Supplementary information

# No evidence of phosphine in the atmosphere of Venus from independent analyses

Supplementary material

S1: Vertical profiles

The temperature profile as presented in "extended Data Figure 8" of G2020a is consistent with the Venus International Reference Atmosphere (VIRA)[30] for mid-latitudes (45 degrees latitude), and also consistent with later observations[31], so is also used in our simulations. The SO_2 profile presented in "extended Data Figure 9" of G2020a is consistent with previous observations[21–23], in particular at altitudes above 70 km relevant to this investigation, and employed in Figure 1 to model a potential SO_2 contamination signature. SO_2 is known to vary significantly in the mesosphere, with peak abundances beyond 1000 ppbv and a minimum at ~80 km in the 10-100 ppbv range[16,18–27]. Orbital missions, which provide global monitoring, observe large variations on timescales of hours to months superimposed on a long-term trend. Mesospheric SO_2 abundances at the time of the JCMT observations in June 2017 thus cannot be constrained by SO_2 abundances measured using ALMA in March 2019, and beam dilution could hinder the detectability of SO_2 in Venus with ALMA[17,32]. We also explored other plausible and reported mesospheric SO_2 profiles as measured by spacecraft[16] and similar to case D of ref [17], with SO_2 of ~30 ppbv at 80 km, and increasing to ~100 ppb at 90 km and reaching ~300 ppb at 95 km. See synthetic spectra for this case in Figure 2.

