

Constraints on Neptune's haze structure and formation from VLT observations in the H-band

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1 ABSTRACT

2 A 1-dimensional microphysics model has been used to constrain the structure
3 and formation of haze in Neptune’s atmosphere. These simulations were cou-
4 pled to a radiative-transfer and retrieval code (NEMESIS) to model spectral
5 observations of Neptune in the H-band performed by the SINFONI Integral
6 Field Unit Spectrometer on the Very Large Telescope (VLT) in 2013. It was
7 found that observations in the H-band and with emission angles $\leq 60^\circ$ are
8 largely unaffected by the imaginary refractive index of haze particles, allowing
9 a notable reduction of the free parameters required to fit the observations. Our
10 analysis shows a total haze production rate of $(2.61 \pm 0.18) \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$,
11 about 10 times larger than that found in Uranus’s atmosphere, and a parti-
12 cle electric charge of $q = 8.6 \pm 1.1$ electrons per μm radius at latitudes between
13 5 and 15° S. This haze production rate in Neptune results in haze optical
14 depths about 10 times greater than those in Uranus. The effective radius r_{eff}
15 was found to be 0.22 ± 0.01 and $0.26 \pm 0.02 \mu\text{m}$ at the 0.1 and 1-bar levels, re-
16 spectively, with haze number densities of $8.48_{+1.78}^{-1.31}$ and $9.31_{+2.52}^{-1.91}$ particles per
17 cm^3 . The fit at weak methane-absorbing wavelengths reveals also the pres-
18 ence of a tropospheric cloud with a total optical depth > 10 at $1.46 \mu\text{m}$. The
19 tropospheric cloud base altitude was found near the 2.5-bar level, although
20 this estimation may be only representative of the top of a thicker and deeper
21 cloud.

22 Our analysis leads to haze opacities about 3.5 times larger than that derived
23 from Voyager-2 observations (Moses et al., 1995). This larger opacity indicates
24 a haze production rate 2 times larger at least. To study this difference haze
25 opacity or production rate, we performed a timescale analysis with our micro-
26 physical model to estimate the time required for haze particles to grow and

1 settle out. Although this analysis shows haze timescales (~ 15 years) shorter
 2 than the time lapsed between Voyager-2 observations and 2013, the solar illu-
 3 mination at the top of the atmosphere has not varied significantly during this
 4 period (at the studied latitudes) to explain the increase in haze production.
 5 This difference in haze production rate derived for these two periods may arise
 6 from: a) the fact that in our analysis we employed spectral observations in the
 7 infrared (H-band), while Moses et al. (1995) used photometric images taken at
 8 5 different filters in the visible. While high-phase-angle Voyager observations
 9 are more sensitive to small haze particles and at altitudes above the 0.1-bar
 10 level, the haze constraints derived from VLT spectra in H-band are limited
 11 to pressures greater than 0.1 bar. As a result of the different phase angles
 12 of the two set of observations, differences in the estimation of M_0 may arise
 13 from the use of Mie phase functions as well. b) our 1-dimensional model does
 14 not account for latitudinal redistributions of the haze by dynamics. A possible
 15 meridional transport of haze with wind velocities greater than $\sim 0.03 \text{ ms}^{-1}$
 16 would result in dynamics timescales shorter than 15 years and thus might ex-
 17 plain the observed variations in the haze production rate during this period.
 18 Compared with our estimations, photochemical models point to even larger
 19 production rates on Neptune (by a factor of 2.4). Assuming that the pho-
 20 tochemical simulations are correct, we found that this discrepancy can be
 21 explained if haze particles evaporate before reaching the tropospheric-cloud
 22 levels. This scenario would decrease the cumulative haze opacity above the
 23 1-bar level, and thus a larger haze production rate would be required to fit
 24 our observations. However, to validate this haze vertical structure future mi-
 25 crophysical simulations that include the evaporation rates of haze particles
 26 are required.

¹ *Keywords:* Neptune; haze microphysics; radiative transfer;

1 Introduction

Analyses of high-phase-angle Voyager-2 images indicated the presence of hazes in the atmosphere of Neptune (e.g. Smith et al., 1989). As in Uranus’s atmosphere, the haze formation is attributed to the condensation of hydrocarbons such as C_2H_6 or C_2H_2 in the stratosphere ~ 0.016 mbar. These hydrocarbons are the product of methane dissociation by solar UV and energetic particles that leads to a number of chemical reactions (Moses et al., 1995, 2018). The condensation of the hydrocarbons into their respective ices results in the formation of the initial haze particles at different altitudes. These initial haze particles, which cannot be detected from ground-based observations due to their small size (radii $< 0.1 \mu m$), can grow in size very quickly by particle coagulation, and then sink deeper into the atmosphere. Estimations of the haze vertical structure (size distribution and density) can be performed using radiative-transfer simulations. For instance, analyses of limb observations taken at different tangent heights can provide strong constraints on the vertical haze density or size distribution. However, our current observations, ground-based telescopes or observations from telescopes in orbit around the earth, do not allow us to perform these kind of analyses and usually the radiative-transfer analyses must assume some properties of the haze such as the vertical distribution. A number of radiative-transfer simulations have been performed to study the aerosol properties in Neptune’s atmosphere (e.g. Karkoschka and Tomasko, 2011; Irwin et al., 2014, 2016a). Although all these analyses required hazes in their respective models to fit the observations, the haze structure (number density and size distribution) used or estimated by their analyses were very different. While Irwin et al. (2016a) analyzed VLT spectra in the H-

1 band (1.4-1.8 μm) using a 2-cloud model compromising a vertically extended
 2 haze layer and a tropospheric compact cloud, Karkoschka and Tomasko (2011)
 3 used a model with different haze layers between the 10 and 0.01-bar levels
 4 without compact clouds to analyse data cube of Neptune acquired with the
 5 Hubble STIS spectrograph. Based on the χ^2 analysis, both models were found
 6 to provide good fits to the respective observations despite the different haze
 7 structure of the models. These different radiative-transfer analyses show the
 8 limitations of determining precisely the scattering and vertical distribution
 9 properties of the haze from remote observations alone.

10 Some previous analyses used haze microphysics simulations to provide con-
 11 straints on the density and size distribution of the haze vertical profiles. For
 12 Uranus's haze, Toledo et al. (2019) combined a microphysical model (Cabane
 13 et al., 1992; Rannou et al., 2004) with the NEMESIS radiative-transfer code
 14 (Irwin et al., 2008) to analyse VLT observations in the H-band. For Neptune's
 15 haze, Moses et al. (1995) analyzed high-phase-angle Voyager-2 images com-
 16 bining models of haze microphysics (Toon et al., 1980) and photochemistry
 17 (Moses et al., 1992) to constrain the haze formation and structure. Although
 18 this analysis provided strong constraints on haze properties, a number of ob-
 19 servations have revealed seasonal changes in Neptune's brightness after the
 20 Voyager-2 observations (e.g. Lockwood and Thompson, 2002; Aplin and Har-
 21 rison, 2016). These changes in brightness, which are associated with changes in
 22 the albedo from hazes and clouds, point out the need of performing new haze
 23 microphysics simulations of the formation and growth of haze in Neptune's
 24 atmosphere. In this work we analysed observations acquired in 2013 using the
 25 SINFONI Integral Field Unit Spectrometer on the VLT in the H-band with a
 26 microphysical code following the approach of Toledo et al. (2019). In order to
 27 fit the spectral observations, the haze microphysical simulations were coupled

1 with NEMESIS. We describe the data utilized in this work, the microphysical
2 model, and radiative-transfer codes in section 2. In sections 3, 4 and 5 we
3 present our model results, compare them to data, and discuss the impact of
4 dynamics on the distribution of the haze in Neptune’s atmosphere.

5 **2 Observations, haze microphysics and radiative transfer**

6 *2.1 Observations*

7 We analyzed observations of Neptune in the H-band ($1.4\text{--}1.8\ \mu\text{m}$) performed
8 by the SINFONI instrument in October 2013 (Irwin et al., 2016a), at the
9 European Southern Observatory (ESO) VLT in Paranal, Chile. These ob-
10 servations were performed with a spatial resolution of about $0.1''$, returning
11 64×64 -spatial-pixel spectral cubes with a spectral resolution of $0.0005\ \mu\text{m}$.
12 Data reduction was carried out with the ESO VLT SINFONI pipeline and ad-
13 ditional photometric corrections as described in by Irwin et al. (2016b) were
14 also included. The reduced data were then averaged with a triangular-shaped
15 instrument function with full width half maximum = $0.002\ \mu\text{m}$, resulting in
16 a final spectral resolution of $R\sim 775$. It was found in previous analyses of
17 Neptune that this spectral resolution provides the best compromise between
18 modeling computational speed, signal-to-noise ratio, and accurate representa-
19 tion of the methane absorption features (Irwin et al., 2014, 2016a).

1 2.2 Haze microphysics and radiative transfer

2 The same 1-dimensional haze microphysics model used in Toledo et al. (2019)
 3 for Uranus was employed in this study to simulate the formation and evolution
 4 of haze in Neptune. This 1-dimensional Eulerian model, first used to study
 5 the haze in Titan’s atmosphere (Cabane et al., 1992; Rannou et al., 2004),
 6 solves the aerosol continuity equation to simulate the time evolution of a given
 7 population of particles, described in terms of particle radius and altitude,
 8 whose production occurs in the stratosphere. The different processes involved
 9 in the time variation of the population of particles are the particle coagulation,
 10 settling and eddy diffusion. The production rate of particles Q at a given
 11 altitude z is parametrized by a sum of Gaussian functions

$$12 \quad Q(z) = \sum_{i=1}^n Q_i \exp\left(-\frac{(z - z_{0i})^2}{2\Delta z_i^2}\right) \quad (1)$$

13 where n represents the number of condensable species (i.e. C_2H_6 , C_2H_2) whose
 14 vertical production region is parametrized by the adjustable parameters z_{0i} (al-
 15 titude of maximum of production) and Δz_i (extension of the haze-production
 16 region). For each condensable species, the term Q_i is given by

$$17 \quad Q_i = \frac{M_{0i}}{(4/3)\pi\rho_i r_0^3} \frac{1}{\sqrt{2\pi}\Delta z_i} \quad (2)$$

18 where M_{0i} is the vertically-integrated production rate ($\text{kg m}^{-2} \text{s}^{-1}$) of species
 19 i , r_0 is the initial model particle radius (set to $0.0012 \mu\text{m}$) and ρ_i is the density
 20 of the material making up the particles (fixed to 700 kg m^{-3}). The parameter
 21 M_{0i} determines the mass of haze produced by the condensation of species i

1 in the stratosphere and its range of values is estimated in our simulations.
 2 In our analysis we have assumed a single condensible species ($n=1$) with z_0
 3 set to the altitude corresponding to the ~ 0.016 -bar level (Moses et al., 1995)
 4 and Δz to 2.5 km. In section 3.4 we will discuss the impact of adding multiple
 5 condensation rates on the haze retrievals. For modelling the coagulation rate in
 6 a given radius bin, we need to specify the sticking coefficient (s) that represents
 7 the probability that two particles will stick together (Pollack et al., 1987). The
 8 value of s for two particles of radius r_1 and r_2 is given by

$$9 \quad s = \exp\left(\frac{-q^2 r_1 r_2}{kT(r_1 + r_2)}\right) \quad (3)$$

10 where T is the temperature, k is the Boltzmann's constant and q is the haze
 11 charge-per-unit-radius parameter (expressed in electrons per micrometre of
 12 radius) which, together with M_0 , is estimated in our simulations. The last
 13 parameter required for the model is the eddy diffusion coefficient (K) whose
 14 profile for Neptune's atmosphere is illustrated in Figure 1. The eddy diffusion
 15 was computed by

$$16 \quad K(p) = K_0 \left(\frac{p}{kT}\right)^{-0.5} \quad (4)$$

17 where the constant K_0 was estimated from K at a given reference pressure
 18 level. The temperature profile used in equation 4, displayed in Fig. 1, is based
 19 on the 'N' profile estimated by radio-occultation from Voyager 2 by Lindal
 20 (1992) with an He:H₂ ratio of 0.177. This He:H₂ ratio results in a helium
 21 volume mixing ratio of 0.15 at altitudes of negligible methane abundance (as-
 22 suming 0.3% nitrogen). For a set of values of M_0 and q the code computes
 23 the vertical haze profiles of number density and size distribution. Since we
 24 are using a 1-dimensional model, latitudinal variations of K and T are not

1 accounted in the simulations. However, in sections 3.3 and 3.5 we will discuss
 2 how variations in the K and T profiles may affect our haze properties estima-
 3 tions.
 4 For simulations of the scattering and absorption of gases and aerosols in Nep-
 5 tune’s atmosphere, the NEMESIS correlated-k radiative transfer and retrieval
 6 code (Irwin et al., 2008) was used. The scattering properties of aerosols were
 7 computed using Mie theory with phase functions approximated at each wave-
 8 length with combined Henyey-Greenstein functions to average over the char-
 9 acteristic “glory” and “rainbow” of pure spherical particles. The same atmo-
 10 spheric model as described in Irwin et al. (2014, 2016a) was used to model
 11 the spectra. The methane abundance profile was set with a deep CH₄ mole
 12 fraction of 4% and the volume mixing ratio limited to a maximum relative
 13 humidity of 60%, as recommended by Karkoschka and Tomasko (2011), but
 14 the stratospheric abundance was set to $(1.5 \pm 0.2) \times 10^{-3}$ as recommended by
 15 Lellouch et al. (2010). The methane absorption in the H-band was computed
 16 using the ‘WKMC-80K’ line database (Campargue et al., 2012). The methane
 17 line data were converted to k-distribution look-up tables covering the spectral
 18 range of the VLT observations and assuming a triangular-shaped instrument
 19 function with full width half maximum=0.002 μm . Collision-induced absorp-
 20 tion by H₂–H₂ and H₂–He were computed using the coefficients of Borysow
 21 et al. (1989), Borysow et al. (2000), and Zheng and Borysow (1995), and an
 22 equilibrium ortho/para-H₂ was assumed at all altitudes and locations. Absorp-
 23 tion by H₂–CH₄ and CH₄–CH₄ collision-induced was also computed (Borysow
 24 and Frommhold, 1987).

