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# **Geophysical Research Letters**



#### RESEARCH LETTER

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#### **Key Points:**

- · Estimation for the first time of the spatial and temporal variability of chemical species due to vertical mixing
- Quantification of the vertical eddy diffusion coefficient, order of magnitude above typical used values
- Cloud-top altitudes change by 0.2– 1 km due to vertical convective mixing and gravity waves

#### **Supporting Information:**

Supporting Information may be found in the online version of this article.

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# Impact of the Turbulent Vertical Mixing on Chemical and **Cloud Species in the Venus Cloud Layer** Maxence Lefèvre , Franck Lefèvre , Emmanuel Marcq , Anni Määttänen,

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**Abstract** The Venusian atmosphere hosts a 10 km deep convective layer that has been studied by various spacecrafts. However, the impact of the strong vertical mixing on the chemistry of this region is still unknown. This study presents the first realistic coupling between resolved small-scale turbulence and a chemical network. The resulting vertical mixing is different for each species; those with longer chemical timescales will tend to be well-mixed. Vertical eddy diffusion due to resolved convection motions was estimated, ranging from 10<sup>2</sup> to 10<sup>4</sup> m<sup>2</sup>/s for the 48–55 km convective layer, several orders of magnitude above the typically used value. In the 48-55 km convective layer, the impact of the small-scale turbulence on the cloud layer boundaries was between 200 m and 1 km. The impact of turbulence on cloud chemistry is consistent with Venus Express/Visible and Infrared Thermal Imaging Spectrometer observations. The observability at the cloud-top of small-scale turbulence by VenSpec-U spectrometer would be challenging.

Plain Language Summary Venus hosts a global sulfuric acid cloud layer between 45 and 70 km. A convective layer is present between roughly 50 and 60 km, with its variability in latitude and local time assessed by observation, with a thicker layer at high latitude and at night. One question that remains unclear is how this turbulence mixes momentum, heat, and chemical species. Especially, the impact of the strong vertical mixing on the chemistry of this region is still unknown. To investigate this topic, we use a convection-resolving model coupled for the first time with a realistic chemical network. The resulting vertical mixing is different for each species: those with longer chemical timescales will tend to be well-mixed. 1D and global circulation models use the so-called vertical eddy diffusion approach to represent turbulent motion, quantified in our model and underestimated in chemistry models. The small-scale turbulence in the cloud layer causes a variation in the altitude of the top and bottom boundaries of the cloud. Our model shows that the impact of turbulence on cloud chemistry corresponds well to what has been observed by satellites. In the future, the EnVision mission will be able to observe chemical species at the small turbulence scales.

#### 1. Introduction

The Venusian cloud layer hosts a convective activity that has been assessed since the beginning of Venus spacecraft exploration (Belton et al., 1976; Rossow et al., 1980). Various Radio occultation experiments studied convective activity (Hinson & Jenkins, 1995; Seiff et al., 1980), measuring a convective layer between 50 and 55 km of altitude. Venus Express and Akatsuki observed this turbulent layer, measuring a strong latitudinal (Tellmann et al., 2009) and local time variability (Imamura et al., 2017) of the depth of the layer. In addition to the convection layer in the deep cloud layer, the venus monitoring camera observed at 365 nm cellular features at the top of the cloud at about 70 km of altitude above the subsolar point, suggesting convective activity (Markiewicz et al., 2007; Titov et al., 2012). A convective layer at this altitude is the main hypothesis for these observed

Regarding Venus cloud chemistry, several 1D models have been developed. The models of Yung et al. (2009), Krasnopolsky (2012), Bierson and Zhang (2020), and Rimmer et al. (2021) used a vertical eddy diffusion value that underrepresents the turbulent activity in the cloud layer (M. Lefèvre et al., 2022), and were not able to reproduce the observed vertical gradient of SO2 in the clouds. The interaction between the turbulence and chemistry has been little studied, and is therefore poorly known. Only McGouldrick and Toon (2008) gave an insight into the change of optical depth due to the convection and gravity waves, using an idealized 2D (zonal/ vertical) representation of the Venus cloud convective layer.

