Above 53 km, the H_2SO_4 supersaturation becomes sufficiently high to suppress the Kelvin effect, allowing all particles to undergo condensational growth (Figure 2.5b). Consequently, the smaller peak clearly seen below 53 km altitude eventually merges into the size distribution of the upper cloud. Below 51 km altitude, all particles start to evaporate because of the increasing H_2SO_4 saturation vapor pressure. The evaporation gradually decreases the droplet number density, leading to the near-complete disappearance of droplets around the cloud base at around 47 km altitude.

Figure 2.5 demonstrates that the size distribution simulated in the present study closely agrees with earlier work by Imamura and Hashimoto (2001). They showed that the size distribution in the lower and middle cloud layers is bimodal at steady-state and that a trimodal distribution can form due to convective mixing. Furthermore, Baker et al. (1999) suggested that the microphysical properties of clouds could be altered by condensation processes initiated by cold downwelling plumes from neutrally stable regions in the cloud layers. This implies that the absence of an explicit connection with convective activity in the current study may explain why a trimodal distribution is not observed in these cloud layers.

The upper haze features a broad, unimodal size distribution with a mode radius of $\sim 0.5 \mu\text{m}$ (Figures 2.5e and 2.5f). This radius possibly represents Mode 2 particles as observed by Wilquet et al. (2009) or the effective radius calculated by Luginin et al. (2016), assuming a unimodal size distribution. The particle population in the upper haze is maintained by the upward transport of particles from the upper cloud layer via eddy diffusion.

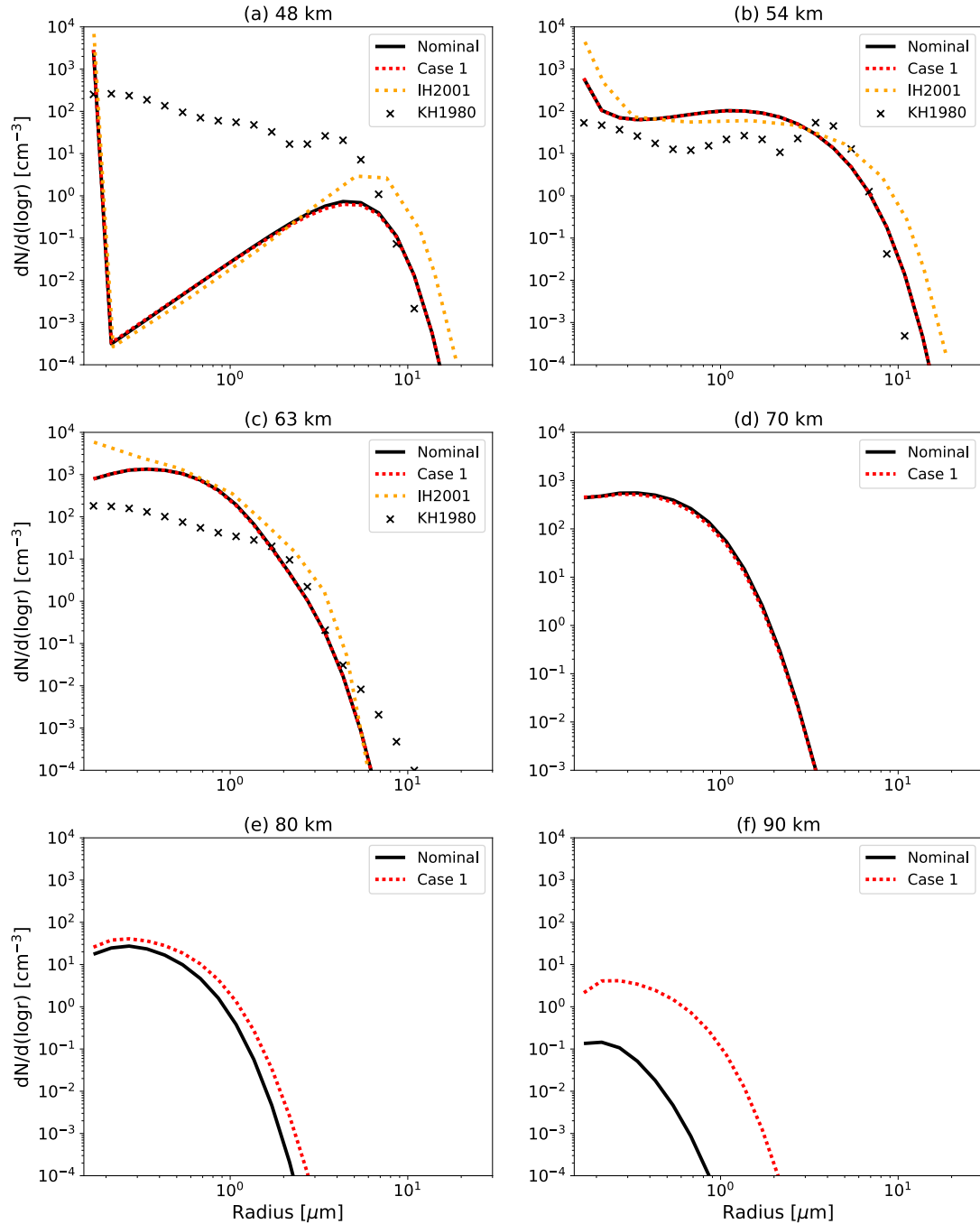


Figure 2.5: The simulated particle size distribution as a function of radius at (a) 48 km, (b) 54 km, (c) 63 km, (d) 70 km, (e) 80 km, and (f) 90 km in the nominal case (black solid line) and Case 1 (red dotted line) in comparison with the previous work by Imamura and Hashimoto (2001) labeled as IH2001. The size distribution obtained by the LCPS (Knollenberg & Hunten 1980) is plotted as KH1980.

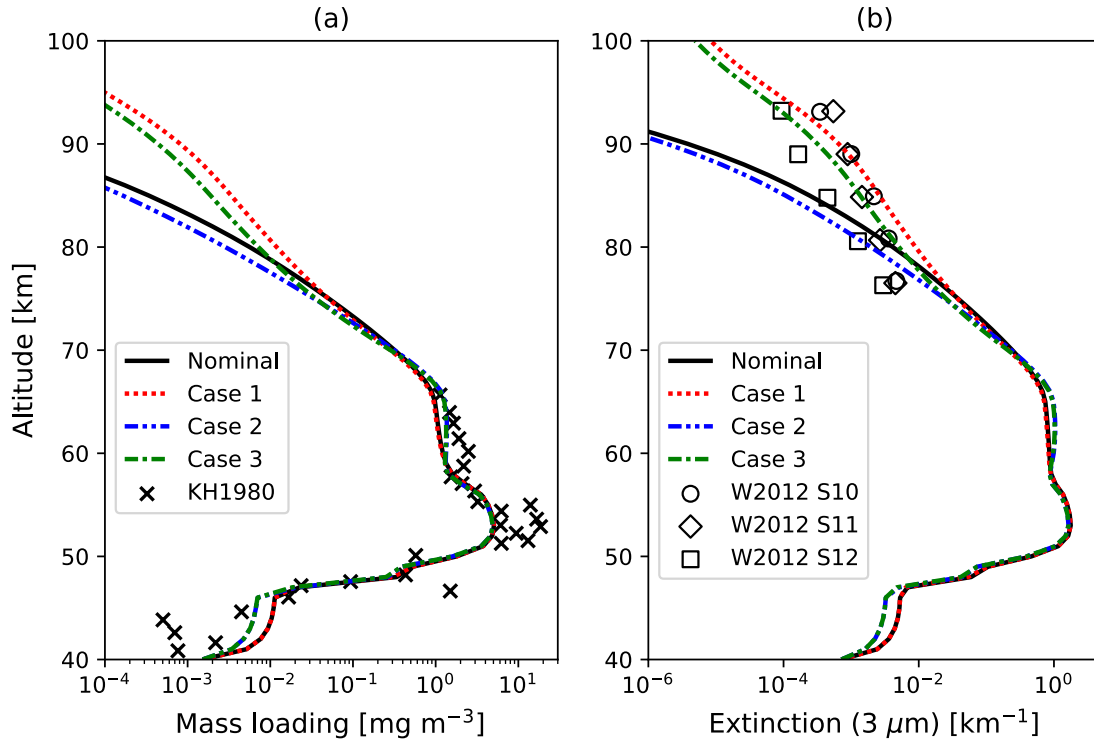


Figure 2.6: (a) Vertical profiles of the simulated mass loading (mg m^{-3}) in the nominal case, Case 1 (red dotted line), Case 2 (blue dashed double dotted line), and Case 3 (green dashed dotted line). The mass loading obtained by the LCPS (Knollenberg and Hunten, 1980) is plotted for comparison as KH1980. (b) Vertical profiles of the simulated extinction coefficient (km^{-1}) at $3 \mu\text{m}$ wavelength in the nominal case (black solid line), Case 1 (red dotted line), Case 2 (blue dashed double dotted line), and Case 3 (green dashed dotted line) in comparison with the extinction coefficient at $3 \mu\text{m}$ wavelength measured by the SPICAV-SOIR (Wilquet et al., 2012) labeled as W2012 (season 10: S10, season 11: S11, season 12: S12).

Changes in the eddy diffusion coefficient above 85 km lead to an enhanced upper haze population. It is evident that the upper haze extends vertically more than in the nominal case because of efficient eddy diffusion transport (Figure 2.4b). The size distribution closely resembles that of the nominal case at 70 km, but the differences between the two cases become increasingly evident at higher altitudes (Figures 2.5d, 2.5e, and 2.5f). The total particle number density of Case 1 is orders of magnitude larger than in the nominal case at 90 km, and larger particles are more abundant in Case 1. This underscores the importance of eddy diffusion transport in shaping the upper haze particle population and its microphysical properties.

Figure 2.6 shows the simulated cloud structure as a function of altitude. To obtain the mass loading, we calculated droplet density of the H_2SO_4 - H_2O solution by interpolating the density of pure H_2O liquid and pure H_2SO_4 liquid based on weight fraction of H_2SO_4 (see Figure 2.9a). For the determination of the extinction coefficient, an open-source software MiePython (Prah, 2023) is used to calculate the Mie scattering extinction efficiency of the aerosols. For the input of Mie scattering

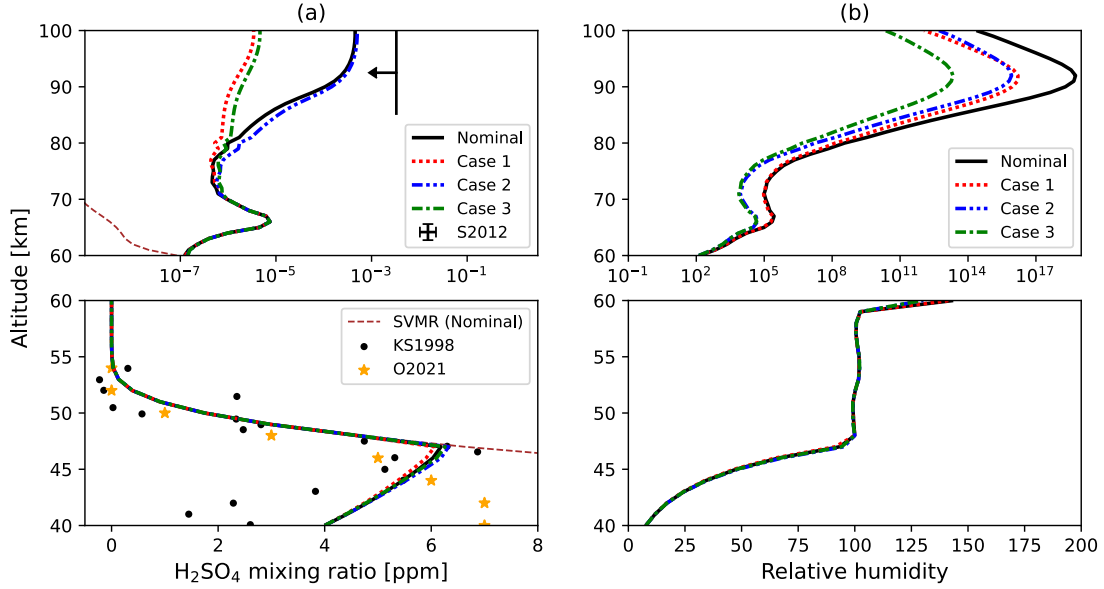


Figure 2.7: (a) Vertical profiles of the simulated H_2SO_4 VMR in the nominal case and Case 1, Case 2, and Case 3 compared with the observations by Magellan radio occultation measurements (Kolodner and Steffes, 1998, labeled as KS1998), VEx/VeRa measurement (Oschlisniok et al., 2021, labeled as O2021), and the upper limit suggested by Sandor et al. (2012) labeled as S2012. The saturation level of H_2SO_4 VMR (SVMR) is also indicated by a brown dashed line for the nominal case. (b) Vertical profiles of the simulated H_2SO_4 relative humidity in the nominal case and Case 1, Case 2, and Case 3.

calculation, we used refractive index from Palmer and Williams (1975) as a function of concentration of H_2SO_4 - H_2O solution. The simulated mass loading in the nominal case is basically consistent with in-situ observations by Knollenberg and Hunten (1980) (Figure 2.6a). The mass loading is highest in the middle cloud layer due to rapid cloud formation induced by convective activity, aligning with in-situ observations. The simulated value is about a factor of 2 smaller than the observed one in the middle cloud layer. The mass loading in the upper cloud is consistent with the observation within a factor of 2, although it is slightly underestimated. The in-situ observations were conducted at the equator, capturing the photochemical production rates of H_2SO_4 vapor and CN in low latitudes. In contrast, our model uses rates from mid-latitudes, which are lower. This likely accounts for the discrepancy between the simulated and observed values.

On the other hand, the nominal case fails to replicate the observed extinction coefficient profile (Figure 2.6b). In the upper haze layer, the extinction coefficient decreases exponentially with altitude, but the simulated gradient is steeper than observed, resulting in values about two orders of magnitude smaller. The discrepancy improves in Case 1 and Case 3 due to the increased eddy diffusion coefficient, leading to a more extended upper haze layer. It is clearly shown that the cases with efficient eddy diffusion is in good agreement with the observed vertical profiles by Wilquet et al. (2012). This suggests that the recently estimated eddy diffusion coefficient (Mahieux et al., 2021) is suitable for the reproduction of the observed upper haze optical properties.

Figure 2.7 and Figure 2.8 show the vertical profiles of condensable gas species. The H_2SO_4 VMR is fixed at 4 ppm at the bottom boundary and takes the maximum value of ~ 6 ppm around the cloud bottom at 47 km for all cases (Figure 2.7a). This peak arises from droplet sedimentation in the lower cloud and subsequent evaporation. Between the cloud bottom and 60 km, the H_2SO_4 VMR aligns with the saturation vapor profile. This pattern agrees with observations from the Magellan mission (Kolodner and Steffes, 1998) and recent VEx data (Oschlisniok et al., 2021), suggesting H_2SO_4 VMR levels of 5-7 ppm in the sub-cloud region at mid-latitudes. Above 75 km, the simulated H_2SO_4 VMR presents two distinct profiles. The first includes the nominal and Case 2 scenarios with a maximum VMR of $\sim 5 \times 10^{-4}$ ppm; the second includes Case 1 and Case 3 with a maximum VMR of $\sim 5 \times 10^{-6}$ ppm in the upper haze layer. The latter cases feature a higher eddy diffusion coefficient above 75 km, leading to a more abundant upper haze particle population. This facilitates condensation onto these particles as the H_2SO_4 vapor is supersaturated. The peak values for both profile types are lower than the upper limit reported by (Sandor et al., 2012), indicating that H_2SO_4 vapor is not a dominant source of sulfur species in the upper haze layer.

The H_2SO_4 VMR is supersaturated above 60 km and reaches its second-highest value at 67 km, due to the photochemical production of H_2SO_4 vapor across all cases. This finding is consistent with Dai et al. (2022), who used a cloud condensation model without detailed microphysics but assumed the same H_2SO_4 production profile as ours. These results indicate that the supersaturation above 60 km is a robust feature across different models when using the photochemical production of H_2SO_4 vapor from the 1-D photochemical model of Krasnopolsky (2012). They further showed that if a Gaussian distribution is assumed for H_2SO_4 production, the H_2SO_4 VMR closely follows the saturation VMR in the upper haze layer. This is because the Gaussian distribution exhibits an extremely low value outside the half-width of the peak production altitude, and loss by H_2SO_4 condensation dominates over the photochemical production. The earlier work by Imamura and Hashimoto (2001), which also assumed a Gaussian profile, did not find supersaturation except near the peak production altitude. This sensitivity of the H_2SO_4 VMR profile to the assumed photochemical production profile suggests it is a critical factor in modeling.

The H_2O vapor VMR in the nominal case monotonically decreases from 40 km to 67 km and stabilizes at ~ 9 ppm above 67 km (Figure 2.8). This trend aligns qualitatively with VEx observations (Cottini et al., 2012; Fedorova et al., 2008, 2016), although the simulated values are slightly higher. The VMR gradient below the cloud base is steeper than in earlier work by Imamura and Hashimoto (2001) and recent microphysics model results reported by McGouldrick (2017) and McGouldrick and Barth (2023). The reason is that we use a lower eddy diffusion coefficient for those altitudes, based on the previous study by Woo et al. (1982). This steeper H_2O VMR gradient helps balance the H_2O vapor supply by upward eddy diffusion flux against loss from chemical reactions. In the lower and middle cloud layers, the gradient is less steep due to stronger eddy diffusion, which acts to flatten the H_2O vapor VMR profile. The mass fraction of liquid H_2O in the droplets increases (corresponding to a decrease in liquid H_2SO_4 mass fraction, as shown in Figure 2.9a), indicating that the uptake of H_2O vapor by the droplets also contributes to the decreasing H_2O VMR from lower to upper cloud layers. Around 67 km, the H_2O VMR experiences a sharp drop due to efficient chemical loss by H_2SO_4 formation (Equation (2.3) and Figure 2.1).

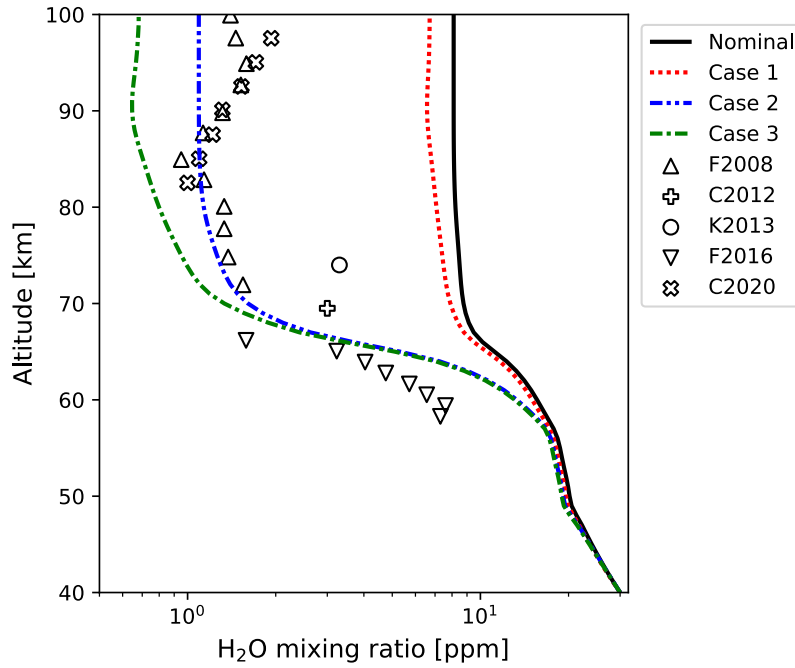


Figure 2.8: Vertical profiles of the simulated H_2O VMR in the nominal case and Case 1, Case 2, and Case 3 compared with observations by SPICAV/SOIR (Fedorova et al., 2008), VIRTIS-H (Cottini et al., 2012), NASA Infrared Telescope Facility (Krasnopolsky et al., 2013), SPICAV/IR (Fedorova et al., 2016), and SPICAV/SOIR (Chamberlain et al., 2020), labeled as F2008, C2012, K2013, F2016, and C2020, respectively.

The H_2O VMR is highly sensitive to the assumed eddy diffusion profile used in the model (Figure 2.8). In Case 2 and Case 3, the H_2O VMR gradient is steeper between 60 and 70 km than in the nominal case and Case 1. Consequently, the H_2O VMR decreases sharply in the upper cloud layer, aligning well with VEx observations (Cottini et al., 2012; Fedorova et al., 2008, 2016) in these two cases. The defining feature of Case 2 and Case 3 is the lower eddy diffusion coefficient between 60 and 70 km, suggesting that the mesospheric H_2O vapor levels are influenced by the efficiency of eddy transport at these altitudes as expected since the chemical loss of H_2O is fixed.

Figure 2.9a shows liquid H_2SO_4 mass fraction in the cloud particles as a function of altitude. Below 50 km, the H_2SO_4 mass fraction exceeds 0.95 and decreases up to 60 km. It then stabilizes at ~ 0.75 from 60 to 75 km in the nominal and Case 1 scenarios, while increasing to ~ 0.8 in Case 2 and Case 3. The simulated values in the cloud layer are consistent with previous observations (Hansen and Hovenier, 1974; Barstow et al., 2012; Cottini et al., 2012) in all cases. In particular, Cases 2 and 3 better match the data from Cottini et al. (2012). This vertical profile is also consistent with reported modeling studies (e.g. Krasnopolsky, 2015; Stolzenbach et al., 2023), although our results are overestimated compared to Stolzenbach et al. (2023) below 60 km. This difference could be caused by the lower temperature bias of their model. Above 75 km, the H_2SO_4 mass fraction decreases again, reaching a minimum around 90 km before rising again up to the model top altitude

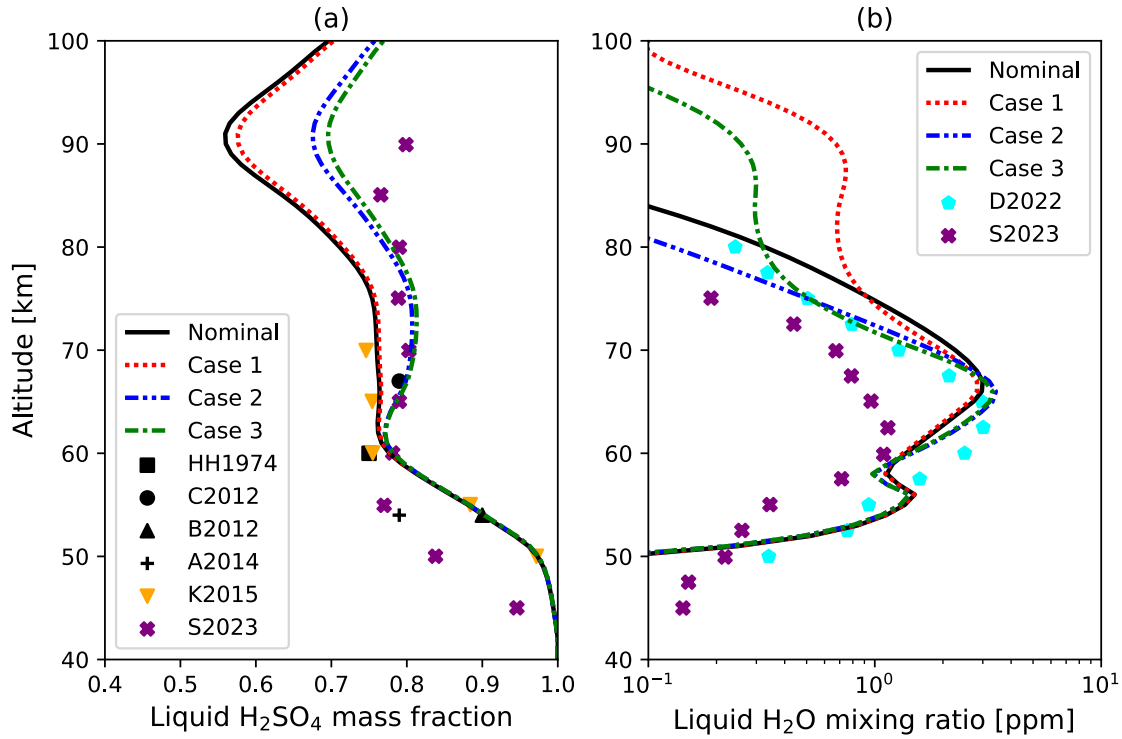


Figure 2.9: (a) Vertical profiles of the simulated liquid H_2SO_4 mass fraction in the nominal case and Case 1, Case 2, and Case 3, in comparison with the modeling studies by Krasnopolsky (2015) and Stolzenbach et al. (2023), labeled as K2015 and S2023, respectively. The observations by Hansen and Hovenier (1974), Cottini et al. (2012), Barstow et al. (2012), and Arney et al. (2014), labeled as HH1974, C2012, B2012, and A2014, respectively. (b) Vertical profiles of the simulated liquid-phase H_2O VMR in the nominal case and Case 1, Case 2, and Case 3 in comparison with the modeling studies by Dai et al. (2022) and Stolzenbach et al. (2023), labeled as D2022 and S2023, respectively.

for all cases. The decrease of the H_2SO_4 mass fraction between 75 and 90 km is attributed to a sharp temperature decrease, as temperature is the key factor determining thermal equilibrium.

The equilibrium condition is also influenced by the background H_2O VMR, as the H_2SO_4 mass fraction adjusts to equilibrium through the uptake or release of H_2O molecules at the particle surface. Therefore, a lower ambient concentration of H_2O molecules results in reduced uptake by the droplets. As shown in Figure 2.8, the H_2O VMR above 60 km is about an order of magnitude lower in Case 2 and Case 3 compared to the nominal case and Case 1. This leads to less H_2O being absorbed into the droplets, and conversely, a higher H_2SO_4 mass fraction within them.

Figure 2.9b shows the H_2O VMR condensed in droplets as a function of altitude. Upon careful examination, the influence of eddy diffusion on the condensed H_2O VMR is noticeable between 60 and 70 km. Specifically, the condensed H_2O VMR is more concentrated around cloud formation altitude around 66 km in Case 2 and 3, due to the reduced eddy diffusion coefficient (see Figure 2.2). Above 70 km, Case 1 and Case 3 exhibit higher levels of liquid H_2O compared to the nominal case

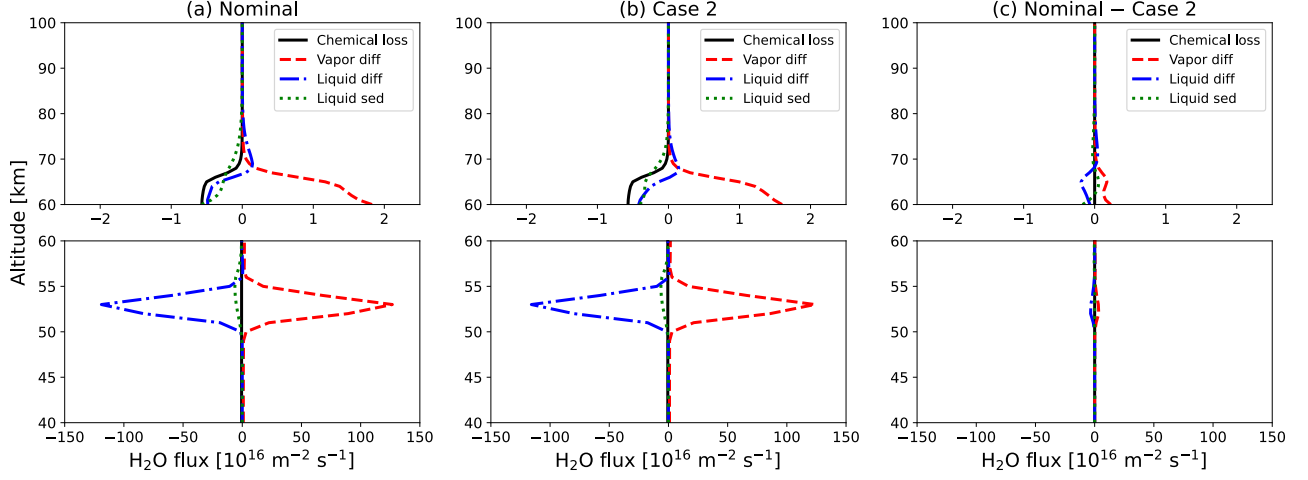


Figure 2.10: Vertical profiles of the simulated cumulative chemical loss of the H_2O vapor (black solid line), H_2O vapor diffusion flux (red dashed line), H_2O liquid diffusion flux (blue dashed dotted line), and H_2O liquid sedimentation flux (green dotted line) in (a) the nominal case and (b) Case 2. (c) Difference of each flux between the nominal case and Case 2 (nominal case minus Case 2 values).

and Case 2. This is attributable to the extended aerosol layer in these two cases facilitated by efficient eddy transport, leading to the upward transport of liquid H_2O in its liquid phase. The condensed H_2O VMR aligns well with the condensation model of Dai et al. (2022) in both Case 2 and Case 3. In their study, the liquid H_2O VMR was ~ 2 ppm in the upper cloud layer, ~ 0.1 ppm at 50 km, and ~ 0.2 ppm at 80 km. Note that the eddy diffusion profile they assumed was $\sim 0.3 \text{ m}^2 \text{ s}^{-1}$ at 40 km, $\sim 1 \text{ m}^2 \text{ s}^{-1}$ at 40-50 km, and $1-6 \text{ m}^2 \text{ s}^{-1}$ at 60-70 km (see Table 2.3), resulting in a similar eddy transport timescale to our model.

A twofold difference in the eddy diffusion coefficient in the upper cloud layer leads to an order-of-magnitude difference in H_2O VMR in the mesosphere, as shown in Figure 2.8. To investigate the sensitivity of H_2O vertical profile with eddy diffusion, we compared the H_2O flux profiles for the nominal case and Case 2 in Figure 2.10; positive fluxes indicate upward transport, and negative fluxes indicate downward transport. Below 60 km altitude, both profiles are identical since the eddy diffusion coefficient is nearly the same at these altitudes. It is noteworthy that the flux balances for the two cases are also similar between 60 and 70 km, even though the eddy diffusion coefficient for Case 2 is twice as large as that of the nominal case. This can be explained as follows. In a steady-state condition, the upward H_2O vapor flux essentially balances with the sum of the liquid diffusion flux, liquid sedimentation flux, and cumulative chemical loss. However, as demonstrated in Figure 2.10c, the difference in liquid flux between the two cases is relatively small, accounting for less than 20% difference in the total sum of the liquid flux and chemical loss. Given that the cumulative chemical loss is a constant parameter in all cases, the sums of the liquid flux and the cumulative chemical loss end up being similar. Consequently, the upward vapor fluxes are also similar, as they are balanced with the downward fluxes.

Table 2.3: Comparison of eddy diffusion in this study and Dai et al. (2022)

Altitude (km)	Eddy diffusion in Case 2 and Case 3 ($\text{m}^2 \text{s}^{-1}$)	Eddy diffusion in Dai et al. (2022) ($\text{m}^2 \text{s}^{-1}$)
40 km	0.1	~ 0.3
50 km	5	~ 0.7
60 km	2	~ 1
70 km	2	~ 6

The two different H_2O VMR trends shown in Figure 2.8 result from the fact that all cases exhibit similar upward H_2O vapor fluxes between 60 and 70 km. In the present study, the species in the gas phase are transported only by eddy diffusion. The H_2O vapor flux due to eddy diffusion $\Phi_{\text{H}_2\text{O}}$ is expressed as $\Phi_{\text{H}_2\text{O}} = K_{\text{eddy}} n_a \frac{\partial f_{\text{H}_2\text{O}}}{\partial z}$, where K_{eddy} is the eddy diffusion coefficient, n_a is the number density of air, $f_{\text{H}_2\text{O}}$ is the H_2O VMR. As shown in Figure 2.10, $\Phi_{\text{H}_2\text{O}}$ does not differ significantly in each case and can be regarded as a constant for simplicity. If K_{eddy} is increased by a factor of 2, the H_2O VMR gradient $\frac{\partial f_{\text{H}_2\text{O}}}{\partial z}$ must be divided by two to yield the same vapor flux $\Phi_{\text{H}_2\text{O}}$. This is why a twofold difference in the eddy diffusion coefficient leads to a significant change in the extent of H_2O vapor depletion in the mesosphere. Consequently, the steep gradient reduces the H_2O VMR in the upper cloud layer, leading to an H_2O VMR that is about an order of magnitude smaller above 70 km in both Case 2 and Case 3.

In the present study, the chemical production/loss rate of condensable gas species is fixed. However, as shown in Figure 2.10, the cumulative H_2O chemical loss also plays a significant role in determining the compensating upward H_2O vapor flux. Namely, if the chemical loss rate is increased and K_{eddy} is fixed, $\frac{\partial f_{\text{H}_2\text{O}}}{\partial z}$ should increase to balance with the chemical loss. Given the amplitude of the cumulative chemical loss compared to the liquid flux components, it is expected that a factor of variation of the loss rate may lead to a considerable change in $\frac{\partial f_{\text{H}_2\text{O}}}{\partial z}$.

The recent cloud condensation model from Dai et al. (2022) showed a similar relationship between eddy diffusion and the H_2O gradient in the upper cloud layer. With the same H_2O chemical loss profile as in our study, the nominal case from Dai et al. (2022) used an eddy diffusion coefficient of $1 \text{ m}^2 \text{s}^{-1}$ at 60 km and $\sim 3 \text{ m}^2 \text{s}^{-1}$ at 65 km. This suggests that the efficiency of eddy transport in their study aligns closely with ours. Their resulting H_2O VMR gradient between 60 and 70 km was steep enough to reduce the H_2O VMR to a few ppm at 70 km. In contrast, when they used the eddy diffusion profile from Bierson and Zhang (2020), which had a value of $\sim 4 \text{ m}^2 \text{s}^{-1}$ between 60 and 70 km, they observed a less steep H_2O gradient. The resulting H_2O VMR exceeded 10 ppm at 70 km. Our results also overestimated the H_2O VMR at 70 km when the eddy diffusion is set to $4 \text{ m}^2 \text{s}^{-1}$ between 60 and 70 km. The similarities between our findings and those from Dai et al. (2022) highlight the critical role of the eddy diffusion coefficient in the upper cloud layer. Furthermore, both

studies concur that an eddy diffusion of $\sim 2 \text{ m}^2 \text{ s}^{-1}$ serves as a threshold beyond which hygroscopic effects become significant.

The influence of eddy diffusion on the chemical production/loss rates should also be explored. It has been established that the vertical profiles of sulfur species are significantly influenced by eddy diffusion. Krasnopolsky (2012) demonstrated that variations in eddy diffusion coefficients above 55 km can lead to a significant difference in SO_2 concentrations in the upper cloud region. Additionally, Bierson and Zhang (2020) found that the altitude and rate of H_2SO_4 production vary considerably with the eddy diffusion coefficient in cloud layers. Given the vital role of the chemical production profile in the formation of the upper cloud and the depletion of H_2O , the H_2SO_4 and H_2O fluxes could be altered by the response of sulfur chemistry to the eddy diffusion. However, here we focus on the impact of changing eddy diffusion on cloud microphysics and its effect on the H_2SO_4 and H_2O fluxes, and the coupling between photochemistry and cloud microphysics is left for future research.

2.2.2 Sensitivity to upper temperature profiles

We examined the sensitivity of aerosol and gas species distributions to variations in the upper temperature profile by comparing VIRA temperature cases (nominal case and Case 3) and SOIR temperature cases (Case 4 and Case 5). The mass loading and extinction coefficient in both scenarios are very similar up to 80 km and are in good agreement with the LCPS observations, as shown in Figure 2.11. Above 80 km, the upper haze is more extended in Case 3 and Case 5 because both cases use the updated eddy diffusion by Mahieux et al. (2021), being consistent with the results shown in Section 2.2.1. However, we observed that the population of the upper haze particles is slightly reduced when using the temperature profile from SOIR. As will be discussed later, this reduction is due to the aerosol evaporation, which is triggered by the warm layer evident in the SOIR temperature profile.

The H_2SO_4 VMR in Case 5, which uses the SOIR temperature profile, closely resembles that of Case 3 up to ~ 85 km altitude but diverges significantly from Case 3 above 85 km (see Figure 2.12a). On the other hand, the H_2SO_4 VMR in Case 4 is larger than the nominal case, but the amplitude is much less pronounced. The saturation H_2SO_4 VMR calculated using the SOIR profile (Case 5) is orders of magnitude higher than that calculated using the VIRA profile, as the maximum temperature of the SOIR profile is up to ~ 50 K higher than that in the VIRA profile. Due to this elevated saturation VMR, the H_2SO_4 vapor is undersaturated in the upper haze layer above ~ 90 km in Case 4 and Case 5 (Figure 2.12b). This undersaturation leads to the evaporation of the upper haze particles, in turn increasing the H_2SO_4 VMR in the SOIR temperature cases. However, the deviation from the VIRA temperature case is more evident in Case 5 than in Case 4 because H_2SO_4 is supplied by the extended upper haze layer. As a result of the combined effect of enhanced eddy diffusion and higher temperature, the H_2SO_4 VMR aligns with the upper limit suggested by Sandor et al. (2012) in Case 5, suggesting that mesospheric H_2SO_4 vapor could be detectable under high-temperature conditions.

Previous photochemical studies suggested that ~ 1 ppm of H_2SO_4 vapor is required to act as a precursor for SO_2 and SO in the upper haze layer (Zhang et al., 2010, 2012). However, the simulated

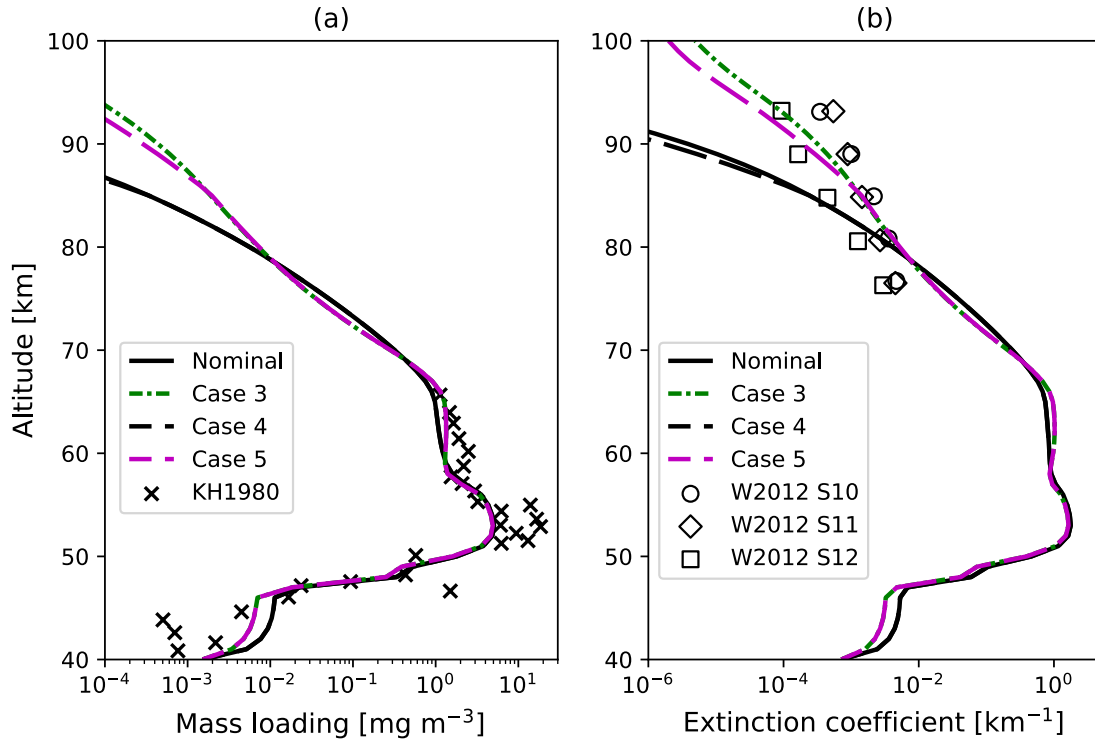


Figure 2.11: Same as Figure 2.6, but the simulated results for Case 4 and Case 5 are added with black and magenta dashed lines, and the Case 1 and Case 2 results are removed for visibility.

values are orders of magnitude lower than this suggested value, even when using the SOIR temperature (see Figure 2.12a). This discrepancy raises the possibility that sources other than H_2SO_4 aerosol may be needed to provide sufficient sulfur, such as elemental sulfur suggested by Zhang et al. (2012).