3 Retrieval analysis

3.1 *H-band observations*

To estimate the haze parameters M_0 and q in Neptune’s atmosphere, we used the haze microphysics and NEMESIS radiative-transfer codes as described in Toledo et al. (2019). VLT H-band spectra were fitted with NEMESIS using an aerosol model comprising a tropospheric cloud and a haze layer whose profiles of particle size and number density are computed with the microphysical model for a set of values of M_0 and q . The cross section (σ), phase function ($p(\theta)$) and single-scattering albedo (ω) of aerosol required for the radiative-transfer simulations were computed using Mie theory for a given aerosol refractive index and size distribution. In previous analyses the imaginary refractive index spectrum of the haze and the tropospheric cloud was fitted. In order to analyse the impact of the imaginary refractive index (k) on the observations, simulations of VLT observations in the H-band for different values of k for the haze particles and a constant effective radius of $0.3 \mu\text{m}$ are shown in Figure 2. These simulations indicate that an increase in k has a little impact on the simulated spectrum. Although at weak methane-absorbing wavelengths (e.g. at $1.585 \mu\text{m}$) some variations can be observed, it is important to note here that the haze microphysics parameters are mainly constrained from the observations at strong methane-absorbing wavelengths (1.46 , 1.64 and $1.7 \mu\text{m}$) since only the high troposphere or stratosphere is sampled. That is to say, the haze microphysics parameters are, in this case, mainly retrieved from the observations for which k does not have a significant effect.

In order to further study the haze scattering properties in the H-band, the

1 spectrum of ω , σ and the scattering cross-section (σ_S) computed for the k
 2 and r_{eff} values used in Fig. 2 are illustrated in Figure 3. This figure indicates
 3 that an increase in k results in a smaller ω (Fig. 3a) and a greater σ (Fig.
 4 3b). The light scattered by the haze in a given level depends on both the
 5 haze number density and the scattering cross-section, defined as $\sigma_S = \sigma \times \omega$.
 6 Fig. 3c shows that in the H-band range, the scattering cross-section is prac-
 7 tically the same for $k=0.0001$, 0.01 and 0.04 . This is due to the fact that in
 8 the H-band range the increases/decreases in ω with k are compensated by the
 9 decreases/increases in σ , resulting in a practically constant $\sigma_S = \sigma \times \omega$. These
 10 results explain the negligible impact of the imaginary refractive index on the
 11 simulated spectrum shown in Fig. 2. Therefore, these results indicate that for
 12 simulating a VLT spectrum in the H-band we can use a constant imaginary
 13 refractive index for haze particles and, therefore, reduce the number of free
 14 parameters in the model.

15 3.2 Aerosol model

16 Similar to previous work (see Irwin et al., 2018), the effective particle radius
 17 and vertical distribution of the tropospheric cloud were fitted in our analy-
 18 sis. The vertical cloud number density was characterised by the total opacity
 19 (τ_{tc}), the fractional scale height (FSH) and the cloud base altitude (CBA).
 20 It is important to note that for haze retrievals we can use a constant imagi-
 21 nary refractive index because the effective radius of particles is smaller than
 22 $0.5 \mu\text{m}$. However, since the radii of the particles of the tropospheric cloud
 23 can achieve values greater than $0.5 \mu\text{m}$, the spectrum of the refractive index
 24 has an important impact on the simulations. Indeed, it can be demonstrated

1 that as r_{eff} increases, the differences between the simulated spectra of Fig.
 2 2 become bigger. Therefore, the spectrum of the imaginary refractive index
 3 for the tropospheric cloud was retrieved, and the real refractive index at a
 4 single wavelength was set to a reference value of 1.4 (see Irwin et al., 2017,
 5 2018); the real part of the refractive index at all other wavelengths was cal-
 6 culated using the Kramers-Kronig relation (see Sheik-Bahae, 2005). The free
 7 parameters in our model are, therefore, M_0 and q for the haze, and τ_{tc} , FSH,
 8 CBA, the size distribution (r_{eff} , ν_{eff}) and the imaginary refractive index for
 9 the tropospheric cloud. Figure 4 shows, as example, the effect of varying the
 10 haze production rate M_0 (upper panel) and the tropospheric cloud opacity τ_{tc}
 11 (lower panel) on the simulated H-band spectrum. These simulations indicate
 12 that the haze production rate has consequences for the whole of the H-band
 13 spectrum measured by VLT. However, this is not the case for the tropospheric
 14 cloud opacity for which we only observe variations in the simulated spectrum
 15 at the weak methane-absorbing wavelengths. These simulations indicate that
 16 by fitting only the observations at strong methane-absorbing wavelengths, we
 17 can derive the haze properties independently of the tropospheric cloud param-
 18 eters.

19 3.3 Haze retrievals

20 To estimate the haze parameters M_0 and q in Neptune with our model, we
 21 chose to analyse VLT observations in regions free of high clouds. Neptune
 22 images are displayed in Figure 5, along with the spectra that result from the
 23 average of the observations in the pixels indicated in red and blue. These pixels
 24 were grouped according to latitude and emission-angle range: the spectrum in

1 red (see Fig. 5b) was computed from all the pixels at latitudes between 0 and
 2 15° S (note that this region is free of high clouds) and with emission angles of
 3 between 20 and 25°, while the spectrum in blue from pixels with emission an-
 4 gles of between 50 and 55° and the same latitude range. To cover a wide range
 5 of emission angles in our analysis, we fitted simultaneously a total of 6 spectra
 6 computed as those shown in the right panel of Fig. 5 but for the emission-
 7 angle ranges [20°-25°], [30°-35°], [40°-45°], [50°-54°], [55°-58°] and [59°-60°].
 8 For these emission-angle ranges we tested the validity of the results shown in
 9 the previous section, and found that to fit these observations a constant imag-
 10 inary refractive index can be used (in this analysis we used a refractive index
 11 of $n=1.4+0.0001i$). However, Irwin et al. (2011, 2016a) found that for limb-
 12 darkening analyses including spectra with emission angles greater than 60°
 13 the haze imaginary refractive index has an impact on the simulated spectra.
 14 Since in this work we are focused on estimating the haze parameters M_0 and
 15 q , our limb-darkening analysis comprises only spectra with emission angles \leq
 16 60°, allowing a notable reduction of the free parameters required to fit the
 17 observations. Figure 6 shows a comparison between observations (thick red
 18 solid lines) and simulations (blue line) performed for $M_0=2.5 \times 10^{-14} \text{ kg m}^{-2}$
 19 s^{-1} and $q=10$ electrons per μm radius. Note that because in this section we are
 20 only interested in estimating the haze parameters, these simulations are only
 21 performed for the observations at strong methane-absorbing wavelengths (in
 22 the spectral ranges 1.46-1.6 μm and 1.65-1.71 μm). Therefore, we can simulate
 23 the observations illustrated in Fig. 6 to find which range of values of M_0 and
 24 q can fit the observations.

25 In order to speed up the simulations, the retrieval procedure makes use of a
 26 pre-computed set of look-up tables of the haze number density and size distri-
 27 bution for a number of combinations of M_0 and q . For each combination of M_0

1 and q , we fitted simultaneously the 6 spectra shown in Fig. 6 with NEMESIS
 2 code at the strong methane-absorbing wavelengths. Figure 7 shows the con-
 3 tour plots of χ^2 in the M_0 - q space of the analysis, along with 3σ significance
 4 level of χ^2 (red solid line). The analysis provides a χ^2 minimum of 423.97,
 5 value that corresponds to a reduced χ^2 (defined as $\chi^2/N_{\text{observations}}$) of 0.64. If
 6 we define the errors of M_0 and q as the ΔM_0 and Δq that give a $\Delta\chi^2=11.8$
 7 (value corresponding to the 3σ significance level), then our analysis provides a
 8 $M_0=(2.61\pm0.18)\times10^{-14}$ kg m $^{-2}$ s $^{-1}$ and a $q=8.6\pm1.1$ electrons per μm radius.
 9 For these retrievals the temperature and eddy-diffusion profiles illustrated in
 10 Fig. 1 were used to simulate the haze microphysics. However, some latitudinal
 11 variations in the values of these parameters could be expected. In order to
 12 study how such variations can affect our estimations of the haze parameters,
 13 the left panel of Figure 8 shows the pressure levels at which the haze scattering
 14 optical depth τ_s are 0.01 and 0.04 for different T profiles. In all of these simu-
 15 lations the only variable that was changed is the temperature profile. These T
 16 profiles are defined as $T=\Delta T+T_0$, where T_0 is the temperature profile shown
 17 in Fig. 1. These variations in the haze optical depth with T can be compared
 18 with those observed when the haze production rate changes (right panel of
 19 Fig. 8). We observe that a variation in M_0 of 7% (note that this is the error
 20 in our estimation of M_0) produces a variation in the pressure level at which
 21 $\tau_s=0.04$ of $\Delta p=\pm \sim 0.4$ bar (see right panel of Fig. 8). We note that to produce
 22 such a change in the pressure level of $\tau_s=0.04$ the temperature must vary by
 23 $\Delta T=\pm \sim 6$ K. Therefore, these simulations indicate that latitudinal variations
 24 in the temperature are not expected to change notably our estimation of M_0
 25 as long as they are smaller than 6 K. A similar analysis was performed for the
 26 eddy-diffusion profile, which simulates the transport of haze particles in the
 27 low stratosphere and troposphere. In this case we multiplied K_0 of equation

4 by different factors, from 0.1 up to 10, to study the variation of the haze
optical depth with the eddy-diffusion profile. These simulations, illustrated in
Figure 9, indicate that the haze optical depth variations with K_0 are smaller
than those observed when M_0 varies by a 7%. Therefore, these simulations
show that latitudinal variations in the values of T and K are not expected to
cause a significant effect on our estimations of the haze parameters.

3.4 *Tropospheric cloud retrievals*

In our prior analysis the haze retrievals were decoupled from the tropospheric
cloud parameters by fitting only the observations at strong methane-absorbing
wavelengths. In our current analysis we fitted the whole VLT H-band spectrum
shown in Fig. 6 to estimate the tropospheric cloud parameters: τ_{tc} , FSH, CBA,
the size distribution (r_{eff} , ν_{eff}) and the imaginary refractive index of cloud
particles. For these retrievals the haze parameters were kept constant to the
values derived in previous section. This analysis provides the best fit shown in
Figure 10 with $\tau_{tc} \sim 12$ at $1.46 \mu\text{m}$, $\text{CBA} \sim 2.5$ bar. Regarding the size of
the particles, presumably made of H_2S ice (Irwin et al., 2018, 2019), we found
that particles with r_{eff} in the range of $2\text{--}2.6 \mu\text{m}$ provide the best fit between
observations and simulations. However, it is important to note here that as
a result of its large opacity these retrievals may be only representative of the
top of a thicker and deeper tropospheric cloud. In order to compare these
retrievals with those given in previous section, Figure 11 shows the vertical
profiles of size, number density and scattering cumulative optical depth of
both the tropospheric cloud and the haze. For comparison purposes, the haze
profiles derived for Uranus (Toledo et al., 2019) are also displayed in Fig.

11. This comparison indicates tropospheric cloud particles on Neptune much bigger in size and number density (for $p > 0.04$ bar), resulting in optical depths of about 250 larger than those due to hazes. Therefore, comparing these results with the haze opacities shown in Fig.8 and Fig. 9, we find that dispersion by cloud particles dominates for altitudes below the cloud top. We also observe that the haze in Neptune is greater in size and number density than the haze in Uranus for $p > 0.02$ bar as a result of the greater haze production rate (M_0 in Neptune is about 10 times bigger). This results from the fact that methane molecules can reach higher altitudes in Neptune than in Uranus because of the higher eddy diffusion coefficient of this planet (Moses et al., 1995). Therefore, this indicates that while the eddy diffusion does not have a major impact on the transport of haze particles in the low stratosphere and troposphere (section 3.3), it does have a major impact on M_0 through the transport of gases in the region of the atmosphere where the methane photolysis takes place (near the 10^{-3} -mbar level). That is to say, greater eddy-diffusion coefficients in the high stratosphere result in a greater haze production rate (parameter estimated in our analysis).