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In order to investigate the turbulent activity inside the Venus cloud layer, we use the limited-area Venus mesoscale model (VMM) large-eddy simulation (LES) mode (M. Lefèvre et al., 2022) adapted from a terrestrial hydrodynamical solver (Skamarock & Klemp, 2008) to simulate only a specified region of the planet at a fine resolution. A complete photochemical scheme (Stolzenbach et al., 2023; Streel et al., 2023) was coupled to the VMM. For the first time in Venusian atmospheric studies, a chemical network is coupled to a 3D convection resolving model, allowing the study of small-scale spatio-temporal variations. We will focus on the main active chemical species present in the cloud: SO<sub>2</sub>, SO, H<sub>2</sub>O and H<sub>2</sub>SO<sub>4</sub> (the latter two in both liquid and gaseous forms).

## 2. Modeling

#### 2.1. IPSL Venus LES Model

Our mesoscale model for Venus is based on the dynamical core of the Advanced Research Weather-Weather Research and Forecast (hereinafter referred to as weather research and forecast) terrestrial model (Skamarock & Klemp, 2008). Alongside solar and infrared heating rates calculated by the Institut Pierre-Simon Laplace (IPSL) Venus planetary climate model (PCM) radiative transfer (Lebonnois et al., 2015) using the cloud model is based on Haus et al. (2014, 2015), a third additional rate representing the heat from the large-scale circulation is imposed. The details of this last rate are described in detail in M. Lefèvre et al. (2017).

#### 2.2. IPSL Photochemistry Model

The LES model is coupled to a photochemistry model described in detail in Stolzenbach et al. (2023). This photochemical model represents the comprehensive chemistries of  $CO_2$  cycle, the sulfur oxidation cycle, and the chlorine cycle for neutral species from 35 to 100 km, meaning from  $6 \cdot 10^5$  Pa–1 Pa and from 460 to 160 K. In addition, a nitrogen photochemical network has been included into the chemistry model (Streel et al., 2023), with the addition of the four nitrogenous species N, N( $^{2D}$ ), NO, and NO<sub>2</sub>. Our chemical network takes into account the 38 species in gas-phase and two liquid-phase species  $H_2SO_4$  and  $H_2O$ , interacting through 123 chemical reactions.

In Stolzenbach et al. (2023), the photolysis calculation was performed off-line with a lookup table that was a function of the  $CO_2$  column,  $SO_2$  column, and solar zenith angle. Currently, following the same strategy as in the Mars version of the PCM (F. Lefèvre, Trokhimovskiy, et al., 2021), photolysis rates for  $CO_2$ ,  $SO_2$ ,  $H_2$  and nitrogen species are calculated on-line.

The vertical profile of sulfur species, especially  $SO_2$ , is an issue that is ubiquitous in Venus chemistry modeling (Bierson & Zhang, 2020; Rimmer et al., 2021; Stolzenbach et al., 2023). The goal of this study is not to reproduce quantitative  $SO_2$  cloud-top abundances, the question of vertical gradient of  $SO_2$  into the clouds being beyond the scope of this paper, but to quantify variabilities due to the interaction between small-scale turbulence and chemical species.

#### 2.3. Microphysics Model

The representation of clouds in the model is based on the hypothesis that clouds are at all times in a state of thermodynamic equilibrium: the condensation/evaporation of  $H_2SO_4$  and  $H_2O$  follows the saturation vapor pressure profile over the calculated equilibrium  $H_2SO_4$  aqueous solution. Clouds in the model are supposed to be at all times in a thermodynamic equilibrium: the condensation/evaporation of  $H_2SO_4$  and  $H_2O$  follow the saturation vapor pressure profile over the calculated equilibrium  $H_2SO_4$  aqueous solution. The partitioning of  $H_2SO_4$  and  $H_2O$  between the gas and liquid phases is calculated at each call to the chemistry subroutine. The cloud particle size distributions for the three modes implemented are prescribed and are the same as in Table 5 from Stolzenbach et al. (2023). A sedimentation flux is also taken into account, based on the Stokes velocity for each particle mode, described in detail in Stolzenbach et al. (2023).

#### 2.4. Simulation Settings

This study is focused on two latitudinal and two local time cases: the Equator and  $75^{\circ}$  at noon and midnight to capture variabilities due to convection activity variation (M. Lefèvre et al., 2018). The M. Lefèvre et al. (2018)'s equilibrium state at the Equator and  $75^{\circ}$  at noon and midnight are used as the initial state with the same resolution and time-step, that is, 400 m over the horizontal domain size of 60 km and 1 s. To ensure that the chemistry is represented adequately in the case of strong vertical wind speed, the photochemistry scheme is called every 10 s,