The H_2O VMR in Case 4 does not deviate from one in the nominal case, as also seen in H_2SO_4 VMR. This is attributed to the lack of aerosols in the upper haze layer in both cases. However, when Case 3 and Case 5 are compared, the H_2O VMR simulated with the SOIR temperature profile diverges notably from that simulated using the VIRA profile, especially above ~ 85 km (Figure 2.3a). In Case 5, the H_2O VMR reaches a minimum value at ~ 85 km and then increases with altitude. This pattern aligns with the H_2O VMR profile observed by SPICAV-SOIR (Fedorova et al., 2008; Chamberlain et al., 2020; Mahieux et al., 2023b, 2024). The increase in H_2O VMR above 85 km can be attributed to the strong dependence of equilibrium conditions on the background temperature. It is important to note that the diverging altitude can vary depending on how the temperature profile is constructed in the model and may also change when taking into account the inter-profile variability discussed in Section 2.1.3.

The equilibrium condition manifests in the liquid H_2SO_4 mass fraction within the droplets. In Case 4 and Case 5, there is a significant increase in the H_2SO_4 mass fraction above 85 km, corresponding to the sharp temperature increase in the SOIR profile (Figure 2.14a). This suggests that the

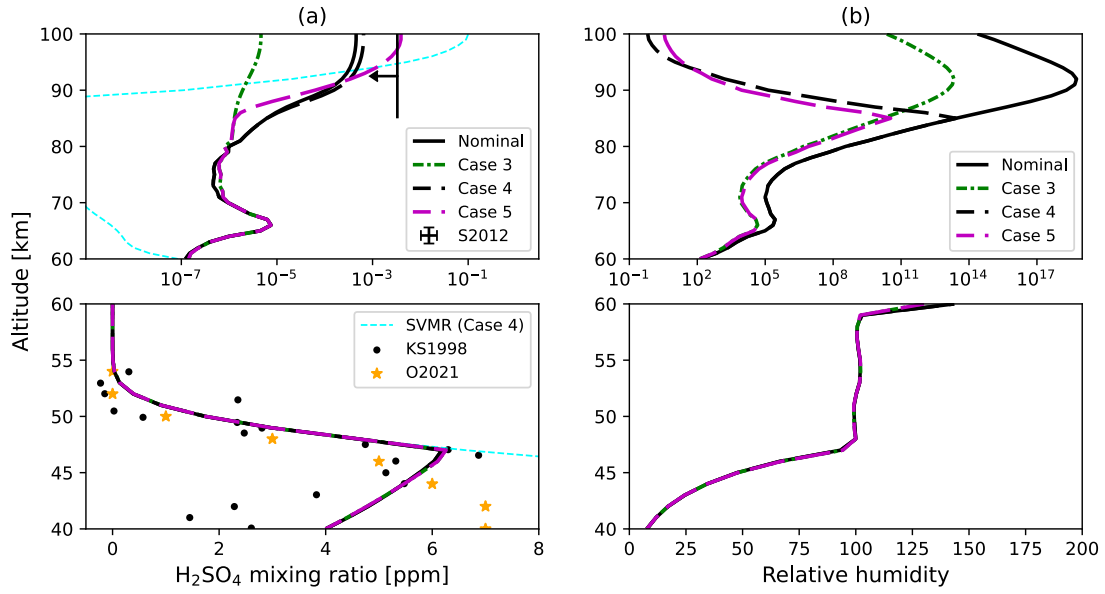


Figure 2.12: Same as Figure 2.7, but the simulated results for Case 4 and Case 5 are added with black and magenta dashed lines, and the Case 1 and Case 2 results are removed for visibility.

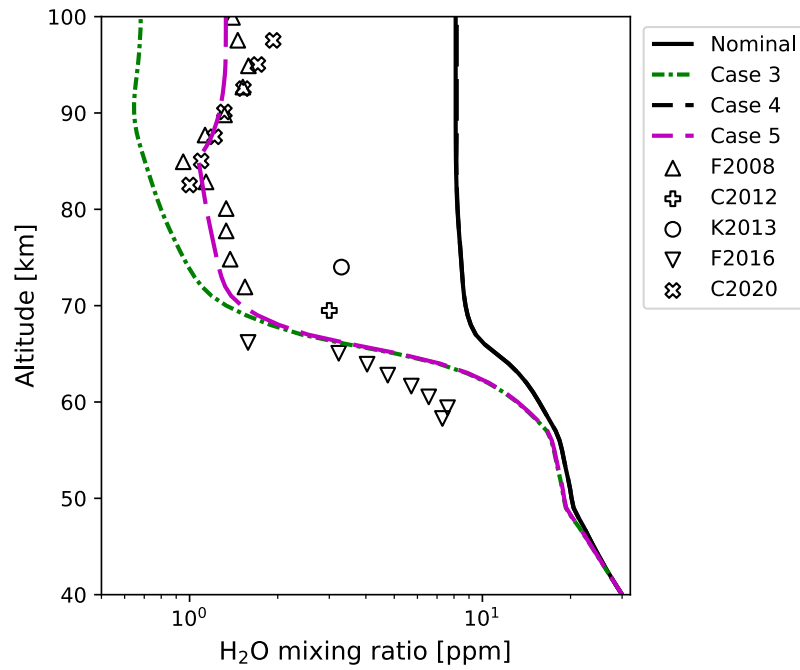


Figure 2.13: Same as Figure 2.8, but the simulated results for Case 4 and Case 5 are added with black and magenta dashed lines, and the Case 1 and Case 2 results are removed for visibility.

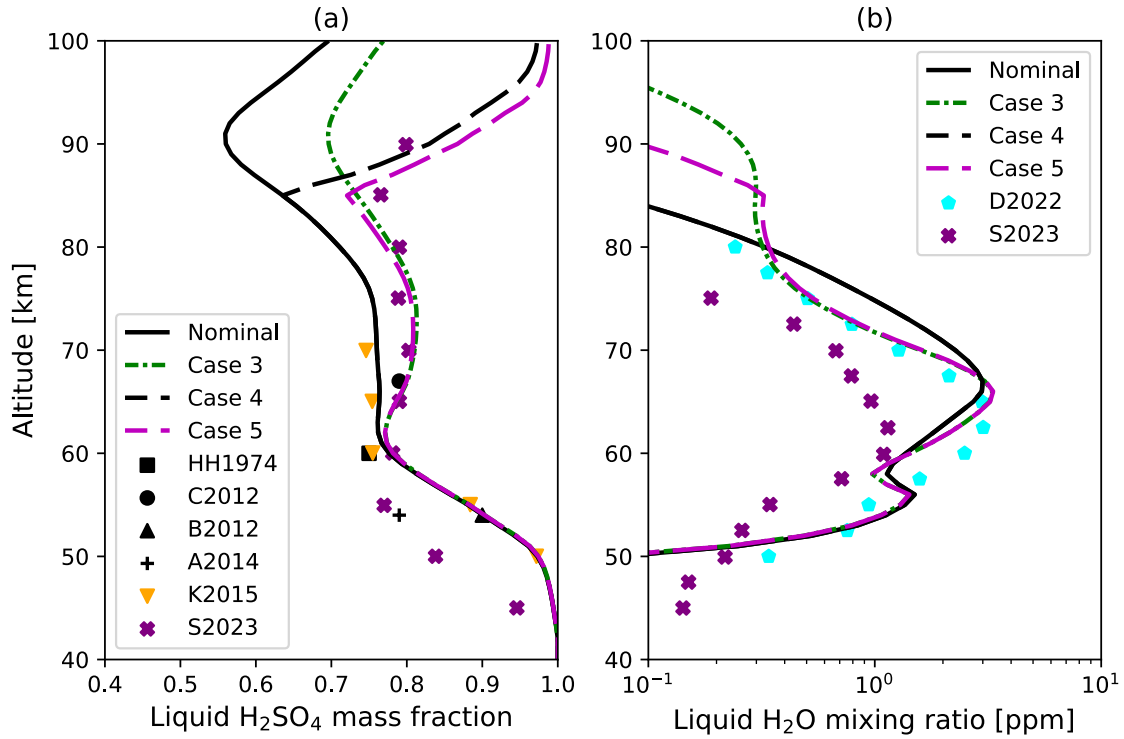


Figure 2.14: Same as Figure 2.9, but the simulated results for Case 4 and Case 5 are added with black and magenta dashed lines, and the Case 1 and Case 2 results are removed for visibility.

droplets cannot maintain a high H_2O content above this altitude. The reduced liquid H_2O VMR in the droplets is corroborated in Case 5 (Figure 2.14b). Specifically, the liquid H_2O VMR experiences a sharp decline above 85 km. This implies that more H_2O molecules exist in the gas phase in the SOIR temperature scenario compared to the VIRA temperature case because the warm layer leads to H_2O evaporation. Thus, the increasing H_2O VMR in Figure 2.13 can be attributed to the release of the H_2O molecules from the upper haze particles that have been transported upward from below 85 km, which is consistent with the recent aerosol-mediated H_2O transport theory proposed by (Mahieux et al., 2024).

2.3 Discussion

2.3.1 Best-fit atmospheric parameters

In Section 2.2.1, we identified a strong dependence of both aerosol and gas species distributions on the eddy diffusion profiles. The aerosol distribution is highly sensitive to the eddy diffusion coefficient above 85 km (Figure 2.6). Results for Case 1 and Case 3, using the eddy diffusion recently estimated by Mahieux et al. (2021), closely match the upper haze distribution observed by SPICAV-SOIR Wilquet et al. (2012). Furthermore, Cases 2 and 3, using lower eddy diffusion coefficients

between 60 and 70 km, are consistent with the observed H₂O VMR distribution reported by VEx observations (Chamberlain et al., 2020; Fedorova et al., 2016, 2008; Cottini et al., 2015). Given that Case 3 successfully reproduces both the observed gas species and aerosol distributions, the present study suggests that the eddy diffusion should be $\sim 360 \text{ m}^2 \text{ s}^{-1}$ in the upper haze layer and $\sim 2 \text{ m}^2 \text{ s}^{-1}$ in the upper cloud layer, when the chemical production/loss rates of condensational species are fixed.

In our study, the eddy diffusion represents global transport processes such as Hadley-type circulation since our model represents the whole atmosphere with a 1-D column. To compare the eddy diffusion coefficient obtained in the present study with those derived from the General Circulation Model (GCM), we employ time scale estimations. The timescale of the eddy diffusion coefficient is given by $\tau = H^2/K_{\text{eddy}}$, where H is the scale height of the atmosphere. On the other hand, the time scale of the global-scale circulation can be estimated by $\tau = H/w$, where w is the typical vertical wind speed. Assuming that eddy transport is mainly driven by the global-scale circulation, the relationship $K_{\text{eddy}} = Hw$ can be derived. Takagi et al. (2018) obtained a zonally and temporally averaged vertical wind speed magnitude on the order of $\sim 0.001 \text{ m s}^{-1}$ at 70 km using AFES-Venus (Atmospheric GCM for the Earth Simulator for Venus). With a corresponding scale height of 5 km, these values yield $K_{\text{eddy}} = 5 \text{ m}^2 \text{ s}^{-1}$, which is within the same order of magnitude as the eddy diffusion coefficient assumed between 60 and 70 km in the present study. Navarro et al. (2021) found a zonally and temporally averaged vertical wind speed magnitude on the order of $\sim 0.1 \text{ m s}^{-1}$ at 90 km using the Institut Pierre-Simon Laplace (IPSL) GCM, and the scale height at the corresponding altitude is ~ 3.7 km. These two values give $K_{\text{eddy}} = 370 \text{ m}^2 \text{ s}^{-1}$, which is equivalent to the eddy diffusion coefficient assumed above 85 km in our study. A GCM study by Sugimoto et al. (2019) showed that vertical eddy diffusion coefficients due to small-scale turbulence should be less than $0.02 \text{ m}^2 \text{ s}^{-1}$ to reproduce realistic superrotation. This value is significantly smaller than the eddy diffusion derived using the above formulation. This could suggest that global-scale circulation effectively represents the eddy diffusion transport in a 1-D column. Specifically, the dynamical time scale obtained in the GCMs are consistent with the time scale required to reproduce observed cloud structure and condensable gas species. Above 85 km, the existence of subsolar-to-antisolar (SSAS) circulation is supported by observations of O₂ nightglow at the antisolar point (Soret et al., 2012) and Venus Thermospheric GCM (e.g. Navarro et al., 2021) (e.g. Hoshino et al. 2013; Brecht et al. 2012; Navarro et al. 2021). Thus, the large eddy diffusion above 85 km may be attributed not only to Hadley-type circulation but also to the influence of SSAS circulation.

The case studies showed that the H₂O VMR profile is highly sensitive to the eddy diffusion coefficient between 60 and 70 km. The eddy diffusion coefficient used in this study is broadly consistent with previous observations and global climate models. Therefore, a thorough examination of the eddy diffusion coefficient in this altitude range is crucial for understanding H₂O transport through the cloud layer. Numerical investigations of the eddy diffusion coefficient can be performed using a General Circulation Model (GCM), as shown by Zhang and Showman (2018). Recently, Fujisawa et al. (2022) achieved a realistic representation of wind and thermal structures using a data assimilation technique that combines AFES-Venus and AKATSUKI observations. Such data assimilation products could serve as valuable resources for deriving an eddy diffusion coefficient that accounts for both global and local transport processes.

In terms of the mesospheric temperature, the SOIR profile is more suitable to reproduce the observed H₂O VMR increase above 85 km (see Figure 2.13). Therefore, the present study suggests that Case 5 provides the best match with observed atmospheric parameters. However, observed temperature profiles appear to differ depending on the instrument used for observation. In general, radio occultation measurements yield lower temperature profiles compared to those obtained through infrared observations (Limaye et al., 2018), as depicted in Figure 2.3a. In addition, Krasnopolsky (2011) argued that high temperatures between 95 and 100 km disagree with the rotational temperature of the O₂ nightglow at 1.27 μm .

This discrepancy could potentially be resolved by measuring the liquid H₂SO₄ mass fraction in upper haze particles. As shown in Figure 2.9a, the liquid H₂SO₄ mass fraction varies significantly depending on the temperature—a variation that should be detectable. Consequently, we can use the H₂SO₄ mass fraction as a proxy to better understand the upper mesospheric temperature profiles and the resulting equilibrium conditions. However, as Mahieux et al. (2023b) pointed out, the temperature difference could be caused by the local time dependence. In that case, the temperature observations can be used as a proxy for the H₂SO₄ mass fraction of aerosols. Regardless of the scenario, acquiring more detailed observations that focus on the local time dependence of mesospheric temperatures is crucial for enhancing our understanding of the microphysical properties of aerosols.

2.3.2 Implication for atmospheric evolution

The H₂O abundance at the homopause regulates the hydrogen (H) escape rate through diffusion-limited flux. Assuming a H₂O vapor abundance of ~ 1 ppm and HCl of ~ 0.5 ppm at the homopause, the diffusion-limited flux is calculated to be $\sim 3 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$ (Catling and Kasting, 2017). This value is consistent with an estimate of the nonthermal escape flux of $\sim 3 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$ (Lammer et al., 2006), indicating that the diffusion-limited flux serves as a good approximation for estimating the present-day H escape flux. Therefore, the H₂O vapor abundance at the homopause is crucial for understanding the current atmospheric evolution of Venus.

The long-term stability of Venus' clouds is still controversial. Bullock and Grinspoon (2001) showed that the clouds would not be stable if the atmospheric SO₂ amount is buffered by carbonate. Conversely, Hashimoto and Abe (2005) argued that the clouds would be stable if the atmospheric SO₂ amount is buffered by pyrite. These contrasting scenarios could lead to significant differences in H₂O loss rates, given that the cloud layer affects the diffusion-limited escape rate. Therefore, the presence of clouds and their impact on mesospheric H₂O abundance should be carefully considered in models of Venus' atmospheric evolution. The present study shows that cloud processes are essential to evaluate the H₂O VMR vertical profile, which is deeply connected to H₂O vapor abundance at the homopause. Consequently, cloud processes cannot be disregarded when deriving the mesospheric H₂O abundance and diffusion-limited escape rate. These cloud effects should be considered not only for the present-day atmosphere but also for the past atmospheric conditions. Furthermore, observations of trace gas species and clouds, possible with the future EnVision mission, will further elucidate the effects of clouds on atmospheric evolution. The VenSpec-H instrument onboard the EnVision spacecraft will observe H₂O abundance below and above the cloud layer with high accuracy,

and Radio Science experiment will obtain liquid H_2SO_4 abundance in the cloud layer (Akins et al., 2023). This information will allow us to investigate the role of clouds on regulating the mesospheric H_2O abundance and its impact on the hydrogen escape.

2.3.3 Bimodal distribution in the upper haze layer

The simulated size distribution in the upper haze layer is unimodal, diverging from the bimodal distribution suggested by VEx observations (Wilquet et al., 2009). Previous microphysics studies (Gao et al., 2014; Parkinson et al., 2015) have also reported unimodal distributions in the upper haze layer at steady state. However, Gao et al. (2014) argued that transient vertical winds could give rise to the observed bimodal distribution. The inclusion of dynamical effects, which are not considered in the present study, may be necessary to accurately reproduce the observed particle size distribution and should be explored in future research.

Homogeneous nucleation processes could play a significant role in the generation of the Mode 1 particles. Dai et al. (2022) obtained a supersaturated H_2SO_4 VMR profile in the upper cloud and haze layer, which aligns with the findings of our study; their results were obtained using a cloud condensation model. They argued that under such supersaturated conditions, homogeneous nucleation is likely to occur. The enhanced CN population could correspond to Mode 1 particles, as observed in the lower and middle cloud layer of recent microphysical model McGouldrick and Barth (2023). This idea is supported by the VIRA temperature cases (nominal, Case 1-3), where the H_2SO_4 vapor is highly supersaturated in the upper haze layer, as shown in Figure 2.7a. The potential for homogeneous nucleation is further explored using a cloud microphysics model that includes CN size distributions in Chapter 4 and 5.

2.4 Chapter Summary

We developed a 1-D cloud microphysics model based on the work of Imamura and Hashimoto (2001) and extended the model-top altitude from 70 km to 100 km. The temperature and pressure profiles are adapted from the mid-latitude profiles from VIRA (Seiff et al., 1985) to represent the global average atmosphere. We updated the production rate of H_2SO_4 vapor using recent photochemical model results (Krasnopolsky, 2012). The simulated size distribution and mass loading are in good agreement with in-situ observations (Knollenberg and Hunten, 1980) and successfully replicate the earlier work by Imamura and Hashimoto (2001). A sharp peak of the H_2SO_4 VMR forms below the clouds, reaching a value of ~ 6 ppm, which is quantitatively consistent with previous observations (Kolodner and Steffes, 1998; Oshlisniok et al., 2021).

To investigate the transport processes of gas species and aerosols, we conducted case studies consisting of four distinct eddy diffusion patterns. These patterns were created using a pair of small and large eddy diffusion coefficients between 60 and 70 km and between 85 and 100 km. When using the recently updated eddy diffusion coefficient from Mahieux et al. (2021) in the upper haze layer (85-100 km), the aerosol extinction coefficient is in good agreement with the SPICAV-SOIR observations Wil-

quet et al. (2012), indicating that efficient dynamical transport is essential for maintaining this layer. The H_2O vapor profile is consistent with the SPICAV-SOIR observations (Fedorova et al., 2008) when the eddy diffusion coefficient in the upper cloud layer is halved relative to previous radio scintillation measurements (Woo and Ishimaru, 1981). The H_2O VMR is highly sensitive to changes in eddy diffusion at these altitudes, as the gradient of the H_2O VMR profile varies significantly depending on the eddy diffusion coefficient. The H_2SO_4 VMR in the upper cloud layer is also highly sensitive to the eddy diffusion coefficient above 85 km, ranging from $\sim 5 \times 10^{-6}$ to $\sim 5 \times 10^{-4}$ ppm. However, the simulated values are orders of magnitude lower than the observational upper limit suggested by Sandor et al. (2012) across cases using the VIRA temperature vertical profile.

To investigate the impact of temperature on gas species and aerosol distributions, we conducted an additional case study using a temperature profile obtained by SPICAV-SOIR (Mahieux et al., 2023b). Due to the presence of a warm layer in the SOIR temperature profile, the H_2SO_4 VMR increases to ~ 3 ppb above 90 km, as the elevated temperature raises the H_2SO_4 saturation pressure. However, when compared to the photochemical modeling results of Zhang et al. (2010), the elevated H_2SO_4 VMR is still insufficient to account for the observed SO_2 inversion layer. In the upper haze layer, the liquid H_2SO_4 mass fraction in the aerosols shows a notable increase compared to the VIRA-based case, and H_2SO_4 gaseous part reaches the upper limit of 3 ppb measured by Sandor et al. (2012).

The present study identifies the best-fit eddy diffusion coefficients as $\sim 360 \text{ m}^2 \text{ s}^{-1}$ above 85 km and $\sim 2 \text{ m}^2 \text{ s}^{-1}$ between 60 and 70 km. Updating the eddy diffusion coefficient in the 60-70 km altitude range is essential for understanding the transport dynamics of H_2O vapor through the cloud layers. In addition, it is also expected that an increase in H_2SO_4 production rate, conversely, H_2O loss rate, results in the depletion of H_2O above the upper cloud layer. Such updates could be achieved through numerical investigations using data assimilation products or future missions. In addition, this study suggests that the H_2O VMR profile is better replicated using the SOIR temperature profile. However, previous observations have shown variability in the temperature profiles in the upper haze layer depending on the instrument used for observation. Measuring the liquid H_2SO_4 mass fraction in droplets could serve as a discriminative factor for validating temperature profiles and establishing the equilibrium conditions in the upper haze layer.

This study shows that the mesospheric H_2O abundance is affected by the transport in the upper cloud layer, which could potentially regulate the diffusion-limited escape rate. Therefore, we suggest that the effects of clouds should be included in models of Venus' atmospheric evolution. In addition, the formation of Mode 1 particles in the upper haze layer could be linked to homogeneous nucleation processes, which are investigated in Chapter 5 of this thesis.

Chapter 3

Development of a Multi-Component Venus' Microphysics Model with a High-Accuracy Condensation Scheme

This chapter is based on the following paper, published in *Earth and Space Science*:

Hiroki Karyu, Takeshi Kuroda, Arnaud Mahieux, Sébastien Viscardy, Anni Määttänen, Naoki Terada, Séverine Robert, Ann Carine Vandaele, and Michel Crucifix (2025). A microphysics model of multicomponent Venus' clouds with a high-accuracy condensation scheme. *Earth and Space Science*, 12 (6): e2025EA004203. 10.1029/2025EA004203

3.1 Model Description

3.1.1 Overview

SPECK computes the vertical distributions of particle components, such as solid CN materials and liquid components, and gas species using the 1-D continuity equation. The particle components are represented by multiple size distributions, each divided into size bins. Interactions between these distributions are also incorporated. The 1-D continuity equation for each aerosol component is expressed as follows (modified from Toon et al. (1988)):

$$\begin{aligned} \frac{\partial c_{q,Ni}(z,t)}{\partial t} = & \frac{\partial}{\partial z} \left[K_{\text{eddy}}(z) n_{\text{air}}(z) \frac{\partial}{\partial z} \left(\frac{c_{q,Ni}(z,t)}{n_{\text{air}}(z)} \right) \right] - \frac{\partial}{\partial z} [w_{\text{sed}}(v_{Ni}, z) c_{q,Ni}(z,t)] \\ & + \left. \frac{\partial c_{q,Ni}(z,t)}{\partial t} \right|_{\text{cond}} + \left. \frac{\partial c_{q,Ni}(z,t)}{\partial t} \right|_{\text{coag}} + \left. \frac{\partial c_{q,Ni}(z,t)}{\partial t} \right|_{\text{nuc}}, \end{aligned} \quad (3.1)$$

where $c_{q,Ni}$ is the concentration (mol m^{-3}) of particle component q in size bin i of distribution N at the altitude z (m) and time t (s), $n_{\text{air}}(z)$ is the air concentration (mol m^{-3}), $K_{\text{eddy}}(z)$ is the vertical eddy diffusion coefficient ($\text{m}^2 \text{s}^{-1}$) of air, and $w_{\text{sed}}(v_{Ni}, z)$ is the sedimentation velocity (m s^{-1}) of aerosol particles with volume v_{Ni} (m^3). The symbol N used in Equation (3.1) refers to the type of particle species, such as CN and H_2SO_4 liquid droplets, and so on. The configuration of particle species and their interactions is described in Section 3.1.3. The sedimentation velocity is expressed as follows (Pruppacher and Klett, 2010):

$$w_{\text{sed}} = \frac{2r_{v_{Ni}}^2 \rho_p g}{9\eta} \left[1 + \text{Kn}(z) \left\{ A + B \exp \left(-\frac{C}{\text{Kn}(z)} \right) \right\} \right], \quad (3.2)$$

where $r_{v_{Ni}}$ is the particle radius (m) corresponding to the volume v_{Ni} , ρ_p is the density (kg m^{-3}) of the particle, $g = 8.7 \text{ m s}^{-2}$ is the gravitational acceleration of Venus, $\eta = 1.5 \times 10^{-5} \text{ kg m}^{-1} \text{s}^{-1}$ is the viscosity of CO_2 gas, $\text{Kn}(z)$ is the Knudsen number for air, and the constants A , B , and C are experimental parameters set to be 1.249, 0.42, and 0.87, respectively, following Kasten (1968). The first term on the right-hand side of Equation (3.1) represents vertical transport by eddy diffusion. The second term accounts for vertical transport due to particle sedimentation. The third term $\left. \frac{\partial c_{q,Ni}}{\partial t} \right|_{\text{cond}}$, the fourth term $\left. \frac{\partial c_{q,Ni}}{\partial t} \right|_{\text{coag}}$, and fifth term $\left. \frac{\partial c_{q,Ni}}{\partial t} \right|_{\text{nuc}}$ are the rates of change ($\text{mol m}^{-3} \text{s}^{-1}$) in particle component q in size bin i of distribution N due to condensation, coagulation, and nucleation, respectively. These microphysics terms will be explained in Sections 3.1.3 and 3.1.4. Similarly, the 1-D continuity equation for gas species is represented as follows:

$$\begin{aligned} \frac{\partial C_q(z,t)}{\partial t} = & \frac{\partial}{\partial z} \left[K_{\text{eddy}}(z) n_{\text{air}}(z) \frac{\partial}{\partial z} \left(\frac{C_q(z,t)}{n_{\text{air}}(z)} \right) \right] \\ & + \left. \frac{\partial C_q(z,t)}{\partial t} \right|_{\text{cond}} + \left. \frac{\partial C_q(z,t)}{\partial t} \right|_{\text{nuc}} + \left. \frac{\partial C_q(z,t)}{\partial t} \right|_{\text{chem}}, \end{aligned} \quad (3.3)$$

where C_q is the concentration (mol m^{-3}) of gas component q , $\left. \frac{\partial C_q}{\partial t} \right|_{\text{cond}}$, $\left. \frac{\partial C_q}{\partial t} \right|_{\text{nuc}}$, and $\left. \frac{\partial C_q}{\partial t} \right|_{\text{chem}}$ are the rates of change ($\text{mol m}^{-3} \text{s}^{-1}$) in gas concentration due to condensation, nucleation, and chemistry, respectively.

We consider three particle components with $q = 1$ for H_2SO_4 , $q = 2$ for H_2O , and $q = 3$ for CN, following previous microphysics studies (Gao et al., 2014; McGouldrick, 2017; McGouldrick

and Barth, 2023). These particle components are further categorized as either condensable or dry species in the model. The condensable species, H_2SO_4 and H_2O , are exchanged between particles and gas phases depending on the saturation vapor pressure. In contrast, the dry species, CN, is non-condensable and remains only within the solid particle phase. The CN material is assumed to be the elemental sulfur and insoluble, following the previous microphysical studies (Gao et al., 2014; Imamura and Hashimoto, 2001; McGouldrick, 2017).

We consider three types of particle distributions based on their components. The first is the CN distribution, which consists solely of the CN particle component, supplied by the formation of elemental sulfur. The second is the sulfuric acid droplet distribution, comprising a binary mixture of H_2SO_4 and H_2O . The third type is the mixed distribution, which includes both CN and sulfuric acid droplets. Mixed particles form either when CN particles become wetted or when CN and sulfuric acid droplets undergo coagulation.

The particle bin structure is constructed based on a volume-ratio size distribution, designed to cover a size range spanning several orders of magnitude (Jacobson, 1997b). The high-edge volume ($v_{i,\text{hi}}$) of the i -th bin is determined by multiplying the low-edge volume ($v_{i,\text{lo}}$) by V_{rat} which is a constant volume ratio between adjacent bins and can take any value greater than one. Accordingly, the volume width of a size bin (dv_i) is expressed as follows:

$$dv_i = v_{i,\text{hi}} - v_{i,\text{lo}} = V_{\text{rat}}v_{i,\text{lo}} - v_{i,\text{lo}} = (V_{\text{rat}} - 1)v_{i,\text{lo}}. \quad (3.4)$$

Throughout the simulation, dv_i , $v_{i,\text{lo}}$, and $v_{i,\text{hi}}$ are fixed, while particle volume v_i can vary between the edges of the respective bins $v_{i,\text{lo}}$ and $v_{i,\text{hi}}$, as explained in the Section 3.1.

3.1.2 Bin scheme

The performance of the sectional microphysics model heavily depends on the choice of bin scheme, as each scheme has its advantages and disadvantages. Figure 3.1 shows three common bin schemes used in aerosol microphysics simulations. The scheme widely used in planetary microphysics models is the full-stationary scheme (e.g. Turco et al., 1979; Toon et al., 1988; McGouldrick and Barth, 2023; Karyu et al., 2024). In this scheme, average particle sizes and bin boundaries are fixed throughout the simulation, making it convenient for nucleation, coagulation, and transport in multiple dimensions. However, one drawback of this scheme is the numerical diffusion of the size distribution during condensation because size growth is treated as Eulerian advection of number density within the size space.

The full-moving scheme is nearly the opposite of the full-stationary scheme, in the sense that the size bin boundaries themselves shift during the growth process (Gelbard, 1990). In a full-moving scheme, the particle radius assigned to each bin increases during growth, while the number density within each bin remains constant (Figure 3.1). This approach can be understood as Lagrangian advection within size space, providing an exact solution for condensational growth. However, this scheme is less practical for multi-dimensional studies, as each grid cell may have a distinct bin structure.

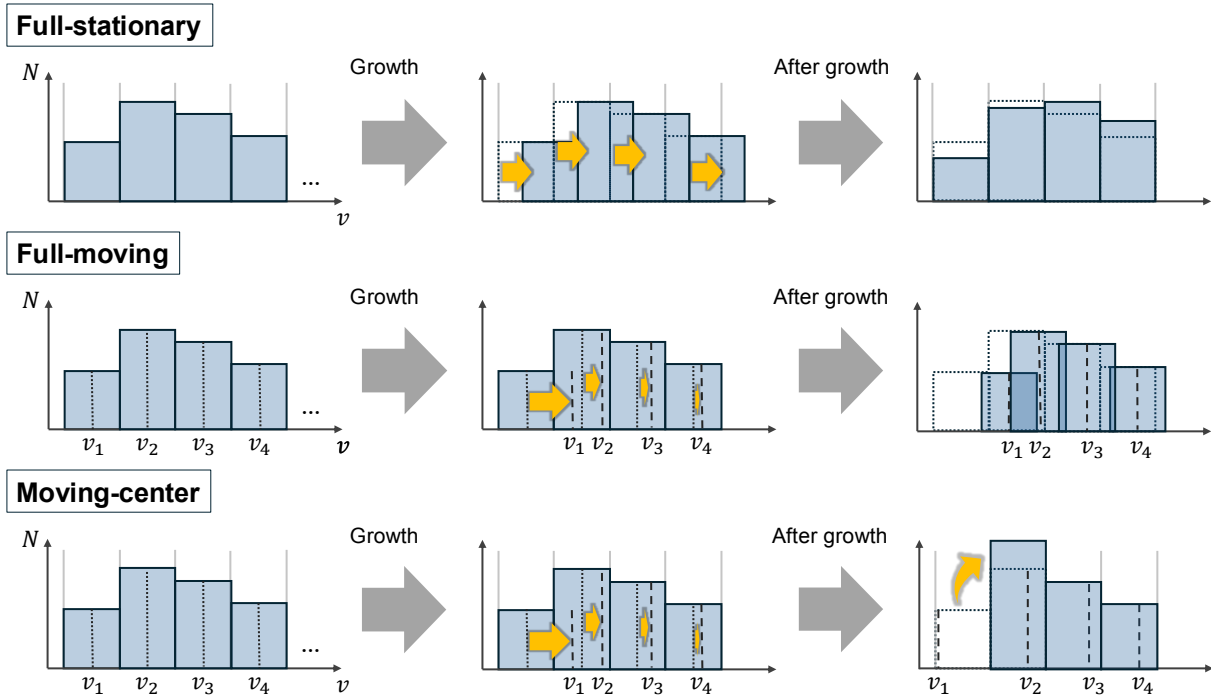


Figure 3.1: A schematic illustration of the full-stationary (top row), full-moving (middle row), and moving-center (bottom row) schemes. The left column represents the initial size distribution as a function of volume, with gray lines marking fixed bin boundaries in the full-stationary and moving-center schemes. Characteristic volumes v_1 – v_4 are assigned to bins in the full-moving and moving-center schemes. The middle column depicts the treatment of growth in each bin scheme: yellow arrows in the full-stationary scheme indicate advection in size space, while in the full-moving and moving-center schemes, they represent growth to the exact volume. The right column shows the predicted size distributions after the growth process. The hatched distribution corresponds to the original size distribution. In the moving-center scheme, all the number density from the first bin is transferred to the next larger size bin when the characteristic volume exceeds the upper boundary of the bin. Note that this figure is intended only to illustrate the calculation concept and does not represent actual calculation results.

The bin scheme applied in this study is the moving-center scheme (Jacobson, 1997b). In the moving-center scheme, the boundaries of each size bin are fixed, but particle size can vary within each bin, as shown in Figure 3.1. As growth occurs, particles are allowed to grow to their exact sizes, which is identical to the full-moving scheme, making the growth calculation equivalent to Lagrangian advection in size space.

Once the particle size surpasses the upper boundary of a bin (corresponding to v_1 in the right column of Figure 3.1), the number density of that bin is transferred to the next larger bin. After this transfer, a pit structure may form within the size distribution due to the emptying of a size bin, creating an artificial pit in the distribution. The volume of the emptied bin (v_1) is then re-initialized to the lower edge of the bin. Meanwhile, the volume of the receiving bin (v_2) is updated to the average of the transferred particle sizes (v_1) and the pre-existing particle sizes (v_2) in that bin. The treatment of evaporation is numerically identical to that of condensation in the moving-center scheme. Thus, when

the particle size crosses the lower boundary of a bin, the number density of that bin is transferred to the next smaller bin.

The volumes of other bins (v_3 and v_4), which do not experience growth or number density transfer, are preserved and integrated until their volumes surpass the boundaries of the respective bins. The formation of the pit is a drawback of the moving-center scheme. However, despite this pit formation, the algorithm has been shown to produce results nearly identical to the full-moving scheme because numerical diffusion is nearly eliminated in this scheme (Jacobson, 1997b, 2005). As a result, the moving-center scheme offers a more accurate solution for condensational growth compared to other sectional bin schemes (Jacobson, 1997b; Zhang et al., 1999). Furthermore, this scheme is easily coupled with nucleation, coagulation, and transport in multiple dimensions, as the bin boundaries remain fixed. Thus, the moving-center scheme can be regarded as a combination of the full-stationary and full-moving schemes.

3.1.3 Condensation scheme

We consider two condensable species, with $q = 1$ for H_2SO_4 and $q = 2$ for H_2O . The condensational growth rate is explicitly calculated for H_2SO_4 . In contrast, the condensation rate of H_2O is determined through thermodynamic equilibrium adjustments. This approach is necessitated by the fact that water condensation on Venus is driven by the strong hygroscopicity of sulfuric acid droplets, which rapidly equilibrate with ambient water vapor. These adjustments depend on the surrounding temperature and available water vapor, following previous microphysical studies (Imamura and Hashimoto, 2001; McGouldrick and Toon, 2007; McGouldrick and Barth, 2023; Karyu et al., 2024). In the moving-center scheme, particle number density is not transferred between bins as size-space advection. Instead, particles grow precisely to their actual sizes during condensation, with number density transfer occurring after updating the size.

The condensational growth of particles and gas conservation are expressed as follows (Jacobson, 1997a):

$$\left. \frac{\partial c_{q,i}}{\partial t} \right|_{\text{cond}} = k_{q,i}(C_q - S_{q,i}C_{q,s}) \quad i = 1, \dots, N_B, \quad (3.5)$$

$$\left. \frac{\partial C_q}{\partial t} \right|_{\text{cond}} = - \sum_{i=1}^{N_B} k_{q,i}(C_q - S_{q,i}C_{q,s}), \quad (3.6)$$

where N_B is the total number of bins, $C_{q,s}$ is the saturation mole concentration (mol m^{-3}) of a vapor over a flat surface, $k_{q,i}$ is the vapor transfer rate (s^{-1}) between the gas phase and all particles of size i , $S_{q,i}$ is the equilibrium saturation ratio of the condensing gas. The saturation vapor pressure for pure H_2SO_4 is taken from Kulmala and Laaksonen (1990), and that for H_2O is taken from Eisenberg and Kauzmann (2005). The vapor transfer rate $k_{q,i}$ is represented as $4\pi n_i r_i D_{q,i,\text{eff}}$, where n_i is the concentration of particles (m^{-3}) of the i -th bin, r_i is the particle radius (m) of the i -th bin, and $D_{q,i,\text{eff}}$ is an effective molecular diffusion coefficient ($\text{m}^2 \text{s}^{-1}$) given by Jacobson (2005) (see also Appendix

B.2 for more details). The equilibrium saturation ratio $S_{q,i}$ is expressed as follows (Seinfeld and Pandis, 2016):

$$S_{q,i} = \exp \left(\frac{2\sigma_p m_w}{r_i RT \rho_{w,i}} + \frac{\mu_q(T, x) - \mu_q^o(T)}{RT} \right), \quad (3.7)$$

where σ_p is the average particle surface tension (kg s^{-2}), m_w is the average molar weight (kg mol^{-1}) of solution, R is the universal gas constant ($\text{J mol}^{-1} \text{K}^{-1}$), T is the temperature (K), $\rho_{w,i}$ is the density (kg m^{-3}) of the solution, $\mu_q(T, x)$ is the chemical potential (J mol^{-1}) of the condensing gas in the H_2SO_4 - H_2O binary solution with H_2SO_4 mole fraction of x at temperature T , and $\mu_q^o(T)$ is the chemical potential (J mol^{-1}) of the pure solution. The first term in the exponential accounts for the Kelvin effect, while the second term represents the activity of the mixed solution. We used the parameterization of Vehkamäki et al. (2002) for σ_p as a function of the temperature and weight concentration of H_2SO_4 in the binary solution. The chemical potentials for the binary solution and pure H_2SO_4 are taken from Zeleznik (1991). The wetting of the CN particles occurs when $C_q > S_{q,i} C_{q,s}$ since detailed heterogeneous nucleation process is not included in this study. Accordingly, particle transfer between different size distributions is also computed. Particles in the CN distribution participate in the mixed size distribution once they obtain liquid component by condensational growth. Conversely, particles in the mixed distribution are transferred to the CN distribution, when they lose all the liquid components.

In general, soluble and insoluble cores play an important role on determining the saturation ratio, as solute reduces the saturation vapor pressure, and the presence of a core modifies the volume of the liquid shell available to the solute (Petters and Kreidenweis, 2007). In the present simulation, we do not take into account these effects, as the CN material is assumed to be perfectly insoluble, and thus, the solute effect is negligible. However, the impact of solubility may become important in future studies if other soluble substances, such as salts, are introduced as additional cloud components.

Equations (3.5) and (3.6) represent $N_B + 1$ ordinary differential equations. The solution for this system is obtained using the Analytical Predictor of Condensation (APC) scheme of Jacobson (1997a, 2002, 2005), introduced in Appendix B.3. The APC scheme conserves mass exactly and does not need iteration. Additionally, the scheme is numerically stable, as the gas concentration remains bounded between 0 and $C_{q,\text{tot}} = C_q + \sum_{i=1}^{N_B} c_{q,i}$ regardless of the time step.

Following the application of the APC scheme, the average volume for each bin is updated. All particle number density is transferred to the adjacent larger (or smaller) bin if the updated volume exceeds the upper (or falls below the lower) boundary of the current bin. When the particle size falls below (or exceeds) the minimum (or maximum) boundary of the size bin grid, the particle number density is recalculated using the smallest (or largest) radius of the size bin grid to ensure mass conservation.