3.5 Haze production analysis

In our previous analysis, we have assumed a simplistic 1-region model for the production of haze. This scheme assumes that species such as C_2H_2 or C_4H_2 condense at the same region of the atmosphere. In order to study the impact of this simplification on the estimation of the haze parameters, we performed microphysics simulations using 3 different models of haze production, whose vertical distribution is characterized by equation 1. The values of p_{0i} and M_{0i}

1 used for each of these models are given in Table 1. Note that while model-1
 2 corresponds to our 1-region model used in the previous sections, model-2 and
 3 model-3 compromise 3 different vertical regions of haze production. The pres-
 4 sure level p_{0i} and value of M_{0i} for each region were chosen according to Moses
 5 et al. (1995). Note that the total haze production M_0 is the same for the 3
 6 models. The profiles of haze number density and optical depth obtained for
 7 these different models are displayed in Figure 12. The different haze produc-
 8 tion regions of model-2 and model-3 can be identified through the maxima
 9 of number density above the 0.1-bar level. Although some variations in the
 10 density profiles can be observed above the 0.03-bar level, the density profiles
 11 below this pressure level converge toward the same solution. To test whether
 12 this different vertical structure above the 0.03-bar level can be significant in
 13 our observations we need to study the cumulative optical depth. In the pre-
 14 vious section we have mentioned that our observations are not sensitive to
 15 variations in the aerosol optical depth smaller than $5 \cdot 10^{-4}$. By studying the
 16 optical depth profiles given in the lower panel of Fig. 12, we observe that the
 17 largest variations in optical depth are given for model-1 and model-2. How-
 18 ever, these differences, only observed for altitudes above the 0.04-bar level,
 19 are in any case smaller than $5 \cdot 10^{-4}$. These results show, therefore, that even
 20 the simplistic 1-region model provides similar results than those given by the
 21 3-region models.

22 Following this study of the impact of using a 1-region model for the produc-
 23 tion of haze, the next step in this analysis is to test the importance of the
 24 radius of the initial haze particles. In all our models of haze production, we
 25 have assumed an initial model radius of $0.0012 \mu\text{m}$. Since our estimations of
 26 haze production rate are given in units of mass, this means that for a bigger
 27 initial model radius the number of initial haze particles must be smaller to

1 keep constant M_0 . The number of haze particles produced in a given region of
 2 the atmosphere depends on the population of nuclei (assuming heterogeneous
 3 nucleation) and the concentration of condensable species. However, the num-
 4 ber concentration of this population of nuclei, as well as its nature, is poorly
 5 constrained, which directly implies the need to assume a value of r_0 . Differ-
 6 ent initial radii may lead to different time rates of coagulation and, hence,
 7 to different haze profiles. As we will discuss in next section, the particle co-
 8 agulation depends on the concentration of particles and the collection kernel
 9 K_c , whose value for particles of radii r_i and r_j is computed assuming classical
 10 assumptions (Fuchs, 1964; Cabane et al., 1992):

$$11 \quad K_c(r_i, r_j) = 4\pi(D_i + D_j)(r_i + r_j)\beta \quad (5)$$

12 where D is the diffusion coefficient and β is a correction factor whose value is
 13 ≈ 1 for $\text{Kn}(\text{Knudsen number}) \ll 1$ or $\beta \approx V_{ij}(r_i + r_j)s/[4(D_i + D_j)]$ for $\text{Kn} \gg 1$,
 14 where V_{ij} is the the mean thermal velocity. Figure 13 shows the variation of
 15 K_c with particle radius at 3 different pressure levels of the haze production
 16 region. These K_c are normalized to the value of K_c for $r=0.0012 \mu\text{m}$. We
 17 observe similar values of K_c for radii between 0.0012 and $0.08 \mu\text{m}$, and a
 18 sharp decrease in K_c for $r > \sim 0.08 \mu\text{m}$. Figure 14 illustrates the results of our
 19 study of the variation of the haze profiles with r_0 by showing the number
 20 density and optical depth computed for a haze production rate of 2.5×10^{-14}
 21 $\text{kg m}^{-2} \text{s}^{-1}$ and different values of r_0 . In these simulations we can observe
 22 that the initial radius only has a significant effect on the haze concentration
 23 in the haze production region. At greater pressures, where the particles are
 24 bigger in size, the concentrations obtained using these different initial radii
 25 are very similar. This results from the fact that for these initial radii, the
 26 values of K_c are very large and similar, leading to very small timescales of

1 coagulation (practically the same); particles grow up in size very quickly in
2 the 3 cases. In these cases the variation in the estimated haze production
3 rate varies in about a 5 %. Similar simulations as those shown in Fig. 14
4 indicate that for initial radius greater than $0.05 \mu\text{m}$, the variations in the
5 haze profiles can be significant with variations in the haze optical depth $>$
6 25 %. However, this upper limit of r_0 represents a extreme case. Therefore,
7 although our model can be simplistic in simulating the vertical region where
8 the haze particles are initially formed, these tests indicate that our estimations
9 are not notably affected by the assumed initial radius or by variations in the
10 altitudes of maximum of production (variations that can be due to changes in
11 the temperature).

12 4 Comparisons with previous works

13 4.1 *Voyager-2 observations*

14 Our estimated haze production rate leads to an opacity of about 3.5 times
15 larger than that derived in Moses et al. (1995) from Voyager-2 observations
16 (1989). This is shown in Figure 15, that compares haze opacity profiles (at two
17 different wavelengths) computed for: a) the haze parameters given in Moses
18 et al. (1995), $M_0=1\times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ and $q=13$ electrons per μm (values
19 referred hereafter as M1995 haze parameters); b) the parameters derived in
20 our analysis (referred hereafter as T2020 haze parameters). For comparison
21 purposes, the optical depth for $M_0=6\times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ and $q=8.6$ electrons
22 per μm is also shown. It is important to note that Moses et al. (1995) used
23 a real refractive index (m) of 1.44 and a density of haze particles $\rho_i=1000 \text{ kg}$

¹ m^{-3} (also referred hereafter as M1995 haze parameters), while in this work
² these parameters were set to $m=1.4$ and $\rho_i=700 \text{ kg m}^{-3}$ (e.g. Karkoschka
³ and Tomasko, 2009; Irwin et al., 2016b; Pollack et al., 1987). In order to
⁴ study the impact of these different opacities on Neptune’s spectra, Figure 16
⁵ shows comparisons between VLT observations (at strong methane-absorbing
⁶ wavelengths) and simulations when the two sets of parameters are employed.
⁷ These results indicate that a reasonably good fit cannot be achieved with
⁸ VLT spectra and M1995 haze parameters (reduced $\chi^2 \sim 10$). Indeed, the haze
⁹ opacity obtained for M1995 parameters is too low to reproduce the reflectance
¹⁰ spectrum in H-band, and thus it is significantly smaller than our estimations.
¹¹ Next step in our analysis is to evaluate these results in terms of M_0 , whose
¹² estimation through the haze opacity requires some assumptions. First among
¹³ them is the real refractive index m whose value, for given profiles of number
¹⁴ density and size distribution, mainly determines the haze opacity. Left panel
¹⁵ of Figure 17 shows the variation of the haze opacity when m is increased,
¹⁶ and the rest of parameters are fixed to T2020 values. For a given M_0 , an
¹⁷ increase in m results in greater opacities. Thus, the larger is the assumed real
¹⁸ refractive index, the smaller the retrieved haze production rate. We examined
¹⁹ the impact of using a real refractive index $m=1.44$, and found that the best fit
²⁰ is achieved for a haze production rate of $\sim 2 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$, about 25 %
²¹ smaller than that obtained for $m=1.4$. Another parameter whose value has a
²² significant impact on the retrieval of M_0 is the density of the haze particles ρ_i .
²³ For a given M_0 , a greater ρ_i results in a fewer number of haze particles created
²⁴ per second, and thus a lower opacity (note that the units of M_0 are kg m^{-2}
²⁵ s^{-1}). Right panel of Fig. 17 shows the variation of the haze opacity when ρ_i is
²⁶ increased and the rest of parameters are fixed to T2020 values. In this case, the
²⁷ larger is the assumed ρ_i value, the smaller the number of particles produced

1 per second, and the larger is the M_0 needed to fit VLT observations. We found
 2 that for $\rho_i=850$ and 1000 kg m^{-3} , the latest the one utilized in Moses et al.
 3 (1995), the best fits are obtained for $M_0= \sim 3.5 \times 10^{-14}$ and $4.5 \times 10^{-14} \text{ kg m}^{-2}$
 4 s^{-1} , respectively. For the rest of model parameters whose values are required
 5 (such as the initial model particle radius or the eddy diffusion coefficient), from
 6 the results shown in section 3.5 and this section we find that their impact on
 7 the estimation of M_0 is negligible compared to that of m or ρ_i . Therefore, based
 8 on these results we conclude that our analysis provides a haze production rate
 9 significantly greater, at least by a factor of ~ 2 , than that derived in Moses
 10 et al. (1995).

11 4.2 Photochemical models

12 In our analysis, a haze production rate of $2.6 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ was derived
 13 for Neptune, about 2.4 times smaller than the $\sim 6 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ value
 14 predicted by the photochemical models (Moses et al., 1995), or about 2.5 times
 15 larger than that derived from Voyager observations. Figure 18a illustrates a
 16 comparison between VLT spectra and simulations performed for a production
 17 rate $M_0=6 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ and the rest of haze parameters set to T2020
 18 values. These results indicate that this haze production rate leads to opaci-
 19 ties not consistent with VLT observations (haze opacity profile illustrated in
 20 Fig. 15). Note that a reduced χ^2 of ~ 40 is achieved. In previous section, we
 21 showed the impact of m and ρ_i on the haze opacity for a given M_0 (see Fig. 17).
 22 Hence, we varied the values of these parameters to study if a better fit can be
 23 achieved. Fig. 18b shows, as example, a comparison similar to that in 18a but
 24 for a greater ρ_i . Although χ^2 remains large, we can see a noticeable improve-

1 ment in the fit (reduced $\chi^2 \sim 4$). As similar improvements can be achieved
 2 by decreasing m (see Fig. 17), we performed a number of fits similar to those
 3 shown in Fig. 18 but for different combinations of these two parameters. Figure
 4 19 illustrates the contour plots of χ^2 in the ρ_i - m space of the analysis, along
 5 with 3σ significance level of χ^2 (red solid lines). Note that for this analysis the
 6 haze production rate is fixed to $M_0 = 6 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$. The contours of χ^2
 7 indicate two minima with m values of 1.3 and 1.35, and densities of 840 and
 8 1000 kg m^{-3} , respectively. In these cases, the reduced χ^2 values obtained are
 9 smaller than 1. Thus, good fits between simulations and VLT observations are
 10 achieved using these m and ρ_i values, and the haze production rate predicted
 11 by the photochemical models. Although it is difficult to place firm constraints
 12 on the density of haze particles, the m values estimated in this analysis can
 13 be compared with the real refractive index of the hydrocarbon ices expected
 14 to form in Neptune's stratosphere. Photochemical simulations indicate that
 15 ethane clearly dominates the haze production rate of the methane photolysis
 16 products (Moses et al., 1995, 2018). Hence, we expect haze particles to have a
 17 real refractive index $m \sim 1.4$, which is characteristic of ethane ice particles.
 18 Since the real refractive index values estimated in the analysis of Fig. 19 are
 19 significantly smaller, we conclude that these solutions are not consistent with
 20 the photochemical simulations.

21 For the haze profiles computed for $M_0 = 6 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$, we also investi-
 22 gated the impact of setting the haze number density to 0 at altitudes below
 23 a certain level in the troposphere (referred thereafter as haze-base pressure
 24 p_{haze}). If haze particles are formed by the condensation of hydrocarbons such
 25 as ethane, then these particles should partially or totally evaporate in the
 26 regions of the atmosphere for which the saturation ratio falls below 1. That
 27 is to say, they may begin to evaporate before the fall to the tropospheric-

1 cloud level. Left panel of Figure 20 shows haze opacity profiles computed for
 2 $M_0=6\times 10^{-14}$ kg m⁻² s⁻¹ and with the haze-base pressure established at 0.36,
 3 0.67 and 1 bar. Note that the haze opacity at pressures greater than p_{haze}
 4 is equal to 0. We performed a number of fits similar to that shown in Fig.
 5 18a but setting the haze-base pressure at different levels. The χ^2 variation of
 6 these fits with p_{haze} is illustrated in the right panel of Fig. 20. We can see that
 7 reasonably good fits are achieved for $p_{haze} \sim 0.55$ bar (reduced $\chi^2 \sim 1.1$). This
 8 requires that haze particles should completely evaporate below this pressure
 9 level. Abundances of ethane in the troposphere indicate saturations below 1
 10 at altitudes below the ~ 0.3 -bar level (Moses et al., 1992). Hence, we conclude
 11 that the scenario of haze particles completely evaporated before reaching the
 12 levels above tropospheric clouds is plausible. This would result in a significant
 13 decrease in the haze opacity with pressure in the subsaturated region, and thus
 14 explain the discrepancies found in the estimation of M_0 . However, to validate
 15 this hypothesis future microphysical simulations that include the evaporation
 16 rates of haze particles are required.