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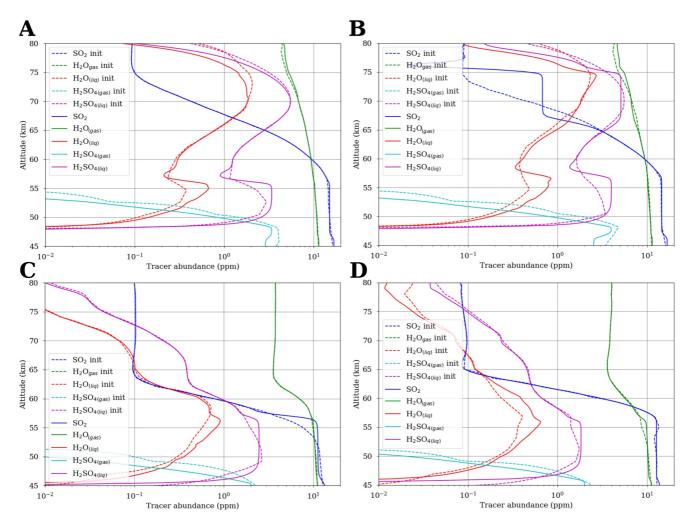


Figure 1. Domain averaged tracer abundance vertical profiles of  $SO_2$  (blue lines),  $H_2O$  in the gas (green lines) and liquid phase (red lines), and  $H_2SO_4$  in the gas (cyan lines) and liquid phase (magenta lines) at the Equator at midnight (a), noon (b), and at  $75^{\circ}$  of latitude at midnight (c) and noon (d). The solid lines represent the model outputs and the dotted lines the initial profile.

which is consistent with characteristic times of microphysical processes (James et al., 1997). Vertical profiles from Stolzenbach et al. (2023) are used as input for the chemistry scheme.

## 3. Vertical Mixing

## 3.1. Global Averaged Results

In the four panels of Figure 1, the convective layer is visible between roughly 48 and 55 km with the low vertical gradient meaning a strong vertical mixing, different for each species. For example, in this region, the vertical mixing ratio gradient of  $SO_2$  is very low, whereas it is slightly larger for water. In M. Lefèvre et al. (2022), the vertical gradient due to mixing was dependent on the ratio between the chemical timescale and the convective layer dynamical timescale of  $1.7 \cdot 10^4$  s. However, both  $SO_2$  and  $H_2O$  vapor have long photochemical timescales, 10 Earth years below 55 km and 2 months above 55 km for  $SO_2$ , and about a decade for  $H_2O$  (all altitudes) (Stolzenbach, 2016). The difference between these two species is that water vapor can condense into droplets. The condensation timescale is around 10 s. The actual lifetime of water vapor is therefore much smaller than its photochemical timescale, leading to a weaker vertical mixing. Above the convective layer, both  $SO_2$  and  $H_2O$  vapor decrease strongly.

The cloud top convective layer is also visible at the Equator at noon (Figure 1-B), with a strong vertical mixing between 67 and 75 km. Like for the 48–55 km convective layer, the effect of vertical mixing is different for each

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species. SO<sub>2</sub> exhibits a weak vertical gradient, whereas for water this gradient is stronger due to condensation. At cloud-top altitudes, around 72 km, SO<sub>2</sub> mixing ratio at noon increases by a factor of four by convective mixing, going from 0.17 ppm at the initial state to 0.7 ppm. For the three other cases, gravity waves are present at cloud-top levels. Contrary to convective motions, these waves, emitted by the convection, generate a much smaller mixing for all species.

Another noticeable feature is the fact that the mixing ratio of condensed species tend to increase in the  $48-55 \,\mathrm{km}$  convective layer, conjugated with the decrease of the condensing vapor below the clouds. The convective motion will transport the condensable species, mainly  $\mathrm{H_2SO_4}$ , from the below the clouds where the abundance is higher to the middle cloud region. The two vapors get depleted below  $48 \,\mathrm{km}$ , whereas the excess condenses in the lower and middle cloud, increasing the opacity of the clouds. This effect is weaker at high latitude due to lower temperatures affecting condensation. The large particles of the mode 3 represent the main source of cloud opacity. With mass coefficients for the different cloud droplet modes (Stolzenbach et al., 2023), the mass column density was calculated (Figure S1 in Supporting Information S1). The mass column at the Equator is 30% larger than the PCM column due to the resolved convection at both local times. If separated between updrafts and downdraft, the difference in the mass column is about 10%. At high latitude, the average increase due to convection is only of 15% compared to the PCM. This difference is due to the condensable reservoir that is much lower at high latitude because of the Hadley Cell circulation (Stolzenbach et al., 2023).