S2: ALMA bandpass corrections

From our first analysis, we note that the quality of the bandpass did improve with the updated JAO scripts, yet the residual spectra still show large fluctuations (see Supplementary Figure 1). In order to correct for this, a 12th order polynomial for the amplitude bandpass calibration was employed in G2020b. We, however, preserved the calibration scheme in the JAO scripts (3rd order polynomial for amplitude bandpass calibration), and removed a 6th order polynomial baseline fit from the final spectrum. The impact of masking the center line region and fitting a high-degree polynomial baseline can be quite problematic, as revealed by the effective disappearance of most of the PH_3 signature between G2020a and G2020b and demonstrated in supplementary figure 2. This was also shown systematically and independently for both the JCMT[13] data and the ALMA[12] data, which both revealed no PH_3 signature. We explored different polynomial fits to the residual data, testing the sensitivity of masking the line center region. We only observe an absorption feature as reported in G2020b when employing a 12th order polynomial, yet a 6th order already appears to provide a relatively good match to the fluctuations in the spectra (Supplementary Figure 1). We finally note that our second and third analyses result in notably flatter final spectra, only needing a 2nd order polynomial to be fit and removed.
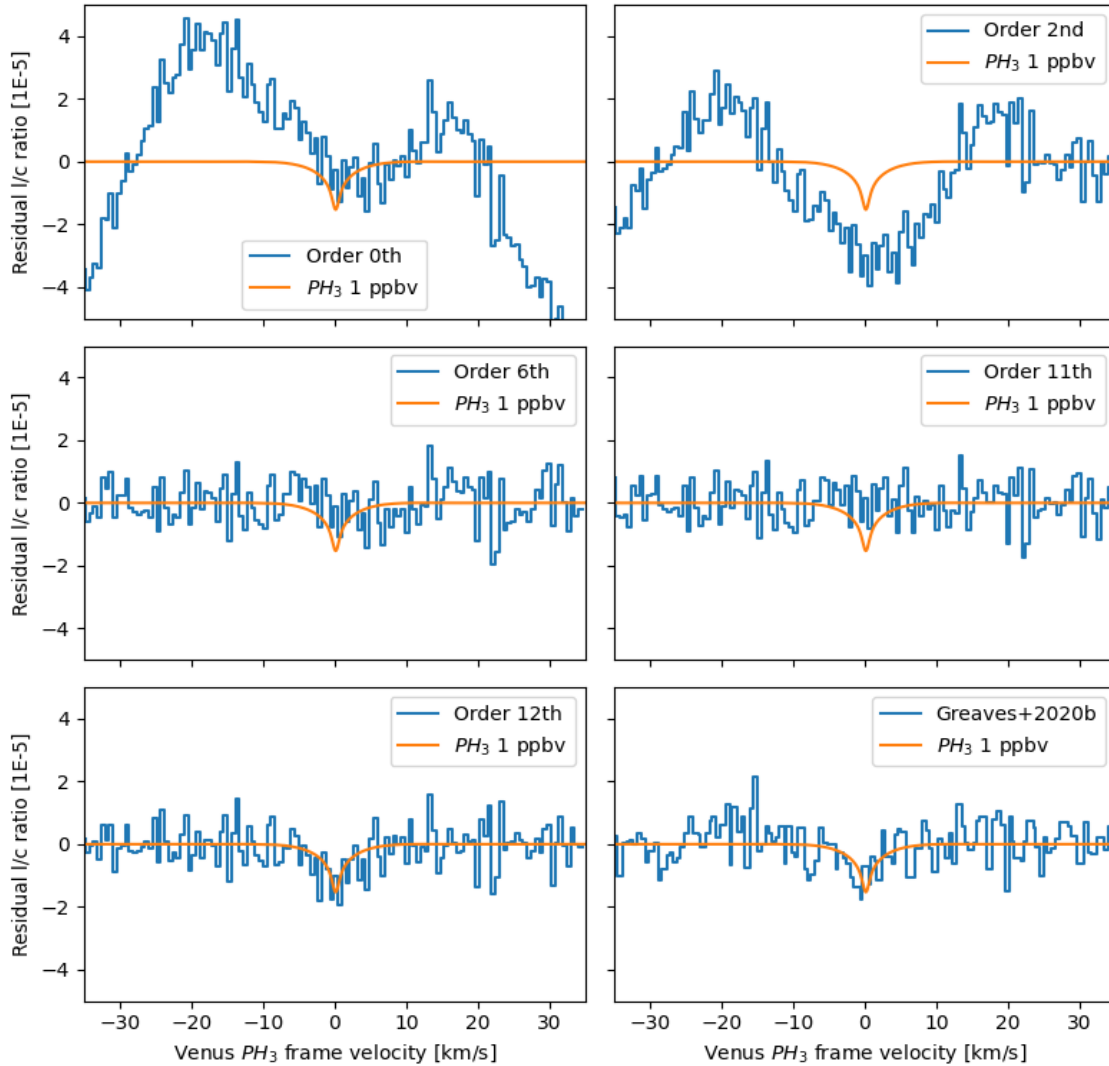
