

1 **Latitudinal variations in methane abundance, aerosol**
2 **opacity and aerosol scattering efficiency in Neptune’s**
3 **atmosphere determined from VLT/MUSE**

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22 **Key Points:**

- 23 • Neptune MUSE visible/near-infrared spectra are well fitted by a simple aerosol
24 model comprised of three distinct layers
- 25 • A darkening of particles at blue-green wavelengths in a deep aerosol layer near 5
26 bar can explain dark spots and the dark ‘South Polar Wave’
- 27 • A brightening of the same particles at red-infrared wavelengths can explain bright
28 zones and spots seen in longwave narrow reflectance peaks

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Abstract

Spectral observations of Neptune made in 2019 with the MUSE instrument at the Very Large Telescope in Chile have been analysed to determine the spatial variation of aerosol scattering properties and methane abundance in Neptune’s atmosphere. The darkening of the South Polar Wave (SPW) at $\sim 60^\circ\text{S}$, and dark spots such as the Voyager 2 Great Dark Spot is concluded to be due to a spectrally-dependent darkening ($\lambda < 650\text{nm}$) of particles in a deep aerosol layer at ~ 5 bar and presumed to be composed of a mixture of photochemically-generated haze and H_2S ice. We also note a regular latitudinal variation of reflectivity at wavelengths of very low methane absorption longer than ~ 650 nm, with bright zones latitudinally separated by $\sim 25^\circ$. This feature, similar to the spectral characteristics of a discrete deep bright spot DBS-2019 found in our data, is found to be consistent with a brightening of the particles in the same ~ 5 -bar aerosol layer at $\lambda > 650$ nm. We find the properties of an overlying methane/haze aerosol layer at ~ 2 bar are, to first-order, invariant with latitude, while variations in the opacity of an upper tropospheric haze layer reproduce the observed reflectivity at methane-absorbing wavelengths, with higher abundances found at the equator and also in a narrow ‘zone’ at 80°S . Finally, we find the mean abundance of methane below its condensation level to be 6–7% at the equator reducing to $\sim 3\%$ south of $\sim 25^\circ\text{S}$, although the absolute abundances are model dependent.

Plain Language Summary

Observations of Neptune in visible light, made with the MUSE instrument at ESO’s Very Large Telescope, reveal the different layers of clouds and gases within this Ice Giant atmosphere, and how they change with height and latitude. The properties of the 1-2-bar layer of methane ice and haze are found to be roughly constant with latitude. However, a diffuse upper layer is thickest at the equator and near the south pole, indicating air rising at mid-latitudes and descending near the equator and poles. Conversely, the distribution of methane between the deep 5-bar clouds (hydrogen sulphide ice and haze) and the middle layers decreased from 6–7% at the equator to $\sim 3\%$ near the south pole, suggesting rising air instead at the equator and descending elsewhere. Finally, a blue-green darkening of the particles in the deep layer can explain Neptune’s dark spots and the dark ‘South Polar Wave’ at 60°S , whereas a brightening of the same particles at red and infrared wavelengths matches occasional discrete deep bright spots and a sequence of previously unnoticed bright ‘zones’, separated by 25° latitude. All this is evidence that the atmospheric circulation changes as a function of height and latitude in complex and surprising ways.

1 Introduction

The study of the aerosols in Neptune’s atmosphere has been transformed in recent years by the analysis of visible and near-infrared multi-spectral imaging data. The retrieval of vertical profiles of aerosols from such data requires knowledge of the vertical and latitudinal distribution of the main gaseous absorber, methane, which for many years, in the absence of any other information, was assumed to be constant at all latitudes. However, by analysing Hubble Space Telescope (HST) Space Telescope Imaging Spectrograph (STIS) observations of Neptune from 2003, Karkoschka and Tomasko (2011) showed that the abundance of methane at equatorial latitudes ($\sim 4\%$) was approximately twice that detected at polar latitudes ($\sim 2\%$), a result that had significant repercussions on the inferred vertical structure of aerosols, which were subsequently found to vary less significantly with latitude. In 2018, Neptune was observed during commissioning operations with the Multi Unit Spectroscopic Explorer (MUSE) Integral Field Unit (IFU) Spectrometer at the Very Large Telescope (VLT) at the European Southern Observatory in Chile. An initial analysis of these data (Irwin, Toledo, Braude, et al., 2019) found a similar lat-