3.1.4 Coagulation scheme

We consider both self-coagulation within the same size distribution and heterogeneous coagulation between different types of size distributions. For example, coagulation within the CN distribution results in the production of larger CN particles and the depletion of smaller CN particles. On the other hand, coagulation between CN and pure sulfuric acid droplets leads to the production of larger mixed droplet particles and the depletion of smaller CN and pure sulfuric acid droplets. The coagulation process is calculated using a volume-conserving semi-implicit scheme for any number N_T of size distributions each with N_B bins, as follows (Jacobson, 2002, 2005):

$$c_{q,N_k,t+\Delta t} = \frac{c_{q,N_k,t} + \Delta t (T_{q,N_k,t+\Delta t,1} + T_{q,N_k,t+\Delta t,2})}{1 + \Delta t T_{q,N_k,t+\Delta t,3}}, \quad (3.8)$$

$$T_{q,N_k,t+\Delta t,1} = \sum_{M=1}^{N_T} P_{N,M} \sum_{j=1}^k \sum_{i=1}^{k-1} f_{N_i,M_j,N_k,t} \cdot \beta_{N_i,M_j,t} \cdot c_{q,N_i,t}, \quad (3.9)$$

$$T_{q,N_k,t+\Delta t,2} = \sum_{M=1}^{N_T} \sum_{I=1}^{N_T} Q_{I,N,M} \sum_{j=1}^k \sum_{i=1}^k f_{I_i,M_j,N_k,t} \cdot \beta_{I_i,M_j,t} \cdot c_{q,I_i,t}, \quad (3.10)$$

$$T_{q,N_k,t+\Delta t,3} = \sum_{M=1}^{N_T} \sum_{j=1}^{N_B} [(1 - L_{N,M})(1 - f_{N_k,M_j,N_k,t}) + L_{N,M}] \beta_{N_k,M_j,t} \cdot c_{q,I_i,t}. \quad (3.11)$$

Here, N_k is the index of the k -th bin in the distribution N , Δt is the simulation time step, $\beta_{i,j}$ is the total coagulation kernel ($\text{m}^3 \text{s}^{-1}$) between size bin i and j . Note that t and $t + \Delta t$ denote the current and advanced time steps, respectively.

The total coagulation kernel is the sum of contributions from Brownian diffusion and gravitational collection, with their formulations detailed in Pruppacher and Klett (2010). Particles formed through the coagulation process often fall between the sizes of adjacent bins. Therefore, partitioning between the adjacent bins is included for volume conservation, and this factor is represented by f_{I_i,M_j,N_k} , which is further expressed as follows (Jacobson et al., 1994):

$$f_{I_i,M_j,N_k} = \begin{cases} \frac{v_{N_{k+1}} - V_{I_i,M_j}}{v_{N_{k+1}} - v_{N_k}} \frac{v_{N_k}}{V_{I_i,M_j}} & v_{N_k} \leq V_{I_i,M_j} < v_{N_{k+1}}, k < N_B \\ 1 - f_{I_i,M_j,N_{k-1}} & v_{N_{k-1}} < V_{I_i,M_j} < v_{N_k}, k > 1 \\ 1 & V_{I_i,M_j} \geq v_{N_k}, k = N_B \\ 0 & \text{all other cases,} \end{cases} \quad (3.12)$$

where $V_{I_i,M_j} = v_{I_i} + v_{M_j}$.

The term T_1 given by Equation (3.9) accounts for the production of larger particles within distribution N from both self-coagulation ($N + N$) and heterogeneous coagulation involving distribution N

(e.g., $N + M$). The term T_2 given by Equation (3.10) represents the production of particles in distribution N resulting from the heterogeneous coagulation of two distinct distributions (e.g., $I + M$). The term T_3 given by Equation (3.11) represents particle loss in distribution N due to both self-coagulation and heterogeneous coagulation.

The three size distributions assumed in this study interact with one another as summarized in Table 3.1. The parameters P , Q , and L account for interactions between different distributions. The CN distribution consists of an involatile CN material, while the sulfuric acid droplet distribution comprises a liquid binary mixture of H_2SO_4 and H_2O . Coagulation within each of these distributions produces larger particles within their respective distributions. In contrast, coagulation between these distributions leads to the formation of larger particles in the mixed distribution. Additionally, any coagulation involving the mixed distribution consistently results in larger particles within the mixed distribution.

Accordingly, the parameters P , Q , and L are defined based on these interactions. The parameter $P_{N,M}$ is set to 1 if particles in distribution N produce larger particles within the same distribution after coagulating with particles in distribution M , and 0 in all other cases. The parameter $Q_{I,M,N}$ is set to 1 if coagulation between particles in distributions I and M results in the formation of larger particles in distribution N , and 0 in all other cases. The parameter $L_{N,M}$ is set to 1 if particles in distribution N are removed from the distribution due to coagulation with particles in distribution M , and 0 for other cases.

In the current framework, three types of particles are considered: CN (Type A), H_2SO_4 – H_2O droplets (Type B), and mixed droplets (Type AB), resulting in $N_T = 3$. Coagulation between particles within the same distribution produces larger particles within that distribution, and thus $P_{A,A}$, $P_{B,B}$, and $P_{AB,AB}$ are set to 1. When particles from different distributions coagulate, two scenarios are possible. In the first, a mixed particle coagulates with either a CN particle or a sulfuric acid droplet to form a larger mixed particle; in this case, $P_{AB,A}$ or $P_{AB,B}$ is set to 1. In the second scenario, a CN particle and a sulfuric acid droplet coagulate to form a new mixed particle, and $Q_{A,B,AB}$ is set to 1. For the corresponding loss processes, such as the coagulation of a CN particle with a sulfuric acid droplet, both $L_{A,B}$ and $L_{B,A}$ are set to 1. This parameter is also set to 1 for homogeneous coagulation, so $L_{A,A}$, $L_{B,B}$, and $L_{AB,AB}$ are likewise all set to 1.

It should be noted that this framework can be applied to any number of distribution by setting up P , Q , and L based on the relationships between independent distributions. While extending the model to incorporate additional size distributions is beyond the scope of this paper, future research could utilize this framework to simulate the mixing of multiple aerosol types, providing a versatile tool for studying complex aerosol interactions.

3.1.5 Benchmark Simulations

The primary objective of this paper is to verify whether the algorithms described above function properly under atmospheric conditions on Venus. To achieve this, we perform both 0-D and 1-D simulations incorporating various processes. In the 0-D simulations, we begin with a simple system

Table 3.1: Coagulation interaction in the simulation. Each type represents a category of aerosol particles distinguished by their chemical composition or initial characteristics. For instance, the coagulation between CN particles (Type A) and sulfuric acid droplets (Type B) results in the formation of mixed particles (Type AB), appearing in the first row and the second column in the table.

Type\Type	CN distribution (A)	Sulfuric acid droplet distribution (B)	Mixed distribution (AB)
CN distribution (A)	A	AB	AB
Sulfuric acid droplet distribution (B)	AB	B	AB
Mixed distribution (AB)	AB	AB	AB

and gradually increase the complexity to ensure that the microphysical scheme operates as expected. The background atmospheric temperature and pressure are set to the equatorial profiles of the Venus International Reference Atmosphere (VIRA) (Seiff et al., 1985) for both 0-D and 1-D simulations, as shown in Figure 3.2a.

The first phase of the 0-D simulations focuses exclusively on particle growth through condensation and evaporation. The results obtained using the moving-center scheme are validated against those of exact solutions calculated with the full-moving scheme. The second phase explores various scenarios involving both growth and coagulation processes to verify the algorithm’s capability to simulate aerosol microphysics across a wide range of complexities.

Two benchmark 1-D simulations are performed to validate our model, including transport processes. In the first benchmark case, we validate SPECK against an existing model that utilizes the full-stationary scheme. The model developed in Chapter 2, (Karyu et al., 2024, hereafter K2024) serves as a proxy for a full-stationary microphysics model. In the second benchmark case, we perform 1-D simulations under the same settings as McGouldrick and Barth (2023), demonstrating a realistic application of our model to the Venus’ cloud system. The eddy diffusion coefficient is set to the same value in the first and second benchmark cases, and these values are taken from McGouldrick and Barth (2023). The profile has a peak value of $60 \text{ m}^2 \text{ s}^{-1}$ at 54 km altitude, a constant value of $2 \text{ m}^2 \text{ s}^{-1}$ above 57 km altitude and $10 \text{ m}^2 \text{ s}^{-1}$ below 48 km altitude (Figure 3.2b). The full profile is given in Figure 3.2b.

3.2 0-D Validation of the Moving-Center Scheme

3.2.1 Condensation alone

We performed a 0-D model experiment to validate the newly introduced condensation scheme using the moving-center scheme. This simulation considers only condensation and evaporation processes, with H_2SO_4 as the sole condensational species for simplicity. The initial conditions include a log-normal size distribution of H_2SO_4 particles corresponding to Mode 1, with 5% supersaturation of H_2SO_4 vapor. The total number of H_2SO_4 in liquid and gas phases remains constant since no chemical

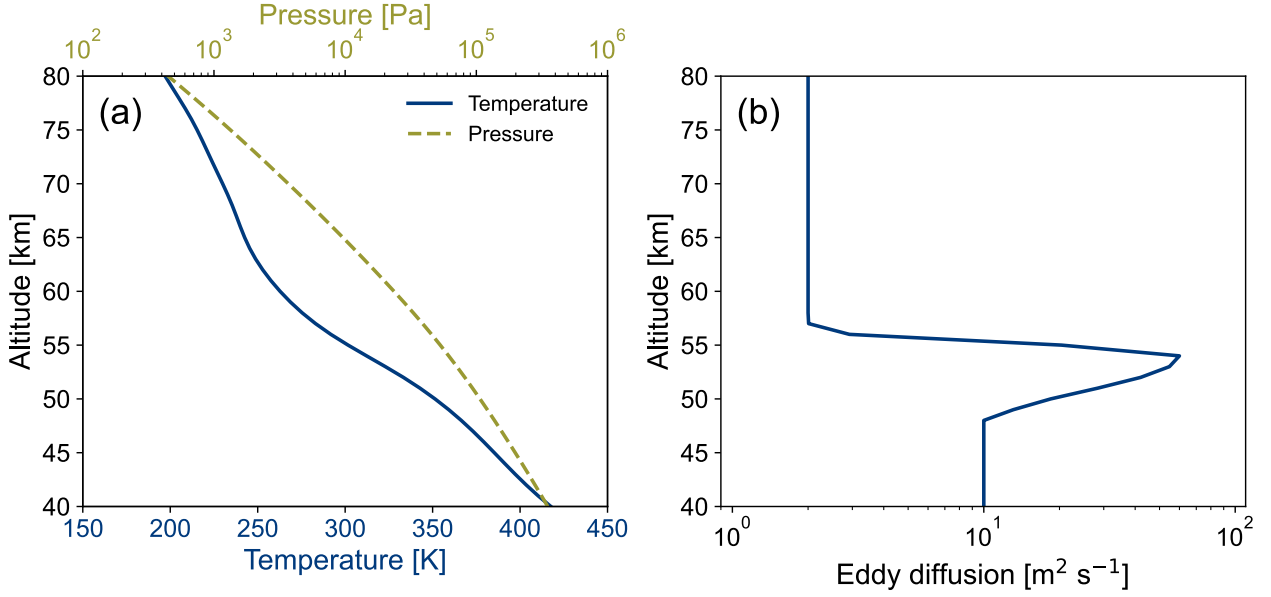


Figure 3.2: (a) Vertical profiles of temperature (blue solid line) and pressure (yellow dashed line) taken from VIRA equatorial profiles (Seiff et al., 1985). (b) Vertical profile of the eddy diffusion coefficient from Mc-Gouldrick and Barth (2023).

production or flux outside of the box is considered. In addition to the moving-center scheme, we performed simulations under the same conditions using the full-moving scheme, which provides an exact solution for condensational growth. The time step was set to 10 s, and the 0-D simulations were run for 1000 s at four different atmospheric conditions corresponding to altitudes of 70, 60, 55, and 50 km, as shown in Figure 3.2a.

The moving-center results are in good agreement with those of the full-moving scheme (Figure 3.3). The system rapidly reaches a near steady-state condition where the ambient vapor pressure equilibrates close to the saturation vapor pressure. Under such conditions, the saturation ratio becomes size-dependent due to the Kelvin effect, which raises the effective saturation vapor pressure over smaller particles. In addition, the rate of change in their volume is much slower for larger particles due to their lower surface-area-to-volume ratio. As a result, small particles experience evaporation, while larger particles are in a state of growth or near-equilibrium.

At 70 km altitude, the results are identical because the condensational mass flux is not significant at this altitude due to the low H_2SO_4 vapor number density of $5.1 \times 10^{12} \text{ m}^{-3}$. Condensation and evaporation significantly affect the size distribution at 60, 55, and 50 km where the H_2SO_4 vapor number densities are 2.4×10^{15} , 5.3×10^{17} , and $5.4 \times 10^{19} \text{ m}^{-3}$, respectively. Evaporation of smaller particles is observed at 60 and 55 km, with slight differences in the timing of evaporation between the two schemes. At 50 km altitude, the effects of growth are more pronounced than at higher altitudes, due to the increased availability of H_2SO_4 vapor under high-temperature conditions. A pit structure

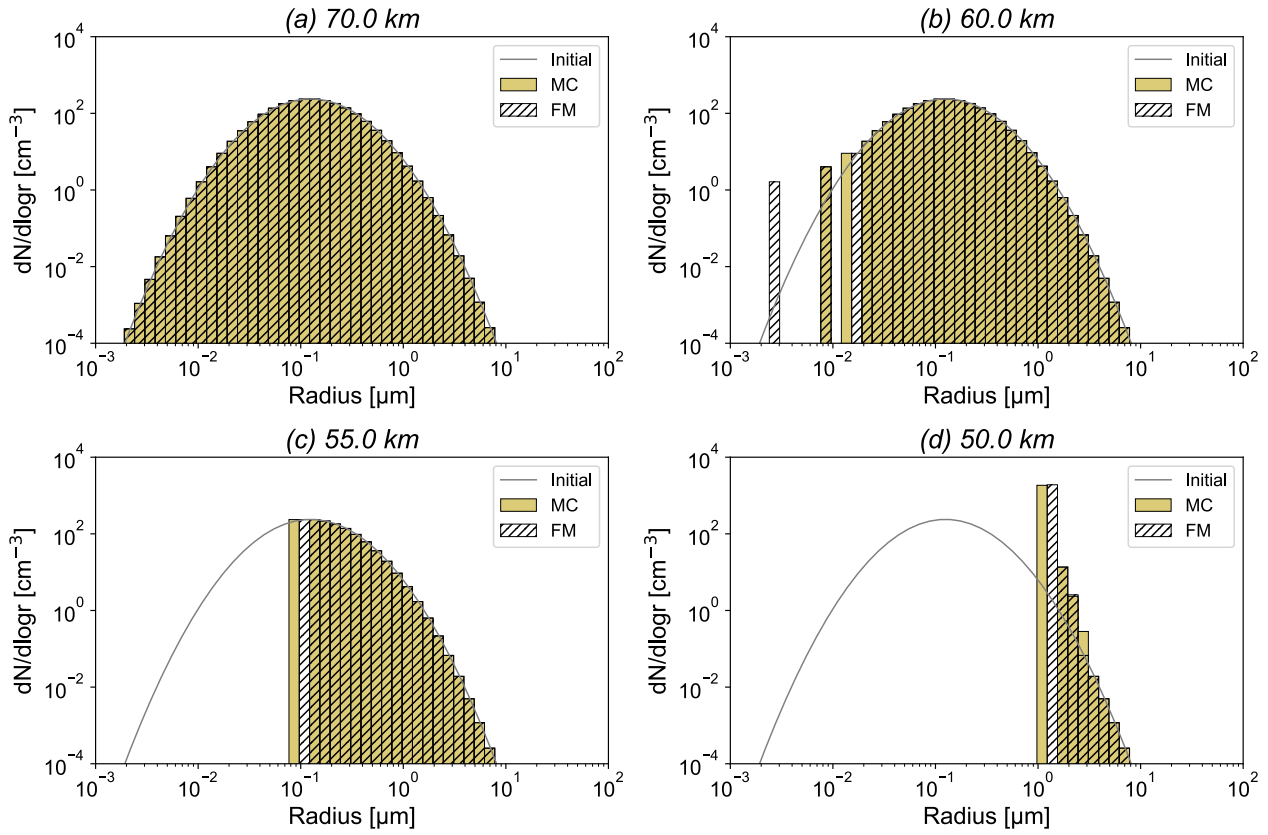


Figure 3.3: Comparison of the moving-center (MC: yellow fill) and full-moving (FM: hatched fill) schemes for H_2SO_4 vapor growth onto aerosol particles at altitudes of (a) 70 km, (b) 60 km, (c) 55 km, and (d) 50 km. The initial size distribution is plotted with a dotted line.

Table 3.2: The normalized relative error of the total number density and mean radius of the moving-center scheme relative to the full-moving scheme.

Altitude (km)	Total number density error	Mean radius error
70	0%	0%
60	11.2%	0.3%
55	14.8%	13.7%
50	3.1%	11.1%

forms between 1 and 2 μm as a result of rapid condensational growth. However, despite the formation of the pit, the results from both methods closely align.

We calculated the normalized relative error of the moving-center scheme relative to the full-moving scheme at the end of the simulation (Table 3.2). The largest error in total number density is 14.8% at 55 km altitude, while the largest error in mean radius is 13.7%, also at 55 km altitude. The mean radius error obtained here is consistent with the 12% error reported in a previous microphysics

algorithm comparison study (Zhang et al., 1999). They also demonstrated that the moving-center scheme yields lower errors than other schemes used in Earth's air-quality models and concluded that the moving-center scheme performs best for condensation. This consistency with previous research strengthens the reliability of the moving-center scheme, especially under conditions relevant to cloud condensation processes.

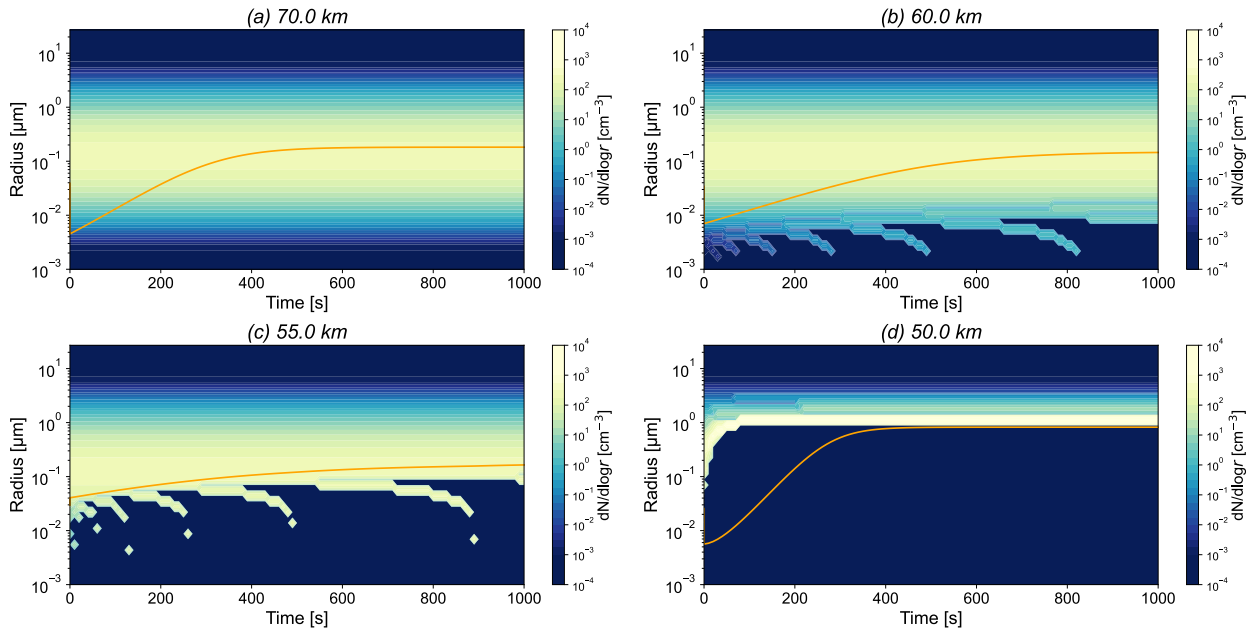


Figure 3.4: Aerosol size distribution as a function of time and radius at altitudes of (a) 70 km, (b) 60 km, (c) 55 km, and (d) 50 km, plotted with critical radius determined by saturation vapor pressure (orange line). The downward (upward) shift of the size distribution corresponds to evaporation (condensation).

Figure 3.4 shows the time series of the particle size distribution at various altitudes. Evaporation occurs at 60 and 55 km altitudes, causing particles at the lower edge of the distribution to shift into smaller size bins. The evaporating particles remain within a single size bin, demonstrating that the moving-center scheme effectively eliminates numerical diffusion. In contrast, Toon et al. (1988) reported that with the full-stationary scheme, the size distribution broadens over time and diverges from the exact solution due to numerical diffusion, particularly when the evaporation rate is rapid. This contrast highlights the benefits of the moving-center scheme, which preserves the integrity of the particle size distribution during rapid evaporation.

On the other hand, given that real atmospheric size distributions are continuous rather than discrete, it may be questionable whether evaporation in nature occurs through such intermittent migrations of isolated size modes toward smaller radii. To examine this, we conducted additional simulations with higher bin resolution (Appendix Figure C.1). These simulation runs demonstrated that particle evaporation proceeds in a more gradual and continuous manner as the resolution increases, consistent with expectations for natural systems. This confirms that the intermittent bin migration observed in the original figure was a resolution artifact, rather than a result of inaccurate evaporation treatment in the model.

The evolution of the critical radius provides further insight into how the particle size distribution is influenced by ambient saturation vapor pressure. The critical radius r_c is determined as follows:

$$r_c = \frac{2\sigma_p m_w}{RT \rho_w \ln(S_q)}, \quad (3.13)$$

where S_q is the supersaturation of the condensing vapor. The critical radius refers to the specific size that a particle must reach in order to continue growing spontaneously. At the beginning of the simulation, high ambient H_2SO_4 vapor pressure results in high supersaturation and a correspondingly small critical radius. As larger particles grow via condensation, they deplete the vapor concentration, leading to a gradual increase in the critical radius. This shift impacts the particle size distribution differently across altitudes due to variations in saturation vapor pressure. At 50 and 55 km, the system responds relatively quickly, and the lower edge of the size distribution closely tracks the critical radius. This behavior is driven by strong condensation and evaporation fluxes associated with high H_2SO_4 saturation vapor pressure. In contrast, at higher altitudes such as 60 and 70 km, where vapor pressure is lower, the response is significantly slower. In particular, the radial growth rate is less than 1 nm h^{-1} for a particle with a radius of $0.01 \text{ }\mu\text{m}$, resulting in no observable changes to the particle size distribution over the simulation timescale.

The pit structure can serve as an indicator of condensation and evaporation. In Figure 3.4b, continuous evaporation occurs around $0.01 \text{ }\mu\text{m}$, causing the size bins to shift toward smaller radii. As the number density in one bin shifts, a pit structure forms in the bin that originally preserved the number density. Subsequently, the number density from the neighboring bin moves to fill the pit. As a result, the pit structure appears to shift toward larger radii during continuous evaporation in a smooth size distribution. On the other hand, rapid condensational growth occurs during the first 200 seconds at 50 km altitude (Figure 3.4d). In this case, the number density distribution shifts toward larger radii. The pit structure created by this process is then replenished by the number density from smaller bins. As a result, the pit structure appears to shift toward smaller radii during condensation growth.

In 0-D box simulations, repeated condensation and evaporation in the moving-center scheme can lead to a narrowing of the aerosol size distribution over time through pit formation, potentially concentrating particles into fewer bins. However, in realistic simulations, such repeated narrowing does not typically occur for the following reasons. First, there are continuous aerosol sources and sinks—such as nucleation, emissions, and photochemical production—help maintain a broad size distribution. Second, multi-dimensional mixing effectively smooths out the pit structures that are more prominent in 0-D models (Mohs and Bowman, 2011). Furthermore, the choice of appropriate bin spacing and resolution further mitigates excessive narrowing. Due to these reasons, many models have successfully implemented moving-center schemes in Earth studies (e.g. Bergman et al., 2012; Fast et al., 2006; Jacobson et al., 2007; Mann et al., 2012; Matsui, 2017), and its applicability to the Venus atmosphere in 1-D is carefully tested in Section 3.3.

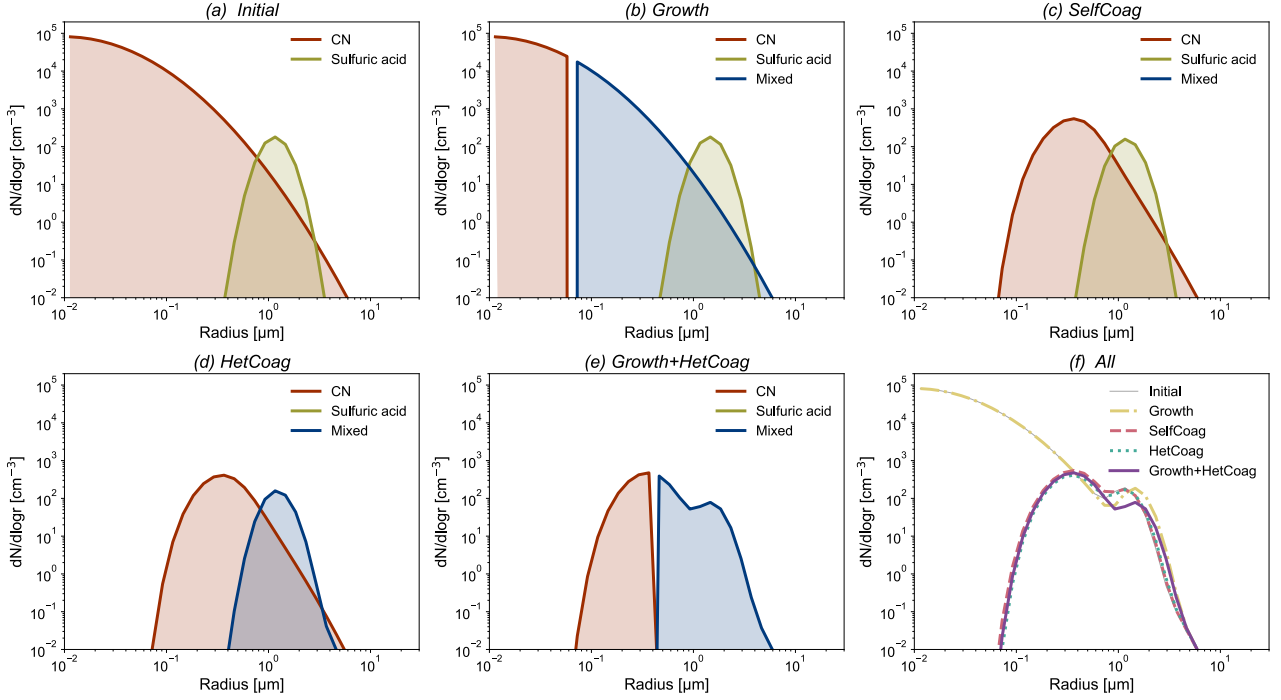


Figure 3.5: Size distributions of CN (red line), sulfuric acid droplet (yellow line), and mixed (blue line) particles at (a) the beginning of the simulation and at the end of simulations with (b) growth alone, (c) self-coagulation alone, (d) heterogeneous coagulation alone, (e) combined growth and heterogeneous coagulation, and (f) a comparison of (a) to (e).

3.2.2 Condensation and coagulation

Next, we performed a 0-D simulation including both condensation and coagulation processes. All processes described in Section 3.1 are incorporated, except for transport, chemical production of H_2SO_4 and CN, and chemical loss of H_2O . This model considers three particle distributions consisting of CN, sulfuric acid droplets, and a mixture of CN and sulfuric acid droplets. The initial particle distribution consists of Mode 1 particles with a radius smaller than $\sim 0.1 \mu\text{m}$ and Mode 2 particles, as shown in Figure 3.5a. Mode 1 particles consist of CN, while Mode 2 particles consist of sulfuric acid droplets. This simulation is analogous to the numerical experiment conducted by Knollenberg and Hunten (1980), but it introduces a Mode 2 particle distribution with a mode radius of $1.16 \mu\text{m}$ to observe particle interactions between independent distributions. Atmospheric temperature and pressure were taken from 60 km altitude in the VIRA profile (Figure 3.2a). The simulation was run for 60 Earth days, following Knollenberg and Hunten (1980). The initial conditions for the condensable species were set to 5% supersaturation for H_2SO_4 and 30 ppm for H_2O . We performed a series of simulations considering different processes: growth alone, self-coagulation alone, heterogeneous coagulation alone, and the combination of condensational growth and heterogeneous coagulation.

The results confirm that the algorithm effectively captures a wide range of particle interaction scenarios. In the growth-only simulation (Figure 3.5b), CN particles larger than $0.05 \mu\text{m}$ are activated and join the mixed size distribution, while sulfuric acid droplets grow slightly from a mode radius of

1.16 μm to 1.47 μm but remain outside the mixed distribution. In the self-coagulation-only experiment (Figure 3.5c), the smaller tail of the Mode 1 distribution significantly decreases due to coagulation, consistent with the numerical experiment by Knollenberg and Hunten (1980). The heterogeneous-coagulation-only experiment shows a similar profile for Mode 1 and Mode 2 particles as observed in the self-coagulation-only case (Figure 3.5d). However, in this scenario, the Mode 2 particles transition into the mixed particle distribution due to heterogeneous coagulation between the CN and sulfuric acid droplet distributions. By the end of the simulation, almost all sulfuric acid droplets have been converted into mixed particles.

When both growth and heterogeneous coagulation are considered, scavenging of Mode 1 particles, a shift in the mode radius of Mode 2 particles from 1.16 to 1.47 μm , and aerosol mixing are observed (Figure 3.5e). This can be interpreted as a combined effect of the growth-only and heterogeneous-coagulation-only experiments. These results indicate that the algorithm effectively captures the complex interactions between growth, coagulation, and mixing processes.

3.3 1-D Simulations

3.3.1 Benchmark simulation I: comparison with the full-stationary scheme

In the first benchmark case, we used the K2024 model developed in Chapter 2 as a proxy for the full-stationary scheme and validated SPECK against the existing model. The K2024 model contains a single size distribution consisting of the H_2SO_4 - H_2O solution and CN material, and hence, the mixing between multi-distributions is not calculated. This simple configuration is also adopted in SPECK for this benchmark case. The size bin ranges from 0.17 μm to 30 μm , and the resolution is varied from low resolution ($V_{\text{rat}} = 4$, $N_{\text{B}} = 12$) to high resolution ($V_{\text{rat}} = \sqrt[8]{2}$, $N_{\text{B}} = 184$) to evaluate the resolution dependence of the moving-center and full-stationary schemes. The simulations are performed under the same conditions.

The model domain extends from 40 to 60 km with a vertical step of 1 km. Photochemical production of H_2SO_4 vapor and CN material is excluded for simplicity. The lower boundary conditions for H_2SO_4 and H_2O are set to 10 ppm and 30 ppm at 40 km, respectively. The CN particle density at the lower boundary is represented by a monodisperse distribution of the smallest particles (0.17 μm) with the particle number density of 40 cm^{-3} , following K2024. At the upper boundary, zero fluxes are assumed for both vapors and particles. The initial condition of all vertical levels is a uniform distribution, set to the value of the lower boundary for vapor and particle species. The timestep is set to 10 s, and the simulation is run for 10^8 s, which is sufficiently longer than the time needed to reach steady state. The simulation results are averaged over the last 10^6 s to compare with the steady-state results obtained by the K2024 model.

Figure 3.6 presents the size distribution at various altitudes for SPECK and the K2024 model with different bin resolutions. In the K2024 results, size distributions tend to broaden at all altitudes when bin resolution is coarse. This broadening effect is particularly evident in the smaller size bins at altitudes of 54 and 56 km, with the lowest bin resolution. Additionally, the upper edge of the size

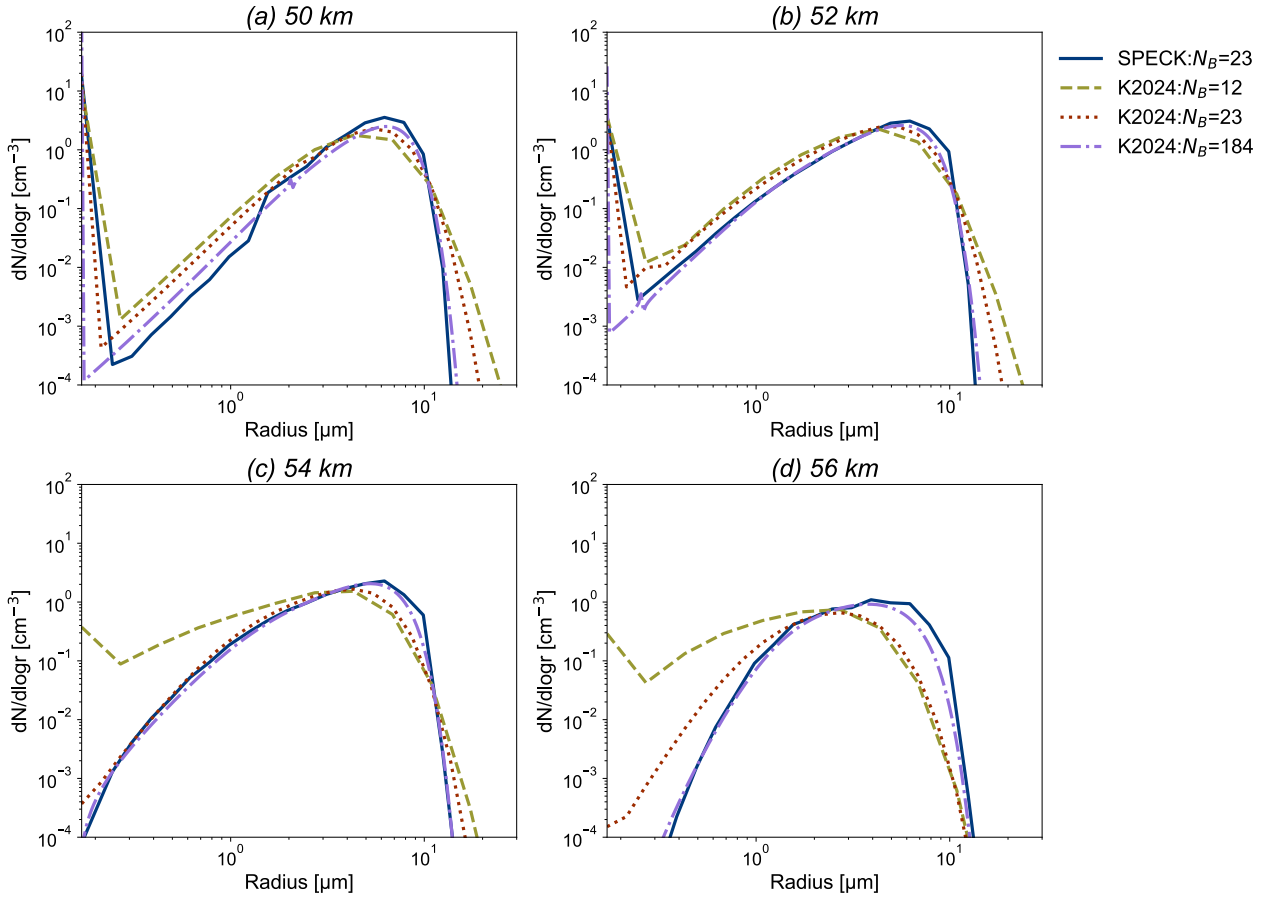


Figure 3.6: Steady-state size distribution at various altitudes calculated by SPECK (blue solid line) and K2024 model with different bin resolutions plotted with yellow dashed line ($N_B = 12$), red dotted line ($N_B = 23$), and purple dot-dashed line ($N_B = 184$). Note that the resolution of SPECK is $N_B = 23$.

distribution also exhibits broadening, especially at 50, 52, and 54 km. As the bin resolution becomes finer, the size distribution narrows. In contrast, SPECK results show less broadening compared to the K2024 model with the same bin resolution ($N_B = 23$). Furthermore, as the bin resolution of the K2024 model increases, its results converge toward those of SPECK.

The close agreement between SPECK and the high-resolution full-stationary scheme highlights the resilience of the moving-center scheme against numerical diffusion. In the full-stationary scheme implemented in K2024, numerical diffusion occurs at lower bin resolutions because size growth is represented as Eulerian advection in size space. Generally, numerical diffusion effects diminish as resolution increases relative to the scale of the physical process, allowing fine structures to be resolved. This trend is evident in Figure 3.6, where the size distribution narrows with increasing resolution. In contrast, the moving-center scheme implemented in SPECK nearly eliminates numerical diffusion by calculating growth using a Lagrangian approach. As a result, the predicted size distribution does not exhibit broadening and closely matches the results of the high-resolution full-stationary

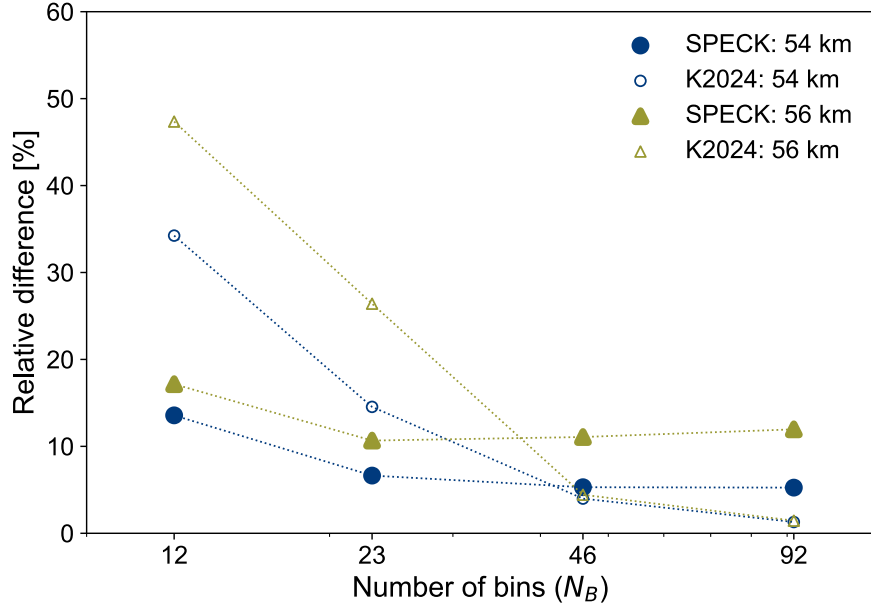


Figure 3.7: Relative differences (%) in the mean radius simulated by SPECK and the K2024 model compared to a reference value at various altitudes as a function of the number of bins. The result of the highest resolution of the K2024 model ($N_B = 184$) is used as a reference value. The four data points at each altitude represent $N_B = 12, 23, 46$, and 92 .

scheme. This demonstrates that SPECK can achieve size distribution predictions comparable to high-resolution full-stationary simulations without requiring fine bin resolution.

It should be noted that the SPECK particle number density at 50 km exhibits a rapid decrease around a $1.5 \mu\text{m}$ radius (Figure 3.6a), which is attributed to the Lagrangian treatment of evaporation. Near the cloud bottom at 50 km, the evaporation timescale is extremely short, and this process is observed as Lagrangian advection of a single size bin toward smaller radii, as illustrated in Figure 3.4c. Consequently, after time averaging, the size distribution at 50 km appears discontinuous around $1.5 \mu\text{m}$, below which particle evaporation dominates. This method of evaporation aligns with the exact solution, as demonstrated in Figure 3.3, and reflects the behavior of real atmospheric processes. In contrast, the K2024 results predict a smooth decrease in number density toward smaller radii. This feature arises from the Eulerian treatment of evaporation, where the scheme handles evaporation through the number density flux between adjacent size bins. As a result, the Eulerian scheme causes particle number density to spread across size bins where particles should not exist, creating a smoother but less physically accurate size distribution.

Figure 3.7 shows the relative differences in the mean radius simulated by SPECK and the K2024 model compared to reference values. The reference value is chosen as the mean radius calculated by the highest resolution ($N_B = 184$) of K2024, serving as the “true” value. The results from the K2024 model with lower resolutions converge toward the reference value as resolution increases, a trend also

observed in Figure 3.6. The relative difference for the K2024 model decreases by nearly half as the resolution is doubled, reflecting the reduced impact of numerical diffusion with increasing resolution.