#### 3.2. Effective Vertical Eddy Diffusion

In order to account for the subgrid scale turbulence, 1D models and 3D GCMs use a diffusion equation as a parametrization, with the vertical eddy diffusion coefficient (or  $K_{zz}$ ) controlling the strength of the mixing processes.

In the Venusian atmosphere, the  $K_{zz}$  parameter has been estimated both from observations and modeling, but is still not well-constrained. The *Pioneer Venus* radio scintillation measurements estimated  $K_{zz}$  between  $2 \cdot 10^{-1}$  m<sup>2</sup> s<sup>-1</sup> at 45 km (Woo et al., 1982) and 4 m<sup>2</sup> s<sup>-1</sup> at 60 km (Woo & Ishimaru, 1981). Using the values of the vertical wind measured in the deep cloud layer by the *VeGa* balloons (Sagdeev et al., 1986),  $K_{zz}$  was evaluated at  $10^3$  m<sup>2</sup> s<sup>-1</sup> at 54 km (Blamont et al., 1986).  $K_{zz}$  was estimated with the vertical profile of  $H_2SO_4$  vapor obtained by Venus Express (VEX)/VeRa radio-occultation between  $10^4$  and  $10^5$  m<sup>2</sup> s<sup>-1</sup> (Dai et al., 2023). Karyu et al. (2024) estimated  $K_{zz}$  around 2 m<sup>2</sup> s<sup>-1</sup> between 60 and 70 km with a 1D chemical model fitting Venus Express observations.

To quantify the vertical mixing of tracers, we define an effective eddy diffusion coefficient  $K_{zef}$  where the interaction of the resolved turbulence, like convection, and chemical processes are taken into account. This coefficient  $K_{zef}$  is calculated as follows:

$$K_{zef} = -\frac{\langle q'w' \rangle}{\partial \langle q \rangle / \partial z} \tag{1}$$

with q a tracer mixing ratio, primed quantities representing perturbations relative to the domain averaged values, and bracket quantities representing domain averaged values. The vertical wind field w' will be the same for each species, but chemical timescale is specific, leading to different q'.

As discussed in the previous section, the effect of vertical mixing is different for each species, and this is clearly seen in the  $K_{zef}$  values shown in Figure 2. For example,  $SO_2$  and  $H_2O$  have different vertical gradients illustrating the difference in the mixing strength, and thus they have different  $K_{zef}$  values, around  $10^3$  m<sup>2</sup> s<sup>-1</sup> for  $SO_2$  and  $10^2$  m<sup>2</sup> s<sup>-1</sup> for  $H_2O$ . The values for the 48–55 km convective layer are quite constant between night and day. Above the convective layer, the turbulence is due mainly to small-scale gravity waves that engender a weak mixing, consistent with the Woo and Ishimaru (1981) measurements. The variability of the diurnal 48–55 km convective activity is visible, with  $K_{zef}$  stronger at night by a factor of two. At 75° degrees of latitude, the vertical mixing is stronger in the convective region by almost a factor 10, with  $K_{zef}$  reaching  $10^3$  m<sup>2</sup> s<sup>-1</sup>. Both below and above the convective layer, gravity waves are imposing a weaker vertical mixing, with values  $10^{-1}$  and 10 m<sup>2</sup> s<sup>-1</sup>. The values inside the 48–55 km convective layer are consistent with in situ measurements (Blamont et al., 1986; Woo et al., 1982; Woo & Ishimaru, 1981). At the Equator at noon, the presence of the cloud-top convective layer

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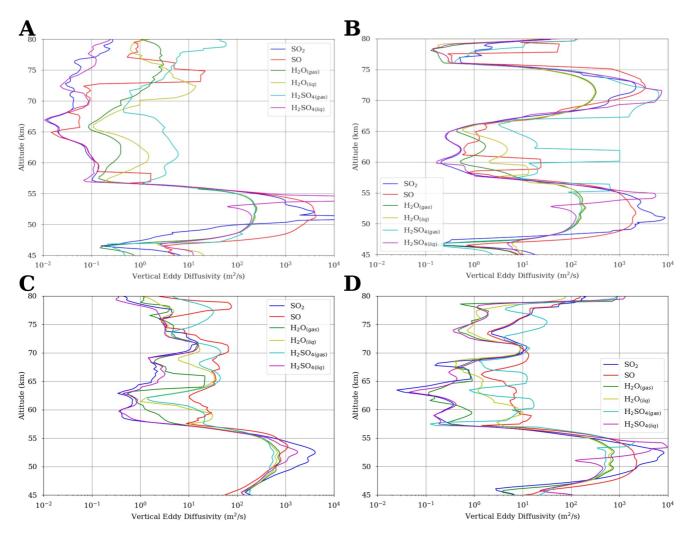