itudinal variation of cloud-top methane mole fraction as the HST/STIS study, and was later revised to include modelling of the limb-darkening effects (Irwin et al., 2021) using the Minnaert approximation (Minnaert, 1941). Irwin et al. (2021) concluded that the ‘deep’ (i.e., at 2–4 bar) mole fraction of methane varied from 4–6% at the equator to 2–4% at polar latitudes, with a boundary at $\sim 30^\circ\text{S}$. Most recently, a joint analysis of HST/STIS, Gemini/NIFS and IRTF/SpeX observations of Neptune and Uranus from 0.3 to 2.5 μm (Irwin, Teanby, Fletcher, et al., 2022) has developed a ‘holistic’ aerosol model of both planets comprised of three basic distinct aerosol layers: 1) a deep H_2S /photochemical-haze aerosol layer with a base pressure $\geq 5\text{--}7$ bar (Aerosol-1); 2) a layer of methane/photochemical-haze just above the methane condensation level at 1–2 bar (Aerosol-2); and 3) an extended layer of small photochemical haze particles extending into the stratosphere (Aerosol-3). For Neptune an additional contribution from upper level (~ 0.2 bar) methane ice clouds was required to match the IRTF/SpeX observations at $\lambda > 1 \mu\text{m}$.

The atmosphere of Neptune occasionally displays dark spots, seen at blue-green visible wavelengths < 650 nm, including the Great Dark Spot (GDS) observed by Voyager 2 in 1989 (Smith et al., 1989), and more recent examples captured in HST Wide Field Camera 3 (WFC3) observations (e.g., Hueso & Sánchez-Lavega, 2019). The most recent dark spot was discovered by HST/WFC3 in 2018 at 23°N and named NDS-2018 (Simon et al., 2019). NDS-2018 was of a similar size to the GDS and subsequently drifted equatorwards, disappearing in 2022 (Wong et al., 2022). As part of a global effort to observe and analyse NDS-2018, Neptune was observed with VLT/MUSE in late 2019 (Irwin et al., 2023). After spatial deconvolution, Irwin et al. (2023) were able to detect the NDS-2018 spot at $\sim 15^\circ\text{N}$ in these observations (the first ground-based detection of such a spot), and were also able to spectrally characterise it at high spectral resolution, the first time that this has ever been achieved for a Neptunian dark spot. Irwin et al. (2023) found that NDS-2018 was caused by a spectrally-dependent ($\lambda < 650$ nm) darkening of the particles in the Aerosol-1 layer at ~ 5 bar and also found a nearby deep bright spot, DBS-2019 at $\sim 10^\circ\text{N}$, which they concluded was caused by a brightening of the same layer at longer wavelengths.