In contrast, the relative difference for SPECK remains consistent across various bin resolutions. Specifically, for bin resolutions greater than $N_B = 23$, the values are very similar, with differences of $<15\%$ at 56 km and $<8\%$ at 54 km. This consistency is due to the resilience of the moving-center bin scheme against numerical diffusion. As a result, SPECK predicts a mean radius more accurately than the K2024 model when the bin resolution is lower than $N_B = 46$. A typical resolution used in microphysics models is $V_{\text{rat}} = 2$, which indicates that the particle volume doubles between adjacent bins, as indicated by Equation (3.4). The resolution corresponds to $N_B = 23$ in this simulation. As shown in Figure 3.7, SPECK achieves closer agreement with the reference value compared to the K2024 model at this typical resolution, highlighting enhanced performance in typical microphysics simulation settings.

3.3.2 Benchmark simulation II: comparison with CARMA

In the second benchmark case, we perform 1-D simulations with the same setting as the recent Venus microphysics study by CARMA (McGouldrick and Barth, 2023). This simulation serves as a realistic application of our model to Venus' cloud system. Two types of simulations were conducted, varying the size bin resolution from nominal ($V_{\text{rat}} = 2$) to low ($V_{\text{rat}} = 4$) resolution cases, with size bins ranging from 10 nm to 30 μm . The former has the same resolution as CARMA, while the latter has a resolution that is half that of CARMA.

The model spans from 40 to 80 km with a vertical grid step of 1 km and considers two types of size distributions consisting of CN and mixed particles. In addition to self-coagulation, heterogeneous coagulation between the two distributions is included. The chemical production of H_2SO_4 vapor is represented by a Gaussian distribution with a column production rate of $6 \times 10^{15} \text{ m}^{-2} \text{ s}^{-1}$, representing $\left. \frac{\partial C_q}{\partial t} \right|_{\text{chem}}$ in Equation (3.3) for $q = 1$. The peak altitude and half width of the Gaussian distribution are 61 km and 2 km, respectively. The loss of H_2O and production of elemental sulfur S is scaled by the following net chemical reaction (Imamura and Hashimoto, 2001), which is introduced in Section 1.3.1 as Equation (1.11) but reintroduced for convenience:



This reaction results in the loss of two H_2O molecules and the production of one S atom per production of two H_2SO_4 molecules. The lower boundary conditions for H_2SO_4 and H_2O are set to 10 ppm and 30 ppm at 40 km, respectively. The concentrations of these gas species are assumed to be zero above the upper boundary. The population of CN particles is fixed to a lognormal distribution at the lower boundary following the LCPS observation by Knollenberg and Hunten (1980), while liquid components are assumed to be zero. The upper boundary conditions of all particles are set to zero flux. We refer the reader to McGouldrick and Barth (2023) for more detailed descriptions of the model and settings.

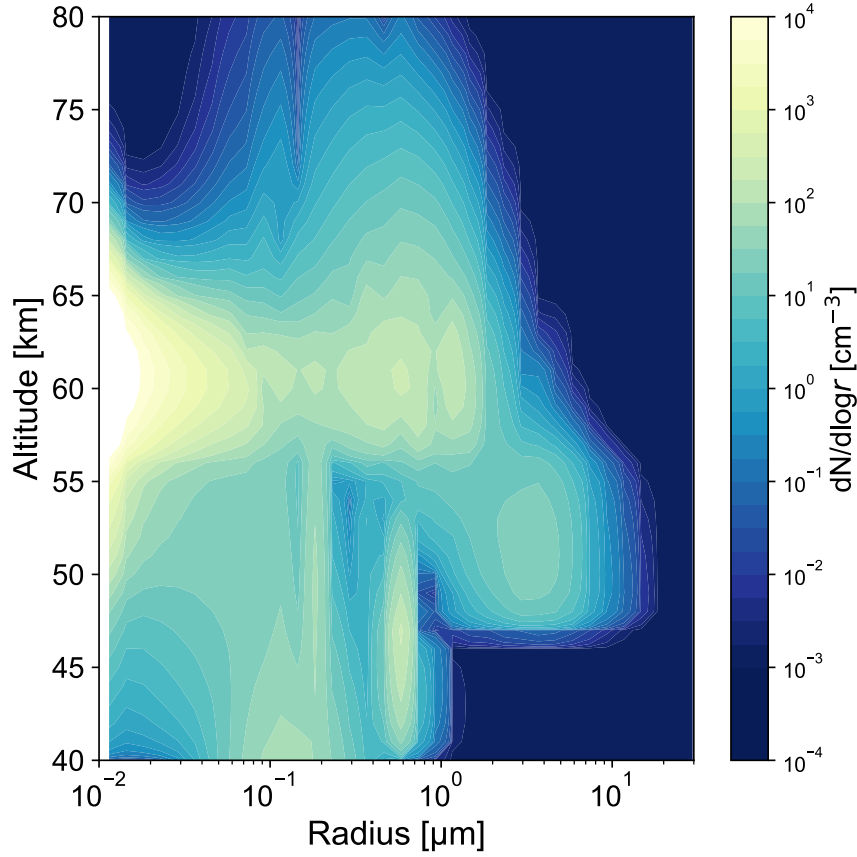


Figure 3.8: Combined size distribution of CN and mixed particles as a function of particle radius and altitude in the nominal resolution case ($V_{\text{rat}} = 2$).

We set the timestep to 10 s and ran the simulation for 10^8 s, which is sufficiently longer than the timescales of the physical processes incorporated within the model, such as eddy diffusion, sedimentation, condensation, and coagulation. We confirmed that the pit formation does not increase in severity as the simulation progresses (Appendix Figures C.2-C.5). Simulation results are averaged over the last 10^6 s of the simulation for comparison with the steady-state results obtained by McGouldrick and Barth (2023).

Figure 3.8 shows the simulated size distribution as a function of particle radius and altitude, and Figure 3.9 shows the size distribution at various altitudes in the nominal resolution case ($V_{\text{rat}} = 2$). The cloud structure can be divided into two distinct layers: a condensational cloud below 57 km and a photochemical cloud above 57 km. The former corresponds to the lower and middle cloud layers observed by the LCPS (Knollenberg and Hunten, 1980), while the latter corresponds to the upper cloud and upper haze. Larger particles, ranging from 1 to 10 μm , are evident in the condensational cloud, corresponding to Mode 2 and Mode 3 particles (Figures 3.9 and 3.9d). These cloud particles form as a result of vigorous condensation driven by the enhanced eddy diffusion. In addition to the larger particles, there is also a small particle population with radii smaller than 1 μm . This population

is composed mainly of CN particles. The CN particles remain unwetted because their radii are smaller than the critical radius defined by the Kelvin effect.

Above the condensational cloud lies the photochemical cloud, which is maintained through the chemical production of H_2SO_4 vapor. The most populated region is around $0.01\ \mu\text{m}$ due to the photochemical production of CN particles (Figures 3.9a and 3.9b). These CN particles grow to approximately $0.1\ \mu\text{m}$ by coagulation, where they overcome the Kelvin barrier and join the mixed droplet size distribution. These droplets grow by condensation and join the condensational cloud through sedimentation, while some particles are carried upward by eddy diffusion and contribute to the upper haze above 70 km. The structure and particle cycles are similar to those described by McGouldrick and Barth (2023), indicating that our model successfully captures the general characteristics of the Venus cloud layers.

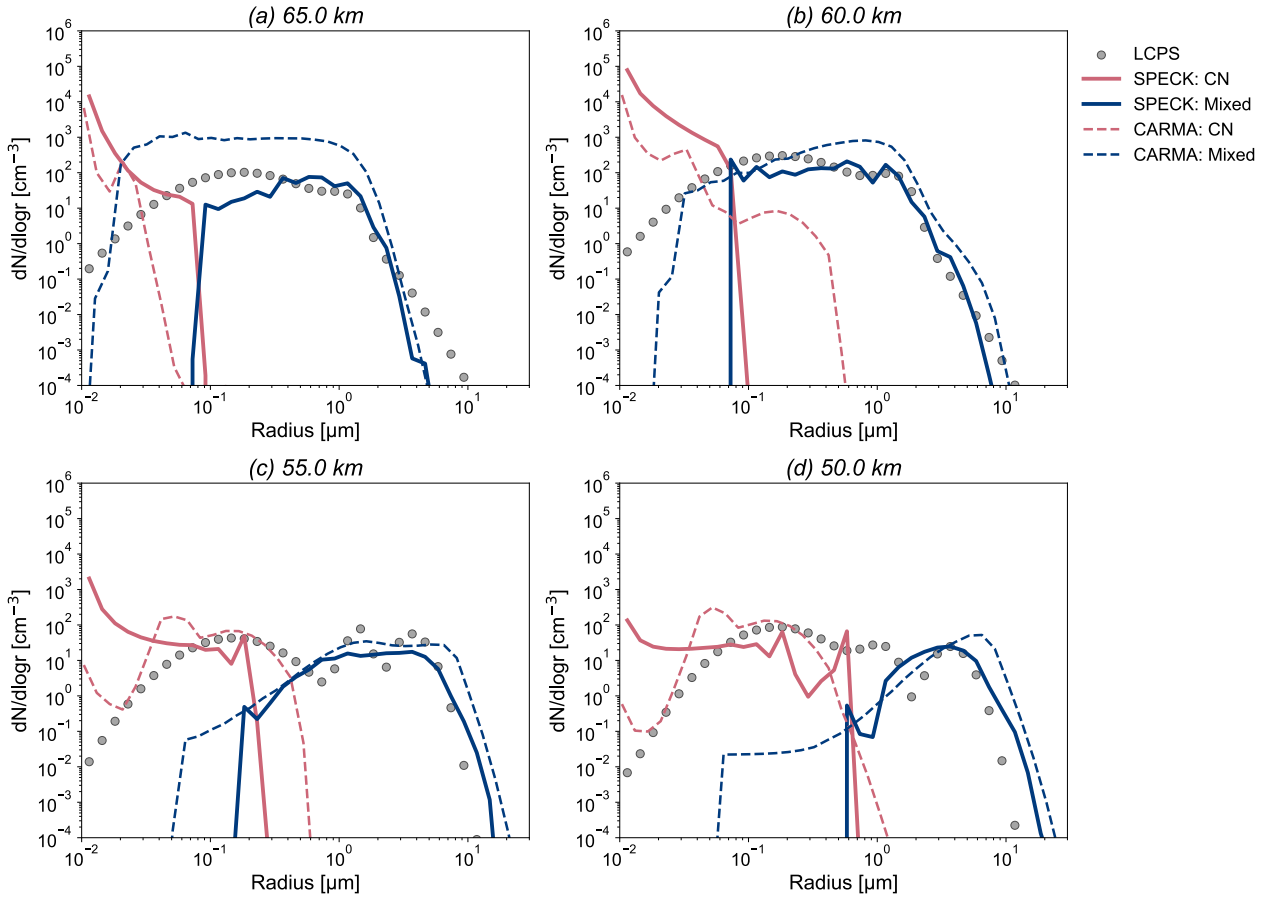


Figure 3.9: Size distribution of the CN (red solid line) and Mixed particles (blue solid line) at altitudes of (a) 65 km, (b) 60 km, (c) 55 km, and (d) 50 km in the nominal bin resolution case ($V_{\text{rat}} = 2$). Equatorial profiles from the CARMA simulation (McGouldrick and Barth, 2023) are also plotted with dashed lines. The simulated distributions are compared with the LCPS in-situ measurements by Knollenberg and Hunten (1980) (gray circles).

The simulated CN distributions exhibit two distinct peaks below 57 km, as shown in Figure 3.8 and Figures 3.9c and 3.9d. These two peaks originate from different altitudes. The peak around $0.6\ \mu\text{m}$ forms when mixed particles evaporate near the cloud base, leaving behind their CN. A similar peak is observed in the CN distribution from CARMA, although the peak radius is somewhat smaller around $0.05\ \mu\text{m}$. In contrast, the peak around $0.2\ \mu\text{m}$ is generated by the evaporation of mixed particles descending from the upper cloud.

The upper cloud region is highly supersaturated with H_2SO_4 vapor, which minimizes the impact of the Kelvin effect. However, when these particles enter the less supersaturated middle cloud, the Kelvin effect becomes significant, causing particles smaller than the critical radius to evaporate. These peaks may be better resolved than in the previous study due to the use of the moving-center bin scheme. As shown in Figure 3.4, the moving-center scheme allows size bins to evaporate to their exact sizes while minimizing numerical diffusion. Consequently, the CN distribution remains confined to a very narrow region without spreading when it is left behind by evaporation.

Figure 3.10 shows the size distribution at various altitudes in the low resolution case ($V_{\text{rat}} = 4$). The simulated distributions are nearly identical to those simulated with nominal bin resolution (Figure 3.9). As already seen in Section 3.3.1, the moving-center scheme predicts similar size distributions regardless of the bin resolution, and this trend is reproduced under more complex settings.

In the current simulation settings, halving the bin resolution reduced the computation time to about 0.16 times that of the nominal resolution, yielding a sixfold efficiency gain. This efficiency is especially critical for 3-D modeling, as fewer size bins lower both the number of tracers and the computational load, making explicit microphysics calculations more feasible. In existing 3-D Venus cloud simulations, microphysics processes are not explicitly calculated, and cloud sizes are represented by three separate modes (Stolzenbach et al., 2023) or four modes (Karyu et al., 2023). The results from SPECK suggest that the number of size bins needed to reasonably simulate microphysical properties can be as low as $N_B = 12$, only three to four times larger than the number of cloud modes currently used in the 3-D models.

Implementing the moving-center scheme in GCMs provides additional advantages that offset the computational cost. By reducing numerical diffusion and accurately representing size distributions with fewer bins, this scheme enables more precise cloud microphysics simulations. This is crucial for understanding cloud dynamics, particle cycling, and radiative effects in Venus's atmosphere. Overall, the moving-center scheme offers an efficient and practical path to incorporating full microphysics in 3-D models, potentially enhancing the predictive capability of Venus GCMs.

3.4 Chapter Summary

We presented a new microphysics model based on the moving-center bin scheme, which effectively eliminates numerical diffusion caused by condensation and evaporation processes. The model also includes interactions between multiple size distributions, making it more versatile for simulating the composition and mixing state of aerosols.

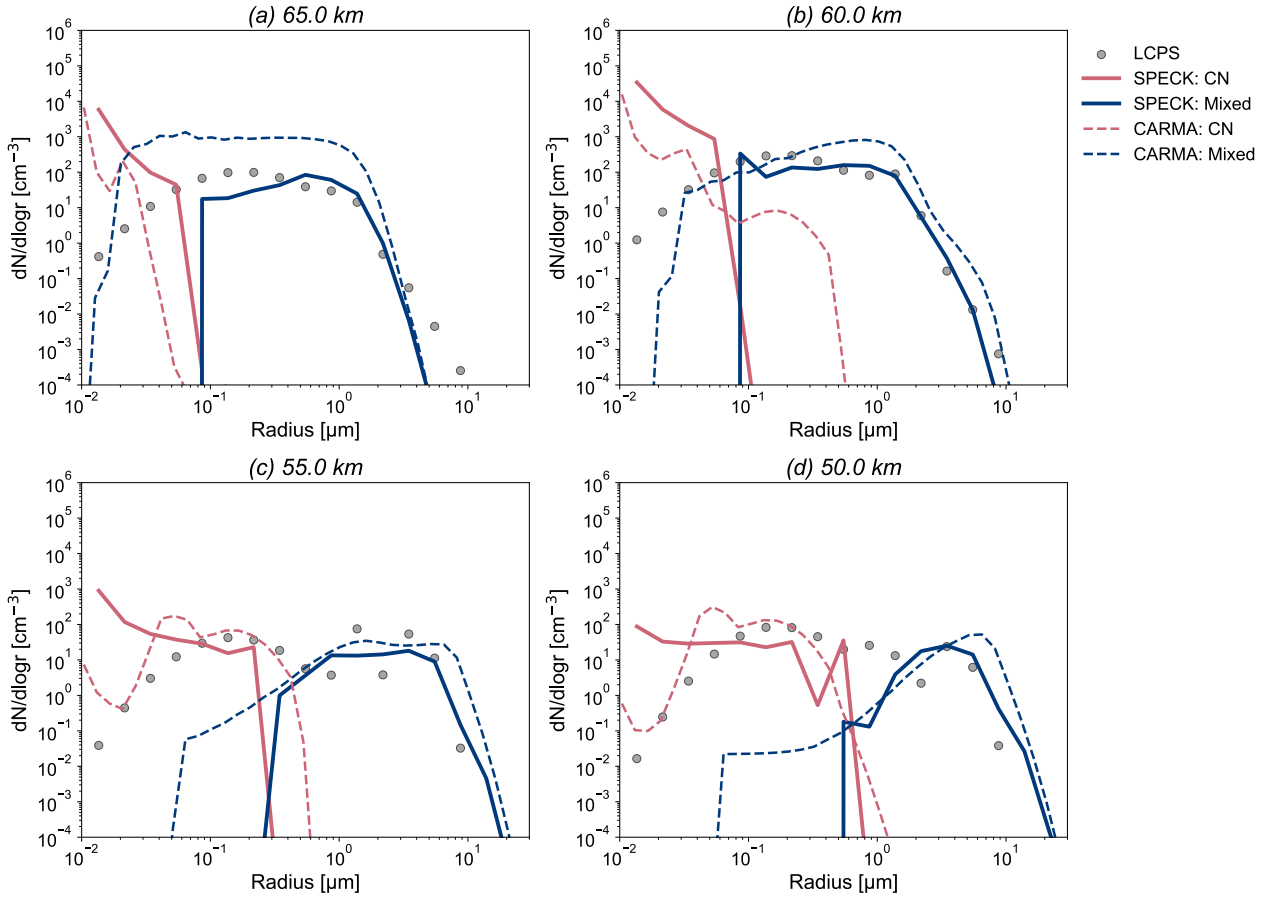


Figure 3.10: Size distribution of the CN (red solid line) and Mixed particles (blue solid line) at altitudes of (a) 65 km, (b) 60 km, (c) 55 km, and (d) 50 km in the low bin resolution case ($V_{\text{rat}} = 4$). Equatorial profiles from the CARMA simulation (McGouldrick and Barth, 2023) are also plotted with dashed lines. The simulated distributions are compared with the LCPS in-situ measurements by Knollenberg and Hunten (1980) (gray circles).

The 0-D simulation results confirm that the microphysics framework is a robust and reliable tool for modeling aerosol microphysics on Venus, particularly in capturing condensation and evaporation dynamics. The 0-D simulation using condensation alone showed nearly identical results with the exact solution within a normalized relative error of less than 15%. Heterogeneous coagulation processes significantly change the composition of aerosols, highlighting the importance of mixing between independent size distributions.

We performed two benchmark simulations to validate SPECK against existing models. In the first benchmark case, we compared our simulation results to those computed using a full-stationary bin scheme (K2024) with varying bin resolutions. We found that SPECK can predict size distributions similar to those obtained with the high-resolution full-stationary scheme, regardless of the bin resolution. In contrast, the size distributions predicted by the K2024 model strongly depend on bin resolution and deviate from the high-resolution results as the resolution decreases. This demonstrates

that SPECK successfully eliminates the effects of numerical diffusion, highlighting its capability for high-accuracy condensation modeling.

In the second benchmark case, we further validated SPECK by comparing our results to those from recent 1-D simulations using CARMA (McGouldrick and Barth, 2023), applying the same simulation settings. The simulated size distributions are in good agreement with CARMA and in-situ LCPS observations, indicating that SPECK can accurately reproduce the complex cloud structure of Venus. Additionally, a simulation performed at lower resolution produced consistent results, demonstrating that SPECK can simulate Venus' cloud microphysics accurately without requiring a large number of bins. Notably, reducing the bin resolution by half resulted in an efficiency increase of more than six times compared to the original, while maintaining simulation accuracy. These findings suggest that SPECK is both efficient and well-suited for future 3-D modeling applications.

Future work can build on this framework to model multi-distribution interactions with high accuracy, offering further insights into complex aerosol processes in the Venus atmosphere. This framework is also applicable to various planetary atmospheres with aerosols, including Mars, Titan, the gas giants, and exoplanets.

Chapter 4

A Cosmic Origin of Venus' Lower Haze

This chapter is based on the following paper, under review in *Nature Astronomy* at the time of writing this thesis:

Hiroki Karyu, Takeshi Kuroda, Anni Määttänen, Arnaud Mahieux, Sébastien Viscardy, Naoki Terada, Séverine Robert, Ann Carine Vandaele, and Michel Crucifix (2025). A cosmic origin of Venus' lower haze. *Nature Astronomy*, under review.

4.1 Model Description

4.1.1 Overview

We used SPECK developed in Chapter 3 to simulate the vertical profiles of cloud particles and condensational vapor species. The model incorporates 45 size bins spanning from 1 nm to 30 μm , with the volume doubling from one bin to the next. The vertical domain extends from 40 km to 100 km altitudes with a 1 km interval. A central feature of SPECK is the implementation of a moving-center bin scheme (Jacobson, 1997b, 2002, 2005) to represent the particle size distributions. This approach is designed to minimize numerical diffusion by treating particle growth in a Lagrangian manner within the size space for each bin. Using this scheme, SPECK has been shown to predict the size distribution with higher accuracy than existing models that calculate the particle growth in an Eulerian manner. Furthermore, SPECK is a highly versatile model, capable of handling multicomponent size distributions, as well as their interactions. This capability allows for the assessment of complex particle composition evolution in the present study. These newly implemented schemes have been rigorously

tested, validated against analytical solutions, and demonstrated to reproduce the observed Venus' cloud structure as measured by the PV LCPS (Karyu et al., 2025).

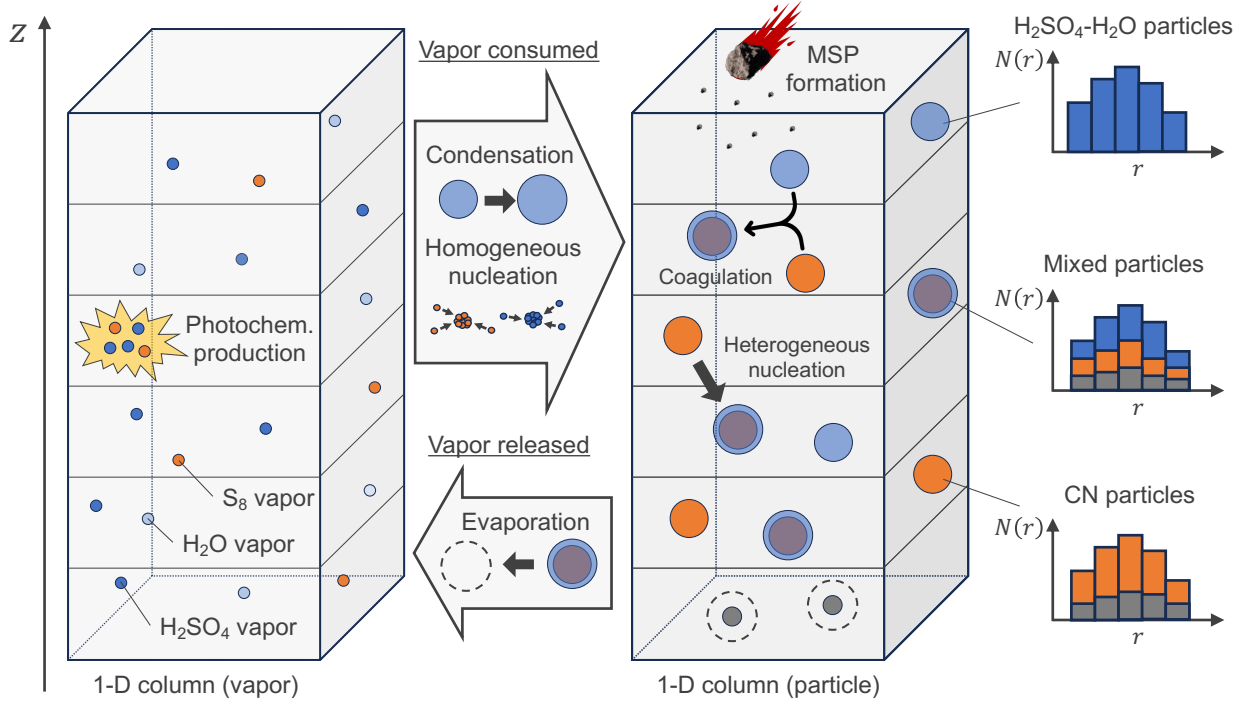


Figure 4.1: Sketch of the model structure and microphysical processes considered in the model.

Figure 4.1 summarizes the model structure and the implemented microphysical processes. For this study, the model, which originally included H₂SO₄ and H₂O (Karyu et al., 2025), has been updated to also include gaseous and solid sulfur allotrope (S₈). In addition, we include olivine (MgFeSiO₄) as an involatile component derived from cosmic dust ablation (detailed in Section 4.1.4). In this configuration, the model includes three gas species (S₈, H₂SO₄, and H₂O) and four particle components (olivine, solid S₈, liquid H₂SO₄, and liquid H₂O), as detailed in Table 4.1. Particles are categorized into three size distributions based on their composition: a CN distribution, an H₂SO₄-H₂O liquid distribution, and a mixed-particle distribution. The specific components of each distribution are also listed in Table 4.1.

4.1.2 Sulfur allotrope microphysics

We incorporate atmospheric and microphysical processes related to sulfur allotropes, such as photochemical production, condensation, evaporation, and nucleation. Sulfur allotropes are generated through the photolysis of SO₂. Based on this assumption, Imamura and Hashimoto (2001) proposed that the production rates of S and H₂SO₄ can be expressed using the following stoichiometric relationship, introduced as Equation (1.8) but reintroduced for the sake of convenience, as follows:

Table 4.1: Summary of gaseous and particle components included in the model

Type	Substance			
	MgFeSiO ₄	S ₈	H ₂ SO ₄	H ₂ O
Gas		○	○	○
CN particle distribution (A)	○	○		
H ₂ SO ₄ -H ₂ O particle distribution (B)			○	○
Mixed particle distribution (AB)	○	○	○	○



Equation (4.1) shows that one S molecule is produced for every two H₂SO₄ molecules produced. Because our model does not compute complex sulfur photochemistry, we parameterized the S production rate using the H₂SO₄ production rate calculated by Krasnopolsky (2012), following Karyu et al. (2024). The H₂SO₄ production is divided by $\cos(45^\circ)$ since Krasnopolsky (2012) used mid-latitude conditions. For simplicity, we also assume that all sulfur allotropes are represented as S₈, as it is the most stable and abundant form under Venus' atmospheric conditions (Krasnopolsky, 2007; Lyons, 2008). As a whole, the chemical production rate of S₈ becomes 1/16 of H₂SO₄.

The saturation vapor pressure of S₈ is formulated according to Onischuk et al. (2016) as follows:

$$P_{\text{sat},\text{S}_8}(T) = \begin{cases} 1.33 \times 10^{11} \exp\left(-\frac{9484.7}{T}\right), & T > 390 \text{ K} \\ 6.60 \times 10^{12} \exp\left(-\frac{11145.1}{T}\right), & T \leq 390 \text{ K}, \end{cases} \quad (4.2)$$

where T is the atmospheric temperature (K). The density ρ_{S_8} and surface tension σ_{S_8} of S₈ are set at 2000 kg m⁻³ (Imamura and Hashimoto, 2001) and 72 mN m⁻¹ (Onischuk et al., 2016), respectively.

The homogeneous nucleation of S₈ is calculated using classical nucleation theory, which is introduced in Section 1.1.2 as Equation (1.1)-(1.3), but reintroduced for convenience:

$$J_{\text{hom}} = 4\pi r_c^2 Z_{\text{homo}} N_v \sqrt{\frac{k_B T}{\pi m_v}} \exp\left(-\frac{\Delta G_{\text{hom}}}{k_B T}\right), \quad (4.3)$$

$$r_c = \frac{2\sigma_v m_v}{\rho_v k_B T \ln S_v}, \quad (4.4)$$

$$\Delta G_{\text{hom}} = \frac{4\pi r_c^2 \sigma_v}{3}, \quad (4.5)$$

where, J_{hom} is the homogeneous nucleation rate (m⁻³ s⁻¹), k_B is the Boltzmann constant (J K⁻¹), N_v is the vapor concentration (m⁻³), m_v is the molecular mass (kg) of vapor, S_v is the saturation ratio of

vapor, and Z_{hom} is the Zeldovich nonequilibrium factor for homogeneous nucleation. The nucleated S_8 particles are introduced into the smallest size bin of 1 nm in the CN size distribution.

Heterogeneous nucleation is included to simulate the activation of CN particles into mixed particles coated with liquid $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$. Several approaches exist to calculate the heterogeneous nucleation rate, including the surface diffusion and the direct vapor deposition approaches (Määttä et al., 2005). The former requires parameters that depend strongly on molecular-scale interactions between the condensate and the nucleating surface (e.g., adsorption energy). However, these parameters are not constrained for a system of solid S_8 and liquid $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$. Therefore, we incorporate the direct vapor deposition approach (Inada, 2002; Määttä et al., 2005), which does not require these unconstrained parameters:

$$J_{\text{het}}(r_{\text{CN}}) = \pi r_{\text{c}}^2 Z_{\text{het}} N_{\text{v}} c_{1,\text{s}} \sqrt{\frac{k_{\text{B}} T}{2\pi m_{\text{v}}}} \exp\left(-\frac{f(r_{\text{CN}}) \Delta G_{\text{hom}}}{k_{\text{B}} T}\right), \quad (4.6)$$

$$c_{1,\text{s}} = \frac{2r_{\text{v}}}{v_{\text{v}}}, \quad (4.7)$$

where J_{het} is the heterogeneous nucleation rate ($\text{m}^{-2} \text{s}^{-1}$), r_{CN} is the radius (m) of a CN particle, r_{v} is the radius (m) of the vapor molecule, v_{v} is the volume (m^3) of the vapor molecule, Z_{het} is the Zeldovich nonequilibrium factor for heterogeneous nucleation, $m = \cos \theta_{\text{c}}$ is the contact parameter that is a function of the contact angle θ_{c} at which an embryo contacts a substrate at the host particle, as previously introduced as Equation (1.6). The factor f is a function of m and the ratio of the radius of the CN to the critical radius $X = r_{\text{CN}}/r_{\text{c}}$, further described as follows:

$$2f = 1 + \left(\frac{1-mX}{g}\right)^3 + X^3 \left[2 - 3\left(\frac{X-m}{g}\right) + \left(\frac{X-m}{g}\right)^3\right] + 3mX^2 \left(\frac{X-m}{g} - 1\right), \quad (4.8)$$

$$g = \sqrt{1 + X^2 - 2Xm}. \quad (4.9)$$

While Equation (1.5) in Section 1.1.2 provides the factor f for a flat surface, the expression here includes the effect of CN's sphericity for greater precision. Since there is no experimental data on the contact angle between S_8 and liquid H_2SO_4 , we set the contact parameter to unity ($m = 1$) for simplicity. This treatment also guarantees consistency with previous microphysics modeling efforts, which assumed perfect wetting of CN (e.g., Imamura and Hashimoto, 2001; Gao et al., 2014; McGouldrick and Barth, 2023; Karyu et al., 2024). The heterogeneous nucleation rate is calculated for the entire size range of the CN distribution. Then, the nucleation probability $P(r_{\text{CN}})$ —the ratio between the number of activated CN (N_{act}) and the total number of CN (N_{tot})—is calculated for each size bin, using the following relationship (Lazaridis et al., 1992):

$$P(r_{\text{CN}}) = \frac{N_{\text{act}}}{N_{\text{tot}}} = 1 - \exp\left(-J_{\text{het}}(r_{\text{CN}}) r_{\text{CN}}^2 \Delta t\right), \quad (4.10)$$

where Δt is the nucleation time (s) set to the model time step of 10 s. At each time step, the number of activated CN are calculated using $P(r_{\text{CN}})$ and transferred from the CN distribution to the mixed droplet distribution.

The condensation and evaporation rates of S_8 are calculated using the APC scheme (Jacobson, 1997b, 2005), as described in Karyu et al. (2025). Following Toon et al. (1982), we assume that CN particles can grow by S_8 condensation, whereas S_8 does not condense onto particles already coated with liquid $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$. This assumption is consistent with observations suggesting that the upper cloud particles have a surface layer of liquid H_2SO_4 (Hansen and Hovenier, 1974; Kawabata et al., 1980; Markiewicz et al., 2018), implying that S_8 condensation onto a liquid surface is ineffective.

4.1.3 Homogeneous nucleation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$

We incorporate the binary homogeneous nucleation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ using the parameterization of Määttä et al. (2018). This parameterization is a significant update from the previous version by Vehkamäki et al. (2002), as its applicable temperature and relative humidity ranges are extended to Venus' atmospheric conditions. The newly formed particles are introduced into the smallest size bin (1 nm) of the $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ droplet distribution, as is the case for the S_8 nuclei.

4.1.4 MSP influx

Meteoritic Smoke Particles (MSPs) are introduced into the CN size distribution as involatile particles composed of olivine (FeMgSiO_4) with a 1-nm radius (Plane et al., 2015; Saunders and Plane, 2006; Frankland et al., 2017). The cosmic dust ablation model predicts that the peak ablation altitude of MSPs is located at ~ 110 km (Carrillo-Sánchez et al., 2020), which is well above the model-top altitude of this study. Thus, we assume a steady state MSP influx at the model-top altitude of 100 km, whose value is scaled by the cosmic dust ablation rate. The cosmic dust ablation rate is not well constrained on Venus, while an ablation model predicts it to be ~ 10 tons day^{-1} (Carrillo-Sánchez et al., 2020). Based on this, we set the nominal cosmic dust ablation flux to be 10 tons day^{-1} and conducted a parameter study ranging from 1 to 1000 tons day^{-1} .

It should be noted that the chemical form of iron compounds within H_2SO_4 droplets in Venus' atmosphere is still a subject of ongoing discussion. Ablated metals are expected to initially form metal carbonates due to the high CO_2 density; these could then react with HCl , which is also abundant above the clouds (Marcq et al., 2018). Therefore, the resulting smoke particles may form from the coagulation of metal chloride molecules. After these particles dissolve in cloud droplets and subsequently evaporate below the cloud deck, the residual particles are likely metal sulfates. However, since these details do not affect the main conclusion of this study, our model assumes a simplified MSP composition of olivine.

4.1.5 Atmospheric conditions

The atmospheric pressure and temperature profiles are taken from the equatorial conditions of the VIRA (Seiff et al., 1985), as shown in Figure 4.2a. The equatorial profiles are chosen for comparison with past probe aerosol measurements since all of them landed in the equatorial region. For vertical transport, we follow the approach of Karyu et al. (2024) and represent all transport processes using eddy diffusion. The Eddy diffusion profile is constructed based on a recent cloud microphysics model study (Karyu et al., 2024), which simultaneously reproduces Venus' cloud structure and condensational vapor profiles from 40 to 100 km, as shown in 4.2b. To reproduce H_2O profile above the cloud top, the eddy diffusion coefficient in the upper cloud layer is set to $2.7 \text{ m}^2 \text{ s}^{-1}$, which is slightly larger than $2 \text{ m}^2 \text{ s}^{-1}$ used in Karyu et al. (2024).

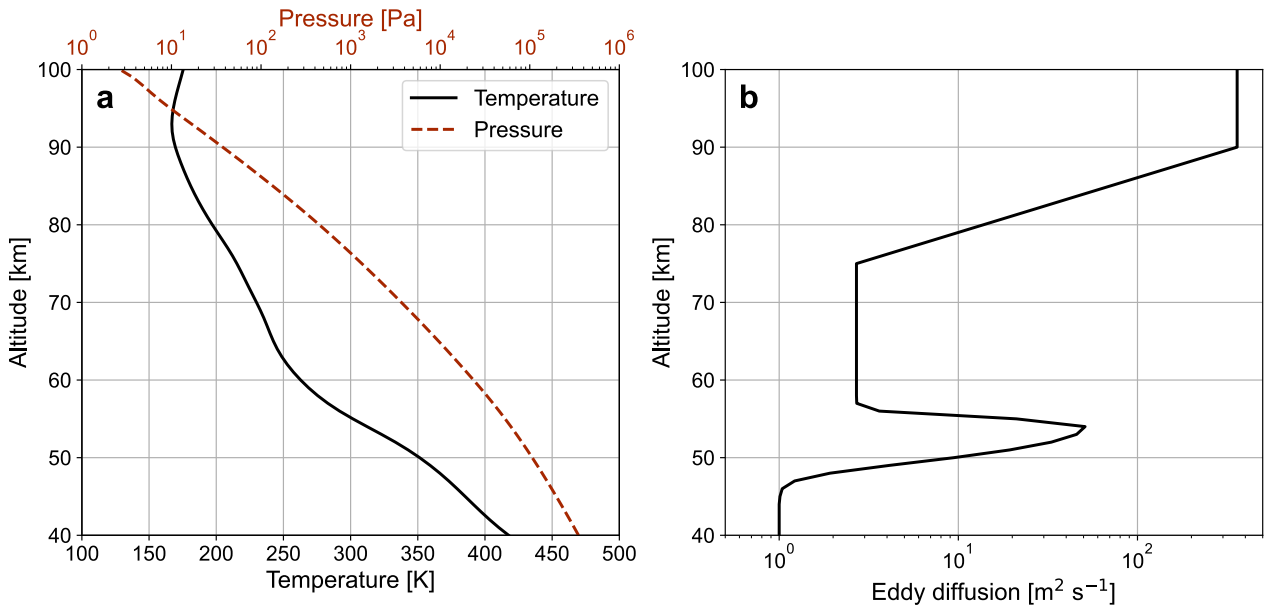


Figure 4.2: (a) Vertical profiles of temperature (black solid line) and pressure (red dashed line) taken from VIRA equatorial profile Seiff et al. (1985). (b) Vertical profile of the eddy diffusion coefficient.

Although previous models included the material transport by the ascending branch of the Hadley circulation as upward vertical wind (Imamura and Hashimoto, 2001), we excluded it for practical reasons. To accurately simulate the VMR of vapor species, the vertical wind profile must satisfy mass conservation (i.e., constant mass flux across the entire altitude range). Consequently, vertical wind speed scales inversely with background air density. While this assumption caused negligible deviations in cloud distributions focused on 40–70 km, our model extends to 100 km, where strictly enforcing mass conservation yields unrealistically high vertical wind speeds. For instance, applying a vertical mass flux of $8 \times 10^{-4} \text{ kg m}^{-2} \text{ s}^{-1}$ —consistent with Imamura and Hashimoto (2001)—results in vertical winds of $\sim 10 \text{ m s}^{-1}$ above 90 km. This is two orders of magnitude higher than Venus GCM predictions (Navarro et al., 2021). Moreover, because this speed exceeds the sedimentation velocity of micron-sized particles, it would artificially sustain large particles in the upper atmosphere. Therefore, we omitted vertical wind effects in this simulation.

4.1.6 Boundary and calculation conditions

The lower boundary condition for gas species is a fixed VMR at 40 km. The VMR of H_2SO_4 in the sub-cloud region has been reported to be approximately 4 ppm (Imamura et al., 2017; Kolodner and Steffes, 1998; Oschlisniok et al., 2021), while that of H_2O is reported to be ~ 30 ppm at altitudes of 30-40 km (Arney et al., 2014; Bézard et al., 2011; Chamberlain et al., 2013; Fedorova et al., 2015; Marcq et al., 2008). Based on these observations, the lower boundary VMRs for H_2SO_4 and H_2O are set to 4 ppm and 30 ppm, respectively. To date, there have been no direct measurements of sulfur allotropes in the cloud layer. However, Krasnopolsky (2007) predicts that ~ 30 ppm of sulfur allotropes are present at 40 km as a result of lower atmospheric thermochemistry. Assuming that these allotropes exist mainly as S_8 (Lyons, 2008), the lower boundary VMR for S_8 is set to 3.75 ppm, which is stoichiometrically 1/8 of the total sulfur atom mixing ratio. For particles, the lower boundary condition is a zero mixing ratio gradient. The upper boundary is a zero-flux condition for all gas species, the droplet particles, and the mixed particles. As noted earlier, the CN particle distribution has the constant input of MgFeSiO_4 as a 1-nm particle at the upper boundary. The simulation is run for 1.5×10^{10} s (1500 Venus days), which is long enough for the system to reach the steady state.