Figure 2. Vertical profiles of the vertical eddy diffusivity ( $m^2 s^{-1}$ ) in the Venus cloud region calculated for SO<sub>2</sub> (blue lines), H<sub>2</sub>O in the gas (green lines) and liquid phase (red lines), and H<sub>2</sub>SO<sub>4</sub> in the gas (cyan lines) and liquid phase (magenta lines) at the Equator at midnight (left) and noon (right). For clarity, the vertical profiles are smoothed. Original profiles are available in Figure S2 in Supporting Information S1.

is visible with an increase of  $K_{zef}$  between 66 and 75 km, from 1 m<sup>2</sup> s<sup>-1</sup> at 65 km to around  $10^4$  m<sup>2</sup> s<sup>-1</sup> at 72 km. These cloud-top values of  $K_{zef}$  are slightly larger than the value for the 48–55 km convective layer. The  $H_2SO_4$  vapor in the 48–55 km convective layer has a  $K_{zef}$  between  $10^2$  and  $10^3$  m<sup>2</sup> s<sup>-1</sup>, more than one order of magnitude lower than the highest estimation from radio-occultations (Dai et al., 2023). Outside the cloud-top convective layer at night, the values of  $K_{zef}$  are consistent with estimation from the modeling of Karyu et al. (2024). Inside the cloud-top convective layer, the value from the resolved convective layer is several order of magnitude higher.

Regarding the vertical mixing parametrization, the value of  $K_{zz}$  in the cloud region is often considered as a tunable parameter. The Krasnopolsky (2012) 1D model uses an eddy diffusion of 1 m<sup>2</sup> s<sup>-1</sup> in the convective layer, whereas Bierson and Zhang (2020) defined several eddy diffusion scenarios for the convective layer mixing parametrization, from 0.1 to 2 m<sup>2</sup> s<sup>-1</sup>. Rimmer et al. (2021) also tested a range of vertical eddy diffusion values in the convective layer, between 0.01 and 1 m<sup>2</sup> s<sup>-1</sup>. These values are lower by several orders of magnitude compared to the turbulent vertical mixing we obtained with the coupled LES-chemistry model in this work, and thus are inconsistent with the present study and the in situ measurements by Blamont et al. (1986), Woo et al. (1982), and Woo and Ishimaru (1981).

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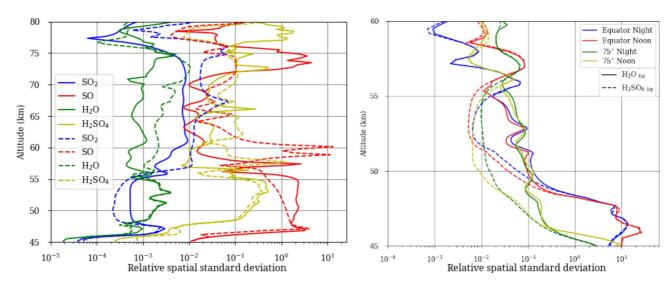


Figure 3. Left: Vertical profiles of the relative standard deviation for  $SO_2$ , SO,  $H_2O$  and  $H_2SO_4$  in gas phase at the equator at night (solid line) and noon (dashed line). Right: Vertical profiles of the relative standard deviation for the condensed phase of  $H_2SO_4$  and  $H_2O$  at the equator (solid line) and 75° (dashed line) for the studied local time.

### 4. Spatial and Temporal Variability

In the 48–55 km convective layer, the spatial standard deviation is larger at night, with more vivid convective motions. At higher altitudes, the standard deviation is again larger in the cloud-top convective layer and due to the trapped gravity waves that have a large amplitude.