Supplementary Figure 1: By masking the center of the spectrum and by fitting a high-degree order polynomial around the line center, features in the core region may be artificially produced (see Supplementary Figure 2). In this figure, we present residual spectra as derived using the JAO scripts, and as presented in G2020b (see bottom/right panel). G2020b employed a bandpass polynomial of 12th order for the amplitude bandpass calibration, instead of the original 3rd order in the JAO scripts. We preserved all parameters as determined by the JAO team, included all baselines, and ultimately obtained the residual as presented in the top/left panel (order 0th baseline removed). We then explored sequentially higher degrees (while masking the center ± 5 km/s) and reached a reasonably good residual at order 6th. The residual with a polynomial of order 11th is quite similar to the 6th order case, and only when employing a 12th order polynomial do we observe a feature as reported in G2020b. In the case of G2020b, the 12th order polynomial was not removed post-processing as shown here, but applied to the visibility bandpass calibration.
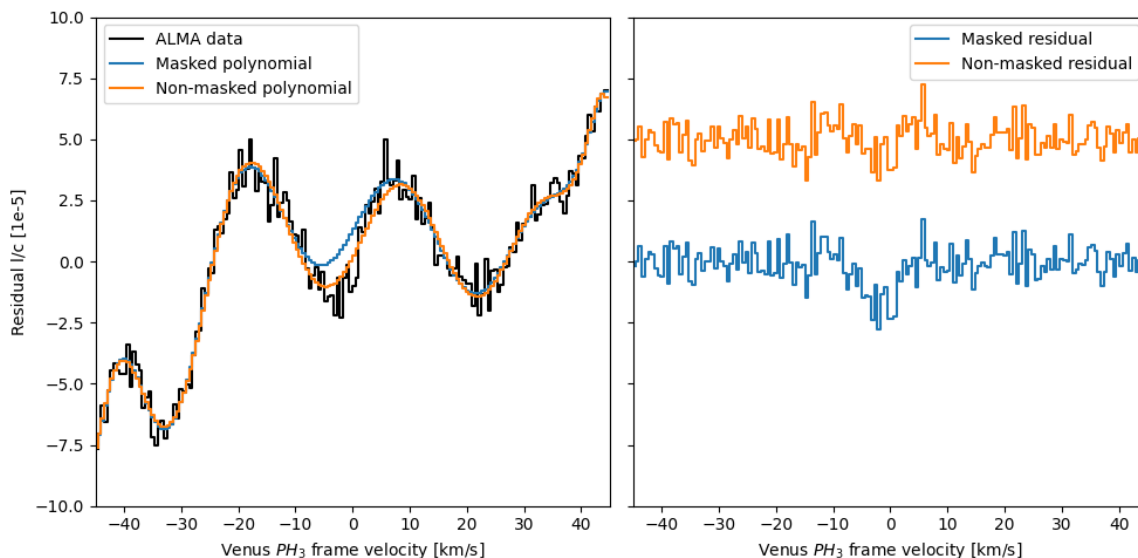
Supplementary Figure 2: Impact of masking the core region while fitting a high-degree polynomial order around the center velocities. **Left:** Continuum subtracted residual spectrum of Venus derived employing the original G2020a scripts (yet disabling self-calibration[9] and including all baselines). Polynomial fits (12th order) are superimposed: "non-masked" trace is a fit to the entire spectrum, while "masked" is a fit excluding the center ± 5 km/s. **Right**: Spectra after subtracting the polynomial fits - The non-masked residual has been offset for clarity. These residuals are derived employing modified scripts as those employed by G2020a, and they are solely to show the complexities on fitting a high-degree polynomial to ripply data.