4.2 Results

The importance of MSP influx in the formation of the lower haze layer is clearly demonstrated. In fact, the lower haze forms only when MSP influx is included in the simulation (Figure 4.3a). Conversely, a sulfur haze does not form beneath the cloud base in either scenario because the sulfur particles evaporate. MSPs injected at the top of the atmosphere are scavenged by sulfuric acid aerosols at high altitudes and descend through the main cloud deck. Upon reaching the cloud bottom, the sulfuric acid evaporates, leaving behind involatile residues originally carried by the MSPs. In contrast, when the MSP influx is not included, the lower haze does not form (Figure 4.3b). Furthermore, our self-consistent modeling of particle formation reveals that sulfur allotrope particles are not stable in the solid phase below the cloud base. This finding raises questions about previous modeling approaches (Imamura and Hashimoto, 2001; Gao et al., 2014; McGouldrick and Barth, 2023; Karyu et al., 2024) in which CN are predefined at the bottom boundary and assumed to be microphysically equivalent to solid sulfur allotrope particles.

The resulting aerosol size distribution at 40 km closely resembles a lognormal distribution (Figure 4.4) and is, in all cases, qualitatively consistent with that inferred from measurements by the PV LCPS (Knollenberg and Hunten, 1980). Although the lower haze number density is an estimated value, it serves as a reliable reference because cloud microphysics models using this number density as a bottom boundary condition have consistently reproduced observed cloud properties (Imamura and Hashimoto, 2001; McGouldrick and Toon, 2007; McGouldrick and Barth, 2023; Karyu et al., 2024). The total haze number density agrees with the LCPS observations within a factor of two in all MSP influx scenarios, with the mode radius increasing with influx.

The formation of the lower haze is governed by a multi-stage process that begins in the upper atmosphere. First, incoming 1-nm MSPs are primarily incorporated into pre-existing sulfuric acid

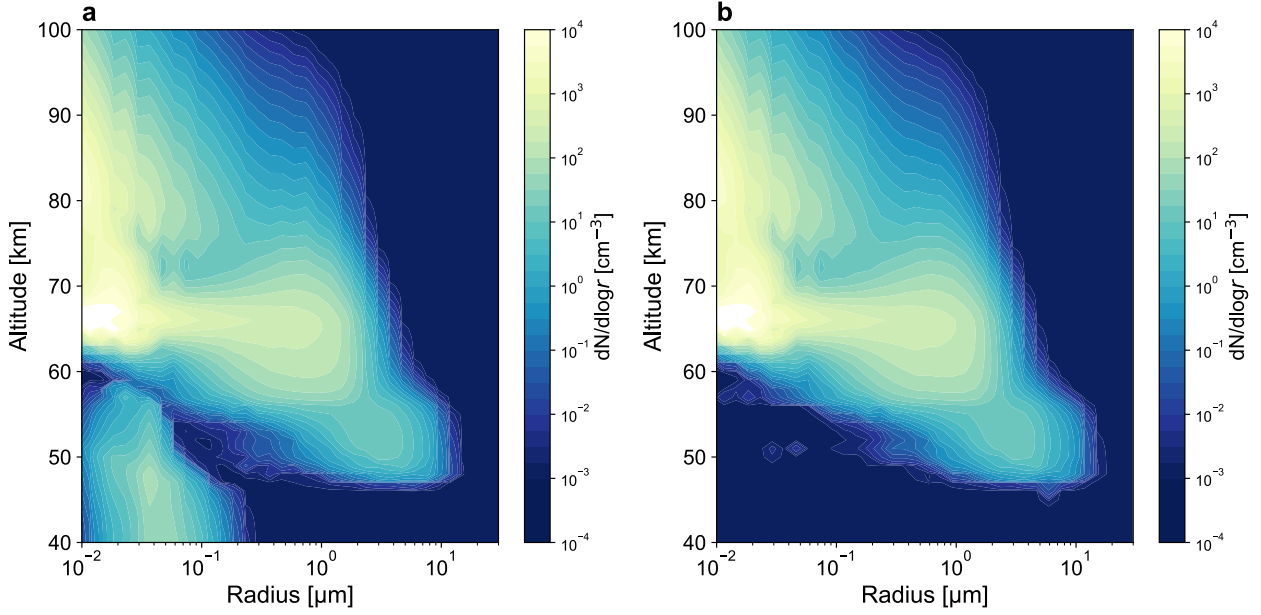


Figure 4.3: Steady-state size distributions of aerosols as a function of particle radius (μm) and altitude (km) obtained from the microphysics simulations with (a) MSP influx of 10 tons day^{-1} and (b) without MSP influx.

aerosols in the upper haze layer. An increase in the MSP influx does not create more particles but rather increases the mass of involatile material within each mixed haze particle. As these mixed particles sediment and cross the cloud base, sulfuric acid evaporates due to high temperature. This process releases the involatile MSP material, and the residual cores are now larger than the initial 1-nm MSPs. Higher initial MSP influxes lead to larger residual cores being released below the clouds. Finally, this population of enlarged, newly released particles accumulate and coagulate, forming the lower haze. The ultimate particle size is determined by a steady state balancing between the mass influx of these residual cores from above and their removal by sedimentation. Consequently, a greater initial MSP influx results in larger cores being released, which in turn coagulate into a lower haze with a larger mode radius.

The physical processes acting on involatile particles released by droplet evaporation at the cloud base are transport and coagulation. Therefore, the particle size distribution is considered to be influenced by these two processes. A critical factor in this context is the transport timescale. Particles continue to undergo coagulation growth until they are transported from the cloud base to the vicinity of 40 km. Consequently, it is hypothesized that the particle number density is determined by the efficiency of coagulation that occurs within the transport timescale.

To evaluate this, we must first derive the particle transport timescale. Below the cloud base, sedimentation speeds for submicron particles range from 10^{-6} to 10^{-5} m s^{-1} . Assuming a vertical distance H of 5 km (defined by the interval between the cloud base and 40 km altitude), the sedimentation timescale τ_{sed} is calculated to be 5×10^8 – $5 \times 10^9 \text{ s}$. This is significantly longer than the eddy

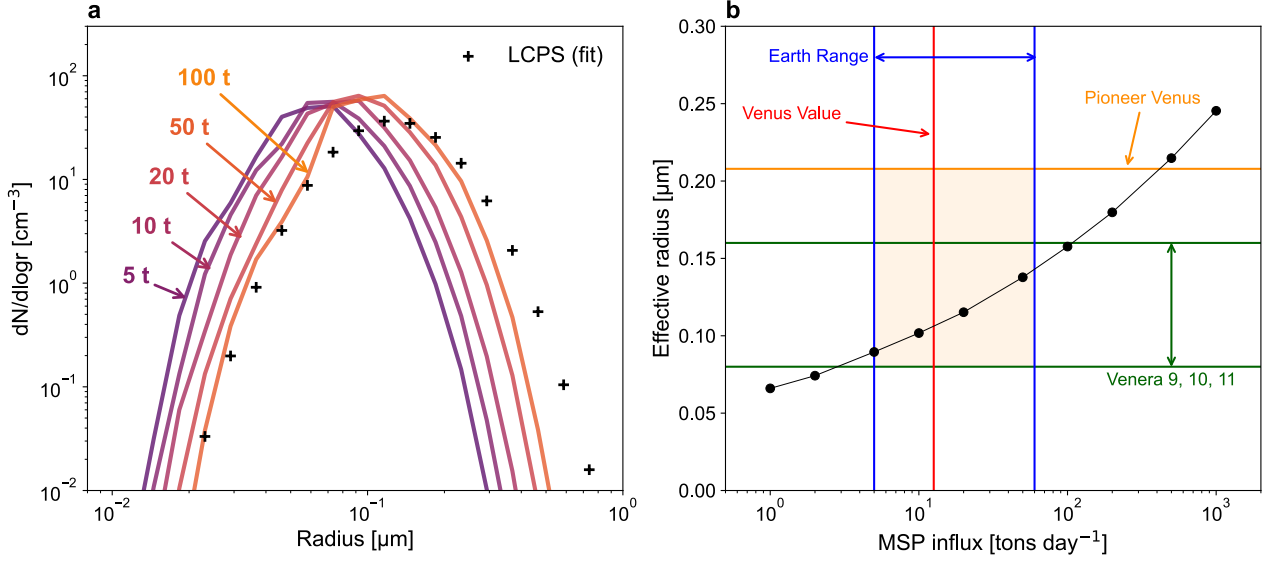


Figure 4.4: (a) Steady-state size distribution of the lower haze particles at the altitude of 40 km obtained from the microphysics simulations with MSP influxes ranging from 5 to 100 tons day⁻¹. The corresponding fluxes are indicated by the colored arrows. The black cross indicates the size distribution estimated from the LCPS measurements (Knollenberg and Hunten, 1980). (b) Effective radius of the lower haze particles at 40 km calculated as a function of MSP influx. The red and blue lines indicate the estimated MSP influx on Venus (Carrillo-Sánchez et al., 2020) and the uncertainty range of MSP influx on Earth (Marsh et al., 2013; Gardner et al., 2014; Plane et al., 2015; Carrillo-Sánchez et al., 2020), respectively. The orange and green lines indicate the effective radius measured by the LCPS (Knollenberg and Hunten, 1980) and the range of effective radius measured by Venera 9, 10, and 11 probes (Marov et al., 1980), respectively. The yellow-shaded area indicates the parameter range that satisfies both the observed effective radii and the uncertainty range of MSP influx.

transport timescale, τ_{eddy} , which is approximately 2.5×10^7 s. Consequently, the effective transport timescale is determined primarily by eddy diffusion.

The coagulation timescale τ_{coag} is estimated by the following relationship (Hashimoto and Abe, 2001):

$$N_p K_{\text{coag}} = \frac{1}{\tau_{\text{coag}}}, \quad (4.11)$$

where N_p is the particle number density (cm⁻³), K_{coag} is the coagulation kernel (cm³ s⁻¹). In this case, timescales τ_{eddy} and τ_{coag} should match because coagulation process takes place during the period of the transport timescale before the particles reach 40 km. Accordingly, by using τ_{eddy} and Equation (4.11), the relationship between number density of particles and the transport timescale can be expressed as follows:

$$N_p = \frac{1}{K_{\text{coag}} \tau_{\text{eddy}}}. \quad (4.12)$$

Adopting a coagulation kernel of $1 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ for $\sim 0.1 \text{ }\mu\text{m}$ particles at an altitude of 40 km (Appendix A) yields an estimated N_p of $\sim 40 \text{ cm}^{-3}$. This result is in good agreement with the simulated values shown in Figure 4.4. In addition, given that K_{coag} decreases with increasing radius towards $\sim 0.1 \text{ }\mu\text{m}$, this formulation explains the slight increase of the peak number density with increasing radius illustrated in Figure 4.4.

The above relationship also implies that transport and coagulation processes govern particle number density, independent of the source particle size. In fact, particle number densities remain consistent across different MSP influx scenarios. Consequently, a higher MSP influx results in an increased particle radius rather than a higher number density. This trend is further illustrated in Figure 4.4.

We converted the modeled size distribution into the effective radius, defined as an area-weighted mean radius (Pruppacher and Klett, 2010), to enable direct comparison with past probe measurements (Knollenberg and Hunten, 1980; Marov et al., 1980) (Figure 4.4b). Observations of the lower haze particles report a range of effective radii. Measurements from the Venera 9, 10, and 11 probes suggest a range of 0.08 to 0.16 μm (Marov et al., 1980), while the Pioneer Venus LCPS instrument reported a larger value of 0.21 μm (Knollenberg and Hunten, 1980). This discrepancy may arise because the PV LCPS value was not measured directly but was instead estimated from a limited number of data points near the instrument's detection limit. Given the scarcity and uncertainties in these observations, we adopt a working range for the lower haze effective radius of 0.08 to 0.21 μm . The corresponding MSP influx required to satisfy this condition ranges from approximately 3 to 300 tons day^{-1} . Since the MSP influx on Venus remains highly uncertain, this influx range can be further constrained by referencing Earth's values as a strong benchmark for a realistic rate. The lower limit of ablation flux, estimated from Earth's sodium layer measurements, is around 5 tons day^{-1} (Marsh et al., 2013; Plane et al., 2015). while the upper limit flux before ablation is approximately 60 tons day^{-1} (Carrillo-Sánchez et al., 2020; Gardner et al., 2014). This range also encompasses the typically estimated Venus' value of about 12 tons day^{-1} (Carrillo-Sánchez et al., 2020). The significant overlap suggests that the observed characteristics of Venus' lower haze can be explained by a realistic MSP influx common to terrestrial planets.

The MSP influx significantly impacts cloud formation in the main cloud deck (Figure 4.5). The lower haze particles, formed from MSPs, are transported upward by eddy diffusion, where they act as efficient CN for $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ droplets. This seeding process substantially increases the total cloud mass loading, with a higher influx resulting in a greater cloud mass (Figure 4.5b). The particle size is an important factor. A higher influx produces larger haze particles which, due to the Kelvin effect (Pruppacher and Klett, 2010), require less supersaturation to become cloud droplets. For instance, compared to the no-MSP case, a 5 tons day^{-1} MSP influx increases mass loading by 18%, while 100 tons day^{-1} results in a 28% increase. This enhancement brings simulations into closer agreement with the LCPS observations (Knollenberg and Hunten, 1980) (Figure 4.5b), highlighting the substantial impact of MSP-enhanced cloud formation.

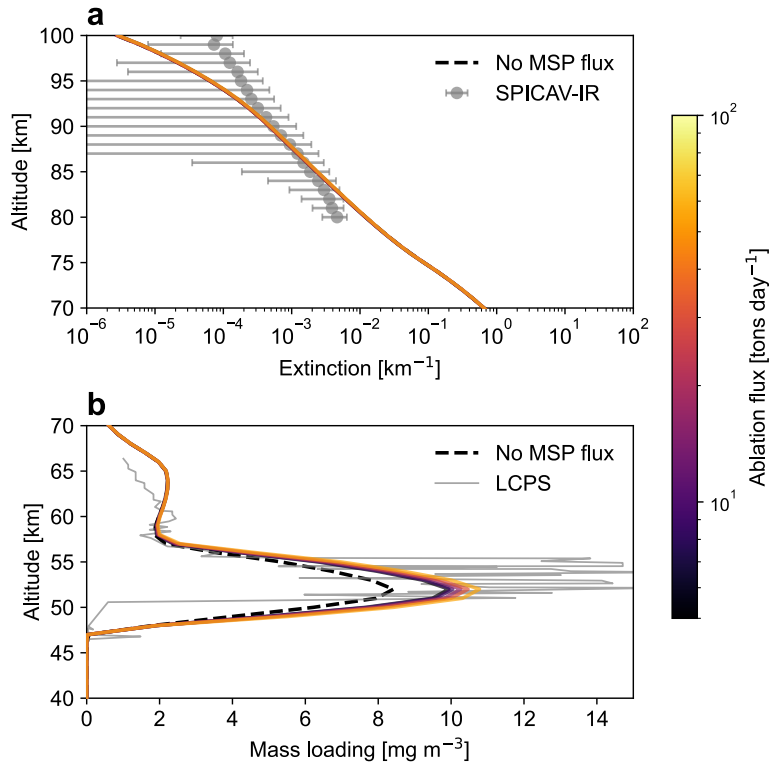


Figure 4.5: (a) Steady-state aerosol extinction profiles at $1\ \mu\text{m}$ wavelength obtained from the microphysics simulations with MSP fluxes ranging from 5 to $100\ \text{tons day}^{-1}$. All the curves are nearly indistinguishable, as their values are approximately the same. The grey circles and error bars indicate observed aerosol extinction at $\sim 1\ \mu\text{m}$ derived from the Spectroscopy for Investigation of Characteristics of the Atmosphere of Venus (SPICAV) data onboard VEx (Luginin et al., 2024) (b) Steady-state aerosol mass loading profiles obtained from the microphysics simulations with MSP influxes ranging from 5 to $100\ \text{tons day}^{-1}$. The grey line indicates the mass loading measured by PV LCPS (Knollenberg and Hunten, 1980). Note that an off-scale mass loading data point from the LCPS data, approximately $90\ \text{mg m}^{-3}$ and peaking around 49 km, is not displayed in the figure for visibility.

In contrast, MSP influx has a negligible effect on the upper haze formation (Figure 4.5a). Cloud formation at these altitudes can be affected by two-component homogeneous nucleation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ and homogeneous nucleation of sulfur allotrope, rather than by heterogeneous nucleation on MSPs. This small contribution of MSPs does not significantly alter the particle size distribution in the upper haze, consistent with a prior modeling study (Gao et al., 2014). Overall, the influence of MSP influx on cloud structure is somewhat counterintuitive: it primarily affects the main cloud deck—far from its initial source—rather than the upper haze at higher altitudes, which is closer to the source.

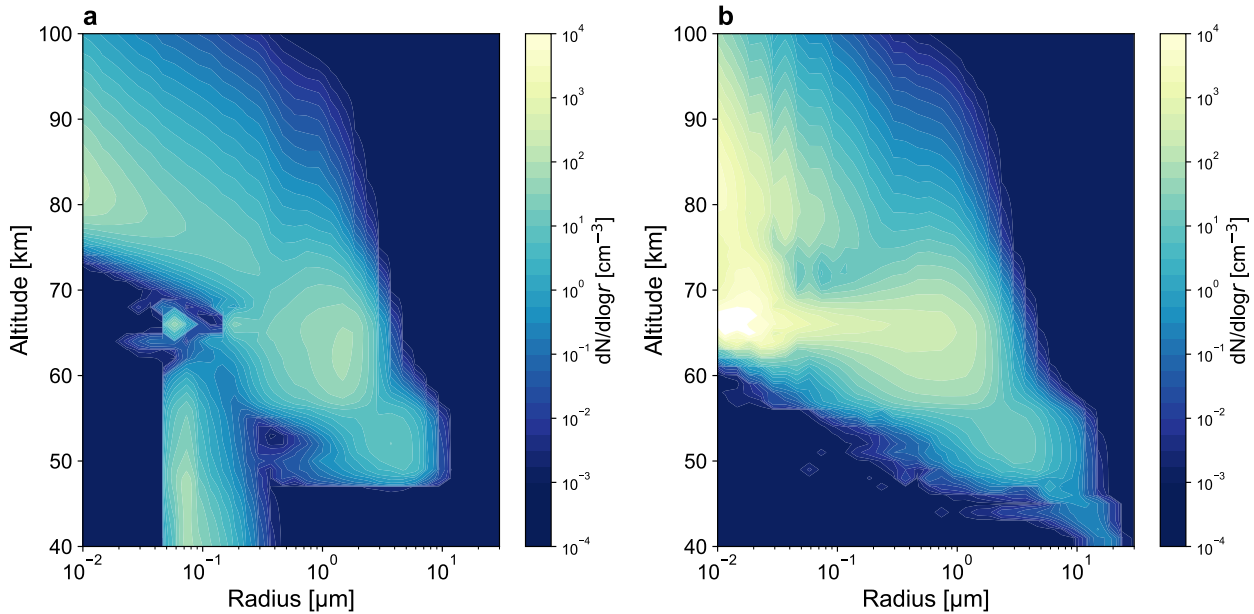


Figure 4.6: Steady-state 2D size distributions of aerosols as a function of particle radius (μm) and altitude (km) obtained from the microphysics simulations (a) with MSP influx of 10 tons day^{-1} but without S_8 vapor, and (b) with S_8 vapor abundance fixed at saturation VMR at the bottom boundary but without MSP influx.

4.3 Discussion

4.3.1 Sensitivity studies

We conducted three sensitivity studies to check the robustness of the conclusions of this study—the lower haze particle formation driven by MSP influx. First, we performed a simulation study that included MSP influx while excluding gaseous and solid S_8 . The purpose of this parameter study is to see whether the lower haze formation occurs without the influence of S_8 particles. The assumed MSP influx is the nominal value (10 tons day^{-1}), and only H_2SO_4 and H_2O are included as vapor species. Consequently, the particle formation is mainly driven by the homogeneous nucleation of H_2SO_4 - H_2O liquid in the upper haze region. The results confirm that the lower haze formation occurs without the influence of S_8 (Figure 4.6a), reinforcing the robustness of the lower haze formation by the MSP influx.

The VMR of S_8 vapor in the lower atmosphere is not observationally constrained. The lower haze particles can still be explained by S_8 particles if they can exist below the cloud deck. Such a condition can be satisfied if S_8 vapor is saturated. Thus, we conducted a parameter study with saturated S_8 VMR at the bottom boundary and without including the MSP influx. Our simulation confirmed that the S_8 particles grow to a radius larger than $10 \mu\text{m}$ if S_8 vapor is saturated at the bottom boundary (Figure 4.6b). The simulated size is too large to account for the observed lower haze particle size distribution. This indicates that the abundant S_8 vapor leads to increased condensation flux onto pre-

existing particles. The enhanced condensation flux makes S_8 particles under the cloud bottom keep growing, instead of remaining in the sub-micron haze size distribution.

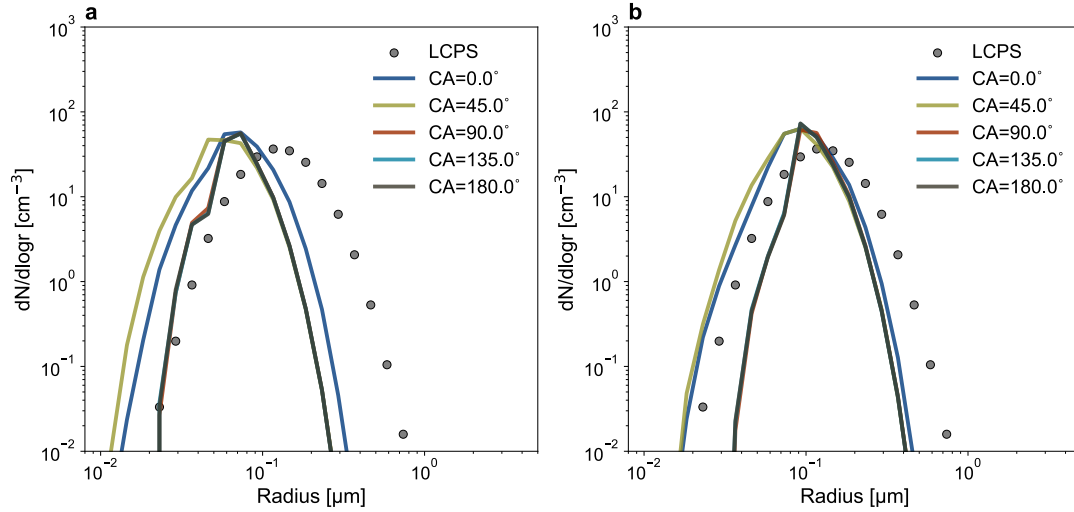


Figure 4.7: a, Steady-state size distributions of aerosols at 40 km as a function of particle radius (μm) obtained from microphysics simulations using different contact angle (CA) ranging from 0° to 180° with an MSP flux of 10 tons day^{-1} . b, Steady-state size distributions of aerosols at 40 km as a function of particle radius (μm) obtained from microphysics simulations using different contact angles (CA) ranging from 0° to 180° with an MSP flux of 50 tons day^{-1} . The LCPS results are also plotted for comparison in both panels.

Although the contact angle between solid sulfur and liquid H_2SO_4 has not been experimentally measured, qualitative evidence suggests that sulfur is not perfectly wettable (Young, 1983). To test the robustness of our findings against this uncertainty, we performed a series of parameter studies using a wide range of contact angles (45° , 90° , 135° , and 180°). We found that the peak of the particle size distribution is almost completely insensitive to the chosen contact angle (Figure 4.7). While the total particle population remained similar across the simulations, the breadth of the size distribution showed slight variations at the smaller end. This indicates that the lower haze still forms with a consistent size distribution, regardless of the assumed wettability of the solid sulfur particles.

4.3.2 Exploration of lower haze sources from the surface

In addition to the cosmic dust, particles from volcanic eruptions or the surface represent another potential source for the involatile material observed in Venus's clouds. To investigate this possibility, we modeled the potential for this dust to form the lower haze using a 1-D transport model based on the SPECK framework.

Our 1-D model simulates the vertical transport and coagulation of dust particles within an altitude range of 0 to 45 km, using a 1 km grid interval. Vertical transport is governed by eddy diffusion and sedimentation. To establish an upper limit for transport efficiency, the eddy diffusion coefficient was set to $1 \text{ m}^2 \text{ s}^{-1}$, an order of magnitude higher than standard values ($\sim 0.1 \text{ m}^2 \text{ s}^{-1}$) used in models of

Venus's lower atmosphere (Krasnopolsky, 2007; Bierson and Zhang, 2020; Rimmer et al., 2021). The coagulation kernel, which accounts for Brownian diffusion and gravitational collection, is identical to that implemented in the main SPECK.

To simulate a volcanic eruption or another elevated dust source, we inject dust particles at an altitude of 10 km. A point source is assumed because Venusian volcanoes are predominantly shield volcanoes, which are associated with mild, effusive eruptions similar to terrestrial hotspots (Head et al., 1992; Ivanov and Head, 2013; Ghail et al., 2024; Filiberto et al., 2025). Furthermore, Venus's dense atmosphere is expected to suppress explosive volcanic activity (Garvin et al., 1982; Thornhill, 1993). The injected particles are assumed to be composed of olivine with a set size of 10 nm. This value is substantially smaller than the typical 1-40 μm range for volcanic ash (Brown et al., 2012); however, this assumption was made to decrease sedimentation velocity, thereby increasing the atmospheric lifetime and transport potential of the particles. Note that volcanic dust typically coagulates, makes large aggregate of a few mm, and fall out of the atmosphere before traveling no longer than 20-30 km of the vicinity of the volcano on Earth (Brown et al., 2012). Thus, we take a very favorable condition for dust transport to optimistically explore the possibility of dust reaching the cloud base altitude. We set the dust injection flux to wide range of parameters from 100 to 1×10^5 tons day⁻¹, which encompasses the typical volcanic ash production rate on Earth of 5.5×10^4 tons day⁻¹. The boundary condition is set to zero flux for the upper boundary and free for the bottom boundary. Simulation is run for 1×10^8 s (~ 10 Venus days), which is sufficient for the model to reach the steady state.

The simulated particle size distributions are summarized in Figure 4.8. Near the source (10 km altitude), volcanic ash initially grows via coagulation driven by high particle number densities. Figure 4.9a demonstrates that as particles transport through the atmospheric scale height (10 km above the source), their number density drops to $\sim 10 \text{ cm}^{-3}$ at 20 km across all flux cases. This mechanism mirrors the process introduced as Equation (4.12). Because number density is regulated by the eddy diffusion timescale and coagulation efficiency, the peak radius at 20 km increases with higher dust injection fluxes.

Simulation results indicate that although dust particles are transported to the cloud base (Figure 4.8), their number density at 40 km remains significantly lower than Pioneer Venus LCPS estimates (Figure 4.9b). Once the number density stabilizes at 20 km (within one scale height), eddy diffusion effectively mixes the particles with the background atmosphere above this altitude. This trend is evident in Figure 4.10, where the particle mixing ratio converges within the 10 km scale height and remains nearly constant thereafter. A constant mixing ratio implies that particle number density decreases exponentially alongside atmospheric density. Consequently, the number density at 40 km is approximately an order of magnitude lower than at 20 km (Figure 4.9), consistent with the pressure difference between these altitudes (3 bar vs. 20 bar).

Although the sedimentation timescale is significantly longer than that of eddy diffusion in the Venus lower atmosphere, sedimentation still induces a slight deviation from the constant mixing ratio, as seen in Figure 4.10. As noted earlier, higher dust injection fluxes yield particles with larger mode

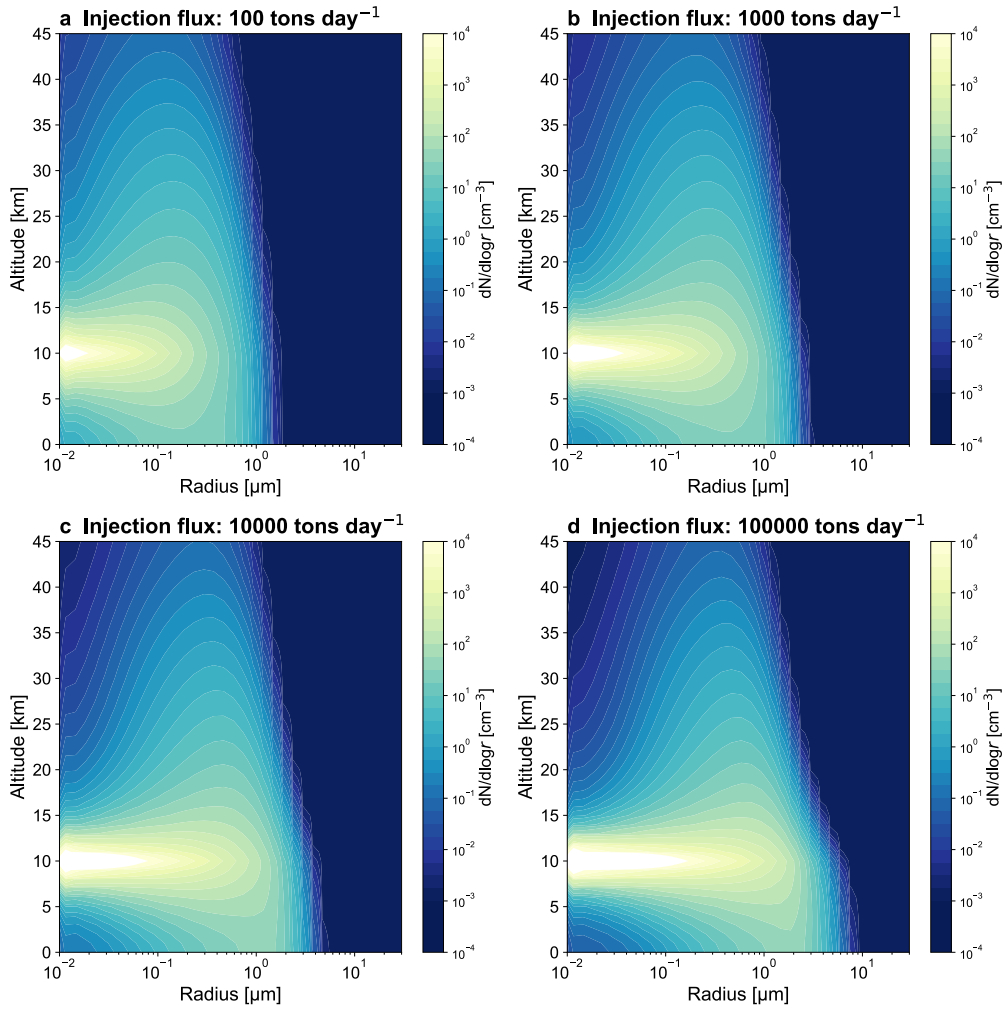


Figure 4.8: Steady-state size distributions of aerosols as a function of particle radius (μm) and altitude (km) obtained from the microphysics simulations with dust injection flux of (a) 100 tons day^{-1} , (b) 1000 tons day^{-1} , (c) 1×10^4 tons day^{-1} , and (d) 1×10^5 tons day^{-1} .

radii. These larger particles are more susceptible to sedimentation, leading to a more pronounced deviation from the constant mixing ratio profile above 20 km.

4.3.3 Exploration of lower haze sources from photochemistry

This section explores photochemical products other than H_2SO_4 and S_8 as a potential source for the lower haze particles. Although various sulfur allotropes exist, our model assumes that gaseous sulfur consists solely of S_8 . However, since each allotrope has a unique saturation vapor pressure (Lyons, 2008), it is possible that allotropes other than S_8 could condense below the cloud base. To investigate this possibility, we compared the VMR of various sulfur allotropes predicted by a chemical kinetic model with their respective saturation vapor pressures (Lyons, 2008) (Figure 4.11). Our analysis focused on the most abundant allotropes at 40 km: S_8 , S_7 , and S_6 , which comprise 80%, 8.5%, and

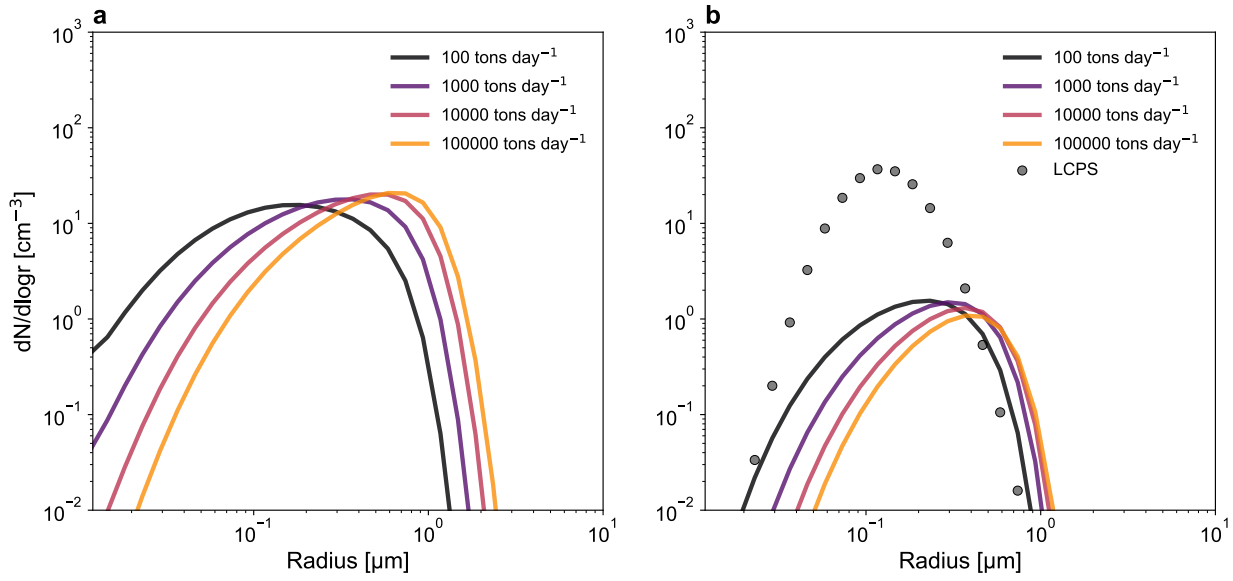


Figure 4.9: Steady-state size distribution of the lower haze particles at the altitude of (a) 20 km and (b) 40 km obtained from the microphysics simulations with dust injection fluxes ranging from 100 to 1×10^5 tons day $^{-1}$.

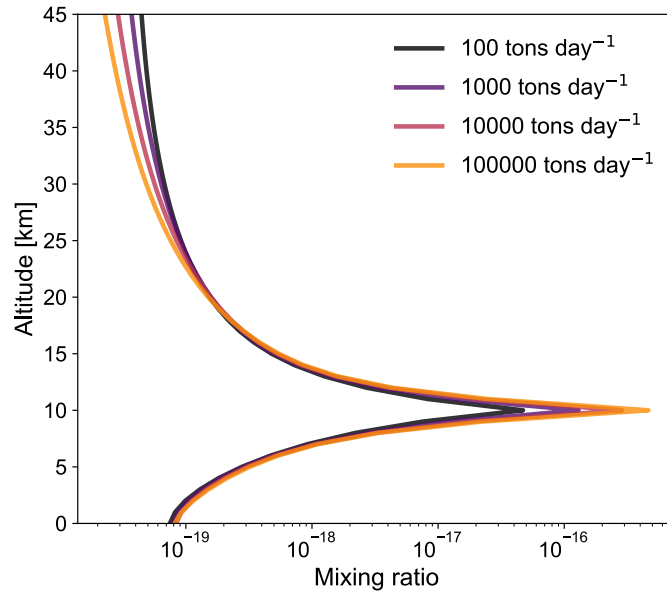


Figure 4.10: Steady-state vertical profiles of particle mixing ratio obtained from the microphysics simulations with dust injection fluxes ranging from 100 to 1×10^5 tons day $^{-1}$.

10.5% of the total sulfur abundance at thermodynamic equilibrium, respectively, for a combined total of 99%. The remaining 1% was attributed to S_5 to estimate its abundance. The results show that none of these selected sulfur allotropes can condense at 40 km, as their VMRs are below the required saturation levels. Furthermore, the abundances of smaller allotropes (S_2 – S_4) are predicted to be less

than 10^{-7} ppm at 40 km and are therefore considered negligible (Krasnopolsky et al., 2013). In conclusion, based on current kinetic chemistry models of the lower atmosphere, the condensation of sulfur allotropes is an unlikely formation mechanism for the lower haze.

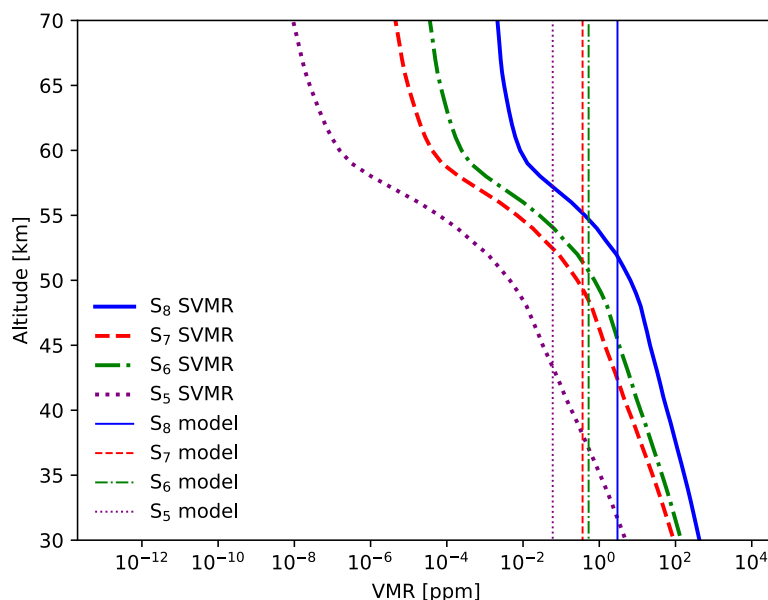


Figure 4.11: Comparison of saturation volume mixing ratios (SVMRs) and predicted atmospheric abundances for major sulfur allotropes. The thick lines show the SVMRs for S_8 (blue solid), S_7 (red dashed), S_6 (green dash-dotted), and S_5 (purple dotted), derived from Lyons (2008). The thin vertical lines indicate the predicted lower-atmospheric abundances for each respective allotrope. As the predicted abundances are significantly lower than the SVMRs, the data suggest that condensation of these sulfur allotropes is not feasible in the lower atmosphere.

Another potential condensate originating from photochemistry is carbon disulfide (CS_2), for which an upper limit of ~ 10 ppb has been reported near the cloud top (Mahieux et al., 2023a). As shown in Figure 4.12, this abundance is lower than the saturation VMR predicted by the Antoine equation (Waddington et al., 1962). Because CS_2 is a photochemical product and its concentration is expected to decrease with decreasing altitude, its condensation below the cloud deck is also considered highly unlikely.

4.3.4 Implication for the unknown UV absorber

Our results suggest that the lower haze particles can provide a source for the mysterious UV-blue absorber on Venus (Pollack et al., 1980). The MSPs introduce various involatile species into Venus' atmosphere, including iron (Fe), magnesium (Mg), and silicon (Si). These species can then accumulate as lower haze particles (Table 4.2). This accumulation of iron provides an explanation for the iron detected by both the VEGA and Venera probes (Krasnopolsky, 1989). Furthermore, recent re-analysis of the mass spectrometer onboard the PV Large Probe indicates that iron sulfate is present in the cloud droplets, likely explained by the inclusion of cosmic material (Mogul et al., 2025). The

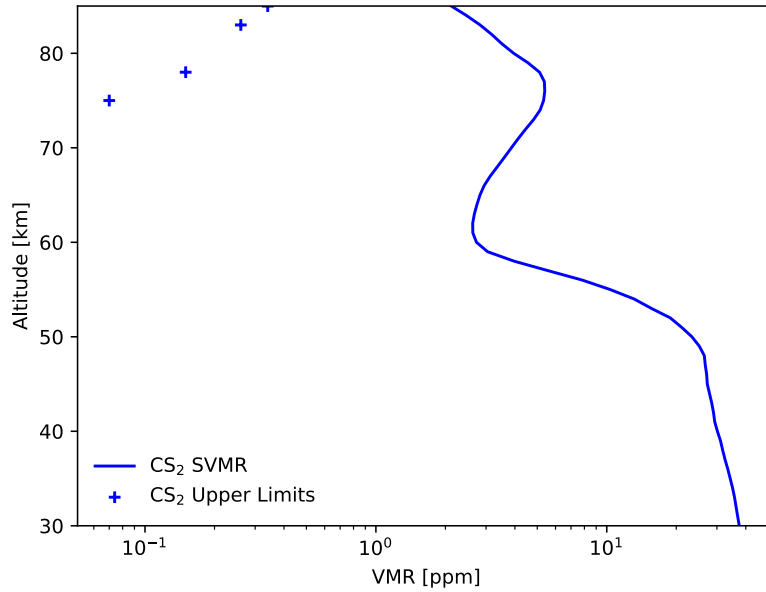


Figure 4.12: Saturation volume mixing ratio of CS₂ (blue solid), calculated with the Antoine equation. The blue crosses indicate the upper limit value of CS₂ VMR (Mahieux et al., 2023a)

present study aligns with this observation. Iron compounds have long been proposed as candidates for the unknown UV-blue absorber on Venus (Zasova et al., 1981). Specifically, proposed mechanisms include iron chloride forming from reactions with hydrogen chloride (Krasnopolsky, 1985, 2017; Egan et al., 2025), or iron sulfates forming in H₂SO₄-H₂O droplets, which has been proposed by a recent study (Jiang et al., 2024). If the haze particles existing between 40 and 57 km, which is prevented from condensation due to their high heterogeneous nucleation barrier (Figure 4.3a), are transported by cloud-penetrative convection (Baker et al., 1999; Lefèvre et al., 2022) and incorporated into H₂SO₄ aerosols, the UV-blue absorption feature can form at the cloud top. This scenario aligns with the previous observations, suggesting that the UV-blue absorber is hidden beneath the upper cloud and transported convectively from the layer below (Titov et al., 2008, 2012). Thus, our work provides the crucial missing link between observational and experimental studies of the UV absorber by identifying a viable physical source of the iron.