17 **5 Timescale analysis and comparisons with Uranus**

18 Once the haze properties were retrieved, our next step is to determine how
 19 temporal variations in the haze production rate (due to variations in the solar
 20 illumination at the top of the atmosphere) can affect the structure (number
 21 density and size distribution) of the haze. To this end, we performed, as in
 22 Toledo et al. (2019), a haze microphysics timescale analysis. We first set M_0
 23 and q to the values estimated in section 3.2, and then we run the model,
 24 started at time $t=0$ with no haze in the atmosphere, until the steady state is

1 achieved (usually for $t > 140$ years). At this moment the haze particle size and
 2 density at a given altitude do not change with time. The haze microphysics
 3 timescale, defined as t_M , at a given altitude of the atmosphere is given by the
 4 time required to achieve the steady state at that altitude. The left panel of
 5 Figure 21 shows the haze number density obtained for $t = 10, 25, 50$ and 180
 6 years. Note that the profile obtained for $t = 180$ years represents the steady
 7 state of these simulations. Comparing the density profiles for $t = 10$ and 180
 8 years, we observe that the 10-years profile converges towards the steady-state
 9 profile for altitudes above the 0.03-bar pressure level. At this altitude, any
 10 change in the haze production rate would result in variations in the haze
 11 properties 10 years after such a change. Proceeding in the same way for the
 12 rest of profiles, we can compute the haze timescale t_M curve as a function
 13 of pressure. These results, along with the timescale of Uranus's haze derived
 14 in Toledo et al. (2019), are displayed in the central panel of Fig. 21. These
 15 simulations indicate that: a) to observe changes in the haze structure (due to
 16 changes in the haze production rate) at the 0.1-bar and 1-bar levels, a time
 17 lapse of 20 and 70 years must occur, respectively; b) at a given altitude, the
 18 haze microphysics timescale in Neptune is shorter than that in Uranus, and
 19 the difference in time between both rates ($\Delta t_M = t_M(\text{Neptune}) - t_M(\text{Uranus})$)
 20 increases with pressure. For instance, while at the 0.01-bar level Δt_M is ~ 10
 21 years, at altitudes below the 0.1-bar level Δt_M is greater than 50 years.
 22 These differences in the haze timescales of both planets arise from the different
 23 time rates of the processes involved in the continuity equation: coagulation,
 24 settling and eddy diffusion. In left panel of Fig. 11 it has been shown that haze
 25 particles in Neptune are greater in size than those in Uranus as a result of
 26 the larger haze production rate. These differences in size lead to different haze
 27 settling-velocities. This can be observed in the right panel of Fig. 21 that shows

1 the haze settling-velocities in both planets for the sizes given in left panel of
 2 Fig. 11. From the maximum of number density, these results indicate similar
 3 settling velocities up to the 0.02-bar level, where the differences in size of haze
 4 particles of both planets become more significant (see left panel of Fig. 11).
 5 From these velocities we can compute the timescale for sedimentation, t_{fall} ,
 6 that is defined as the time required for haze particles to fall a distance H :

$$7 \quad t_{fall} = \frac{H}{v_{fall}} \quad (6)$$

8 The velocities v_{fall} as a function of pressure are given in Fig. 21c and the
 9 distance H is the pressure scale height ($H=29$ km for Neptune and $H=30$ km
 10 for Uranus). The timescales for sedimentation computed from equation 6 are
 11 illustrated in Figure 22a. These results indicate: a) t_{fall} is very similar in both
 12 planets for altitudes above the 0.02-bar level; b) t_{fall} in Neptune is smaller
 13 than t_{fall} in Uranus for $p > 0.02$ bar; c) the difference between t_{fall} in Nep-
 14 tune and Uranus increases with pressure. By comparing the haze timescales
 15 displayed in the central panel of Fig. 21 with the t_{fall} rates given in Figure
 16 22a, we can observe, for pressures greater than 0.03 bar, the high correlation
 17 between the differences in t_{fall} of both planets and the differences in the haze
 18 timescales Δt_M discussed above.

19 Although the vertical profiles of v_{fall} can explain the increase of Δt_M with
 20 pressure, it is important to note that above the 0.03-bar level any $\Delta t_M > 0$
 21 cannot be explained by t_{fall} since the value of this time rate above that level is
 22 very similar in both planets. The density profiles displayed in central panel of
 23 Fig. 11 show that the 0.03-bar level corresponds closely to the lower boundary
 24 of the haze production region, where the particles grow in size very quickly
 25 due to coagulation. It is reasonable to believe, therefore, that above the 0.03-
 26 bar level, the differences in the haze microphysics timescales of both planets

1 are mainly due to the different time rates of coagulation. The timescale of
 2 coagulation, t_{coag} , is defined as the time required to reduce the haze number
 3 density by 50 %. This time rate depends on the collection kernel K_c , cal-
 4 culated for spherical particles using classical assumptions (Fuchs, 1964), and
 5 the haze number density. To estimate t_{coag} we started with the haze given by
 6 the steady profiles, and then we run our haze microphysics model up to the
 7 time for which the haze concentration has been reduced to 50 % at all the
 8 altitudes. For a given altitude, t_{coag} is the exact time at which the density is
 9 reduced by 50 %. In order to ensure that any changes in the haze number
 10 density are only due to coagulation, these simulations are performed with the
 11 haze production rate, the eddy-diffusion profile and the settling velocities set
 12 to $M_0=0$, $K=0$ and $v_{fall}=0$. Fig. 22b shows the coagulation timescales for
 13 Neptune and Uranus as a function of pressure. Note that because both the
 14 number density and K_c changes with pressure, t_{coag} varies with altitude; at
 15 higher altitudes where the number density is large and the haze radii small
 16 t_{coag} takes smaller values and, hence, it is at these altitude where coagulation
 17 dominates the temporal variations of the haze concentration $C(r,z)$. We also
 18 observe that t_{coag} is about 10 times smaller in Neptune than in Uranus at the
 19 altitudes where the haze is produced as a result of its bigger haze concen-
 20 tration ($M_0(\text{Neptune}) \sim 10 \times M_0(\text{Uranus})$). Therefore, these results show that
 21 above the 0.03-bar level the haze timescale t_M is dominated by the particle
 22 coagulation, and this time constant is smaller in Neptune as a result of its
 23 bigger haze production rate (about 10 times).
 24 The last process involved in the continuity equation, and therefore in the time
 25 variation of $C(r,z)$, is the eddy diffusion. The time rate of eddy diffusion, de-
 26 fined as t_{diff} , represents the time required for haze particles to be mixed over

1 a distance H and it is given by

$$2 \quad t_{diff} = \frac{H^2}{K} \quad (7)$$

3 where H is the pressure scale height and K is the eddy diffusion coefficient
 4 given in Fig. 1. The timescales of eddy diffusion computed from equation 7
 5 are displayed in 22c. These results indicate that as a result of the bigger eddy-
 6 diffusion coefficient, the eddy mixing in Neptune's atmosphere is about 10
 7 times more efficient (in terms of t_{diff}) than that in Uranus's. For a given alti-
 8 tude, however, the importance of the eddy diffusion coefficient to the temporal
 9 variation of $C(r,z)$ is determined by the values of the time rates t_{diff} , t_{coag} and
 10 t_{fall} . For instance, if we find that $t_{diff} \gg t_{coag}$ and t_{fall} , then the eddy mix-
 11 ing does not have a major impact on vertical structure of the haze. In order
 12 to study the 3 processes simultaneously, Fig. 22d compares the time rates of
 13 t_{diff} , t_{coag} and t_{fall} in Neptune's atmosphere. This comparison indicates that
 14 for $p < 0.04$ bar (haze production region), t_{coag} is much smaller than t_{fall} and
 15 t_{diff} . At these altitudes the haze number density of particles with $r \sim r_0$ is
 16 limited by coagulation after ~ 1 day of being produced. For these radii the
 17 particles remain at the same altitude and undergo changes mainly due to par-
 18 ticle coagulation ($t_{coag} \ll t_{fall}, t_{diff}$). As the haze particles grow in size, particle
 19 removal due to sedimentation takes place, limiting the number of particles of
 20 a given size. We can observe that the altitude for which $t_{fall} = t_{coag}$ is at the
 21 0.04-bar level. This level establishes the altitude at which particles with radii
 22 of $\sim 0.2 \mu\text{m}$ are transported by sedimentation a distance H in about the time it
 23 takes for coagulation to reduce the concentration in about 50 %. Therefore, at
 24 altitudes below the 0.04-bar level, it is the settling velocities v_{fall} that mainly
 25 control the time variation of $C(r,z)$. As in Uranus' atmosphere (Toledo et al.,
 26 2019), the timescale of eddy diffusion was found to be much bigger than t_{fall}

1 and t_{coag} at all the altitudes, indicating the little impact of this parameter on
 2 the time variation of $C(r,z)$. We also tested the effect that would result from
 3 an increase in the eddy diffusion near the tropopause (instead of the decrease
 4 illustrated in Fig 1), and we found that our results would not change as long
 5 as K values are smaller than $\sim 10000 \text{ cm}^2 \text{ s}^{-1}$.
 6 Our timescale analysis shows that the timescales for aerosols to grow and fall
 7 are of about 15 and 30 years at the observable pressure levels in the atmo-
 8 spheres of Neptune and Uranus, respectively. The timescale analysis reveals
 9 that the time lapsed between both observations is long enough to observe
 10 changes in the haze structure due to microphysics. However, this time lapse
 11 represents only a $1/7$ of a Neptunian year and for which the solar insolation
 12 has not changed significantly at the studied latitudes. Therefore, a signifi-
 13 cant increase of the haze production rate is not expected, due to variations
 14 in the solar insolation at the top of the atmosphere. The different values of
 15 M_0 estimated in Moses et al. (1995) and this work may arise from the differ-
 16 ent kind of data used to constrain the haze parameters. While Moses et al.
 17 (1995) analyzed photometric images taken at 5 different filters in the visible,
 18 in the present work observations in the infrared were used. As demonstrated
 19 in section 3.1, the choice of using observations in the H-band is based on the
 20 fact that the observations are practically independent of the haze imaginary
 21 refractive index for the emission-angle ranges analysed in section 3.2, allowing
 22 a notable reduction of the number of free parameters. However, this differ-
 23 ent spectral range used to constrain the haze properties may lead to different
 24 estimations of M_0 (visible observations are more sensitive to small particles).
 25 These discrepancies between both analyses can be studied in future works by
 26 applying our model to visible observations or comparing our estimations of M_0
 27 with photochemistry simulations since, as pointed by Moses et al. (1995), the

1 haze production rate estimated in their analysis is substantially lower (about
2 5 times) than the predictions from photochemical simulations (Moses et al.,
3 1992, 1995; rom, 1993).

4 Finally, we can also consider the possibility of temporal and latitudinal varia-
5 tions of the haze production rate as a result of other factors such as a merid-
6 ional transport of aerosols from regions of upwelling to regions of subsidence.
7 This implies that the timescales for a stratospheric meridional transport of
8 haze particles are smaller than those given for microphysics (or in terms of
9 velocities that the stratospheric meridional winds are faster than the haze
10 settling velocities). The timescale for a haze particle to be transported a dis-
11 tance equal to the radius of Neptune (R) can be estimated using the following
12 expression

$$13 \quad t = \frac{R}{w} \quad (8)$$

14 where w is the meridional wind. From Eq. 8 and the timescales shown in Fig.
15 21, we can compute for each altitude the equivalent wind velocity (V^*) that
16 represents the wind velocity for which the timescale for haze meridional trans-
17 port equals the microphysics timescale. If the meridional wind velocity is larger
18 than the equivalent wind velocity, then the haze spatial distribution and its
19 temporal evolution are mainly dominated by dynamics. Figure 23 shows the
20 haze microphysics timescales and the equivalent wind velocities as a function
21 of the pressure, and where we observe that if the meridional wind velocities
22 are larger than $\sim 0.03 \text{ ms}^{-1}$ then the dynamics is the main factor controlling
23 the haze distribution in the atmosphere and therefore, the cause of the varia-
24 tions in M_0 . Estimations from feature tracking indicate that meridional wind
25 velocities may exceed this velocity at some latitudes (Tollefson et al., 2018).
26 However, these estimations are limited to the altitudes of clouds and may not

1 be representative of a meridional stratospheric circulation. Therefore, to fully
 2 study whether the haze spatial distribution and temporal evolution are dom-
 3 inated by dynamics, GCM simulations are required. These simulations will
 4 determine the role played by the global circulation in the distribution of
 5 the haze over the planet.