In addition to capturing the reflection spectra of NDS-2018 and DBS-2019, the 2019 VLT/MUSE data also show distinct latitudinal variations over a wide range of wavelengths (473 – 933 nm), in particular the South Polar Wave (SPW) at $\sim 60^\circ\text{S}$, first seen in Voyager 2 images in 1989 (Smith et al., 1989), which is dark at blue-green wavelengths, but invisible at longer wavelengths. The spectral features of the SPW are very similar to those of dark spots and Karkoschka (2011a) concluded that it is caused by a darkening of particles at pressures > 3 bar, which was later confirmed by Irwin, Teanby, Fletcher, et al. (2022). The SPW has been visible ever since the Voyager 2 flyby in HST imaging observations and these observations are reviewed in detail by Karkoschka (2011b). The SPW is found to have a southern boundary at $\sim 70^\circ\text{S}$, and a clearer northern boundary that varies with longitude as a wavenumber-1 disturbance of amplitude $\sim 5^\circ$, and whose northern extent varies with time from $50\text{--}55^\circ\text{S}$. The SPW feature seems to be related to the South Polar Feature (SPF) near 70°S , which are short-lived, bright clouds that change on a time scale of hours (Hammel et al., 1989). Karkoschka (2011b) notes that the SPF longitude coincides with that of the northernmost extent of the SPW, which suggests there is a dynamical link. The features may also have been dynamically linked with the second Voyager dark spot (DS2) (Sromovsky et al., 1993) and Karkoschka (2011b) suggest that these features are static with respect to a new coordinate system for the deep rotation of Neptune with a rotation rate of 15.9663 hours, compared with 16.108 hours derived from Voyager radio data (Lecacheux et al., 1993).

While the 2019 VLT/MUSE data show the SPW very clearly, they also detected latitudinal variations at several other wavelengths that have not been fully noted before. In this paper, we further analyse the 2019 VLT/MUSE data to revise our understand-

131 ing of the latitudinal variation in aerosol properties and methane abundance in Neptune’s
 132 atmosphere.

133 2 Observations

134 The observations of Neptune were recorded in October and November 2019 using
 135 the Multi Unit Spectroscopic Explorer (MUSE) Integral Field Unit (IFU) spectrome-
 136 ter (Bacon et al., 2010) at the Very Large Telescope (VLT) of the European Southern
 137 Observatory at La Paranal, Chile. MUSE records ‘cubes’ of data, where each point in
 138 the 300×300 ‘spaxel’ image contains a complete spectrum covering ~ 3700 wavelengths
 139 from 473 to 933 nm at a spectral resolution of 2000 – 4000. The observations were recorded
 140 in Narrow-Field Mode, which has field of view of $7.5'' \times 7.5''$ and each ‘spaxel’ is of size
 141 $0.025'' \times 0.025''$ (equivalent to $530 \text{ km} \times 530 \text{ km}$ on Neptune’s disc). To improve the
 142 signal-to-noise ratio, and also to make the observations consistent with our only avail-
 143 able source of methane absorption data (Karkoschka & Tomasko, 2010), the spectra were
 144 averaged with a triangular instrument lineshape of Full-Width-Half-Maximum (FWHM)
 145 2 nm, sampled every 1 nm (to achieve Nyquist sampling), reducing the number of wave-
 146 lengths from ~ 3700 to 459.

Table 1. VLT/MUSE Neptune observations (Narrow-Field Mode).

Obs. ID	Date	Time (UT)	airmass	seeing (arcsec)	Observing Conditions (and any features present)
1A	Oct. 17th 2019	00:10:35	1.208	0.88	Poor
1B	Oct. 17th 2019	00:15:20	1.194	0.84	Poor, incomplete
1C	Oct. 17th 2019	00:20:09	1.181	0.90	Poor
1	Oct. 17th 2019	00:24:51	1.170	0.69	Poor
2	Oct. 17th 2019	03:47:29	1.150	0.63	Moderate
3	Oct. 17th 2019	03:52:12	1.160	0.74	Moderate
4	Oct. 17th 2019	03:56:55	1.171	0.76	Moderate
5	Oct. 17th 2019	04:01:38	1.183	0.69	Moderate
6	Oct. 18th 2019	00:01:20	1.223	0.67	Clear, NDS-2018 and DBS-2019
7	Oct. 18th 2019	00:18:08	1.176	0.62	Clear, NDS-2018 and DBS-2019
8	Oct. 18th 2019	00:22:53	1.165	0.58	Clear, NDS-2018 and DBS-2019
9	Oct. 18th 2019	00:27:36	1.154	0.55	Clear, NDS-2018 and DBS-2019
10	Oct. 18th 2019	00:32:19	1.144	0.56	Clear, NDS-2018 and DBS-2019
11	Oct. 18th 2019	03:26:42	1.118	0.62	Clear/Moderate
12	Oct. 18th 2019	03:32:01	1.127	0.76	Clear/Moderate
13	Oct. 18th 2019	03:37:33	1.138	0.70	Clear/Moderate
14	Oct. 18th 2019	03:42:53	1.149	0.70	Clear/Moderate
15	Nov. 13th 2019	02:31:22	1.232	0.93	Poor
16	Nov. 13th 2019	02:36:05	1.248	1.00	Poor/Moderate
17	Nov. 13th 2019	02:40:48	1.264	0.97	Poor/Moderate
18	Nov. 13th 2019	02:35:33	1.281	0.99	Poor/Moderate