Table 4.2: The mass density of involatile species deposited as the lower haze particles at 40 km.

MSP influx	Olivine (mg m ⁻³)	Fe (mg m ⁻³)	Mg (mg m ⁻³)	Si (mg m ⁻³)
5 tons day ⁻¹	1.1×10^{-4}	3.0×10^{-5}	1.3×10^{-5}	1.5×10^{-5}
10 tons day ⁻¹	1.5×10^{-4}	4.6×10^{-5}	2.0×10^{-5}	2.3×10^{-5}
20 tons day ⁻¹	2.6×10^{-4}	7.2×10^{-5}	3.1×10^{-5}	3.6×10^{-5}
50 tons day ⁻¹	4.6×10^{-4}	1.3×10^{-4}	5.5×10^{-5}	6.4×10^{-5}
100 tons day ⁻¹	7.2×10^{-4}	2.0×10^{-4}	8.5×10^{-5}	9.8×10^{-5}

Our findings show that MSPs are critical to Venus' climate, operating through two primary mechanisms. First, MSPs strengthen the planet's greenhouse effect by enhancing the main cloud deck and trapping outgoing thermal radiation (Limaye et al., 2018; Haus et al., 2016). Given the atmosphere's low stability, even a slight change in the heating rate can readily destabilize the cloud layer (Limaye et al., 2018). The subsequent heating at the cloud base drives strong convection, which further contributes to cloud formation, consequently creating a positive feedback loop (McGouldrick and Toon, 2007). Second, the MSPs provide iron compounds, which can serve as a UV absorber in the Venus atmosphere (Pollack et al., 1980). In fact, half of the incident solar energy is deposited between 57 and 70 km and causes strong heating (Limaye et al., 2018), exciting thermal tides and maintaining the atmospheric super-rotation that effectively transports heat from the dayside to the nightside (Hornouchi et al., 2020). Collectively, these effects originating from MSPs potentially affect the radiative budget of Venus' atmosphere by influencing both outgoing thermal and incoming solar radiation, and thus, its climate.

The upcoming missions will offer the opportunity to investigate the characteristics of the lower haze layer in greater detail. In particular, the Venus Mass Spectrometer (VMS) on DAVINCI is designed to detect compounds across a broad mass range, from 2 to 550 Da (Garvin et al., 2022). The masses of gaseous species that would sublime from MSP-related compounds fall within this detectable range, which would provide direct evidence of their composition and abundance. More specifically, the potential for detecting iron as iron chloride is noteworthy, as its abundance in the gas phase is predicted to reach up to 17 ppb due to sublimation from the solid phase (Krasnopolsky, 2017). The detection of such species by VMS will be crucial for confirming the origin of the lower haze layer and assessing its potential as a UV-blue absorber.

4.3.5 Implication for other planets

Finally, we discuss the implications of our findings for other planets. While the well-studied upper hazes on solar system gas giants are composed of organic photochemistry products, the microphysical processes below the main cloud decks are poorly understood, particularly regarding the origin and composition of CN (Yair et al., 1995). On these planets, involatile particles are likely not effectively scavenged from the atmosphere because, similar to Venus, cloud particles eventually evaporate completely below the cloud base (Atreya and Wong, 2005). Equilibrium cloud condensation models predict that water cloud droplets form near the cloud base on Jupiter, Saturn, and Neptune, where atmospheric temperatures exceed ~ 300 K (Atreya and Wong, 2005). At such high temperatures, water cannot efficiently nucleate on its own and thus requires pre-existing CN. Significant cosmic dust injection has been estimated on gas giants (Moses and Poppe, 2017; Poppe, 2016). Drawing from our Venus simulations, the cosmic dust ablation would lead to the formation of sub-micron particle layers below the cloud base. These sub-micron particles deposited under the cloud deck would be transported upward again by vigorous convection and contribute to water droplet formation (Sugiyama et al., 2014). This mechanism should be particularly important for Jupiter, which is estimated to receive an even greater flux of cosmic dust ablation than Venus (Poppe, 2016).

On planets like Earth, where surface liquid water exists, rain constantly scavenges involatile particles from the air (Pruppacher and Klett, 2010). However, as observed in our Solar System, planets with surface liquid water are truly unique; most planets possess dry atmospheres where liquid droplets evaporate before reaching the surface (Sánchez-Lavega et al., 2004). In such worlds, the lower haze originating from cosmic dust can accumulate in the hot lower atmosphere, enhancing cloud formation by acting as efficient CN. This would significantly impact the planet's climate, as clouds exhibit a strong radiative effect (Pierrehumbert, 2010). Furthermore, iron compounds within the cloud particles can alter their optical properties, including UV light absorption (Jiang et al., 2024). These effects establish cosmic dust as an essential component of planetary climates, a role that is also likely to be significant for exoplanets.

4.4 Chapter Summary

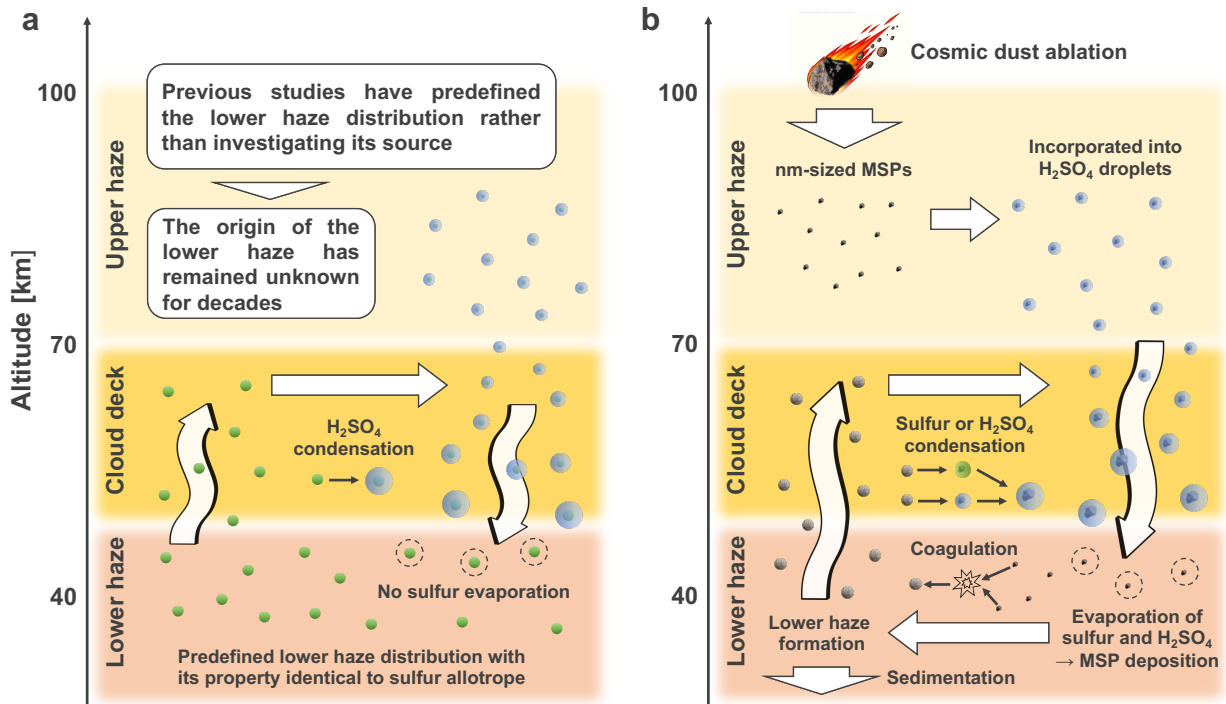


Figure 4.13: (a) Sketch of the cloud cycle on Venus simulated by previous microphysics models (Imamura and Hashimoto, 2001; McGouldrick and Toon, 2007; Gao et al., 2014; McGouldrick and Barth, 2023) with predefined lower haze distribution. (b) Sketch of the cloud cycle on Venus simulated by this study with the MSP injection and the subsequent lower haze formation.

We simulated lower haze formation by implementing sulfur allotrope microphysics and, most importantly, the MSP influx from cosmic dust ablation into SPECK. As shown in Figure 4.13a, previous models imposed the lower haze as a boundary condition, whereas our model aims to reproduce this layer without it.

Our results indicate that the lower haze can form from a constant MSP input. In contrast, the lower haze cannot form from sulfur allotropes, as they completely evaporate below the cloud base. The proposed formation mechanism is summarized in Figure 4.13b. First, MSPs are incorporated into H_2SO_4 droplets in the upper haze layer. These MSP-containing droplets sediment through the cloud layer and evaporate upon reaching the cloud base. Upon this evaporation, the involatile MSP-derived material is released, and these particles then coagulate to form the lower haze.

The simulated size distribution at 40 km is similar to a lognormal distribution, consistent with previous estimates based on PV LCPS observations. Assuming an MSP influx range consistent with those on terrestrial planets, the effective radius of the lower haze falls within the 0.08-0.21 μm range observed by previous probe measurements. Therefore, we conclude that cosmic dust ablation is a viable source for the lower haze. Our findings have important implications for the Venus climate system. First, by seeding the main cloud, the lower haze increases mass loading by 20-30% compared to a no-haze scenario. This enhances cloud deck heating and drives convection, further reinforcing cloud formation and creating a positive feedback loop. Second, the MSP-derived iron can serve as the unknown UV absorber, as recent experimental studies show that iron compounds formed in H_2SO_4 droplets closely match the observed UV absorption feature. Therefore, the MSP influx and resulting lower haze formation can affect radiative transfer by both thickening the clouds and providing UV-absorbing material.

Chapter 5

Origin of Venus' Clouds

This chapter is based on the following paper, to be submitted to the Planetary Science Journal:

Hiroki Karyu, Takeshi Kuroda, Anni Määttänen, Arnaud Mahieux, Sébastien Viscardy, Naoki Terada, Séverine Robert, Ann Carine Vandaele, and Michel Crucifix (2025). Origins of Venus' Clouds Investigated with a Cloud Microphysics Model. *The Planetary Science Journal*, to be submitted.

5.1 Model Description

5.1.1 Overview

To simulate the vertical profiles of cloud particles and condensable vapors on Venus, we use the 1D cloud microphysics model SPECK (Karyu et al., 2025), developed in Chapter 3. The microphysical processes in this model are identical to those in Chapter 4, which include sulfur allotrope microphysics, various nucleation processes, and olivine as an involatile particle component. We therefore detail only the deviations from the previous chapter below. The first major difference is the treatment of the lower haze. As shown in Chapter 4, only the cumulative effect of the MSP influx is important, and its contribution to upper haze formation is negligible. Therefore, the lower haze is not formed from MSP influx in this chapter but is instead imposed at the lower boundary. The second major difference is the latitudinal setting. This chapter focuses on the global cloud structure, whereas the previous chapter focused on the equator to allow comparisons with probe measurements predominantly taken in that region. Therefore, we use a mid-latitude condition (45°) to represent the global average structure, which is the same treatment as Chapter 2.

5.1.2 Atmospheric conditions

This study focuses on the globally averaged distributions of particles and gas species. The atmospheric pressure and temperature profiles are taken from the mid-latitude (45°) conditions of the VIRA (Seiff et al., 1985), as shown in Figure 5.1a. For vertical transport, we follow the approach of Karyu et al. (2024) and represent all transport processes using eddy diffusion. We constructed the eddy diffusion profile for the upper cloud and upper haze layers based on the findings of Karyu et al. (2024). They found that an eddy diffusion coefficient of $\sim 2 \text{ m}^2 \text{ s}^{-1}$ in the upper cloud is required to reproduce the observed H_2O VMR profiles. They also showed that a more recently estimated coefficient of $\sim 360 \text{ m}^2 \text{ s}^{-1}$ (Mahieux et al., 2021) accurately reproduces the observed haze extinction profiles. Following previous studies (James et al., 1997; McGouldrick and Barth, 2023; McGouldrick and Toon, 2007), we use a large eddy diffusion coefficient in the lower and middle cloud layers to account for convective activity. Below the cloud base, the coefficient is set to $1 \text{ m}^2 \text{ s}^{-1}$. The complete eddy diffusion profile is shown in Figure 5.1b and is consistent with recent 3-D large-eddy simulations (Lefèvre et al., 2022, 2024).

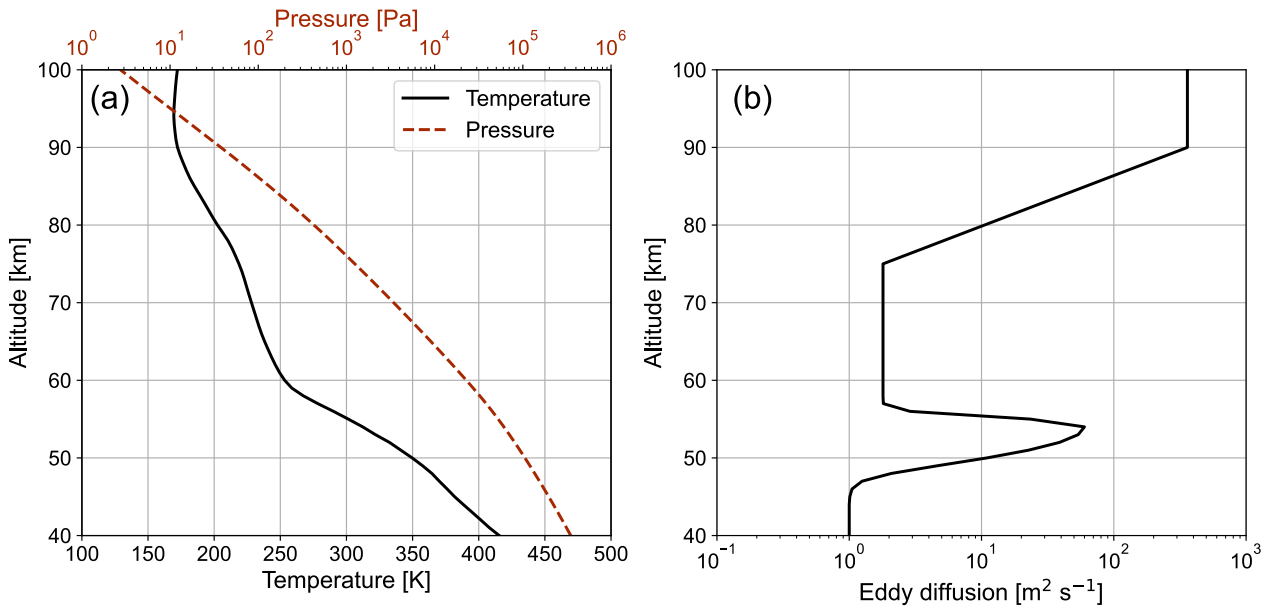


Figure 5.1: (a) Vertical profiles of temperature (black solid line) and pressure (red dashed line) taken from VIRA mid-latitude (45°) profile Seiff et al. (1985). (b) Vertical profile of the eddy diffusion coefficient.

5.1.3 Boundary conditions

The lower boundary condition for gas species is fixed at the same value as Chapter 4 at 40 km. For particles, the lower boundary condition is a zero mixing ratio gradient, except for those in the CN distribution. The CN size distribution at the lower boundary is fixed to a lognormal distribution with a total number density of 200 cm^{-3} , a mode radius of $0.125 \mu\text{m}$, and a standard deviation of 2.16, following McGouldrick and Barth (2023). In addition, at the bottom boundary, the lower haze

particles are assumed to consist solely of olivine. This assumption is based on the results of the previous Chapter 4. The upper boundary is a zero-flux condition for all species (both gases and particles).

5.1.4 Case studies

To investigate the contribution of each nucleus type to cloud formation, we designed several case studies, each incorporating a different set of microphysical processes or particle components. The nominal case includes all processes and components described above and serves as a benchmark for this study. We then performed a series of sensitivity simulations, systematically excluding key processes—such as $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ homogeneous nucleation, S_8 vapor formation, and the inclusion of lower haze particles—to isolate the effect of each nuclei source on the resulting cloud structure. These experimental settings are summarized in Table 5.1. For each case, we performed a simulation for 5×10^8 s (equivalent to 50 Venus years) and present the steady-state results in the next section.

Table 5.1: List of case studies

Simulation name	$\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ homo. nuc.	S_8 vapor	Lower haze
Nominal	○	○	○
No $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ homo. nuc.		○	○
No S_8 vapor	○		○
No lower haze	○	○	

5.2 Results

5.2.1 Nominal case

The nominal case reproduces the typical Venus cloud structure, which consists of different particle size modes at various altitudes (Figures 5.2a and 5.3). In the lower haze layer (40–47 km), the particle size distributions are mainly characterized by a mode radius of 0.1–0.2 μm , and particles larger than 1 μm are also present above 47 km. These distributions are broadly consistent with observations from the PV LCPS (Knollenberg and Hunten, 1980), although the trimodal structure is not apparent in our simulation. This is likely due to the absence of transient vertical wind in our simulation, since Imamura and Hashimoto (2001) showed that the vertical wind induces the transition from bimodal to trimodal structure in the lower and middle clouds. In the upper cloud layer (57–70 km), both micron-sized and sub-micron particles are dominant, as observed by the PV LCPS and the OCPP (Kawabata et al., 1980). Above 70 km, the upper haze layer consists of sub-micron particles, consistent with observations from SPICAV aboard VEx (Luginin et al., 2016, 2024; Wilquet et al., 2009). The overall simulated cloud structure in Figure 5.2a is also consistent with recent microphysics model results (Karyu et al., 2024; McGouldrick and Barth, 2023).

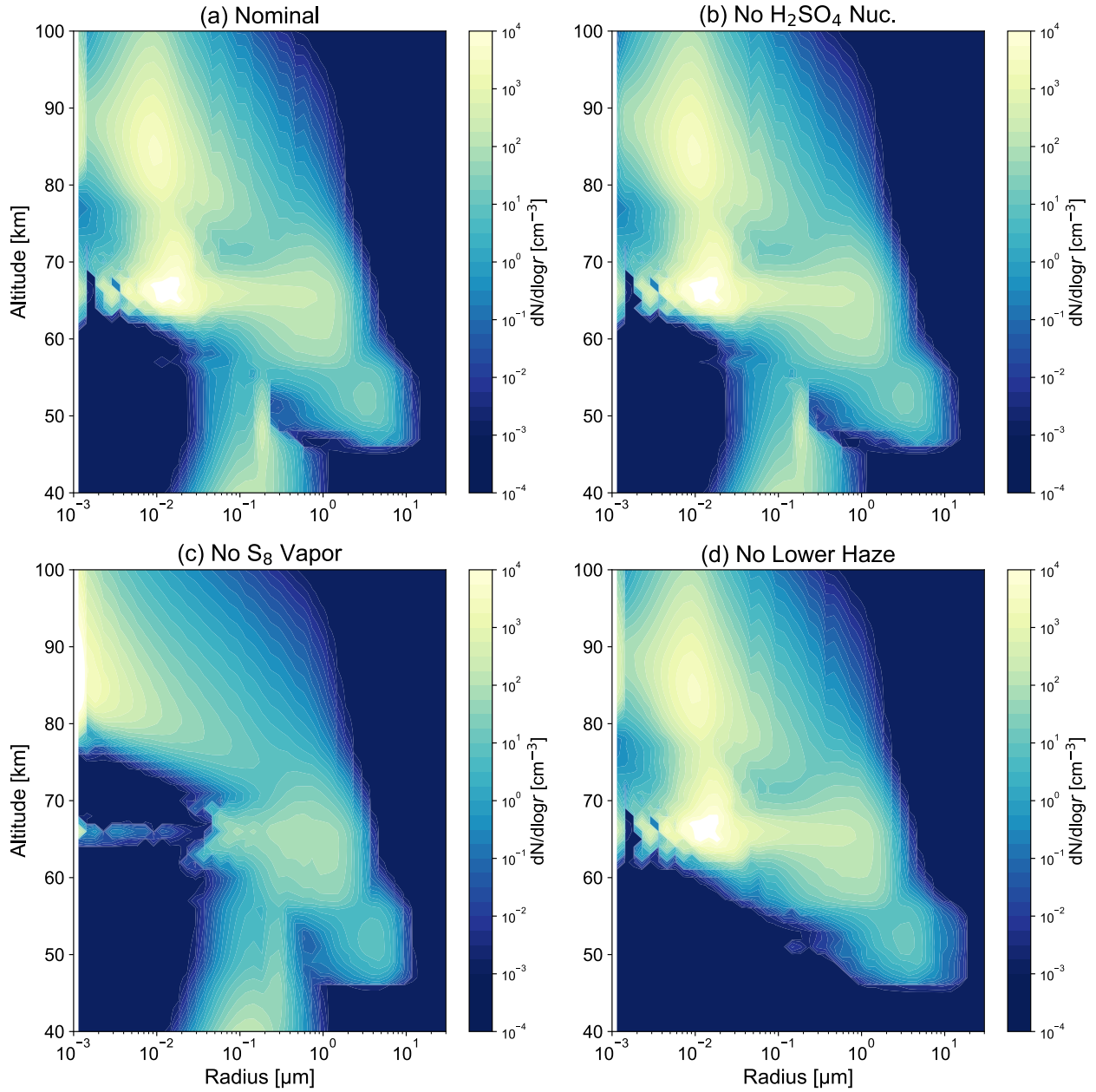


Figure 5.2: Simulated particle size distribution as a function of particle radius and altitude simulated in (a) Nominal, (b) No $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ homogeneous nucleation, (c) No S_8 vapor, and (d) No lower haze cases. The discontinuous size distribution below $0.1 \mu\text{m}$ between 60 and 70 km is an artifact due to the condensation scheme treated in Lagrangian manner (see, Karyu et al., 2025).

Significant amounts of CN particles form in the lower and middle cloud layers (Figures 5.3a and 5.3b). The sub-micron CN particles originate from the lower haze supplied from the bottom boundary, while micron-sized CN particles form as S_8 vapor condenses onto these preexisting haze particles. These CN particles can be further activated by heterogeneous nucleation of H_2SO_4 and transferred to the mixed particle size distribution. Sulfur allotropes have long been proposed as a

component of the lower and middle clouds based on gas chromatography experiments from the Vega probes (Krasnopolsky, 1985, 1989; Mukhin et al., 1987), and our results support these earlier findings.

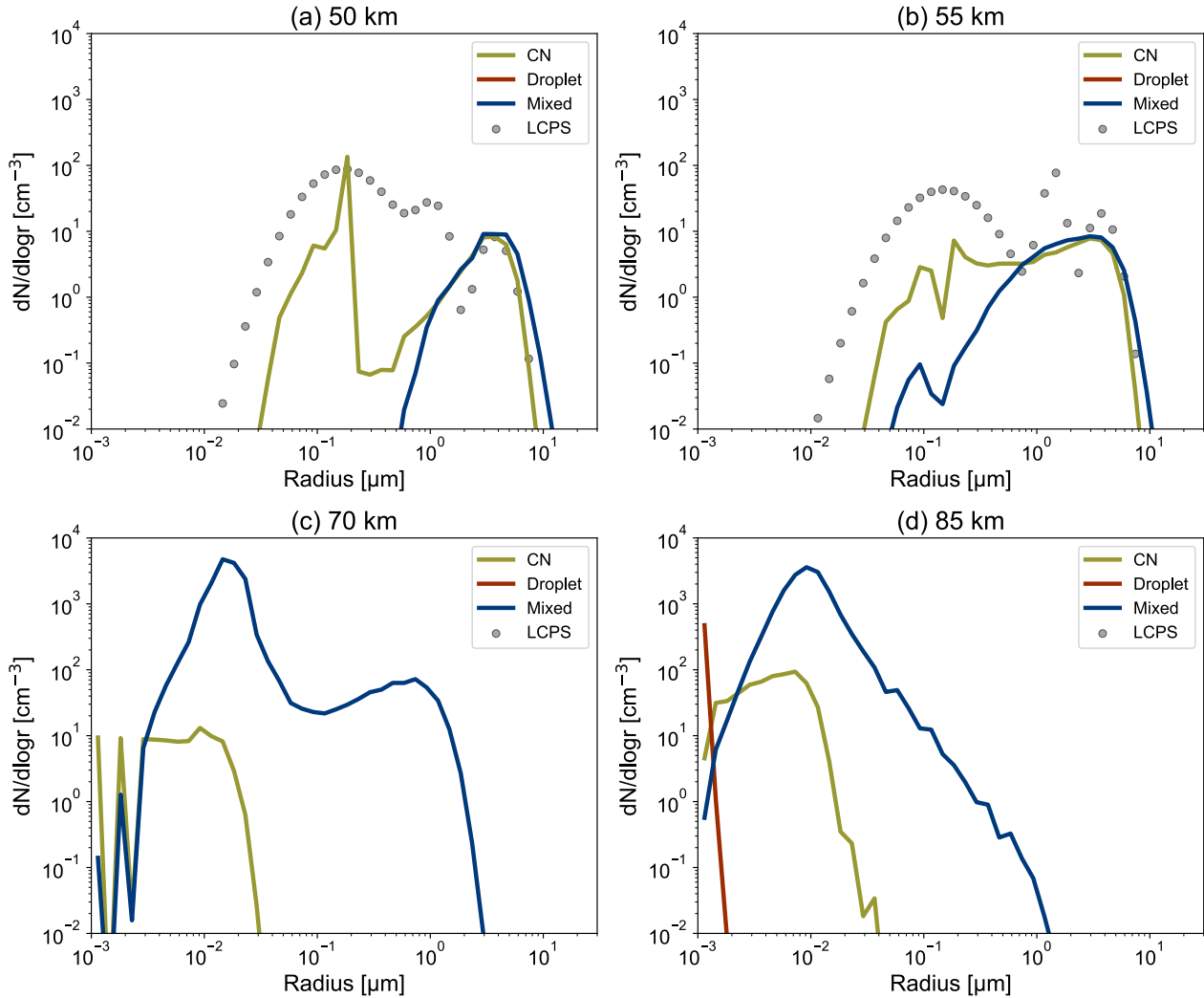


Figure 5.3: Simulated size distribution of the CN particles (yellow), H₂SO₄-H₂O particles (red), and mixed particles (blue) at altitudes of (a) 50 km, (b) 55 km, (c) 70 km, and (e) 85 km in Nominal case. The simulated distributions are compared with the LCPS in-situ measurements by Knollenberg and Hunten (1980) (gray circles). The red line is rarely seen in the plot because the H₂SO₄-H₂O homogeneous nucleation only occurs at high altitudes.

The simulated H₂SO₄ VMR profile is consistent with previous observations (Kolodner and Steffes, 1998; Imamura et al., 2017; Oschlisniok et al., 2021) (Figure 5.4a). The profile closely follows the saturation VMR from 48 to 60 km, where the photochemical production of H₂SO₄ becomes effective. Above 60 km, the H₂SO₄ vapor is supersaturated, which is consistent with recent cloud modeling studies incorporating the same H₂SO₄ production profile (Dai et al., 2022; Karyu et al., 2024; Shao et al., 2024).

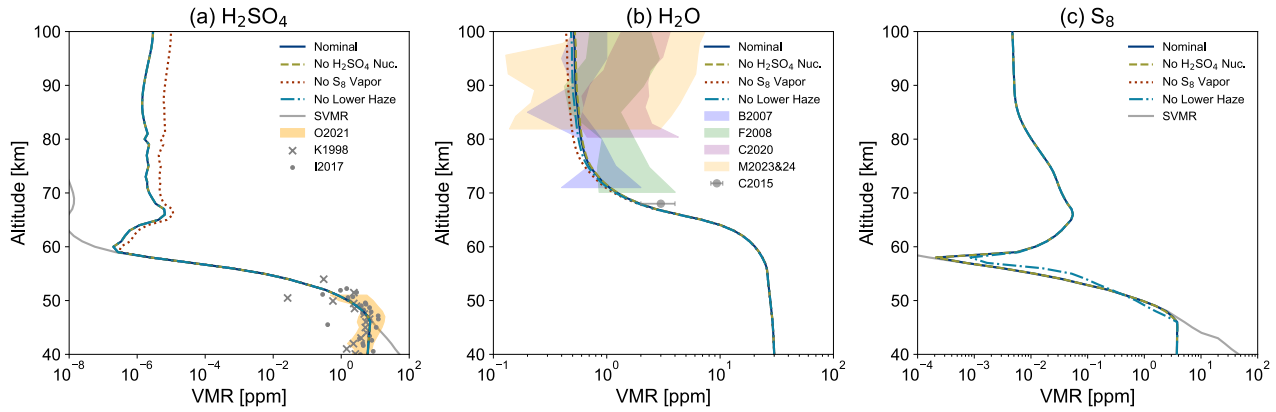


Figure 5.4: Simulated vertical profiles of (a) H_2SO_4 vapor VMR, (b) H_2O vapor VMR, and (c) S_8 vapor VMR in each case, plotted with observed values. H_2SO_4 vapor observations are taken from Oschlisniok et al. (2021) (O2021), Kolodner and Steffes (1998) (K1998), and Imamura et al. (2017) (I2017). H_2O observations are taken from Bertaux et al. (2007) (B2007), Fedorova et al. (2008) (F2008), Chamberlain et al. (2020) (C2020), Mahieux et al. (2023b), Mahieux et al. (2024) (M2023&24), and Cottini et al. (2015) (C2015). Saturation vapor mixing ratio is also plotted with gray line for H_2SO_4 and S_8 .

The H_2O VMR profile is in good agreement with the VEx observations (Chamberlain et al., 2020; Cottini et al., 2015; Fedorova et al., 2008; Mahieux et al., 2023b, 2024) (Figure 5.4b). The rapid decrease in H_2O VMR between 60 and 70 km is caused by a combination of photochemical loss and the downward flux of liquid H_2O driven by droplet sedimentation (Karyu et al., 2024).

The S_8 VMR profile is undersaturated below 48 km, closely follows the saturation curve from 48 to 59 km, and becomes supersaturated above 59 km (Figure 5.4c). Its peak VMR of ~ 50 ppb is located at an altitude of approximately 67 km. The cloud base for both H_2SO_4 and S_8 is located at 48 km, as both vapors are undersaturated below this altitude. These characteristics of the cloud base are consistent with the findings of Krasnopolsky (2016), who studied S_8 condensation using a simplified model.

The nuclei formation via homogeneous nucleation and subsequent cloud formation occur above 60 km (Figure 5.5a). The H_2SO_4 - H_2O homogeneous nucleation rate peaks at approximately 85 km. This is a combined result of H_2SO_4 supersaturation due to photochemical production and lower temperatures in the mesosphere. The S_8 homogeneous nucleation rate peaks at around 67 km (Figure 5.5b), overlapping with its photochemical production rate profile. The heterogeneous nucleation rate of H_2SO_4 - H_2O onto S_8 particles shows a nearly identical profile to the S_8 homogeneous nucleation rate, indicating that cloud droplets form immediately after the initial S_8 particles are generated. These newly formed cloud droplets then grow further via H_2SO_4 condensation. This combined process provides an efficient mechanism for generating cloud droplets in the upper cloud layer, a result also reflected in Figure 5.2a.

Although both H_2SO_4 - H_2O and S_8 homogeneous nucleation are active in the upper haze layer, S_8 nuclei contribute more to haze formation. As shown in Figures 5.4a and 5.4c, the S_8 vapor abun-

dance is significantly greater than that of H_2SO_4 . This is partly due to the model assumption that prohibits S_8 condensation onto sulfuric-acid-coated particles. Consequently, S_8 nuclei grow faster than H_2SO_4 - H_2O nuclei through condensation (Figure 5.3d). These S_8 particles are then activated via heterogeneous nucleation and transferred to the mixed-particle size distribution, creating a peak in its number density at approximately $0.01\ \mu\text{m}$. In contrast, sulfuric acid particles grow more slowly by condensation and are therefore scavenged by larger particles before reaching sizes that would significantly affect the size distribution.

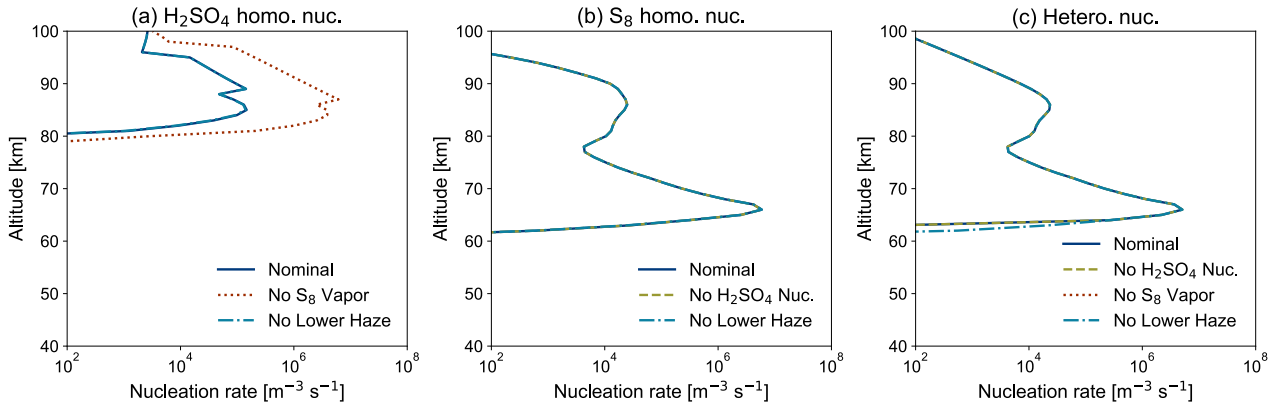


Figure 5.5: Simulated vertical profiles of (a) H_2SO_4 - H_2O homogeneous nucleation rate, (b) S_8 homogeneous nucleation rate, and (c) heterogeneous nucleation rate of H_2O - H_2O onto S_8 particles in each case. Note that H_2SO_4 homogeneous nucleation rate is not shown for the case study without H_2SO_4 - H_2O homogeneous nucleation and that S_8 homogeneous nucleation rate is not shown for the case study without S_8 vapor.

The simulated mass loading is also consistent with the PV LCPS observations (Figure 5.6a). The first mass loading peak, centered at approximately 52 km, is formed by condensation of H_2SO_4 vapor supplied from the sub-cloud region. The second peak, around 65 km, results from S_8 homogeneous nucleation (Figure 5.5b) and subsequent particle growth via H_2SO_4 condensation; this process is a direct consequence of photochemistry in the upper cloud layer. This overall cloud formation mechanism is consistent with previous microphysics simulations (Imamura and Hashimoto, 2001; James et al., 1997; McGouldrick and Toon, 2007).

For comparison with remote sensing observations, we derived the effective radius, defined as the area-weighted mean particle radius (Figure 5.6b). The simulated profile is consistent with the in-situ data from PV LCPS (Knollenberg and Hunten, 1980), showing an effective radius of $\sim 4\ \mu\text{m}$ in the lower and middle cloud layers and $\sim 1\ \mu\text{m}$ in the upper cloud layer. Furthermore, the simulated effective radius of $\sim 1\ \mu\text{m}$ at the cloud-top altitude ($\sim 70\ \text{km}$) agrees with previous remote sensing measurements (Kawabata et al., 1980; Markiewicz et al., 2018; Satoh et al., 2021).

For comparison with observations from the SPICAV instrument suite (UV, IR, and SOIR) on-board VEx, we derived extinction coefficients at three representative wavelengths: 200 nm, $1\ \mu\text{m}$, and $3\ \mu\text{m}$ (Luginin et al., 2016, 2024; Wilquet et al., 2009, 2012). The simulated extinction coefficients are consistent with the observations at each wavelength within observational uncertainty (Figure 5.7).

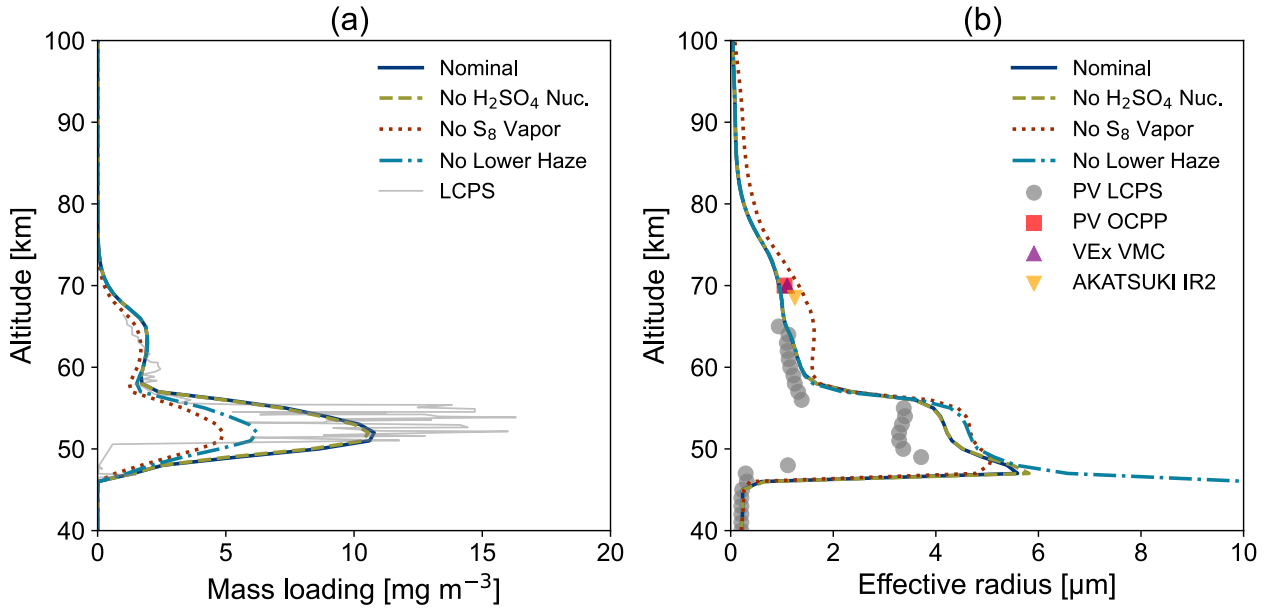


Figure 5.6: Simulated vertical profiles of (a) mass loading and (b) effective radius in each case, plotted with observed values. The mass loading observation is taken from the PV LCPS measurements (Knollenberg and Hunten, 1980). The effective radius observations are taken from PV LCPS (Knollenberg and Hunten, 1980), PV OCPP (Kawabata et al., 1980), VEx VMC (Markiewicz et al., 2018), and AKATSUKI IR2 (Sato et al., 2020).

SPICAV-UV is sensitive to sub-micron particles, while SPICAV-IR and -SOIR are sensitive to micron-sized particles. Therefore, the close agreement between our results and these multi-wavelength observations indicates that the model accurately represents particle abundances across a broad size range, from sub-micron to micron scales.

Table 5.2 lists the simulated cloud top altitude and the optical thickness at a $1 \mu\text{m}$ wavelength. The cloud top is located at 70 km, which is in agreement with observed mid-latitude values ranging from 68 to 74 km at near-infrared wavelengths (Haus et al., 2014, 2013; Ignatiev et al., 2009; Satoh et al., 2021). The total cloud optical thickness is 32.3, consistent with typical values of 25 to 40 reported by near-infrared observations (Grinspoon et al., 1993; Haus et al., 2013, 2014; Satoh et al., 2009, 2021). Furthermore, the optical thickness of the upper cloud is consistent with the range of 15 to 20 reported by Grinspoon et al. (1993).