6 **6 Conclusions**

7 We have used a 1-dimensional microphysical model to simulate the haze for-
 8 mation and evolution in Neptune’s atmosphere. These simulations were cou-
 9 pled with the NEMESIS radiative-transfer code to model infrared data in
 10 the H-band from VLT/SINFONI acquired in 2013. The choice of using obser-
 11 vations in the H-band to constrain the haze properties is based on the fact
 12 that these observations for the emission-angle ranges analysed are not signif-
 13 icantly affected by the imaginary refractive index of haze particles, reducing,
 14 therefore, the number of free parameters required to fit the observations. The
 15 main goal of combining microphysics and radiative-transfer simulations was to
 16 better estimate the haze vertical profiles of size distribution and number den-
 17 sity, which are extremely challenging to constrain from radiative-transfer and
 18 ground-based observations alone, and to derive the timescales of microphysics.
 19 Our simulations show a haze production rate of $M_0 \sim (2.61 \pm 0.18) \times 10^{-14}$ kg
 20 $\text{m}^{-2} \text{s}^{-1}$ and a particle electric charge of $q \sim 8.6 \pm 1.1$ electrons per μm radius at
 21 latitudes between 5 and 15° S. This haze production rate is of about ten times
 22 greater than that derived for Uranus’s atmosphere. Our analyses also indicate
 23 the presence of a tropospheric cloud with a total optical depth greater than 10
 24 at 1.46 μm (about 250 times greater than the haze optical depth). The cloud

1 base, which was found near the 2.5-bar level, may indicate only the top of a
 2 thicker and deeper cloud as a result of the large retrieved opacity.
 3 The haze opacity derived in this work is about 3.5 times larger than that ob-
 4 tained by Moses et al. (1995) from Voyager-2 observations in 1989. In terms of
 5 M_0 , we found that this larger opacity indicates a production rate at least two
 6 times larger than the one derived from Voyager-2 observations. The analysis of
 7 section 5 indicates that the haze microphysics timescales are of about 15 years
 8 at the 0.1-bar pressure level. As a consequence, temporal variations in the haze
 9 production rate due to seasonal variations in the solar illumination at the top
 10 of the atmosphere might affect the haze structure during the time lapsed be-
 11 tween Voyager-2 and 2013. Although this seasonal variability may explain the
 12 variations of M_0 , the elapsed time between these observations represents only
 13 a $\sim 1/7$ of a Neptunian year with a monthly mean solar insolation that has
 14 not changed significantly at the studied latitudes. The different estimations
 15 of M_0 may be due to: a) in our analysis we used infrared data to constrain
 16 the haze properties, while Moses et al. (1995) analyzed photometric images at
 17 5 different filters in the visible. While high-phase-angle Voyager observations
 18 are more sensitive to small haze particles and at altitudes above the 0.1-bar
 19 level, the haze constraints derived from VLT spectra in H-band are limited
 20 to pressures greater than 0.1 bar. As a result of the different phase angles of
 21 the two set of observations, differences in the estimation of M_0 may arise from
 22 the use of Mie phase functions as well. b) the transport of haze particles from
 23 different regions of the planet. Meridional winds greater than the equivalent
 24 wind velocities illustrated in Fig. 23 may result in temporal and latitudinal
 25 variations in the haze structure and, hence, in M_0 . To further address this
 26 point, however, GCM models capable of modelling the transport of haze and
 27 the photochemistry are required. These simulations compared or coupled with

1 haze microphysics will determine how efficient dynamics is in distributing the
2 haze in the atmosphere.

3 Although the haze production rate estimated in this work is significantly larger
4 than that from Voyager observations, photochemical models point to even
5 larger production rates on Neptune (by a factor of 2.4). Assuming that the
6 photochemical simulations are correct, we found that this discrepancy can be
7 explained if haze particles evaporate before reaching the tropospheric-cloud
8 levels. This scenario would decrease the cumulative haze opacity above the
9 1-bar level, and thus a larger haze production rate would be required to fit
10 VLT observations. However, to validate such a haze vertical structure future
11 microphysical simulations that include the evaporation rates of haze particles
12 are required.

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Table 1

Values of p_{0i} and M_{0i} (see equations 1-2) used in model-1, model-2 and model-3 for the simulation of the haze production region.

	Model-1	Model-2	Model-3
Region-1	$p_{01}=0.016\text{bar}$	$p_{01}=0.016\text{bar}$	$p_{01}=0.016\text{bar}$
Region-2	-	$p_{02}=0.008\text{bar}$	$p_{02}=0.009\text{bar}$
Region-3	-	$p_{03}=0.011\text{bar}$	$p_{03}=0.011\text{bar}$
Region-1	$M_{01}=M_0$	$M_{01}=0.8\times M_0$	$M_{01}=0.81\times M_0$
Region-2	-	$M_{02}=0.1\times M_0$	$M_{02}=0.16\times M_0$
Region-3	-	$M_{03}=0.1\times M_0$	$M_{03}=0.03\times M_0$

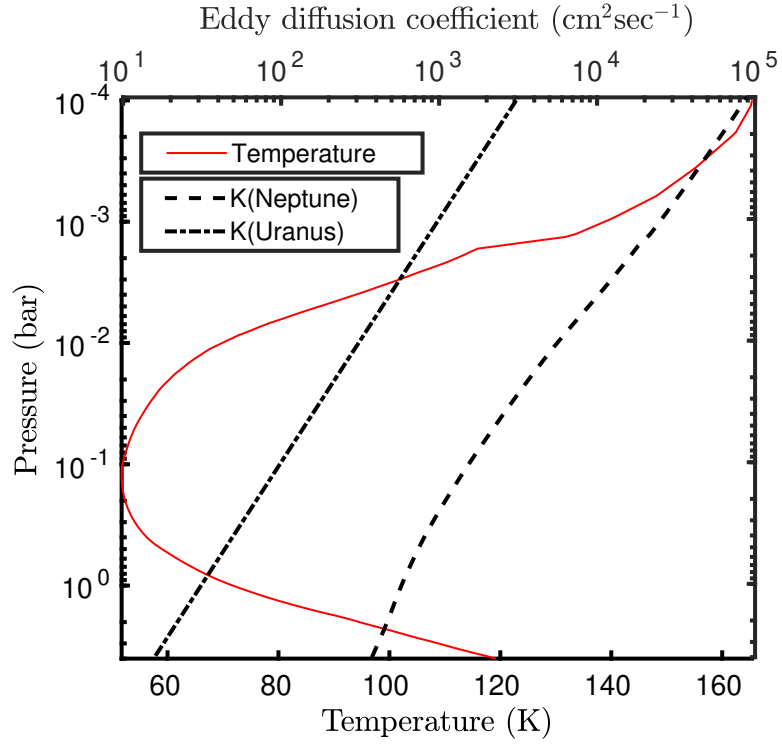


Fig. 1. Variation of temperature and eddy diffusion coefficient with height in Neptune's atmosphere. For comparison purposes, the eddy diffusion coefficient in Uranus's atmosphere is also shown.

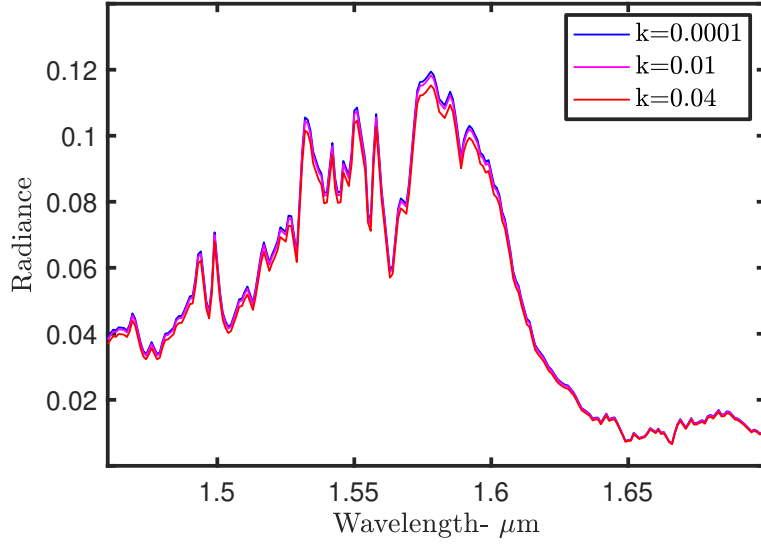


Fig. 2. H-band spectrum ($\mu\text{W cm}^{-2} \text{ sr}^{-1} \mu\text{m}^{-1}$) simulated for a haze $r_{eff}=0.3 \mu\text{m}$ and different values of the haze imaginary refractive index. The tropospheric cloud properties derived in section 3.3 were used for these simulations.

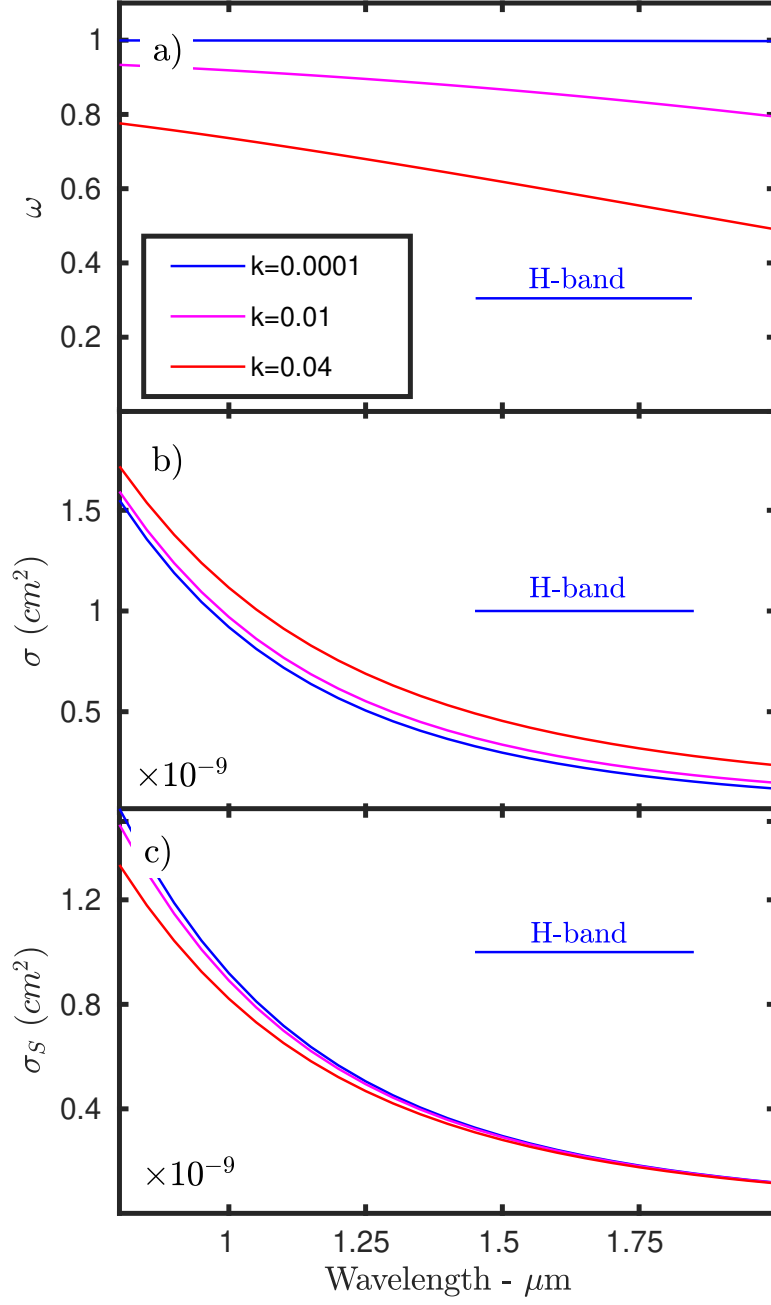


Fig. 3. Spectrum of the single-scattering albedo (panel a), cross section (panel b) and scattering cross-section (panel c) computed with Mie theory for a $r_{eff}=0.3 \mu\text{m}$ and different values of the imaginary refractive index. In all the case the real refractive index was set to 1.4.

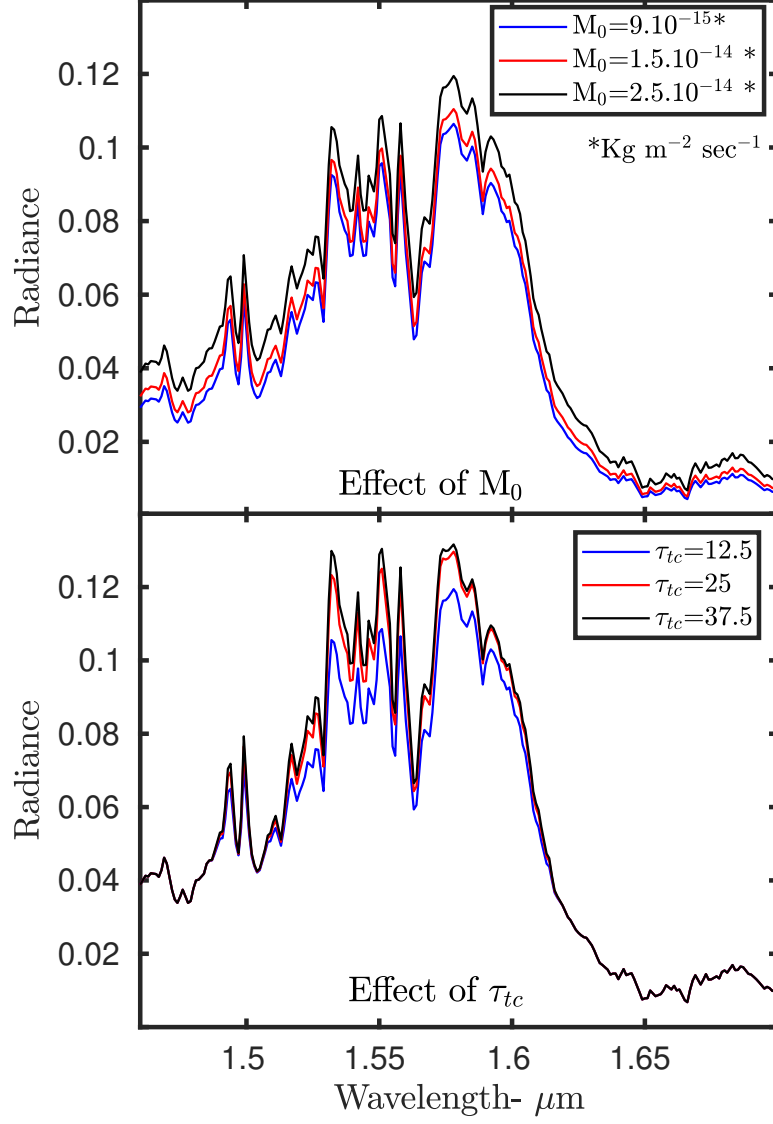


Fig. 4. Variation of the VLT simulated spectrum ($\mu\text{W cm}^{-2} \text{sr}^{-1} \mu\text{m}^{-1}$) with the haze production rate (upper panel) and the tropospheric cloud opacity (lower panel).