S3: Validation of the ALMA analysis by interpreting other nearby lines

We independently analyzed the ALMA data using our calibration scripts for the region near the SO_2 line at 267.537458 GHz and the HDO (J=$2_{2,0}$-$3_{1,3}$) line at 266.16107 GHz (see Fig. Supplementary Figure 3). We do not detect SO_2, and estimate a mesospheric abundance <10 ppbv, which is consistent with the reported range of variability observed of mesospheric SO_2 (see S1). Furthermore, beam dilution could hinder the detectability of SO_2 in Venus[17,32]. We have a detection of HDO, and when assuming a D/H of 200[1] we estimate a mesospheric value of ~60 ppbv for water, in agreement with previous findings[23].
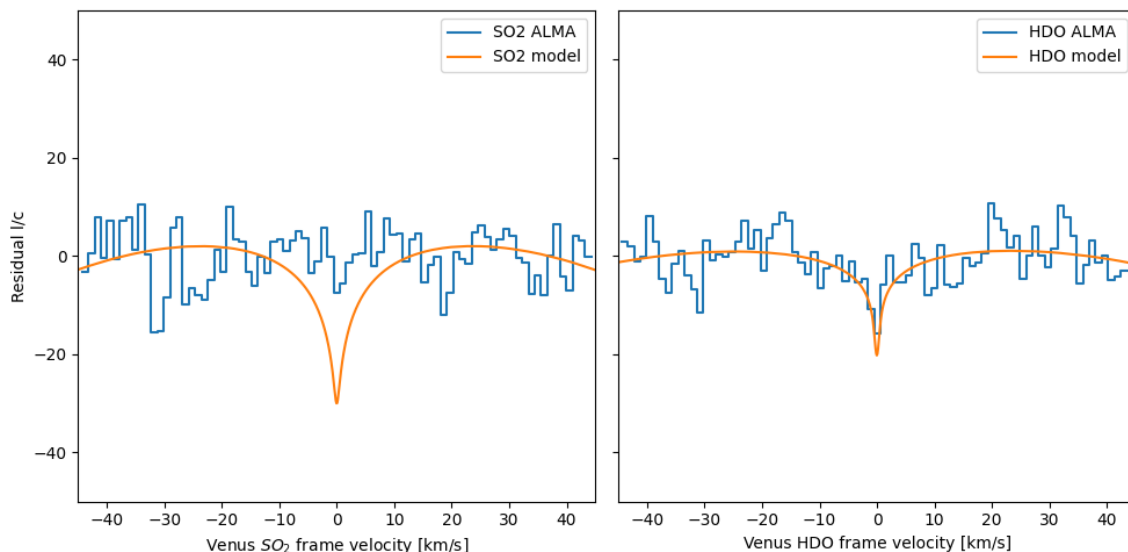
Supplementary Figure 3: Comparison between models and ALMA data, for the SO_2 ($J=13_{3,11}-13_{2,12}$) transition at 267.537458 GHz and for the HDO ($J=2_{2,0}-3_{1,3}$) transition at 266.161070 GHz. **Left**: Our independently processed ALMA data for the SO_2 line, and "SO_2 model" is a synthetic spectrum modeled employing the VIRA45 T/P profile and mesospheric (70-90 km) abundance of SO_2 of 10 ppb. **Right**: Our analysis of the ALMA data for a nearby HDO line, and "HDO model" is a synthetic spectrum as modeled adopting a D/H of 200[20], and a plausible H_2O abundances of ~60 ppbv in the mesosphere (70-100 km). A 2nd order polynomial was removed from each spectrum (while masking the ± 5 km/s region).

S4: Altitude of the probed narrow molecular absorptions

The altitude from which a specific absorption/emission originates is related to the spectroscopic parameters of the targeted line and those of competing radiatively-active species in this spectral region. Going deeper into the Venusian atmosphere, the pressure increases, and so does the Lorentzian width of the lines. For instance, when considering the linewidth of 0.186 cm^{-1}/atm for PH_3, at 70 km (3.4×10^{-2} atm) the line would be 213 km/s at 267 GHz, much broader than the narrow window region of ± 5 km/s or ± 10 km/s used to search for PH_3. For the spectral range shown in Figures 1/2 and in the figures in G2020a/b/c, the spectral line would appear completely flat, since it encompasses a spectral range much broader than shown. Since a polynomial is removed for pixels beyond the core region, any PH_3 information beyond this velocity would also be removed. A PH_3 linewidth of 20 km/s would occur at 3.2×10^{-3} atm (81 km), so in principle information below this altitude would be removed. Furthermore, as we go deeper into the atmosphere, collision-induced-absorptions and the broad wings of other strong submillimeter absorbers (e.g., CO_2, SO_2, H_2O) dominate over the PH_3 signatures, further masking any potential signatures in these regions.

In order to quantify the specific altitude in which PH_3 would produce a detectable absorption in the residual data, we synthetized spectra using the linewidth considered in G2020a/b (0.186 cm^{-1}/atm) at different altitudes for a 10 km layer of PH_3. The model spectra for each altitude are shown in Supplementary Figure 4, with the bottom altitude listed in the y-axis. The figure