N.B., the Observation ID mostly relates to order of observation in our data set. Exposure time for all observations was 120s. Obs ‘6’ is highlighted as this was the main set used in this study.

147 Neptune was observed on several occasions in this programme (summarised in Ta-
 148 ble 1), where five exposures on October 18th (Observations 6 – 10), included the NDS-
 149 2018 dark spot. Although these data were recorded with the GALACSI adaptive optics
 150 system (Stuik et al., 2006), using a laser guide star, the achieved spatial resolution near
 151 500 nm was insufficient to resolve the faint NDS-2018 feature, which has a diameter of

152 $\sim 0.2''$ and has low contrast. However, with the development of a novel deconvolution
 153 technique, MODIFIED-CLEAN, the deconvolved ‘cubes’ had sufficient discrimination to de-
 154 tect and spectrally characterise the NDS-2018 dark spot and also a nearby deep bright
 155 spot DBS-2019 (Irwin et al., 2023). The appearance of Neptune at several wavelengths
 156 in the best-resolved deconvolved cube in this set, ‘Obs-6’ is shown in Fig. 1. Irwin et al.
 157 (2023) found that the NDS-2018 dark spot (visible at 551 nm in Fig. 1) is formed by a
 158 spectrally-dependent ($\lambda < 650$ nm) darkening of the aerosols in the deep Aerosol-1 layer
 159 at ~ 5 bar, presumably coincident with the H_2S condensation level. Irwin et al. (2023)
 160 also found a new deep bright spot near NDS-2018, visible in the 831-nm image, which
 161 they named ‘Deep Bright Spot - 2019’ (DBS-2019), and which, from the narrowness of
 162 its reflectance peaks, was determined to be caused by a spectrally-dependent brighten-
 163 ing of the same ~ 5 -bar Aerosol-1 layer at wavelengths longer than ~ 650 nm.