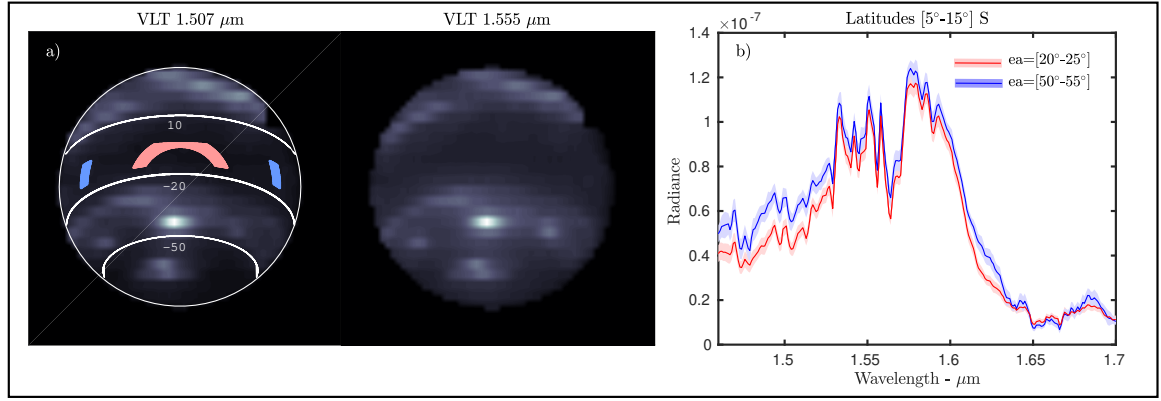


Fig. 5. (a) Very Large Telescope (VLT) images of Neptune at 1.507 and 1.555 μm made in October 2013. The areas highlighted in red and blue indicate pixels at latitudes between 5° and 15° S and with emission angles of between 20 and 25° (red), and of between 50 and 55° (blue). (b) Spectra ($\mu\text{W cm}^{-2} \text{sr}^{-1} \mu\text{m}^{-1}$) obtained from the average of the observations lying in the red area and in the blue area. The shaded errors represent the 1-standard deviation.

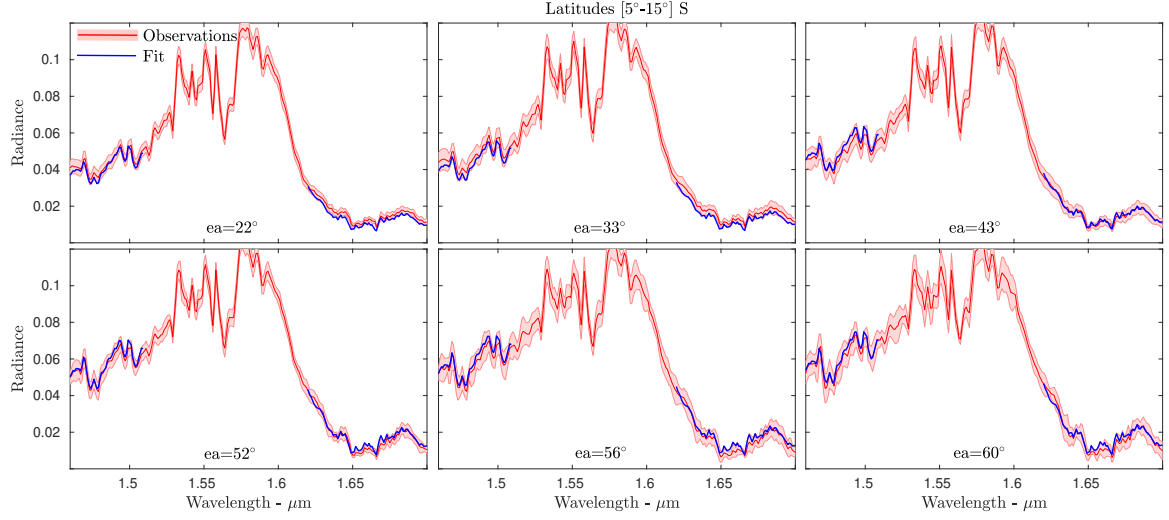


Fig. 6. Comparisons between simulations and observations at latitudes between 5° and 15° S. The thick red solid lines represents VLT spectra computed as in Fig. 5 but for the emission angle (ea) ranges $[20^\circ, 25^\circ]$, $[30^\circ, 35^\circ]$, $[40^\circ, 45^\circ]$, $[50^\circ, 54^\circ]$, $[55^\circ, 58^\circ]$, $[59^\circ, 60^\circ]$. The blue lines represent the simulations performed for a haze production rate of $2.5 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ and an electric charge of $q=10$ electrons per μm radius.

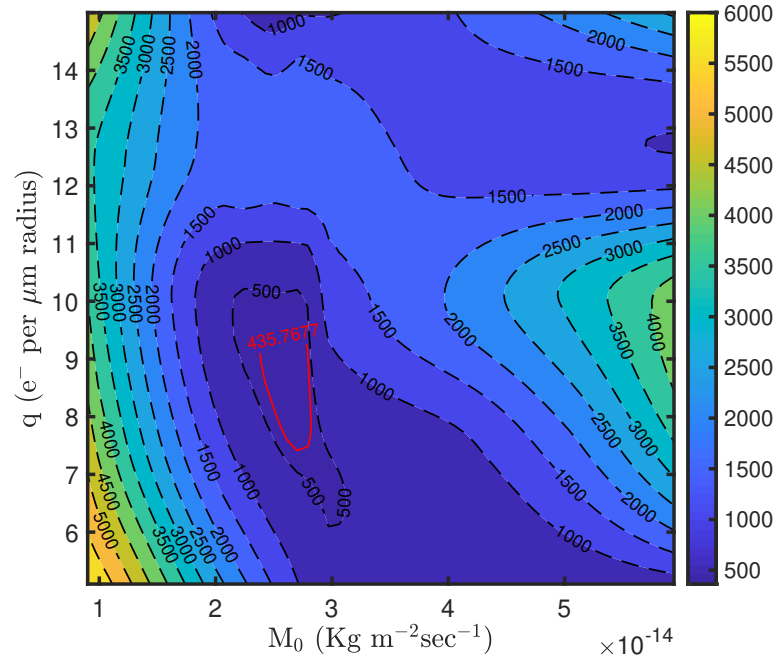


Fig. 7. Contours of χ^2 in the M_0 - q parameter space for the spectra given in Fig. 6. The red solid line shows the 3σ significance level.

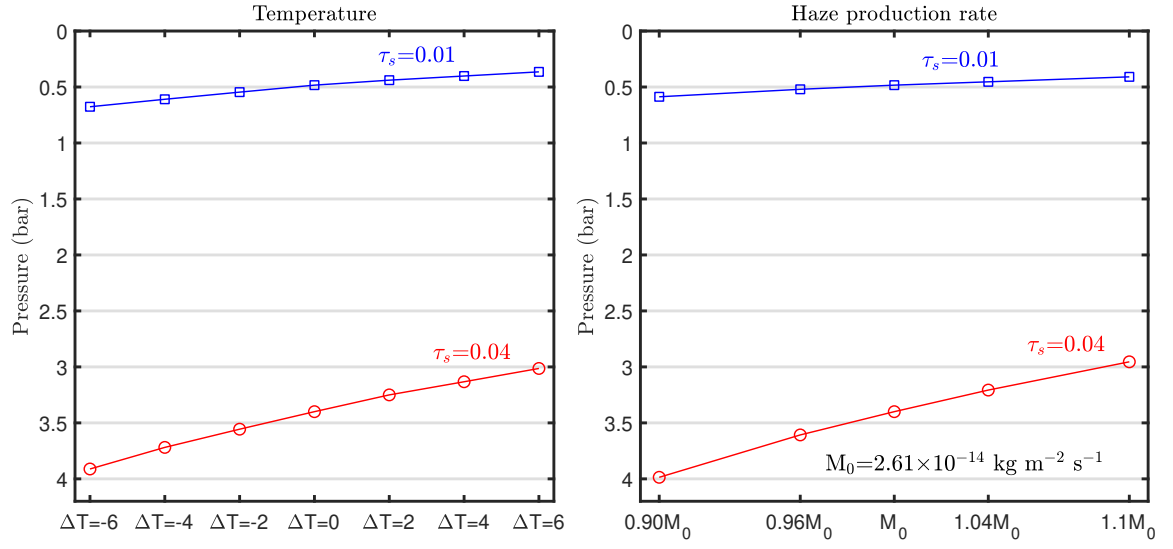


Fig. 8. Pressure level at which the haze cumulative optical depth τ_s is 0.01 (blue line) and 0.04 (red line) for different ΔT (left) and haze production rates M_0 (right).

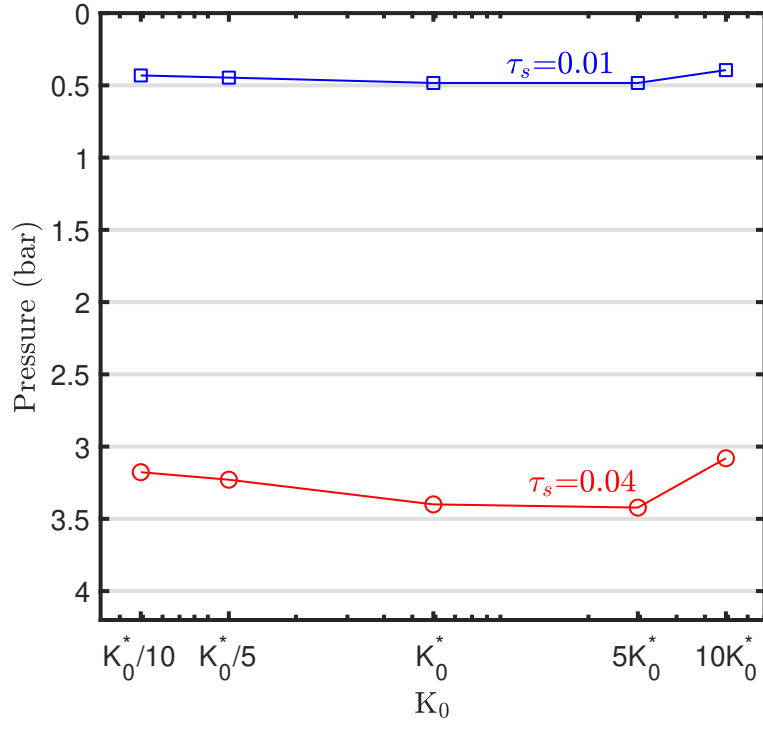


Fig. 9. Pressure level at which the haze cumulative optical depth τ_s is 0.01 (blue line) and 0.04 (red line) for different values of K_0 (see equation 4). K_0^* represents the value used for the profile shown in Fig. 1.

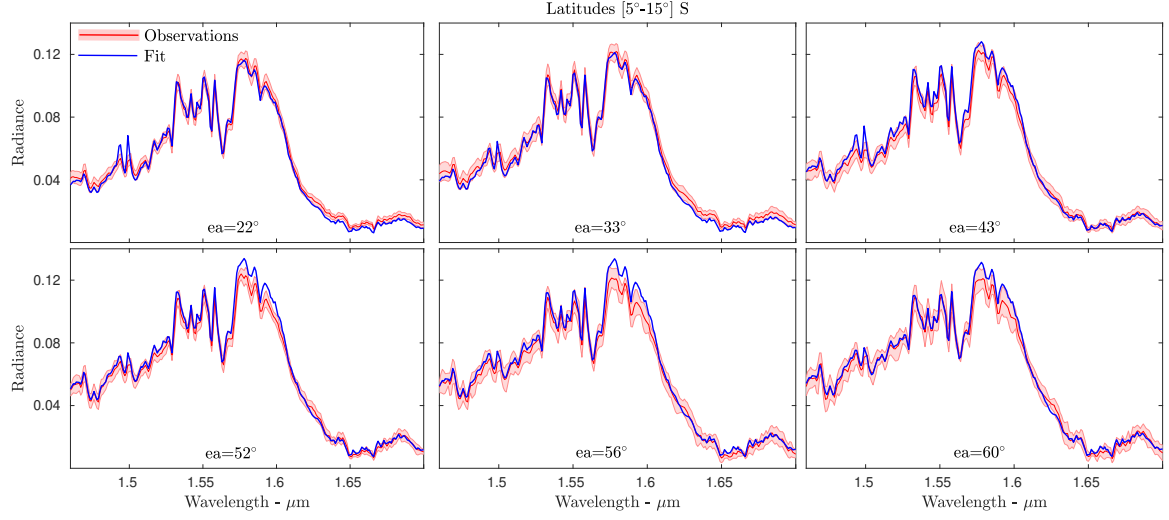


Fig. 10. Comparisons between simulations and observations at latitudes between 5° and 15° S. The thick red solid lines represents VLT spectra illustrated in Fig. 6, and the blue lines are simulations obtained from the optimal solutions of the tropospheric cloud parameters and the haze parameter derived in section 3.3.