 $SO_2$  exhibits the lowest standard deviation in the 48–55 km convective layer due to a very strong vertical mixing seen in Figure 1. On the contrary, SO exhibits the highest spatial variabilities. At cloud-top altitudes, SO has a lifetime of about 30 min due to quick photolysis processes. This lifetime is of the same order of magnitude as the dynamical timescale of the cloud-top convection, leading to high spatial variability (M. Lefèvre, Trokhimovskiy, et al., 2021). In the 48–55 km convective layer, the lifetime of SO is much longer due to the lack of photons at these altitudes, but the vertical gradient is strong, going at the Equator from  $10^{-10}$  ppm at 48 km to  $10^{-4}$  ppm at 55 km at night and from  $10^{-5}$  ppm at 48 km to  $10^{-3}$  ppm at 55 km at noon. There will be SO-poor ascending plumes and SO-rich descending plumes, leading to a high spatial variability due to convective mixing.

The spatial variability for the condensed species is shown in Figure 3-right. The variability is higher at lower latitudes due to the Hadley circulation that generates a strong latitudinal gradient for the gas phase of  $H_2O$  and  $H_2SO_4$ . The variability is also stronger for condensed water above the 48–55 km convection layer because of higher mixing ratio of water at this altitude range. The higher variability of the condensed species inside the 48–55 km exhibits the strong non-linearity of the condensation processes.

The dynamical timescale of the deep and cloud-top convective layer can be calculated as  $\tau_{dyn} = H/\sigma_w$ , with H being the scale height and  $\sigma_w$  the spatial standard deviation of the vertical wind at a given altitude. M. Lefèvre et al. (2022) estimated this timescale to be 17,000 s at 50 km and 6,000 s at 70 km. Coupled with the zonal wind flow, using high-frequency over several hours, the temporal variability timescale of chemical species is approximately 20 min inside the 48–55 km convective layer with an Eulerian perspective.

#### 5. Impact on Cloud Boundary Altitudes

Convective layers are present at the bottom of the cloud layer, both at night and day, and at cloud-top altitudes at the subsolar point only, and can therefore impact altitudes of the cloud boundaries. For the bottom altitude, it is pretty straightforward to define, since the abundance of condensed species drops drastically to zero below the  $H_2SO_4/H_2O$  saturation level. However, for the cloud top, it is more subtle as the condensed species abundance decreases more gradually with increasing height. In order to calculate the cloud top elevation  $Z_{top}$ , we resolve the following equation:  $\int_{Z_{top}}^{\infty} x(z) \rho_{gas}(z) dz = M_0$  where  $M_0$  is the condensed species mass column density yielding an optical depth equal to one at 250 nm, x the mass mixing ratio of condensed species, and  $\rho_{gas}$  the density of the gas.

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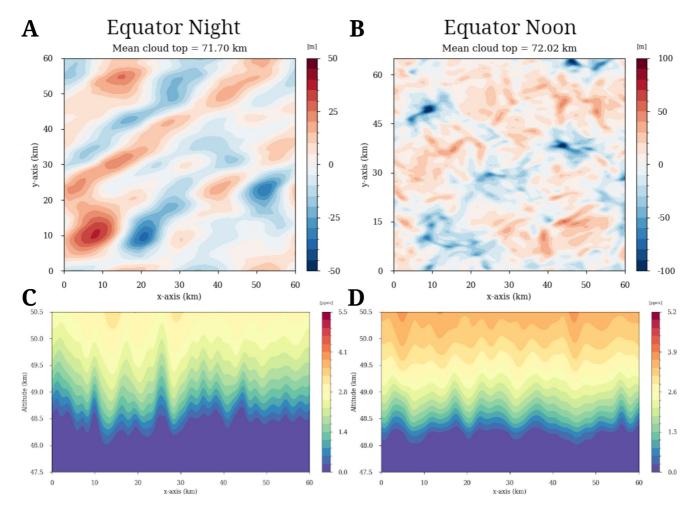


Figure 4. Top: Instantaneous cloud top altitude at the equator at midnight (left) and noon (right). Bottom: Instantaneous vertical cross-sections of gaseous  $H_2SO_4$  at the equator at midnight (left) and noon (right).

For cloud top altitudes, using the radiative transfer model from Marcq et al. (2020),  $M_0$  is equal to 0.29 pr- $\mu$ m at 250 nm.

There is only a limited diurnal variability of cloud-top and cloud-bottom altitudes, around 71 and 48 km respectively. At  $75^{\circ}$  (not shown), the cloud top is reaching altitudes of 61 km and the cloud bottom is around 45 km. These values are consistent with observations (Barstow et al., 2012; Ignatiev et al., 2009). However, at the cloud top there is a strong difference of horizontal structure of the cloud-top altitude. At the Equator at noon, the subsolar convective layer generates a displacement of the cloud of around 200 m, with  $\sim$ 20 km diameter convective updraft cells with narrow downdrafts. For the other cases, gravity waves at night induce a much smaller cloud-top variability visible in Figure 4-A, no larger than 100 m, with the 30 km horizontal wavelength waves visible (Peralta et al., 2008; Piccialli et al., 2014).