Overall, the nominal model shows good agreement with previous observations and numerical modeling across a range of metrics. This indicates that the self-consistent particle formation framework presented in this study—which directly calculates vapor-to-particle conversion—can successfully simulate a realistic Venus cloud structure.

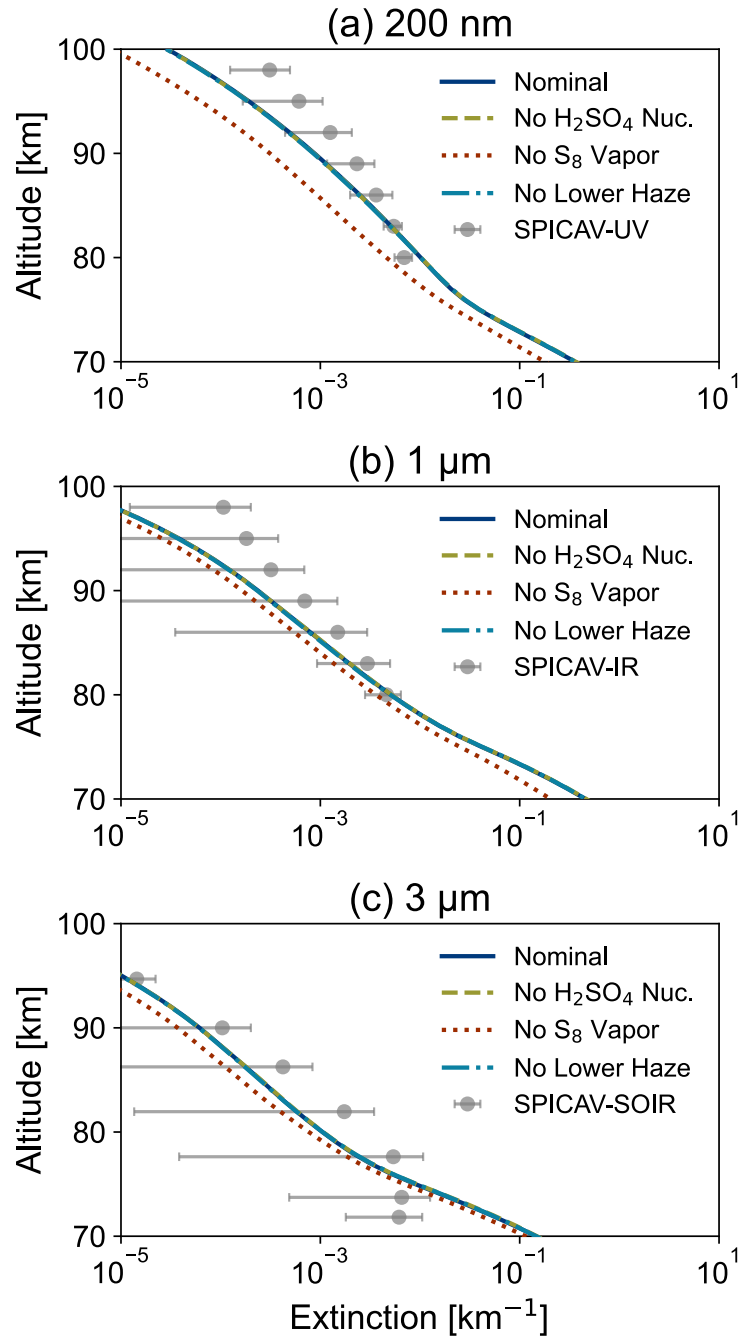


Figure 5.7: Simulated vertical profiles of upper haze extinction calculated using (a) 200 nm, (b) 1 μm , and (c) 3 μm for each case, plotted with observed values. The observations of 200-nm and 1- μm extinction profiles are taken from SPICAV-UV and -IR measurements (Luginin et al., 2024), The observations of 3- μm extinction profiles are taken from SPICAV-SOIR measurements (Wilquet et al., 2009, 2012).

Table 5.2: Summary of cloud top altitude and cloud opacity at 1- μm wavelength

Case	Cloud top altitude (km)	Lower and middle cloud opacity	Upper cloud opacity	Total opacity	Relative change ^d
Nominal	70	13.7	17.3	32.3	-
No H ₂ SO ₄ nuc.	70	13.3	17.3	31.9	1%
No S ₈ vapor	68	7.0	10.6	18.2	44%
No lower haze	70	8.1	17.4	26.8	17%
Observations	68–74 ^a	-	15–20 ^b	25–40 ^c	-

^a Ignatiev et al. (2009), Haus et al. (2013), Haus et al. (2014), Sato et al. (2020).

^b Grinspoon et al. (1993).

^c Grinspoon et al. (1993), Satoh et al. (2009), Satoh et al. (2021), Haus et al. (2013), Haus et al. (2014).

^d Relative change of total optical thickness compared to the nominal case.

5.2.2 No H₂SO₄ Nucleation

As shown in Figure 5.2b, excluding H₂SO₄-H₂O homogeneous nucleation does not significantly change the particle size distribution in the nominal case. As a result, the simulated vapor profiles (Figure 5.4), mass loading (Figure 5.6a), and effective radius (Figure 5.6b) are nearly identical to those of the nominal case. The upper haze profiles are also similar, supporting the conclusion that the majority of haze particles form via S₈ homogeneous nucleation and subsequent heterogeneous nucleation of H₂SO₄ onto S₈ CN. Consequently, the cloud optical thickness is nearly the same as that of the nominal case (Table 5.2). These results are consistent with previous studies showing that, under classical nucleation theory, homogeneous nucleation of H₂SO₄-H₂O plays only a minor role in Venus' cloud formation (McGouldrick and Barth, 2023).

5.2.3 No S₈ vapor

This scenario resulted in a particle size distribution completely different from that of the nominal case (Figures 5.2c and 5.8). The primary reason for this difference is the absence of S₈ homogeneous nucleation in the upper cloud layer. In this region, particle number density significantly decreases (Figure 5.8c), particularly for particles smaller than $\sim 0.07 \mu\text{m}$. This scarcity of small particles also reduces the number density in the middle and lower cloud layers, since a substantial fraction of these clouds is seeded by particles settling from the upper cloud layer. The resulting decrease in cloud mass is evident at all altitudes (Figure 5.6a).

Consequently, the total optical thickness only reaches 18.2, approximately half the value of the nominal case (Table 5.2). This value falls well outside the observed range of 25 to 40 for Venus' total optical thickness. This suggests that if the Venus cloud system were composed solely of H₂SO₄ and

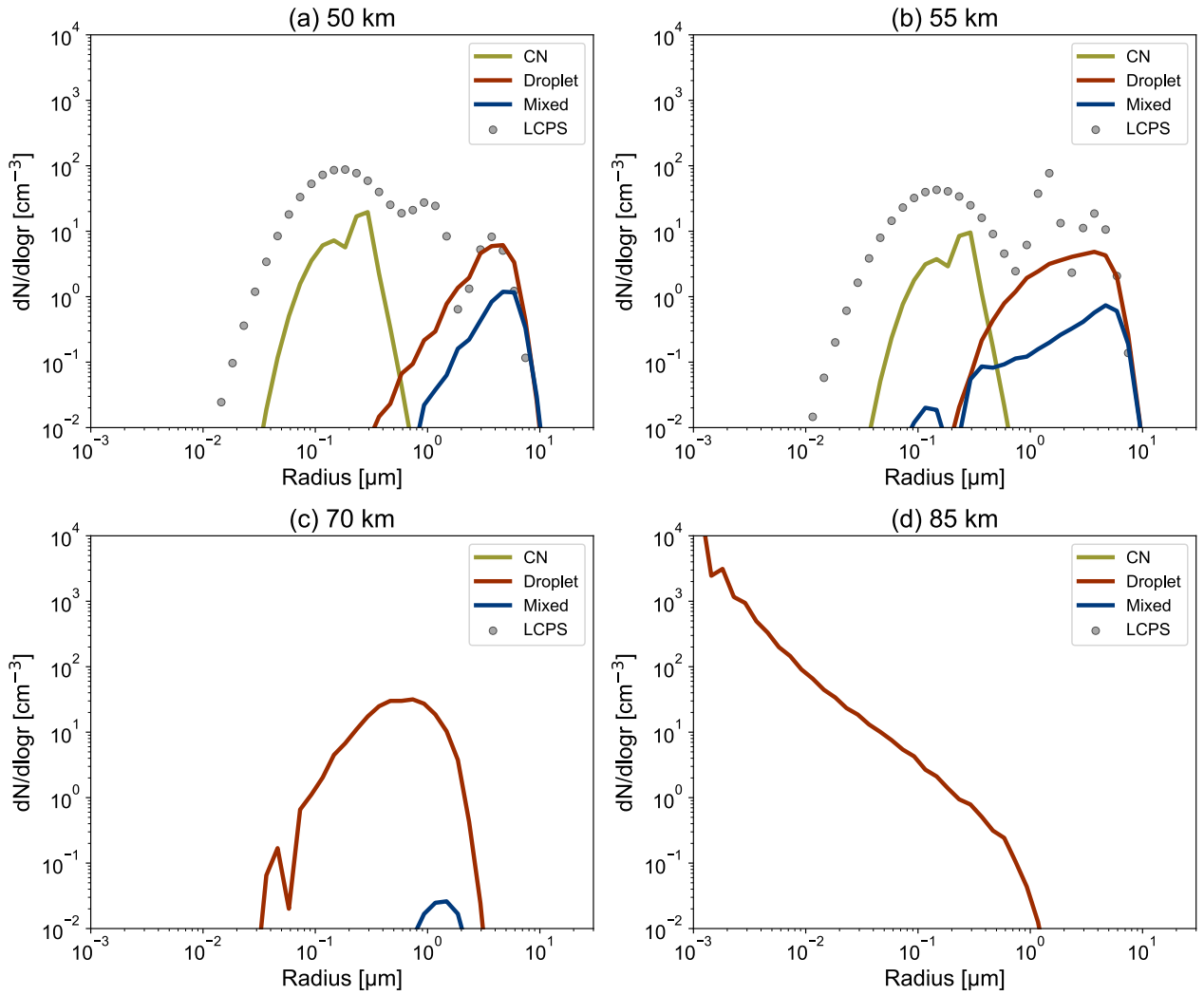


Figure 5.8: Simulated size distribution of the CN particles (yellow), $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ particles (red), and mixed particles (blue) at altitudes of (a) 50 km, (b) 55 km, (c) 70 km, and (e) 85 km in the case without S_8 vapor. The simulated distributions are compared with the LCPS in-situ measurements by Knollenberg and Hunten (1980) (gray circles).

H_2O , it would be impossible to explain the observed cloud properties. Thus, homogeneous nucleation of S_8 appears to be necessary to reproduce the observed cloud optical thickness.

In this simulation, the upper haze still forms (Figure 5.7) from homogeneous nucleation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ (Figure 5.5a). This nucleation process is enhanced by more than an order of magnitude compared to the nominal case because more H_2SO_4 vapor is available in the upper haze region (Figure 5.4a). The increased H_2SO_4 vapor is caused by a weakened condensation sink due to the smaller cloud amount simulated in this case study. Enhanced nucleation produces a peak in particle number density at 1 nm, with a gradual decrease toward larger sizes (Figures 5.2c and 5.8d), indicating a distinct steady-state size distribution for the upper haze in the absence of S_8 . In this scenario, the haze extinction

coefficient is generally consistent with the observations (Figure 5.7), although the 200-nm profile is slightly lower than the observed values. This slight discrepancy at 200 nm is due to the absence of sub-micron particle formation initiated by S_8 nucleation. In summary, while a reasonable amount of upper haze may still form without S_8 , the main cloud layers cannot be sustained.

5.2.4 No lower haze

This simulation allows us to assess the contribution of the lower haze to cloud formation. Without CN supplied from below the cloud deck, mass loading in the lower and middle cloud layers decreases by approximately half compared to the nominal case, and the effective radius increases (Figure 5.6b) because the available H_2SO_4 vapor is partitioned among fewer particles. The optical thickness of the lower and middle clouds also decreases by about half, resulting in a 17% decrease in the total optical thickness compared to the nominal case (Table 5.2). This suggests that nearly half of the cloud mass in the lower and middle layers originates from the lower haze, while the rest is supplied by particles falling from the upper cloud. The near-total absence of sub-micron particles in the lower and middle cloud layers (Figure 5.2d) indicates that S_8 particles descending from the upper cloud layer are not recycled as CN after they evaporate at the cloud base. In previous studies (Gao et al., 2014; Imamura and Hashimoto, 2001; Karyu et al., 2024; McGouldrick and Barth, 2023), the microphysical properties of photochemical CN and lower haze particles were not separated from one another. Instead, the photochemical CN (assumed to be S_8) were treated as involatile and were therefore recycled through droplet evaporation and subsequent upward transport. Our results call this assumption into question, suggesting that if the photochemical CN are indeed composed of volatile sulfur allotrope, they are not a sustainable source of recycled nuclei for the lower clouds.

5.3 Discussion

5.3.1 Cloud formation scenario

In this section, we estimate the contribution of each nucleus type to the total cloud optical thickness by comparing the results of our case studies (Table 5.2). The contribution of H_2SO_4 - H_2O homogeneous nucleation is minimal, as its exclusion changes the total optical thickness by only 1% compared to the nominal case. In contrast, removing S_8 vapor decreases the total optical thickness by 44%. However, this value cannot be interpreted as the direct contribution of S_8 , because the removal of S_8 enhances H_2SO_4 nucleation, which then partly compensates for the loss of S_8 -initiated cloud formation.

The direct contribution can be assigned to the lower haze. Removing this source decreases the total optical thickness by 17%. This can be considered a direct measure of the lower haze's contribution because its supply to the lower clouds is decoupled from the nucleation processes occurring in the upper atmosphere (McGouldrick and Toon, 2007; McGouldrick, 2017). Given that our study considers only three types of CN, the contribution of S_8 can be inferred by subtracting the contributions from the other two sources from the total. The combined contribution of H_2SO_4 nuclei (1%) and the lower

haze (17%) is 18%. Therefore, the remaining 82% of the total optical thickness can be attributed to cloud formation initiated by S_8 nucleation.

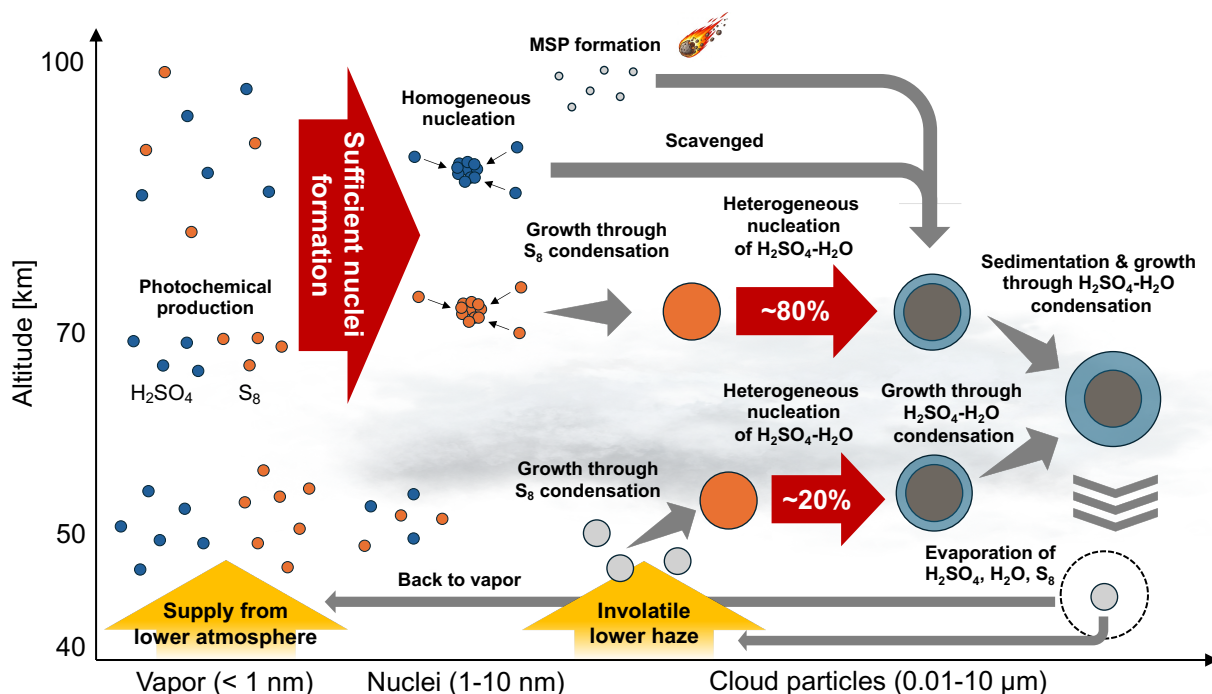


Figure 5.9: Sketch of the cloud cycle proposed in this study. Note that H_2O is an important condensable species as well as H_2SO_4 , but it is not included in the illustration to reduce the visual complexity.

Based on our findings, we propose the following cloud formation scenario, summarized in Figure 5.9. The primary source of CN is the homogeneous nucleation of S_8 vapor in the upper cloud layer. These CN are immediately activated into $H_2SO_4-H_2O$ droplets via $H_2SO_4-H_2O$ heterogeneous nucleation due to high supersaturation of H_2SO_4 , and this source accounts for ~80% of the total optical thickness of the cloud. These droplets grow via $H_2SO_4-H_2O$ condensation to become ~1 μm particles and then sediment into the middle cloud layer. As they keep sedimenting, the droplets grow larger, eventually becoming the large Mode 3 particles observed in the lower and middle clouds.

The lower haze particles also play a crucial role, acting as CN for the lower and middle cloud layers and accounting for ~20% of the total optical thickness. These lower haze particles are originally supplied by cosmic dust ablation and the subsequent MSP formation, as shown in Chapter 4. The MSPs are incorporated into H_2SO_4 droplets in the upper haze layer and sediment through the haze and cloud layers within these droplets. Upon reaching the cloud bottom through sedimentation, both H_2SO_4 and S_8 evaporate, leaving behind only the involatile olivine originating from the MSP and eventually form the lower haze particles. The lower haze particles are transported into the cloud layer and recycled as CN.

Our results also suggest that S_8 nucleation is important to form the upper haze, where the newly formed S_8 particles grow to ~0.01 μm and provide an efficient source of CN for H_2SO_4 droplets. On

the other hand, the upper haze formation via $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ homogeneous nucleation cannot be ruled out since the upper haze extinction profile roughly agrees with the previous observations without S_8 nucleation, as demonstrated in Section 5.2.3.

5.3.2 Implication for cloud particle composition

Our nominal model shows that a significant amount of solid S_8 particles can form within the cloud layer, which may provide an important insight into the observations from the PV LCPS instrument. Knollenberg and Hunten (1980) reported the existence of Mode 3 particles with high aspect ratios in the lower and middle clouds, suggesting they were made of a solid crystalline material. Sulfur allotropes are a strong candidate for this material, as they exist as a solid phase under the temperature conditions of the Venus' clouds (Lyons, 2008). Our model shows that solid particles can grow to the size of Mode 3 particles (Figure 5.4a and 5b). This is further supported by gas chromatography experiments from the Vega probe missions, which suggested the presence of reduced sulfur components in the clouds (Krasnopolsky, 1985, 1989; Mukhin et al., 1987). Therefore, we propose that the crystalline, high-aspect-ratio particles observed by PV can be composed of solid sulfur allotropes.

5.3.3 Open questions and future perspectives

Our results indicate that S_8 is a key component of Venus' clouds and that the observed cloud structure cannot be reproduced without their contribution. However, the physical properties of both gaseous and solid sulfur allotropes (not only S_8 but also S-S_7) are poorly constrained, especially under Venus' atmospheric conditions. To better understand the cloud formation processes on Venus, this knowledge gap must be addressed. We have identified the following key areas where experimental data are critically needed:

- (i) Saturation vapor pressure at low temperatures. The saturation vapor pressure of S_8 used in current models is an extrapolation from room-temperature experimental data. Since the homogeneous nucleation rate strongly depends on the degree of supersaturation, this uncertainty may result in order-of-magnitude differences in the calculated rates. As our work shows, homogeneous nucleation is most important at temperatures far below room temperature, where experimental data are currently missing and urgently needed.
- (ii) Contact angle of H_2SO_4 on solid sulfur. This parameter is crucial for accurately simulating the heterogeneous nucleation process, as it determines how efficiently a CN can be activated into an H_2SO_4 droplet. This effect is therefore critical for simulating the initial step of droplet formation throughout the cloud layers. To date, no experimental data for this contact angle are available, despite its importance. Young (1983) qualitatively showed that sulfur allotropes are insoluble in sulfuric acid, suggesting a contact parameter less than unity. Therefore, in addition to experiments, a parameter study against the contact parameter should be the focus of future work to understand the connection between the sulfur allotrope's physical properties and the observed cloud structure.

- (iii) Condensation of sulfur allotropes onto H_2SO_4 droplets. Whether sulfur can condense onto existing H_2SO_4 droplets is critical for simulating both the cloud composition and the abundance of sulfur allotrope gases. In our model, the high abundance of S_8 vapor above 60 km (Figure 5.4) partly reflects the assumption that this process is prohibited. Although previous observations suggest that upper cloud particles are spherical and coated in liquid H_2SO_4 (Hansen and Hovenier, 1974; Kawabata et al., 1980; Markiewicz et al., 2018), another study argued for the possibility of S_8 condensation in the cloud layers (Young, 1983) in the lower and middle clouds. To constrain the composition of Venus' clouds, the possibility of sulfur condensation onto liquid H_2SO_4 particles should be explored in future experimental studies.

In addition to these physical properties, sulfur chemistry plays an important role in cloud formation. Lyons (2008) shows that the saturation vapor pressure of different sulfur allotropes can vary by orders of magnitude; specifically, smaller molecules (e.g., S_2 , S_3) have a lower saturation vapor pressure and can therefore nucleate or condense more efficiently. Since our model assumes that all sulfur consists of S_8 and does not include detailed sulfur chemistry, the nucleation rate may be underestimated. Furthermore, our model uses a simple stoichiometric production ratio of one sulfur molecule per two H_2SO_4 molecules (Imamura and Hashimoto, 2001), whereas photochemical models predict that the production rate of sulfur aerosol is similar to H_2SO_4 in the upper cloud region (Zhang et al., 2012). Therefore, investigating the connection between sulfur chemistry and cloud microphysics is a critical next step.

Finally, we outline prospects for upcoming Venus missions. The VenSpec spectrometer suite on board ESA's EnVision mission (ESA, 2023) plans to observe Venus' clouds at different wavelengths ranging from UV to near infrared. VenSpec-U (Conan et al., 2024) will observe the UV spectra at the cloud top with high resolution to elucidate the nature of the unknown UV absorber (e.g., Pollack et al., 1980), possibly related to the abundance of sulfur allotropes (Toon et al., 1982). VenSpec-H will observe the cloud layer at near infrared wavelength, including polarization signals from the cloud top (Neefs et al., 2025). As previously demonstrated (Hansen and Hovenier, 1974; Kawabata et al., 1980), polarization data can be used to derive the size and refractive index of the cloud particles, which is primary information to understand cloud microphysics and composition. Our model provides a detailed dataset of cloud microphysical properties and composition simulated from the most fundamental processes, enabling future comparisons with these observations to constrain Venus' cloud cycle.

Furthermore, the mass spectrometer onboard NASA's DAVINCI probe will observe trace gas abundances, including H_2SO_4 and sulfur allotropes (S-S_8) (Garvin et al., 2022), both primary components of Venus' clouds. In particular, as demonstrated in our study, sulfur allotropes play a key role in cloud formation. Therefore, constraining their abundance throughout the cloud deck will help us understand the cloud formation processes, since there has been no direct measurement of sulfur allotropes in the cloud layer.

5.4 Chapter Summary

We investigated the fundamental origins of Venus clouds by developing a 1-D microphysics model that self-consistently simulates cloud formation from the vapor phase, including S_8 . By comparing simulations with different sources of cloud nuclei—photochemically produced S_8 , H_2SO_4 , and the lower haze layer—we assessed their relative contributions to the global cloud system.

Our nominal model incorporates all three CN sources. It successfully reproduces a wide range of observational constraints, including the cloud structure, gas abundance profiles, mass loading, and optical thickness. The key findings from our sensitivity studies reveal a clear hierarchy among the cloud formation mechanisms. We found that homogeneous nucleation of H_2SO_4 - H_2O has a negligible impact on the overall cloud mass. In contrast, the lower haze particles serve as an important source of CN for the lower and middle cloud decks, contributing $\sim 20\%$ of the total cloud optical thickness. Our results demonstrate that the homogeneous nucleation of S_8 vapor in the upper atmosphere is the dominant mechanism responsible for sustaining Venus's massive cloud deck, accounting for an estimated $\sim 80\%$ of the total cloud mass. Without this photochemical source of S_8 particles, the simulated cloud optical thickness is reduced by nearly half, well below observed values. This finding establishes that sulfur allotropes are a critical driver of the entire Venus cloud cycle. Furthermore, our results suggest that a significant amount of sulfur allotropes can condense in the lower and middle cloud layers. As sulfur allotropes likely exist in the solid phase, this may provide an explanation for crystalline particles with high aspect ratios found by the PV LCPS.

Future progress in understanding Venus's atmosphere will depend heavily on laboratory experiments to better constrain key physical and chemical properties of sulfur allotropes under Venus' conditions. Specifically, measurements of saturation vapor pressure at low temperatures, the contact angle of H_2SO_4 - H_2O on solid sulfur, and the potential for S_8 to condense onto sulfuric acid droplets are urgently needed to reduce uncertainties in microphysical models and refine our understanding of the cloud life cycle.

Furthermore, by comparing our cloud model with future observations, we can advance our understanding of Venus' cloud formation. Key parameters for comparison with our simulations include cloud microphysical properties and composition measured by the VenSpec suite onboard the EnVision mission and sulfur allotrope abundances measured by the DAVINCI probe.

Chapter 6

Conclusions and Future Perspectives

6.1 General Conclusions

The main goal of this thesis was to elucidate the origins of Venus clouds by answering the three questions posed in Chapter 1. To do this, we first needed to constrain eddy diffusion coefficients to accurately simulate the cloud and condensable species profiles. We also needed to develop a cloud microphysics model that can simulate microphysical processes of multi-component cloud particles with high accuracy.

In Chapter 2, we constrained the eddy diffusion coefficients by conducting parameter studies based on eddy diffusion coefficients observed at different altitudes. We found that the eddy diffusion coefficient should be around $\sim 2 \text{ m}^2 \text{ s}^{-1}$ between 60 and 70 km, and $\sim 360 \text{ m}^2 \text{ s}^{-1}$ above 85 km. This profile served as a reference in the following Chapters 4 and 5. In addition, we found that coupling between eddy transport and condensation/evaporation of cloud particles changes H_2O and H_2SO_4 abundances in the upper haze layer, potentially affecting atmospheric evolution and chemistry.

In Chapter 3, we developed a new cloud microphysics model based on the moving-center bin structure, named SPECK, to accurately simulate multi-component cloud microphysics. We thoroughly validated microphysical schemes implemented into this model in 0-D settings against the exact solution. Furthermore, we reproduced Venus cloud structure by incorporating the same settings as used in a state-of-the-art cloud microphysics model (McGouldrick and Barth, 2023). The model successfully reproduces the Venus cloud structure with lower resolution, suggesting that SPECK is applicable to future 3-D modeling.

Thanks to the development of SPECK, we finally were able to incorporate microphysical processes and new components that have not been included in existing models. The first one is S_8 and its microphysics, which has a key role in producing CN from vapor phase through homogeneous nucleation. The second one is inclusion of involatile substance, which is necessary to produce the lower haze layer without imposing it at the lower boundary. By including these processes, we set out to answer three questions posed at the beginning. In the following, we answer these questions based on

our findings from this thesis.

1. What is the origin of the lower haze?

In Chapter 4, we identified MSPs originating from cosmic dust ablation as a viable source of the lower haze. We simulated the lower haze formation by assuming constant MSP source from the upper atmosphere. The resulting microphysical characteristics were consistent with observations. We also excluded the possibility of other sources, such as dust or ash from the surface or volcanoes and photochemical products.

2. Can a sufficient amount of nuclei form by homogeneous nucleation?

In Chapter 5, we found that the combination of nuclei source hypothesized in Venus atmosphere— S_8 homogeneous nucleation, binary H_2SO_4 - H_2O homogeneous nucleation, and lower haze particles—successfully reproduces the observed Venus cloud structure. In particular, significant amount of S_8 nuclei can form in the upper cloud layer, and the subsequent heterogeneous nucleation of H_2SO_4 - H_2O efficiently seed the upper cloud particles.

3. What is the relative contribution of each type of nucleus?

Additionally, Chapter 5 revealed the hierarchy among different nuclei sources. The most significant one is S_8 which makes up $\sim 80\%$ of the observed cloud opacity. The second most important one is the lower haze particles seeding the remaining $\sim 20\%$ of the clouds. Finally, we found that the effect of binary H_2SO_4 - H_2O homogeneous nucleation is negligible, only contributing $\sim 1\%$ of cloud opacity.

The complete cycle of the Venus clouds is shown in Figure 5.9. The continuous photochemical supply of S_8 and subsequent homogeneous nucleation is necessary to form the observed thick upper cloud layer. On the other hand, H_2SO_4 - H_2O binary homogeneous nucleation plays a negligible role in the bulk cloud formation. The fact that the lower haze particles contribute to 20% of the observed cloud thickness on global average identifies cosmic dust as an essential part of Venus' climate, which has been overlooked in previous studies.

New questions emerged from this thesis as well. In Chapter 2, we learned that the interaction between aerosols and background atmosphere significantly changes H_2O abundance in the upper haze layer, consistent with recent observations by Mahieux et al. (2024). This may affect the atmospheric evolution through changing hydrogen escape rate from the top of atmosphere. However, to what extent aerocols can affect the climate evolution remains unknown and should be investigated in the future.

In Chapter 4, we suggest that the lower haze formation can also occur on other planets, especially on Jupiter. To what extent the lower haze formation can affect cloud formation in planetary atmospheres needs to be answered. This can be modeled by applying SPECK to other planets and investigated in the future, as described in the following Section 6.2.2.

In Chapter 5, we found that the contribution of sulfur allotropes is crucial for the cloud formation. We also pointed out that there are some missing important parameters that can affect the cloud microphysics processes, such as saturation vapor pressure and contact angle against liquid sulfuric acid. In addition, chemical reaction rates related to sulfur allotrope chemistry are poorly constrained, and estimated values are used in current modeling (Mills and Allen, 2007). Therefore, we recommend future experimental or theoretical work to better constrain these microphysical and chemical properties. Accordingly, the effects of these parameters on cloud structure should be investigated with SPECK in the future.

6.2 Future Perspectives

6.2.1 Synergies with future observations

Our model is the only existing Venus cloud microphysics model that include sulfur allotropes and MSP generated by cosmic dusts. This compositional addition can be an advantage to interpret future observations. We showed that the upper cloud particles form through heterogeneous nucleation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ on sulfur allotrope cores in Chapter 5. Hence, investigation of sulfur allotrope inclusion in the cloud droplets is necessary to validate our theory. Sulfur allotropes have a refractive index of 1.94 in near-infrared region (Sasson et al., 1985), which is significantly higher than that of H_2SO_4 being ~ 1.45 (Palmer and Williams, 1975). Thus, the observed real part of refractive index should be higher than H_2SO_4 if the upper cloud is made of a mixture of H_2SO_4 and sulfur allotropes. Polarimetric measurement is an effective way to determine the refractive index of particles (Bohren and Huffman, 2008). VenSpec-H onboard EnVision probe will observe polarization signals from the Venus atmosphere in near-infrared wavelengths (Neefs et al., 2025). Thus, the next step towards the comparison with future measurement will be simulation of the polarization signal from the particle composition and distribution obtained by SPECK.

The observation of gaseous sulfur allotropes is also a critical step to understand the Venus cloud system, as demonstrated in Chapter 5. The future DAVINCI mission plans to observe sulfur allotropes with mass spectrometer while descending through the atmosphere (Garvin et al., 2022). The sulfur allotrope abundance in the upper cloud can be used to estimate the homogeneous nucleation rate and compared with the present thesis results. On the other hand, sulfur allotrope abundance in the lower atmosphere will be a necessary information to set up the boundary condition used in SPECK, since the current version relies on the estimate from kinetic chemistry model (Krasnopolsky, 2007; Krasnopolsky et al., 2013).

Our model may be able to contribute to the identification of the unknown UV absorber. To date, many substances have been proposed based on their spectral match, including sulfur allotropes (Hapke and Nelson, 1975; Toon et al., 1982) and iron compounds (Zasova et al., 1981; Jiang et al., 2024; Egan et al., 2025). While previous studies have prioritized spectral fitting in selecting candidate substances, few have discussed the origins of these materials, except for sulfur allotropes (Toon et al., 1982). In Chapter 4 and 5, we showed that both iron compounds and sulfur allotropes can exist in the Venus clouds, based on plausible sourcing mechanism. To confirm if these sources can reproduce the observed UV absorption features, our simulation should be compared with the observed UV spectra from previous observations (Pollack et al., 1980; Pérez-Hoyos et al., 2018) as well as future observations performed by VenSpec-U (Conan et al., 2024).

6.2.2 Beyond Venus

The cloud microphysics model developed in this study, SPECK, is highly versatile and can be applied to any cloud systems by adding condensable species unique to each planet. The application to Mars

cloud system consisting of H_2O , CO_2 , and dust particles is ongoing and yielding promising results (Sato et al., 2025).

As discussed in Chapter 1, cloud system sourced by photochemistry is thought to be a common regime among observed planets, and Venus can be regarded as a prototype. On these outer planets, photochemically-generated hydrocarbon haze can serve as CN, and main condensable vapors (e.g., ammonia, ammonium hydrosulfide, water) condense on these CN (West et al., 2004). Here, the role of hydrocarbon to the condensable vapors are to what sulfur allotropes to sulfuric acid on Venus. Thus, cloud formation mechanism can be investigated by applying SPECK to outer planets. In addition, the lower haze formation on gas giants and ice giants should be tested in future studies, as its universality is suggested in Chapter 4. To date, the most common approach to studying outer planet clouds is equilibrium condensation models (Atreya et al., 1999; Sánchez-Lavega et al., 2004; Atreya and Wong, 2005), which neglect the detailed treatment of hydrocarbon CN and the resulting particle diversity. Applying SPECK to these outer planets will therefore be an important step toward general understanding of the cloud formation mechanism.

6.2.3 Future development: from photochemistry to climate

Our ultimate goal—though beyond the scope of this thesis—is to reproduce planetary climate from first principles with basic parameters such as basic atmospheric composition and host star spectra. As discussed in Chapter 1, clouds play a pivotal role to control the climate and atmospheric composition of planetary atmospheres. While the development of SPECK is necessary to assess the feedback of clouds in such ultimate model, there are still some critical steps that should be addressed in future.

The first one is coupling with chemistry. In this thesis, we developed SPECK, which can simulate cloud formation from the vapor phase via homogeneous nucleation. In other words, SPECK can simulate the cloud structure once the atmospheric composition is derived. However, since photochemistry determines this composition, simulating the cloud structure from a more fundamental level requires coupling the cloud microphysics model with a photochemical model. Photochemical and Radiation Transport Model for Extensive Use (PROTEUS) is a highly versatile photochemical model designed for application to various planets (Nakamura et al., 2023), developed at Tohoku University. Coupling SPECK with PROTEUS will be an ideal solution for connecting cloud microphysics with photochemistry and placed as a next step.

In addition to photochemistry, multiphase chemistry is an essential component of atmospheric chemistry. On Earth, it greatly impacts both atmospheric composition (e.g., ozone depletion by polar stratospheric clouds) and cloud composition (e.g., acid precipitation) (Seinfeld and Pandis, 2016; Abbatt and Ravishankara, 2023). In contrast, this field remains underexplored in planetary science, though it is slowly gaining attention (Delitsky and Baines, 2023; Lasne, 2021). A key challenge is constraining chemical reaction rates occurring in planetary aerosols because they cannot be sampled and directly measured for chemical analysis unlike Earth. However, recent progress on our experimental work is changing this situation. For example, Ubukata et al. (2025) measured the in-droplet rate of SO_2 under Venus cloud conditions, using Electrodynamic Balance (EDB) chamber developed

at Tohoku University. Collaboration with such experimental activities will enable the construction of multiphase chemistry models for planetary atmospheres.

The second one is coupling with radiative transfer. In the current version of SPECK, the temperature profile is fixed. Consequently, the derived cloud structure is simply an output for a given temperature condition. However, in the real atmosphere, clouds and the temperature profile are coupled, providing mutual feedback through radiative transfer. Planetary climates can be understood as the quasi-steady state resulting from these coupled processes. Thus, coupling SPECK with a radiative transfer model is necessary to understand how planetary climates are shaped.

In summary, reproducing planetary climates from first principles requires starting with basic atmospheric composition and simulating the interactions among photochemistry, cloud microphysics, and radiative transfer. Such a development will help elucidate Venus's complex climate, allowing us to address the questions listed below.

Is the current Venus climate the only possible solution?

The reason why the thick Venus clouds can reside within 50 and 70 km is because the atmospheric temperature is sufficiently low for H_2SO_4 to condense in these altitudes, which is, in turn, an result of high albedo maintained by the presence of the clouds. This indicates that if Venus' climate starts from cloud-free atmosphere, the atmospheric temperature is likely significantly higher and thick cloud may be impossible to form. This means that Venus had to have a specific initial condition to have the current climate—for example, a temperate climate gradually warming up to the current state, instead of having a super-hot atmosphere in the first place. If we can identify this kind of dichotomy in Venus' atmosphere, we may be able to constrain the past climate condition of Venus. This can also support the future observation of volcanic degassing by VenSpec-H onboard EnVision (Robert et al., 2025), since the degassed component can be used to constrain the past atmospheric condition of Venus (Grinspoon, 1993; Constantinou et al., 2025).

What is the cause of climate variation on Venus?

A decadal-scale Venus climate variation has been observed in planetary albedo (Lee et al., 2015, 2019), H_2O (Encrenaz et al., 2020), SO_2 (Marcq et al., 2020), H_2SO_4 (Oschlisniok et al., 2012), and zonal wind (Khatuntsev et al., 2022). Recently, a decadal-scale variation in temperature profiles is also confirmed, and its amplitude is five times larger than that of Earth (Ando et al., 2025). These variations are possibly interconnected through feedback between atmospheric chemistry, cloud microphysics, and radiative transfer (Lee et al., 2019; Ando et al., 2025). However, the connection between these atmospheric processes remains unknown since existing Venus climate models has not integrated these components in one framework. The future climate model including photochemistry, cloud microphysics, and its radiative effects, can investigate this phenomena through identifying feedback loops existing in Venus' atmosphere.

Appendix A

Coagulation

This section provides the formulation of the coagulation kernel used in this thesis. The coagulation kernel, denoted as $K(r_i, r_j)$ (or simply $K_{i,j}$), describes the rate at which two particles of radii r_i and r_j collide and coalesce. The evolution of the particle number density $n(r, t)$ is governed by the following coagulation equation (Pruppacher and Klett, 2010):

$$\frac{\partial n(r, t)}{\partial t} = \frac{1}{2} \int_0^r K(r - r', r') n(r - r', t) n(r', t) dr' - n(r, t) \int_0^\infty K(r, r') n(r', t) dr'. \quad (\text{A.1})$$

Particle collisions are driven by several mechanisms, the most important being Brownian diffusion (caused by random particle motion) and gravitational collection (caused by differential settling velocities). These two processes are standard in existing Venus cloud microphysics models (e.g. McGouldrick and Barth, 2023). While other mechanisms are considered on Earth—such as Van der Waals forces (important for fractal geometries) or turbulent inertial motion and shear (important for particles $>10 \mu\text{m}$)—these are less relevant for Venus. Therefore, we include only the two primary processes: Brownian diffusion and gravitational collection.