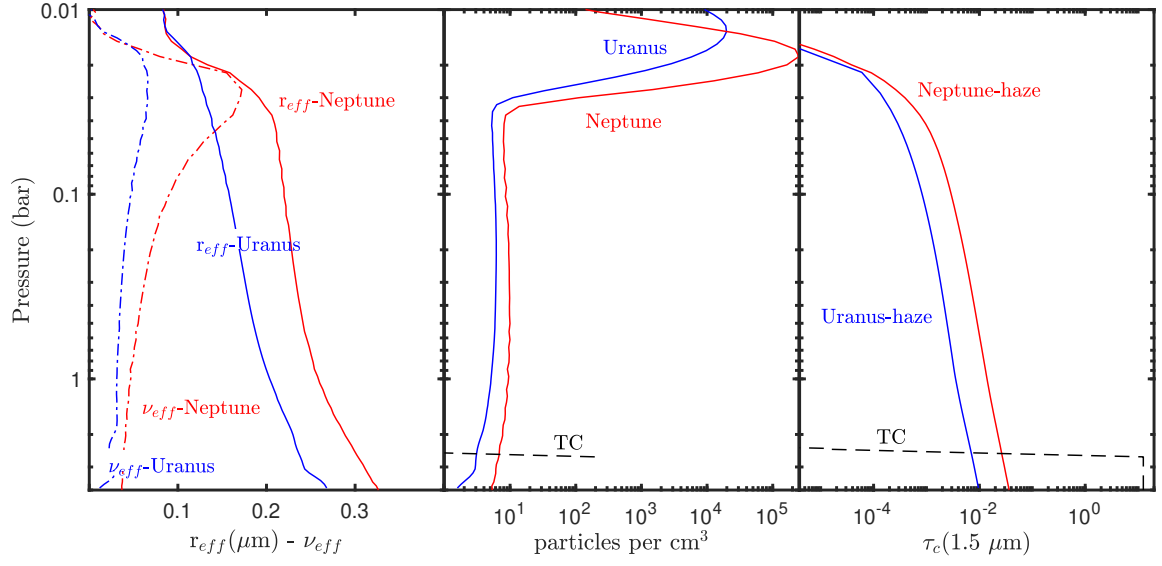


Fig. 11. Aerosol vertical profiles of size distribution (left panel), number density (central panel) and cumulative optical depth (right panel) derived in our analysis. For comparison purposes, the haze profiles in Uranus derived in Toledo et al. (2019) are also shown. The black dashed lines in the central and right panels indicate the number density and opacity of the tropospheric cloud (TC).

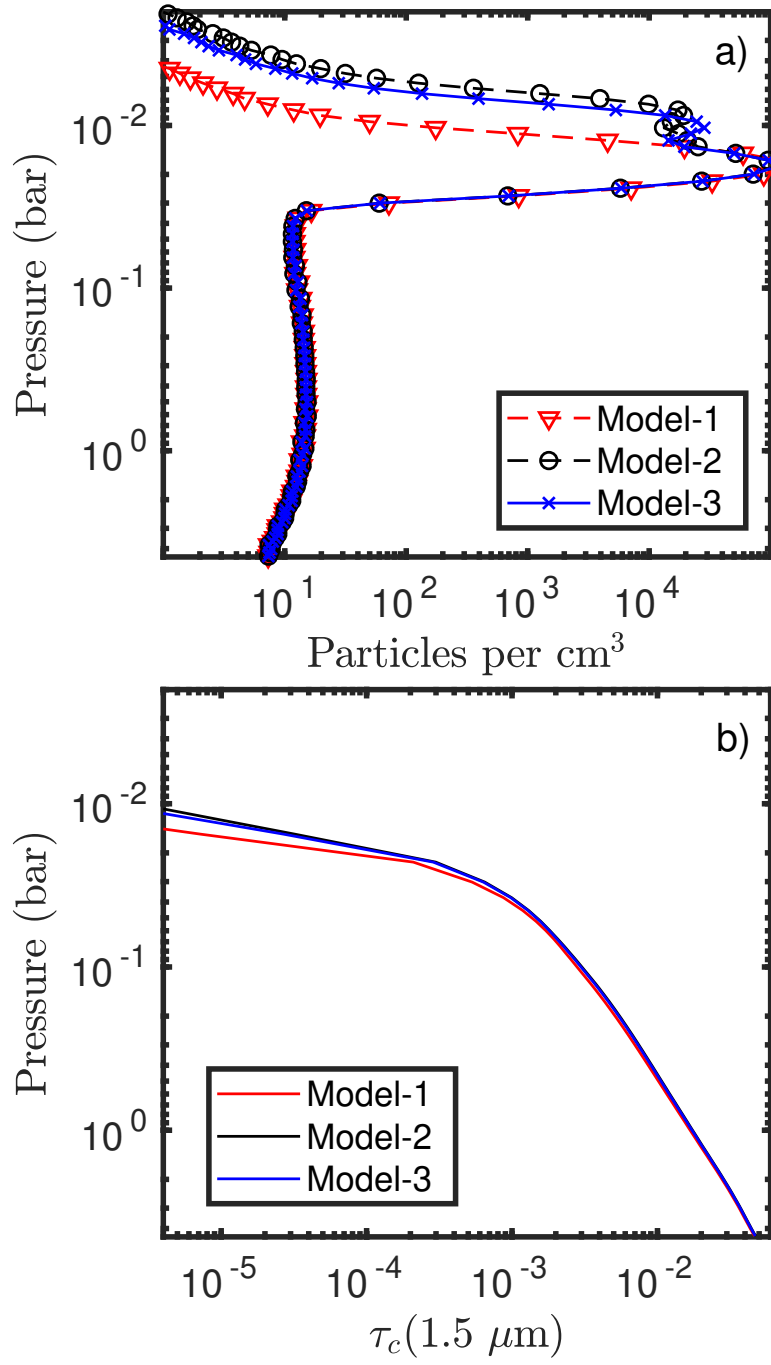


Fig. 12. Haze profiles of number density (upper panel) and cumulative optical depth (lower panel) computed with our haze microphysical model for the haze production models illustrated in Table 6. The optical depth was computed assuming a constant refractive index of $n=1.4+0.0001i$.

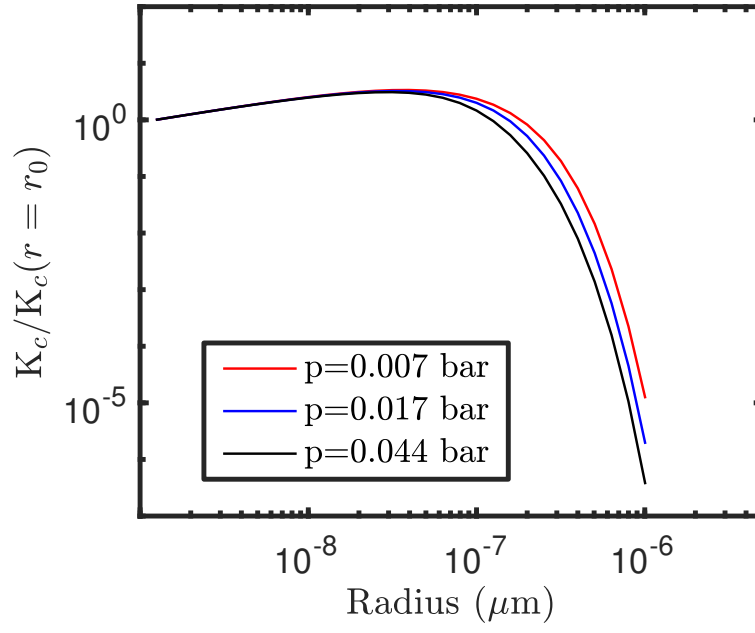


Fig. 13. Variation of the collection kernel K_c with particle radius at 3 different pressure levels of the atmosphere. The values are normalized to the value of K_c for $r=r_0$.

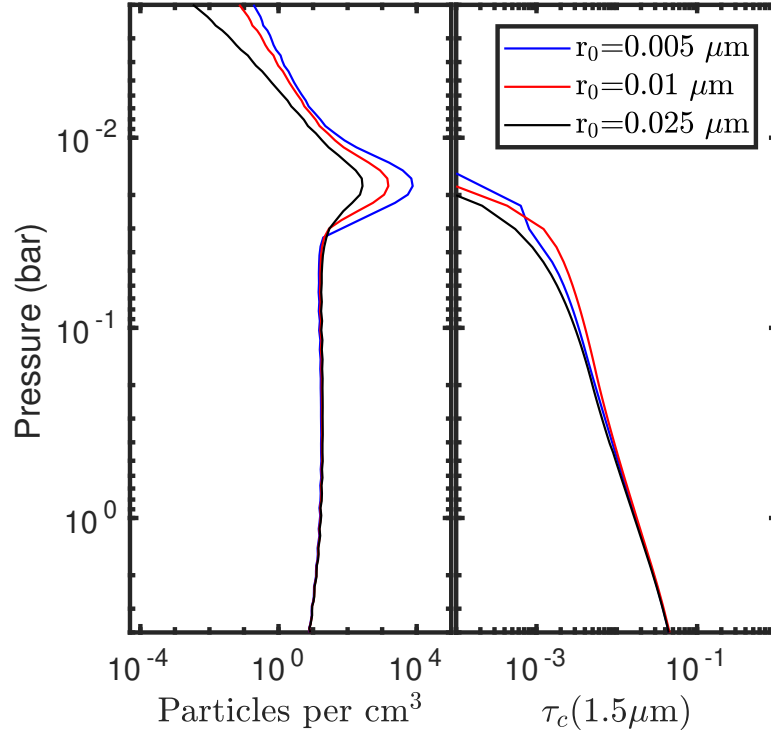


Fig. 14. Haze profiles of number density (left panel) and cumulative optical depth (right panel) computed with our haze microphysical model for different initial model radii r_0 and a haze production rate of $2.5 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$.

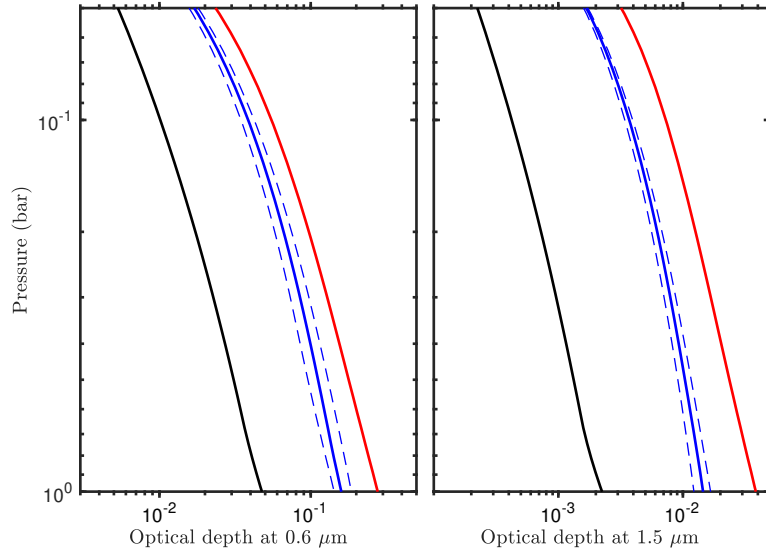


Fig. 15. Cumulative optical depth at 0.6 and 1.5 μm computed for the haze parameters derived in section 3.3 (blue solid lines with errors represented by dashed blue lines) and for M1995 haze parameters (black solid lines). For comparison purposes, the optical depth obtained for a haze production rate of $6 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ is also given (red solid lines).

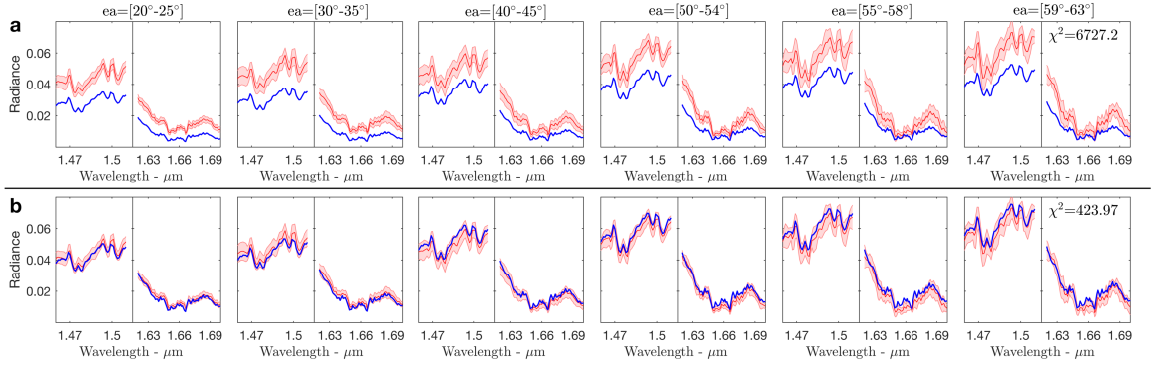


Fig. 16. Comparison between simulations and observations at latitudes between 5° and 15° S. The thick red solid lines represent the VLT spectra illustrated in Fig. 6, and the blue lines are simulations performed for M1995 haze parameters (a) and for the haze optimal solution derived in section 3.3 (b)

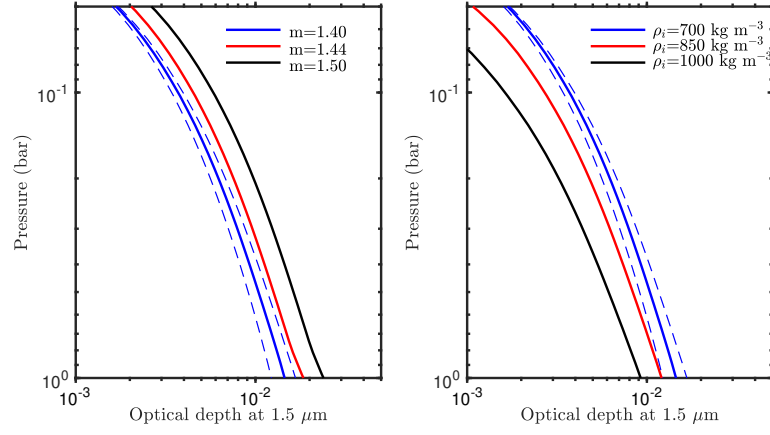


Fig. 17. Variation of the haze optical depth with the real refractive index (left panel) and the density of the haze particles (right panel). In both cases the blue solid line with errors (dashed lines) represents the profile obtained for T2020 parameters.