At the cloud bottom, the convective layer is present at all local times and latitudes but is slightly more intense at night and at high latitudes (M. Lefèvre et al., 2018), leading to a stronger vertical variability of the cloud bottom. At the Equator, the amplitude of the variations is around 600 m at night against 300 m at noon, with structures of 5 km wide. At 75° (not show), with a stronger convective layer, the amplitude of the variation of the cloud bottom reaches 600 m at noon against almost a 1 km at night, with horizontal structure over 10 km wide.

#### 6. Discussion

VeGa 1 and VeGa 2 measurements of SO<sub>2</sub> (Bertaux et al., 1996) and H<sub>2</sub>O vapor (Surkov et al., 1986) in lower clouds are the only ones with a high vertical resolution. Inside the convective layer, SO<sub>2</sub> exhibits strong and

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opposite vertical gradients and values that differ by a factor of three, incompatible with a well-mixed species. For  $H_2O$  vapor, a small vertical gradient between 50 and 60 km was observed, compatible with the presence of a convective layer, but the mixing ratio measured is several orders of magnitude higher than measurements obtained later from remote sensing orbiters (Marcq et al., 2023).

The large probe cloud particle size spectrometer experiment on Pioneer Venus (Knollenberg et al., 1980) and VENERA 9, 10, and 11 (Marov et al., 1980) measured cloud microphysical properties, such as extinction coefficient, scattering coefficient, backscattering, and number density with high vertical sampling. The probes showed that these properties are almost constant in the middle cloud region, approximately between 51 and 57 km. This corresponds to the region of the well-mixed liquid phase of H<sub>2</sub>SO<sub>4</sub>, suggesting that the middle cloud is strongly influenced by vertical mixing due to convection.

Ground telescopes have observed local time variations of  $SO_2$  faster than few hours (Encrenaz et al., 2013, 2016), with an increase of a factor 5, indicating convective activity. The local injection of  $SO_2$  by cloud-top convection is one of the main hypotheses for the long-term variations of  $SO_2$  at cloud top (Marcq et al., 2013, 2020). The observed plumes are located near the Equator but are not increasingly present around noon (Encrenaz et al., 2019), therefore not consistent with the cloud-top convection presented here. In Figure 1, the vertical mixing at the subsolar point can increase the  $SO_2$  mixing ratio by a factor 4. A study with a global model is necessary to test this hypothesis.

VEX/Visible and Infrared Thermal Imaging Spectrometer (VIRTIS) measured  $H_2O$  vapor inside the convective-like features at the subsolar point (Cottini et al., 2012), showing a variability inferior to 1 ppm and a variability of the cloud-top altitude around 500 m. Our model exhibits a variability of 0.3 ppm and a cloud-top variability around 200 m. The model underestimates these variabilities, but the result is consistent with convective cells at the lower edge of their estimated diameter (Markiewicz et al., 2007). The convection-induced cloud-top altitude variability is consistent with the Akatsuki measurement for the convective-like cells (Sato et al., 2020). Short-scale spatial and temporal (inferior to the 4 days atmospheric rotation rate) variability of  $SO_2$  column density was observed with SPICAV (Vandaele et al., 2017), suggesting of turbulence activity. Radio occultation onboard Venus Express also observed gaseous  $H_2SO_4$  (Oschlisniok et al., 2021), with variability larger than the one predicted by the model that cannot be accounted for turbulent processes. Oschlisniok et al. (2021) also reported radio occultation measurements of  $SO_2$  but the spatial and temporal resolution is too coarse to be able to compare it with our model outputs. VIRTIS measured middle and lower cloud features, and a larger variability was observed at low latitudes (McGouldrick et al., 2008), an associated timescale of 30 hr corresponding to an eddy diffusion of 250 m<sup>2</sup> s<sup>-1</sup> (McGouldrick et al., 2012). These two observations are consistent with the resolved variability and vertical mixing of cloud droplets in the convective layer (Figure 3-right and Figure 2).