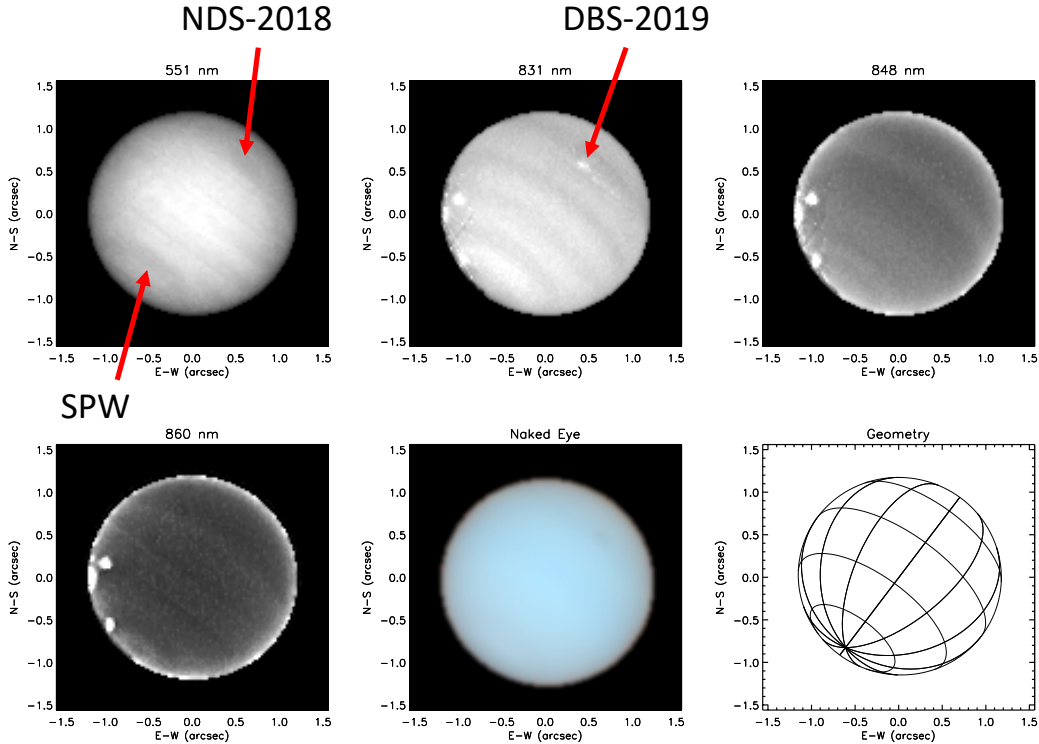


Figure 1. Deconvolved MUSE ‘slices’ at 551, 831, 848 nm, and 860 nm, respectively, from the ‘Obs-6’ cube. Also shown is the modelled appearance of Neptune to the naked eye, reconstructed from the MUSE observations using standard colour-matching functions and correctly accounting for gamma corrections. A reference latitude and longitude grid, with spacing of 30° in each direction, is also shown. In the 511-nm and naked-eye images the South Polar Wave (SPW) is just visible at bottom left, while the dark spot NDS-2018 can just be seen at top right. The DBS-2019 bright spot is only visible at longer wavelengths of extremely low methane absorption, such as 831 nm shown here. The 848 and 860 nm images, at methane-absorbing wavelengths, probe the haze high in the atmosphere, with 860 nm probing slightly higher and revealing a south polar collar at 80°S . The bright spots on the left edge of Neptune’s disc at longer wavelengths are upper tropospheric methane ice clouds at 0.1 - 0.6 bar.

164 While Irwin et al. (2023) describe the characterisation and interpretation of the NDS-
 165 2018 and DBS-2019 discrete features, the MUSE data also have sufficient spatial and spec-

166 tral resolution to determine latitudinal variations of aerosol opacity and methane abun-
 167 dance. These latitudinal changes are clearly visible in Fig. 1, including the dark SPW
 168 at 60°S at short wavelengths (here at 551 nm), but also prominent banded features at
 169 longer continuum wavelengths (here at 831 nm), and a small bright collar (80°S) seen
 170 about the south pole at 860 nm, which at a wavelength of strong methane absorption
 171 must be caused by increased aerosol abundance high in the atmosphere. The banding
 172 seen at 831 nm is also just visible in recent 845-nm HST/WFC3 images (e.g., Chavez
 173 et al., 2023), which covers the same reflectance peak, but at much lower spectral reso-
 174 lution ($\Delta\lambda \sim 84$ nm). However, the banding is particularly prominent at this wavelength,
 175 which is at the centre of reflectance peak where we can see to deep pressures in the at-
 176 mosphere, and the narrowness of the spectral signature of these bands, discussed later,
 177 indicate that they are likely caused by variations of the deep aerosol layers.