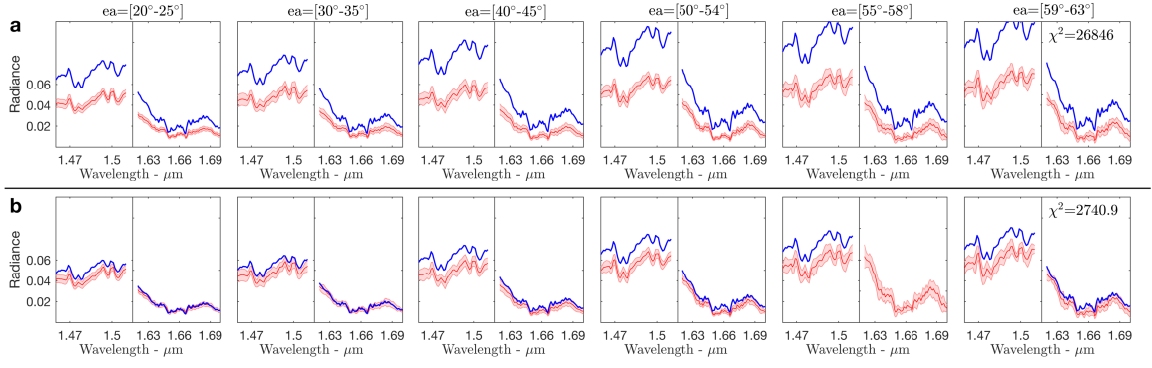


Fig. 18. Comparison between simulations and observations at latitudes between 5° and 15° S. The thick red solid lines represent the VLT spectra illustrated in Fig. 6, and the blue lines are simulations performed for a haze production rate of 6×10^{-14} $\text{kg m}^{-2} \text{s}^{-1}$ and a density of haze particles ρ_i of 700 kg m^{-3} (a) and 1000 kg m^{-3} (b). In both cases the real refractive index was set to 1.4.

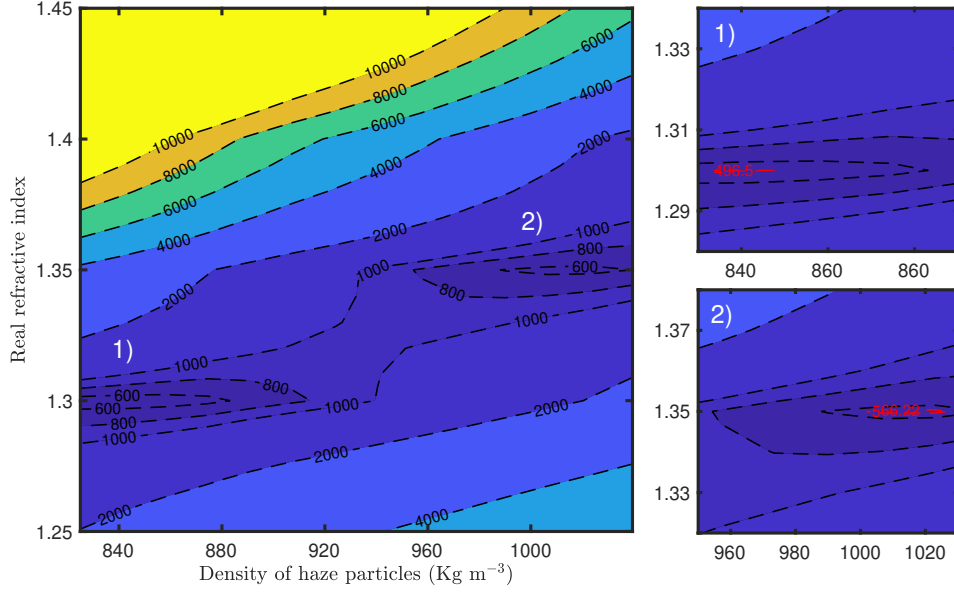


Fig. 19. Contours of χ^2 in the ρ_i - m parameter space for the spectra shown in Fig. 6. For all the simulations the haze production rate was fixed to $6 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$. The red dashed lines show the 3σ significance levels of the two minima.

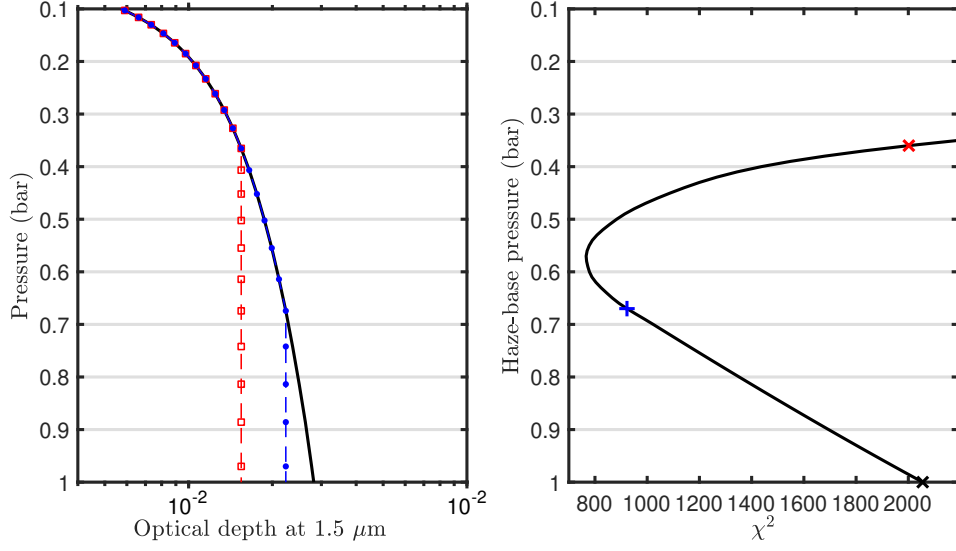


Fig. 20. Left) Cumulative optical depth at 1.5 μm computed for a haze production rate of $6 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ and setting the haze-base pressure at 0.36 bar (red curve), 0.67 bar (blue curve) and 1 bar (black curve). The rest of haze parameters were set to T2020 values. Right) Variation of χ^2 with the haze-base pressure. The haze parameters were set to the values used in Fig. 18a, and the crosses indicate the χ^2 values achieved for the profiles of left panel.

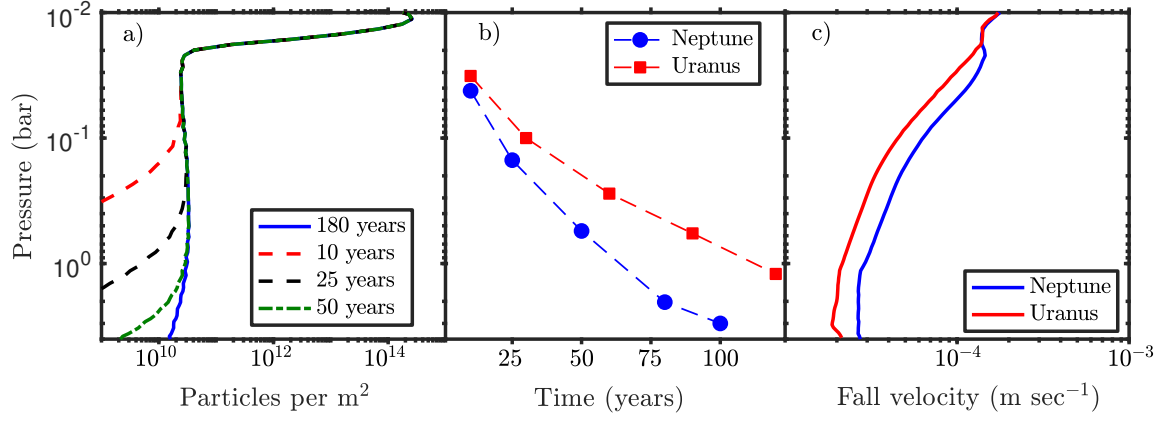


Fig. 21. (a) Haze density profile computed for Neptune's atmosphere with our haze microphysical model for different time scales using a haze production rate of $M_0 2.5 \times 10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ and $q=10 \text{ e}^-$ per μm radius. (b) Variation of haze microphysics timescale with pressure for a haze production of 2.5×10^{-14} (Neptune) and $3 \times 10^{-15} \text{ kg m}^{-2} \text{ s}^{-1}$ (Uranus). (c) Variation of fall velocity with altitude in Neptune and Uranus for the haze particles radius illustrated in left panel of Fig. 11.

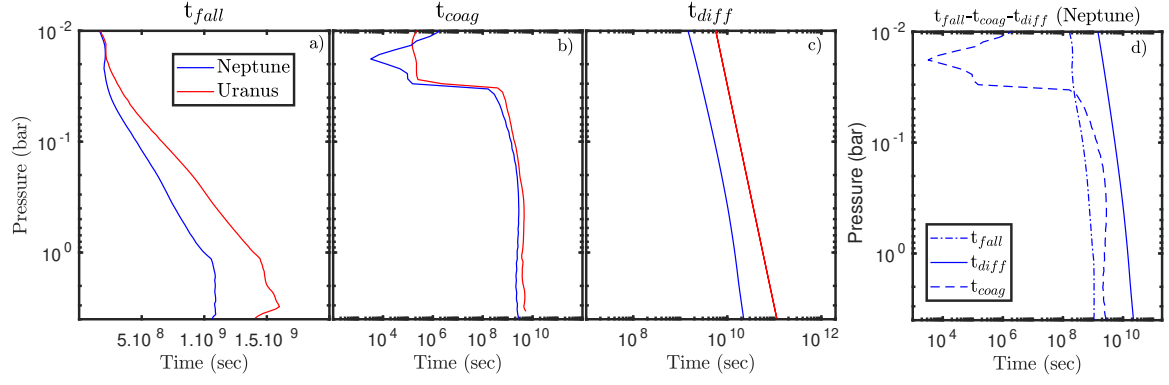


Fig. 22. Comparison of the sedimentation (panel a), particle coagulation (panel b) and eddy diffusion (panel c) timescales in Neptune and Uranus computed for a haze production of 2.5×10^{-14} (Neptune) and $3 \times 10^{-15} \text{ kg m}^{-2} \text{ s}^{-1}$ (Uranus). Panel d shows a comparison between these timescales for Neptune.

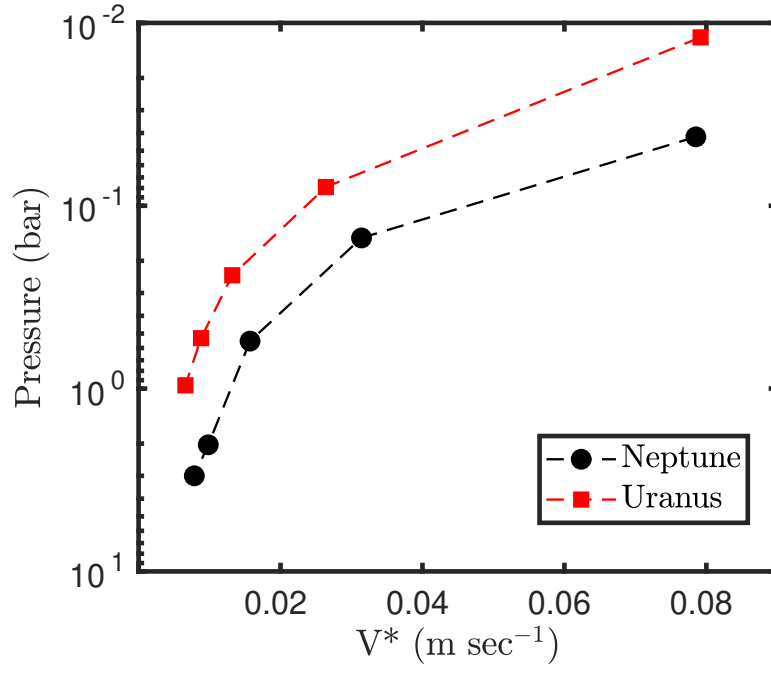


Fig. 23. Variation of the equivalent wind velocity with altitude in Neptune and Uranus computed from the timescales shown in Fig. 21. Note that the equivalent wind velocities in Neptune are greater than in Uranus as a result of its smaller t_M .