NASA's VERITAS and DAVINCI and ESA's EnVision are future space missions that will study cloud chemistry. The DAVINCI mission (Garvin et al., 2022) will use the mass spectrometer VMS and the laser Spectrometer Venus Tunable Laser Spectrometer (VTLS) to measure, H<sub>2</sub>O, SO<sub>2</sub>, OCS and CO from 64 km to the ground with high vertical sampling, and potentially record turbulent activity. The EnVision spacecraft will host a UV spectrometer VenSpec-U (Marcq et al., 2021) to measure SO<sub>2</sub>, SO:SO<sub>2</sub> ratio, and the unknown UV absorber at cloud-top altitudes. An IR spectrometer VenSpec-H will measure H<sub>2</sub>O, HDO, OCS, CO, and SO<sub>2</sub> between 65 and 80 km (Robert et al., 2019). Figures S3 and S4 in Supporting Information S1 show respectively SO<sub>2</sub> and the SO:SO<sub>2</sub> mixing ratio maps at cloud-top altitudes at the subsolar point. Convective cells are visible in SO<sub>2</sub> and SO with different structures due to chemical lifetime differences. The variability in SO<sub>2</sub> would be challenging to measure, and no convective structure is discernible for the SO:SO<sub>2</sub> ratio with a 12 km resolution. Figure S5 in Supporting Information S1 shows maps of CO, OCS, H<sub>2</sub>O and HCl at cloud-top altitudes at the Equator at night and noon at the model resolution. The spatial resolution of VenSpec-H will be close to 100 km, limiting the turbulent activity's impact on observations. A radio-science experiment will also be present (Dumoulin et al., 2020), measuring convective layer depth and H<sub>2</sub>SO<sub>4</sub> content in vapor and liquid from 35 to 55 km.

#### 7. Conclusion

In this study, we present the first coupling between a resolved turbulence model and a chemical model for the Venus' atmosphere.

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The impact of the strong vertical motion on different chemical species is estimated. The engendered vertical mixing is different for each species. Molecules with long chemical timescales will tend to be very well-mixed. For water, the condensation being a fast process, the vertical mixing inside the 48-55 km convective layer is weaker than for SO<sub>2</sub>. This convective layer tends to increase the optical depth of the clouds, by bringing H<sub>2</sub>SO<sub>4</sub> vapor-rich air from below the cloud, the vapor condensing into droplets during the ascent. The increase of opacity of the clouds due to this increase was quantified.

With the resolved convective motions, an estimation of the vertical eddy diffusion is possible. Values from 10<sup>2</sup> to 10<sup>4</sup> m<sup>2</sup> s<sup>-1</sup> for the 48–55 km convective layer are calculated. This range of values is several orders of magnitude larger than the values generally used in 1D chemistry modeling. In order to represent with accuracy the convective mixing inside the cloud layer, realistic values of vertical eddy diffusion must be implemented.

With a simplified microphysics, for the first time, the impact of the small-scale turbulence on the cloud layer boundaries was estimated. At the bottom of the cloud layer, convective motions are ubiquitous, generating altitude variation of between 300 m and 1 km depending on the convective strength. At the cloud-top, the presence of gravity waves emitted by the 48-55 km convective layer generates only small variation of the cloud top altitude inferior to 100 m. At the subsolar point, the cloud-top convective layer generates displacements of the cloud top of the order of 200 m.

The convection-induced cloud-top altitude variability in the model is consistent with measurements by Venus Express and Akatsuki. The impact of turbulence on cloud chemistry is consistent with some past observations. In situ measurement of clouds properties in the lower and middle cloud shows constant profiles consistent with the strong mixing of H<sub>2</sub>SO<sub>4</sub> liquid phase by the convection in this region.

The observability of the small-scale turbulence was assessed. EnVision with VenSpec-U would be able to resolve the spatial variability of  $\mathrm{SO}_2$  due to convection at cloud-top altitudes.

This first version of the coupled LES-chemistry-cloud model will be improved in the future. The equilibrium cloud parameterization used here will be replaced by a full microphysical model for the clouds that accounts for nucleation and growth of cloud droplets (Määttänen et al., 2023).

Regarding the sulfur and water cycle, several species like S<sub>n</sub>, H<sub>2</sub>S can act as buffers of sulfur species and could be added into the chemical network. These two examples will also be measured by DAVINCI. The current formalism of the radiative transfer in the model does not account for cloud radiative feedback. This effect should be taken into account in future studies to provide a more realistic description of the Venusian cloud dynamics.

#### **Data Availability Statement**

The Simulation outputs used to obtain the results in this paper are available in the open online repository M. Lefèvre (2024).

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