demonstrates that these observations only sample PH_3, if present, above 75 km in altitude. The other linewidth suggested in G2020a/b of 0.286 cm^{-1}/atm requires an absorption region even higher in the atmosphere (>80 km). This is consistent with the findings and modeling presented in Lincowski+2021[17].
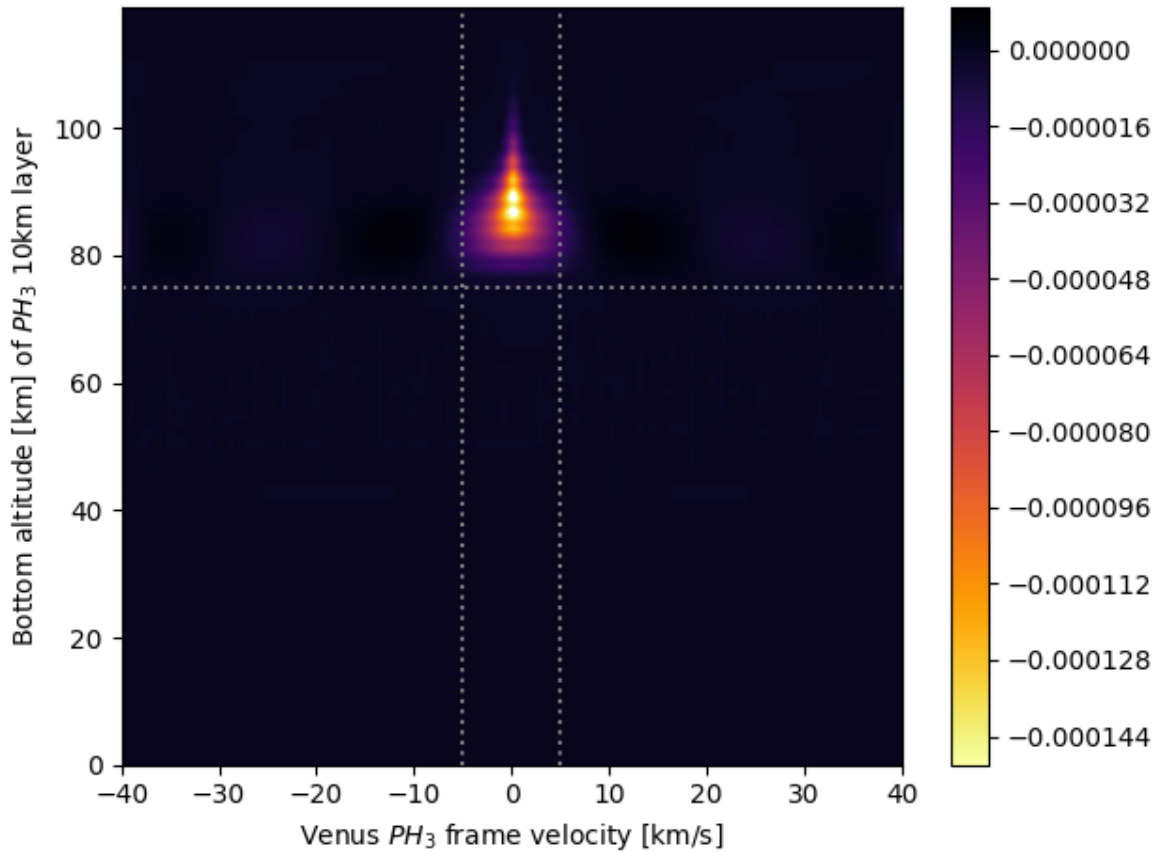
Supplementary Figure 4: Sensitivity analysis of the recoverable PH_3 J=1-0 signature as a function of velocity and altitude on Venus. This was computed by synthesizing spectra of a hypothetical 10 km-deep layer of PH_3 located at different altitudes. The vertical axis defines the lower bound of the layer, with the horizontal dotted line marking the 75 km altitude. The vertical lines denote the ± 5 km/s window around the line center of the transition. A 6th order polynomial was removed for each spectrum, while also masking the ± 5 km/s center region, as performed to the ALMA data presented in Figure 2, Supplementary Figures 1 and 2, and the figures in G2020a/b. A linewidth of 0.186 cm^{-1}/atm was considered as in G2020a/b.