A.1 Brownian Diffusion

Particles suspended in the atmosphere constantly move irregularly because of random bombardments of air molecules, called Brownian diffusion. This random motion leads to scattering and collision of particles. This process is referred to as Brownian coagulation. The form of the Brownian diffusion coagulation kernel K depends heavily on the Knudsen number (Kn_a), defined as follows (Pruppacher and Klett, 2010; Jacobson, 2005):

$$\text{Kn}_{a,i} = \frac{\lambda_a}{r_i}, \quad (\text{A.2})$$

where λ_a is the mean free path of the gas molecules, and r_i is the particle radius. The mean free path of the gas molecules is the average distance that an air molecule can travel before colliding another air molecule, described as follows:

$$\lambda_a = \frac{2\eta_a}{\rho_a \bar{v}_a}, \quad (\text{A.3})$$

where η_a is the dynamic viscosity of air ($\text{kg m}^{-3} \text{s}^{-1}$), ρ_a is the density of air (kg m^{-3}), $\bar{v}_a$ is thermal speed of an air molecule. The thermal speed of an air molecule is expressed as follows:

$$\bar{v}_a = \sqrt{\frac{8k_B T}{\pi \bar{M}}}, \quad (\text{A.4})$$

where T is the air temperature (K), and $\bar{M}$ is the average weight of an air molecule (kg).

A.1.1 Continuum Regime ($\text{Kn}_a \ll 1$)

In the continuum regime, particles are large compared to the mean free path of the gas, and thus, the motion is diffusive. The collision rate is derived from the diffusion flux of particles towards a central absorbing sphere. In this case the coagulation kernel for the continuum regime is given as follows:

$$K_{i,j}^B = 4\pi(D_i + D_j)(r_i + r_j), \quad (\text{A.5})$$

where, r_i, r_j are the radii of the colliding particles, and D_i, D_j are the particle diffusion coefficients ($\text{m}^2 \text{s}^{-1}$). The particle diffusion coefficient is described as follows, using the Stokes-Einstein relation:

$$D_i = \frac{k_B T}{6\pi\eta_a r_i} G_i(\text{Kn}_{a,i}), \quad (\text{A.6})$$

where G_i is the Cunningham slip-flow correction, serving as the correction of the Stokes drag law for non-continuum effects. This parameter is further described as follows:

$$G_i(\text{Kn}_{a,i}) = 1 + \text{Kn}_{a,i} \left[A + B \exp\left(-\frac{C}{\text{Kn}_{a,i}}\right) \right]. \quad (\text{A.7})$$

Standard coefficients for air typically used are derived by Kasten (1968): $A = 1.249$, $B = 0.420$, $E = 0.87$.

A.1.2 Free Molecular Regime ($\text{Kn}_a \gg 1$)

In the free molecular (or kinetic) regime, particles are small compared to the mean free path. They behave like gas molecules, traveling in ballistic trajectories between collisions. The kernel is derived from the kinetic theory of gases, as follows (Jacobson, 2005):

$$K_{i,j}^B = \pi(r_i + r_j)^2 \sqrt{\bar{v}_{p,i}^2 + \bar{v}_{p,j}^2}, \quad (\text{A.8})$$

$$\bar{v}_{p,i} = \sqrt{\frac{8k_B T}{\pi \bar{M}_{p,i}}}, \quad (\text{A.9})$$

where $\bar{v}_{p,i}$ is the average thermal speed (m s^{-1}) of particle with mass $\bar{M}_{p,i}$.

A.1.3 Transition Regime ($\text{Kn}_a \approx 1$)

For particles intermediate in size ($\text{Kn} \approx 1$), neither the continuum nor the free molecular expressions are accurate. Fuchs (1964) proposed an interpolation formula for combining these two regimes. To do so, he considered a region of thickness δ as a function of the mean free path λ_p . He assumed that particles are transported by Brownian diffusion outside this region δ , while inside δ they undergo thermal motion like molecules. He derived the Brownian coagulation rate in the transition regime as follows.

$$K_{i,j}^B = \frac{4\pi(D_i + D_j)(r_i + r_j)}{\frac{r_i + r_j}{r_i + r_j + \sqrt{\delta_i^2 + \delta_j^2}} + \frac{4(D_i + D_j)}{\sqrt{\bar{v}_{p,i}^2 + \bar{v}_{p,j}^2}(r_i + r_j)}}, \quad (\text{A.10})$$

$$\delta_i = \frac{(2r_i + \lambda_{p,i})^3 - (4r_i^2 + \lambda_{p,i}^2)^{3/2}}{6r_i \lambda_{p,i}} - 2r_i, \quad (\text{A.11})$$

$$\lambda_{p,i} = \frac{8D_i}{\pi \bar{c}_i}. \quad (\text{A.12})$$

This equation simplifies to Equation (A.5) for small Knudsen numbers (continuum regime) and to Equation (A.8) for large Knudsen numbers (free molecular regime). Therefore, this expression is used for the Brownian diffusion coagulation kernel in this thesis.

A.2 Gravitational Collection

When particles with different sedimentation velocities fall, one may catch up to the other, resulting in a collision. This process is called the gravitational collection, formulated as follows (Pruppacher and Klett, 2010):

$$K_{i,j}^G = E_{\text{coll},i,j} \pi(r_i^2 + r_j^2) |V_{f,i} - V_{f,j}|, \quad (\text{A.13})$$

$$E_{\text{coll},i,j} = \frac{r_i^2}{2(r_i^2 + r_j^2)}, \quad (\text{A.14})$$

where $E_{\text{coll},i,j}$ is the collision efficiency between particles with radius r_i and r_j , and $V_{f,i}$ is the sedimentation velocity (m s^{-1}) of particle with radius r_i , introduced as Equation (3.2). As evident in Equation (A.13), the kernel reduces to 0 when two particles have the same sedimentation speed because one particle cannot catch up with the other in this case.

A.3 Coagulation Kernel under Venus' Atmospheric Conditions

The combined effect of the Brownian diffusion and the gravitational collection are expressed as the sum of both kernels (Jacobson, 2005):

$$K_{i,j} = K_{i,j}^B + K_{i,j}^G. \quad (\text{A.15})$$

Figure A.1 plots the coagulation kernel values calculated via Equation (A.15) under Venusian atmospheric conditions. Temperature and pressure profiles are from the VIRA mid-latitude data (Seiff et al., 1985). We selected reference particles with radii of $\sim 0.1 \mu\text{m}$ and $\sim 1 \mu\text{m}$, as these sizes dominate the observed distribution on Venus (Knollenberg and Hunten, 1980). For submicron particles ($\sim 0.1 \mu\text{m}$), Brownian diffusion dominates at all altitudes, regardless of the colliding particle's size. Gravitational collection is negligible here because the cross-sectional area decreases with particle radius. In contrast, gravitational collection accounts for a significant portion of the kernel when both colliding particles are larger than $1 \mu\text{m}$. As shown in Figure A.1, the coagulation kernel for micron-sized particles is dominated by Brownian diffusion when colliding with particles smaller than $\sim 1 \mu\text{m}$, and by gravitational collection for those larger than $\sim 1 \mu\text{m}$.

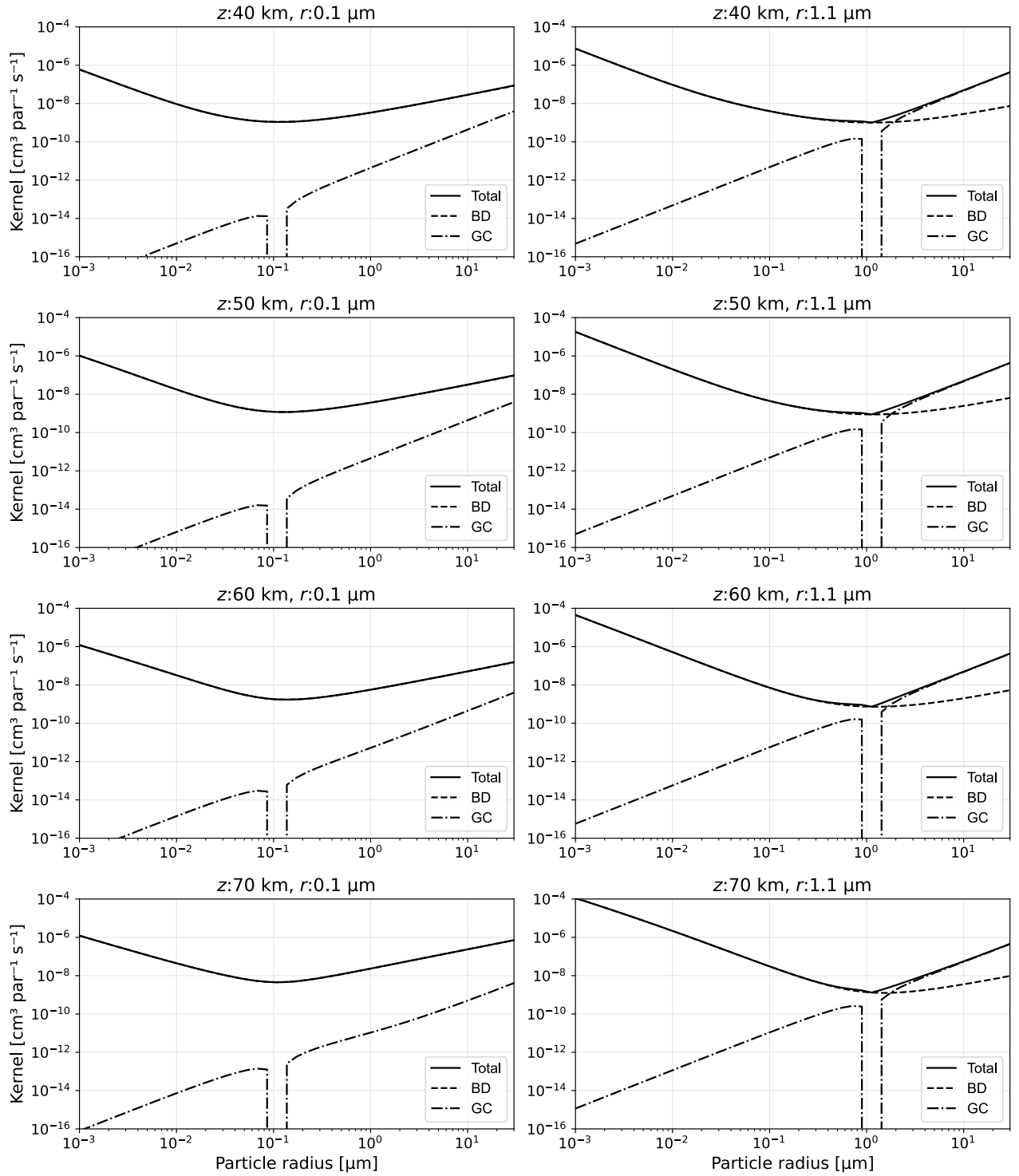


Figure A.1: Coagulation kernel at different altitudes and radii of colliding particles used in this thesis as a function of radius. “BD” and “GC” represent the coagulation kernel calculated based on Brownian diffusion and gravitational collection, respectively, and “Total” represents the sum of BD and GC. The dip of the GC kernel at 0.1 μm in the left column and 1.1 μm in the right column is created because the falling speed of the colliding particles is zero.

Appendix B

Condensation and Evaporation

B.1 Mass Flux to and from a Single Particle

The mass flux (kg s^{-1}) to or from a single particle made of substance q with size i is expressed as follows (Jacobson, 2005):

$$\frac{dm_i}{dt} = \frac{4\pi r_i D'_{q,i} (p_q - p'_{q,s,i})}{\frac{D'_{q,i} L_{e,q} p'_{q,s,i}}{\kappa'_{a,i} T} \left(\frac{L_{e,q} m_q}{RT} \right) + \frac{RT}{m_q}}, \quad (\text{B.1})$$

where r_i denotes the particle radius (m), $D'_{q,i}$ is the effective molecular diffusion coefficient ($\text{m}^2 \text{s}^{-1}$) of condensable species q onto a particle of size r_i , p_q is the ambient partial pressure (Pa) of the species, $p'_{q,s,i}$ is the saturation vapor pressure (Pa) over the particle surface, $L_{e,q}$ is the latent heat (J kg^{-1}), $\kappa'_{a,i}$ is the thermal conductivity of the moist air ($\text{J m}^{-1} \text{s}^{-1} \text{K}^{-1}$), R is the universal gas constant ($\text{J mol}^{-1} \text{K}^{-1}$), T is the air temperature (K), and m_q is the molar mass (kg mol^{-1}) of the condensable gas. This equation indicates that when the ambient partial pressure of species q exceeds the saturation vapor pressure, the mass flux is positive, leading to condensation. Conversely, if the partial pressure is lower than the saturation point, the mass flux becomes negative, resulting in evaporation. In the Venusian atmosphere, the latent heat of sulfuric acid is negligible. This assumption simplifies Equation (B.1) to:

$$\frac{dm_i}{dt} = \frac{4\pi r_i m_q D'_{q,i} (p_q - p'_{q,s,i})}{RT}. \quad (\text{B.2})$$

The diffusion coefficient of condensable species is further expressed as follows:

$$D'_{q,i} = D_q \omega_{q,i}, \quad (\text{B.3})$$

where D_q is the molecular diffusion coefficient ($\text{m}^2 \text{s}^{-1}$), and $\omega_{q,i}$ is the correction factor for collision geometry. The molecular diffusion coefficient is expressed as follows:

$$D_q = \frac{5}{16N_A d_q \rho_a} \sqrt{\frac{RT m_a}{2\pi} \left(\frac{m_q + m_a}{m_q} \right)}, \quad (\text{B.4})$$

where N_A is the Avogadro number, d_q is the collision diameter (m) of gas molecule q , ρ is the air density (kg m^{-3}), m_a is the molar weight of air (kg mol^{-1}). On the other hand, $\omega_{q,i}$ is described as follows:

$$\omega_{q,i} = \left[1 + \left\{ \frac{1.33 + 0.71 \text{Kn}_{q,i}^{-1}}{1 + \text{Kn}_{q,i}^{-1}} + \frac{4(1 - \alpha_{q,i})}{3\alpha_{q,i}} \right\} \text{Kn}_{q,i}^{-1} \right]^{-1}, \quad (\text{B.5})$$

where $\text{Kn}_{q,i}$ is the Knudsen number of the air molecule, defined as in Equation (A.2), and $\alpha_{q,i}$ is the accommodation coefficient of the gas molecule on the particle surface. The accommodation coefficient represents the ratio of gas molecules that stick to the particle surface to the total number of molecules colliding with it and is set to unity in this study.

The saturation vapor pressure over a particle, $p'_{q,s,i}$, differs from that over a flat surface, $p_{q,s}$, due to surface curvature—a phenomenon known as the Kelvin effect. The relationship between the saturation pressure over a curved surface and that over a flat surface is described as follows:

$$\frac{p'_{q,s,i}}{p_{q,s}} = \exp \left(\frac{2\sigma_p m_p}{r_i RT \rho_p} \right), \quad (\text{B.6})$$

where σ_p denotes the surface tension (J m^{-2}) of the particle, m_p is the mass (kg) of particle, ρ_p is the density (kg m^{-3}) of the particle.

B.2 Mass Flux to and from a Group of Particles

The condensation or evaporation rate of a single particle was derived in Section B.1. We now extend this framework to a system comprising multiple particles and ambient gas to calculate the vapor exchange between the gas and condensed phases in a cloud microphysics model. First, we need to convert Equation (B.2) into volume-concentration form. The volume of a single particle with radius r_i is $v_i = 4\pi r_i^3/3$. Given a particle number density n_i (par m^{-3}), the total volume concentration of particles with individual volume v_i is $v_i = n_i v_i$. Using this relationship and Equation (B.2), the condensation rate for a population of particles of size i can be expressed as:

$$\frac{dv_{q,i}}{dt} = n_i^{2/3} (48\pi^2 v_i)^{1/3} D'_{q,i} \frac{m_q}{\rho_{p,q}} (C_q - C'_{q,s,i}), \quad (\text{B.7})$$

where C_q is the number density (mol m^{-3}) of condensable gas molecule q calculated as $C_q = p_q/RT$, and $C'_{q,s,i}$ is the number density (mol m^{-3}) of condensable gas molecule q calculated as $C'_{q,s,i} = p'_{q,s,i}/RT$. We further simplify this Equation by defining the number concentration of condensed phase $c_{q,i}$ (mol m^{-3}), mass-transfer coefficient $k_{q,i}$ (s^{-1}), and effective saturation ratio $S'_{q,i}$ as follows:

$$\frac{dc_{q,i}}{dt} = k_{q,i}(C_q - S'_{q,i}C_{q,s}), \quad (\text{B.8})$$

$$c_{q,i} = \frac{\rho_{p,q}V_{q,i}}{m_q}, \quad (\text{B.9})$$

$$k_{q,i} = n_i^{2/3} (48\pi^2 v_i)^{1/3} D'_{q,i} = 4\pi r_i n_i D'_{q,i}, \quad (\text{B.10})$$

$$S'_{q,i} = \frac{C'_{q,s,i}}{C_{q,s}}, \quad (\text{B.11})$$

where $C_{q,s}$ is the saturation vapor pressure (Pa) of pure substance over a flat surface. On the other hand, the ambient vapor pressure also changes in response to the condensation growth of particles. This effect can be expressed as follows:

$$\frac{dC_q}{dt} = - \sum_{i=1}^{N_B} [k_{q,i}(C_q - S'_{q,i}C_{q,s})], \quad (\text{B.12})$$

where N_B is the total number of particle bins. Equations (B.8) and (B.12) comprises $N_B + 1$ ordinary differential equations (ODE).

B.3 Solutions to Growth Equations

The concentrations of condensed and vapor phases in the cloud microphysics model are determined by numerically solving this ODE system. Condensation or evaporation rates can be extremely rapid, depending on the saturation ratio of the condensable species. In such scenarios, solving Equations (B.8) and (B.12) with an explicit scheme may yield non-physical negative concentrations for C_q or $c_{q,i}$. Consequently, implicit schemes are preferred for solving ODEs in cloud microphysics models. The Analytical Predictor of Condensation (APC) scheme is an efficient implicit method for solving this system (Jacobson, 1997a, 2002). To implement this method, we first discretize Equations (B.8) and (B.12) using variables at the future time step $t + \Delta t$ and the current time step t , as follows:

$$\frac{c_{q,i,t+\Delta t} - c_{q,i,t}}{\Delta t} = k_{q,i}(C_{q,t+\Delta t} - S'_{q,i}C_{q,s,t}), \quad (\text{B.13})$$

$$\frac{C_{q,t+\Delta t} - C_{q,t}}{\Delta t} = - \sum_{i=1}^{N_B} [k_{q,i}(C_{q,t+\Delta t} - S'_{q,i}C_{q,s,t})]. \quad (\text{B.14})$$

The solution of Equation (B.13) can be derived by time integration as follows:

$$c_{q,i,t+\Delta t} = c_{q,i,t} + \Delta t k_{q,i}(C_{q,t+\Delta t} - S'_{q,i}C_{q,s,t}). \quad (\text{B.15})$$

To derive the concentration of gas phase C_q , we use the following relationship for mass conservation:

$$C_{q,t+\Delta t} + \sum_{i=1}^{N_B} c_{q,i,t+\Delta t} = C_{q,t} + \sum_{i=1}^{N_B} c_{q,i,t} = C_{\text{tot}}. \quad (\text{B.16})$$

By substituting Equation (B.15) into (B.16), we get the following expression for the gas concentration at the advanced time step:

$$C_{q,t+\Delta t} = \frac{C_{q,t} + \Delta t \sum_{i=1}^{N_B} k_{q,i} S'_{q,i} C_{q,s,t}}{1 + \Delta t \sum_{i=1}^{N_B} k_{q,i}}. \quad (\text{B.17})$$

We must account for scenarios where gas or condensed phase concentrations drop below zero. To prevent these non-physical results, we impose a limit on the gas concentration defined as $C_{q,t+\Delta t} = \min(C_{q,t}, C_{\text{tot}})$. This threshold is substituted into Equation (B.15) to derive the final aerosol concentration. Furthermore, because Equation (B.15) may also yield negative values, we apply two additional constraints to the condensed phase concentration as follows:

$$c_{q,i,t+\Delta t} = \max(c_{q,i,t+\Delta t}, 0), \quad (\text{B.18})$$

$$c_{q,i,t+\Delta t} = \frac{C_{q,t+\Delta t} - C_{q,t} + \sum_{i=1}^{N_B} \max(c_{q,i,t} - c_{q,i,t+\Delta t}, 0)}{\sum_{i=1}^{N_B} \max(c_{q,i,t+\Delta t} - c_{q,i,t}, 0)} (c_{q,i,t+\Delta t} - c_{q,i,t}). \quad (\text{B.19})$$

The first limit states that the concentration of the condensed phase must not fall below zero. The second limit is calculated after solving the first limit for all size bins and serves as the correction for the condensed phase in cases where the first limit was operated. Since these limits avoid concentrations from falling below zero in any condition, the APC scheme is unconditionally stable.

Particle volumes are updated using Equation (B.9) after applying the APC scheme. Since the APC scheme calculates growth based on the mass flux to individual particles, it demonstrates high compatibility with Lagrangian bin schemes, including both moving-center and full-moving approaches.

This scheme can be extended to treat any number of aerosol size distributions simultaneously, which can be highly valuable for planetary aerosol systems where more than two condensational gases coexist in the atmosphere.

Appendix C

Examination of Numerical Artifact Caused by the Moving-Center Scheme

Figure C.1 presents the results of 0-D simulations performed with different bin resolutions ranging from $N_B = 45$ to $N_B = 360$, using the same calculation settings as those described in Section 3.2.1 of the main text. When the resolution is lower, the evaporation occurs intermittently. As the resolution is increased, the particle evaporation proceeds in a smoother, more continuous manner, which is consistent with natural systems.

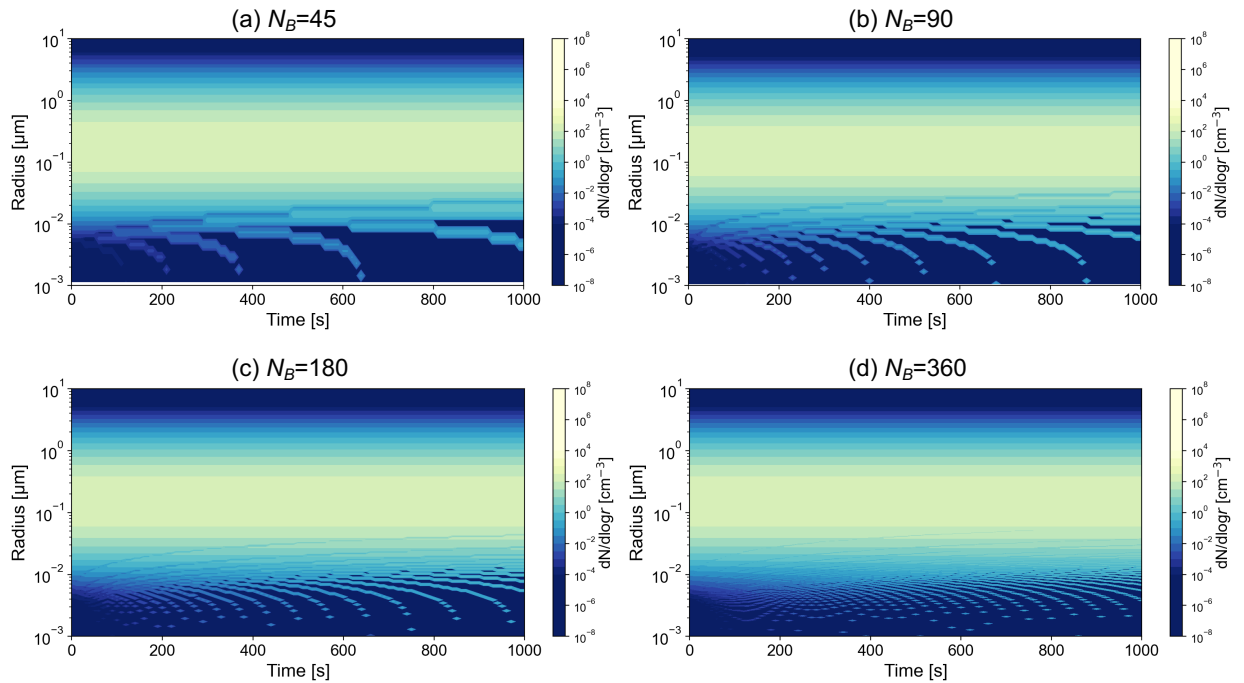


Figure C.1: Aerosol size distribution as a function of time and radius at 60 km with different bin resolution (a) $N_B = 45$, (b) $N_B = 90$, (c) $N_B = 180$, (d) $N_B = 360$.

Figures C.2–C.5 show snapshots taken every 1000 seconds during the last 9000 seconds of the simulation. The size distribution remains stable in the steady state, with minimal pit formation. We focus on altitudes of 54, 48, 47, and 46 km, where condensation peaks near 54 km (main cloud deck) and evaporation intensifies near the cloud base (48–46 km). At 54 and 48 km, occasional pits appear due to the bin scheme’s discreteness, but these have little impact on the overall size distribution. At 47 km (the cloud bottom), the distribution evolves dynamically as particles evaporate and pits form. Although the distribution appears irregular, this is expected in the transition region. Despite this, the overall size distribution remains consistent over time, and the pits do not amplify or propagate into adjacent grid cells. At 46 km, below the cloud base, particles fully evaporate and the size distribution disappears. This steady pattern persists over time.

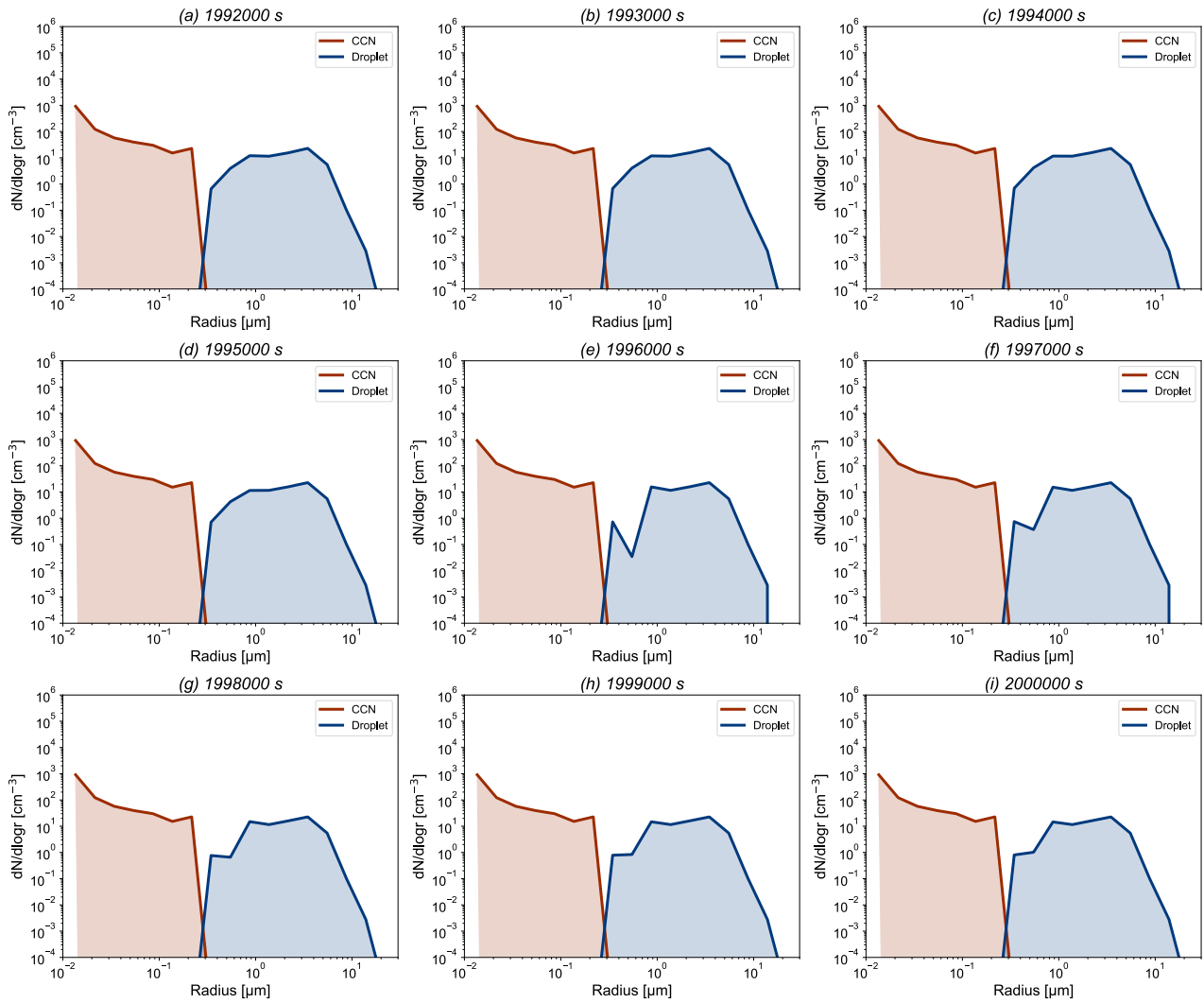


Figure C.2: Time evolution of the size distribution at 54 km. The simulation output is plotted over the final 9000 seconds with an interval of 1000 seconds.

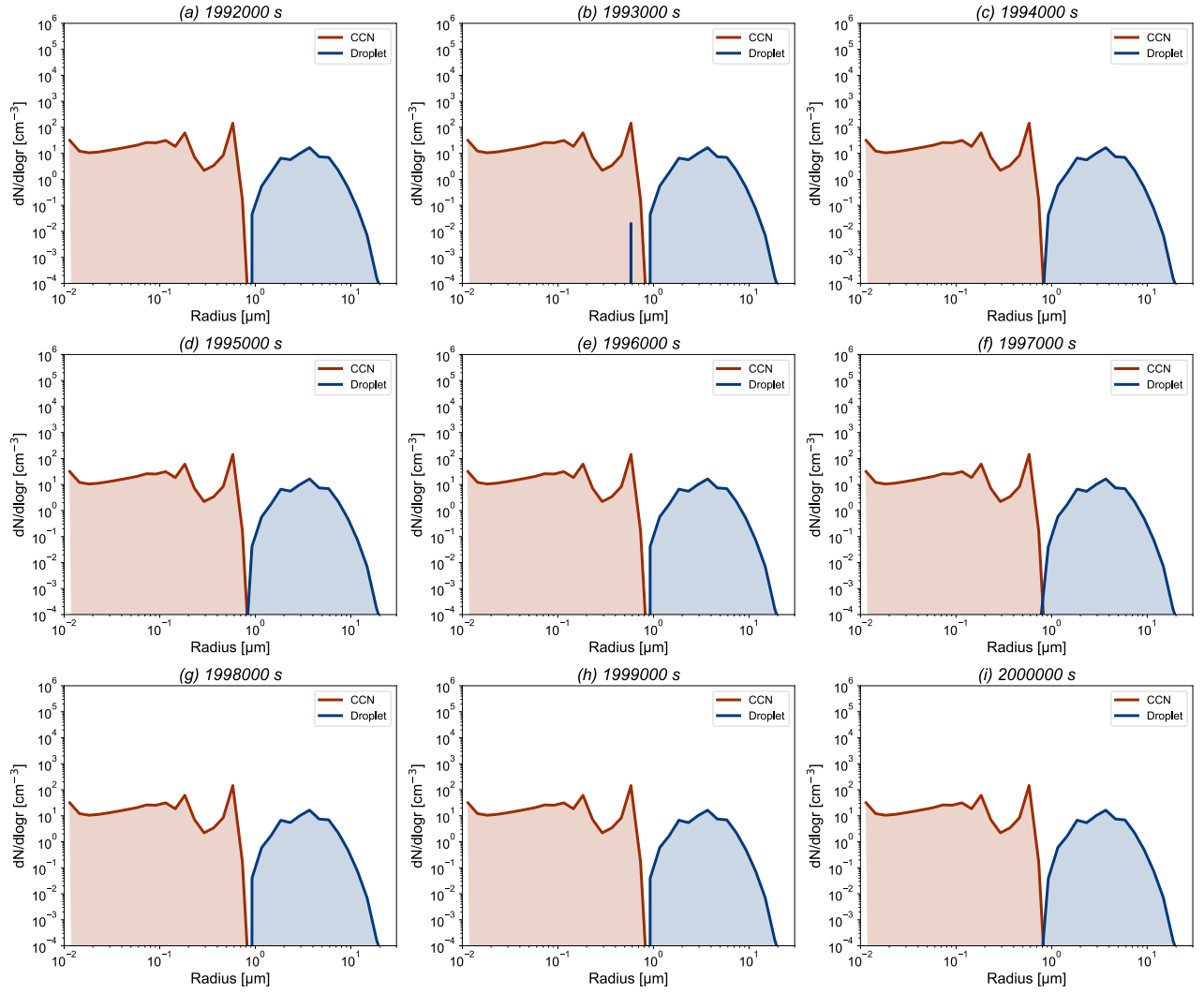


Figure C.3: Time evolution of the size distribution at 48 km. The simulation output is plotted over the final 9000 seconds with an interval of 1000 seconds.

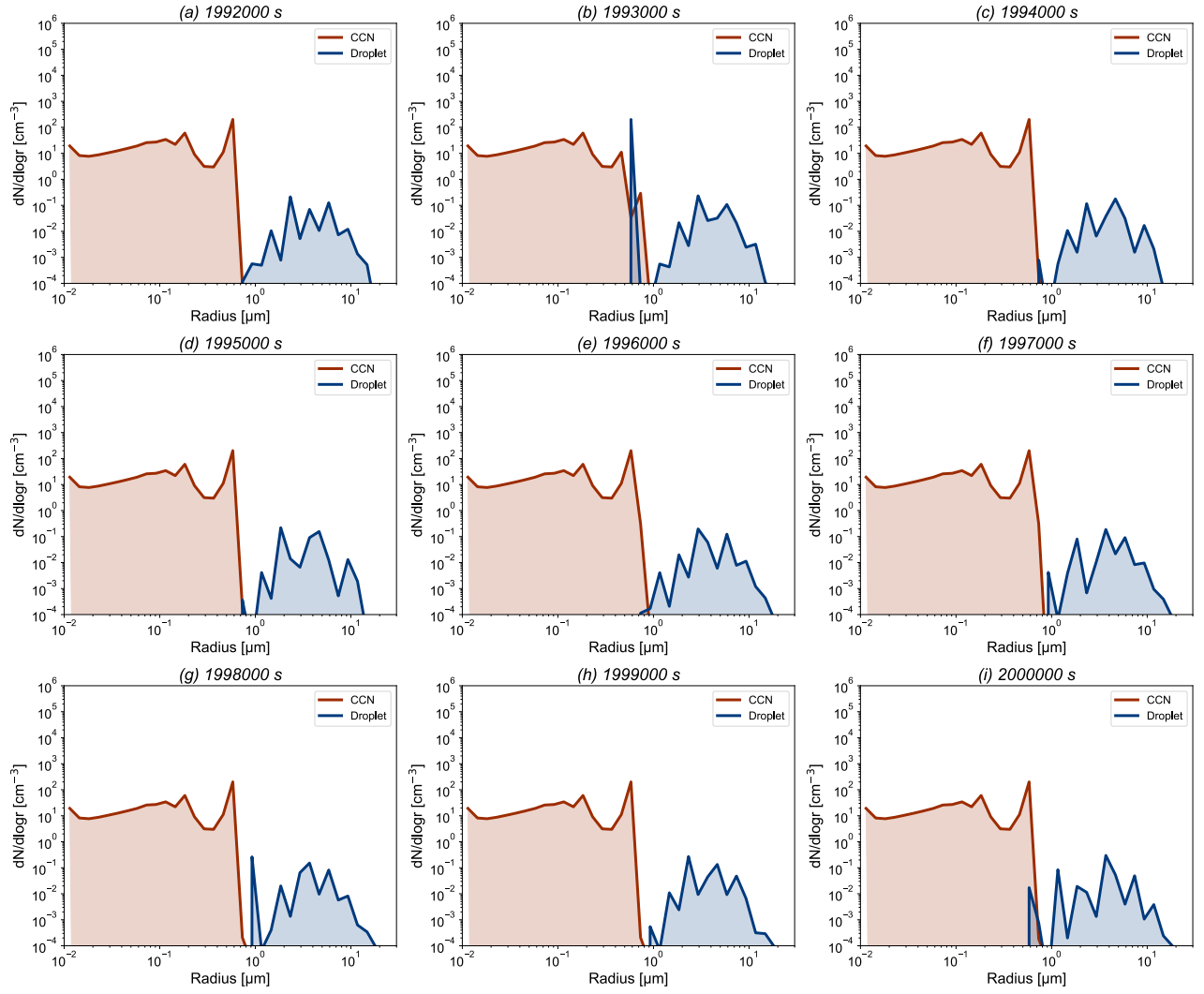


Figure C.4: Time evolution of the size distribution at 47 km. The simulation output is plotted over the final 9000 seconds with an interval of 1000 seconds.

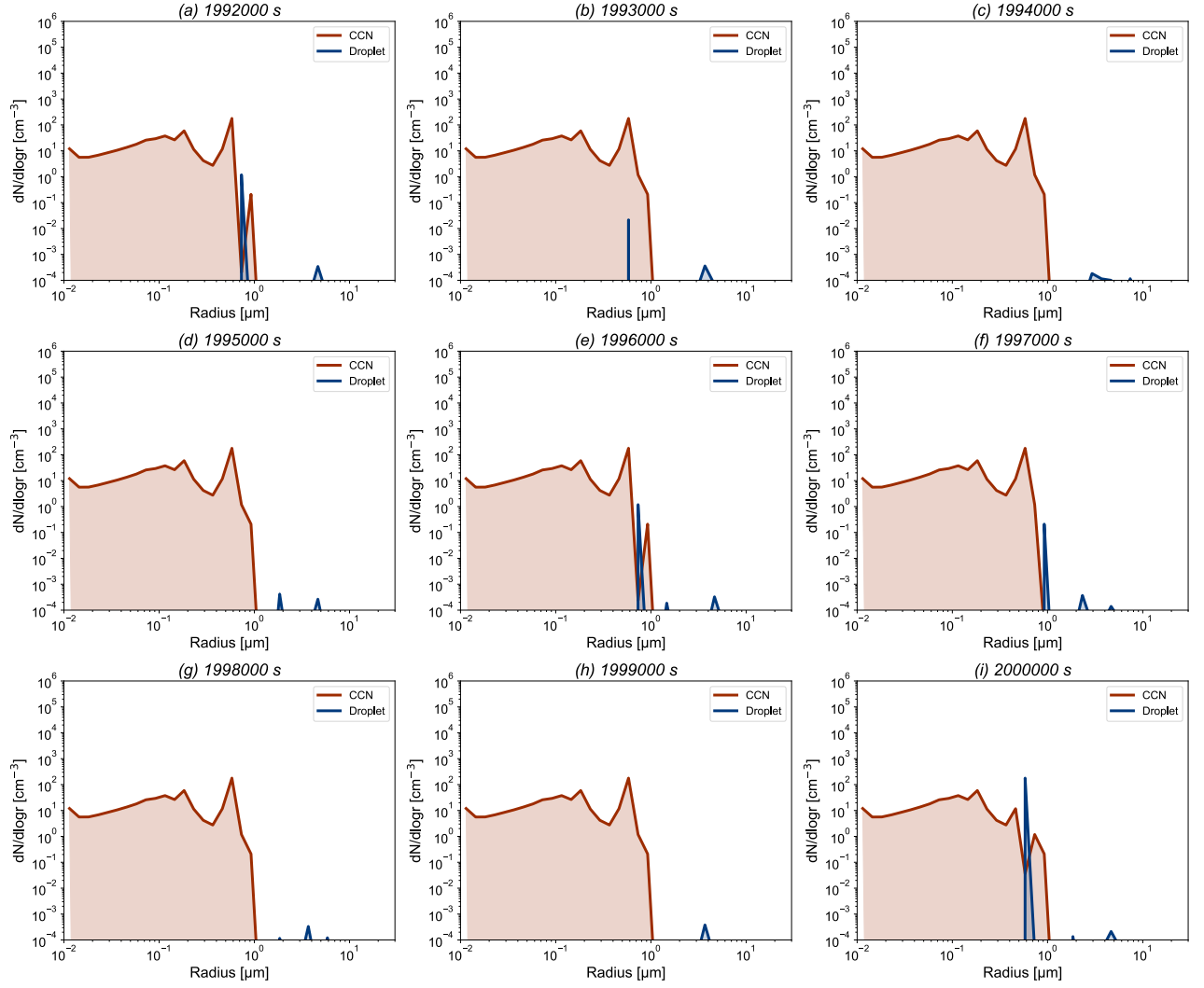


Figure C.5: Time evolution of the size distribution at 46 km. The simulation output is plotted over the final 9000 seconds with an interval of 1000 seconds.

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