178 Although the MUSE observations were photometrically corrected by observing a
 179 standard star shortly before viewing Neptune, uncertainty remained in the absolute cor-
 180 rection. Hence, to ensure the disc-averaged MUSE spectrum was consistent with the disc-
 181 averaged spectrum of Neptune measured in 2003 with HST/STIS (Karkoschka & Tomasko,
 182 2011), the data were scaled to give the same reflectivity as HST/STIS in the equatorial
 183 region of 10°S – 10°N. Long-term records of the disc-integrated blue and green magni-
 184 tudes of Neptune (Lockwood, 2019) reveal no notable changes between 2003 and the 2016,
 185 which justifies this simple scaling and which also has the merit of allowing direct com-
 186 parison with retrievals from the HST/STIS observations.

187 3 Analysis

188 The analysis in this paper follows on from a combined analysis of HST/STIS, IRTF/SpeX,
 189 and Gemini/NIFS observations of both Uranus and Neptune by Irwin, Teanby, Fletcher,
 190 et al. (2022), which we will refer to henceforth as ‘IRW22’ or the ‘holistic’ model. Given
 191 that we do not know the composition of the aerosols in Neptune’s atmosphere, and do
 192 not have a definite expectation of the volume mixing ratio profile of the main visible/IR
 193 absorber, methane, the simultaneous retrieval of aerosol and methane abundance pro-
 194 files is a degenerate problem, even when analysing the 800 – 860 nm region that can dif-
 195 ferentiate between aerosols and methane abundance (Karkoschka & Tomasko, 2011). As
 196 a result, since previous studies have concentrated on particular narrow wavelength ranges,
 197 this has led to a multitude of similar, but not wholly consistent aerosol/methane solu-
 198 tions. One way to reduce the degeneracy is to analyse simultaneously as wide a wave-
 199 length range as possible in order that the sizes of the scattering particles can be better
 200 constrained (small particles will have cross-sections that fall as $1/\lambda^4$, while larger par-
 201 ticles will have cross-sections that vary more slowly with wavelength). The wavelength
 202 range analysed by IRW22 was 0.3 – 2.4 μm , which allowed good discrimination against
 203 particle size. However, even when analysing a wide range of wavelengths, there are still
 204 multiple solutions as we do not know the composition of particles and so we must try
 205 to infer the scattering properties (in this case the opacities and single-scattering albedos)
 206 directly from the data. IRW22 addressed this degeneracy by attempting to fit sim-
 207 ultaneously not only the disc-averaged reflectance spectrum over this range, but also
 208 how the spectrum varied with solar and viewing zenith angles, through fitting their ‘limb-
 209 darkening’ or ‘centre-to-limb’ functions. This was done by analysing all observations on
 210 the planets’ discs and quantifying how the reflectance at each wavelength, (I/F) , var-
 211 ied towards the limb of the planet. IRW22 found that this variation was well approx-
 212 imated by the Minnaert approximation (Minnaert, 1941):

$$(I/F) = (I/F)_0 \mu_0^k \mu^{k-1}. \quad (1)$$

213 Here, μ and μ_0 are the cosines of the viewing and solar zenith angles, respectively,
 214 $(I/F)_0$ is the fitted nadir reflectance, and k the fitted limb-darkening parameter. The

215 reflectance, I/F , is the observed radiance at each location, I , divided by the radiance
 216 reflected from a perfectly-reflecting Lambertian surface at the same point, and in this
 217 study we used the solar spectrum of Chance and Kurucz (2010). IRW22 estimated the
 218 scattering properties of the particles in the different aerosol layers using Mie theory and
 219 fitted for their complex refractive index spectra. Note that this analysis assumes that
 220 the particles in a certain layer, which might be a mixture of ice and haze particles of var-
 221 ious size distributions, can be approximated as being composed of a single size distri-
 222 bution of particles, with a single complex refractive index spectrum. The imaginary re-
 223 fractive index spectra were treated as free parameters to be fitted, while the real parts
 224 were computed using a Kramers-Kronig analysis, assuming $n_{real} = 1.4$ at 800 nm; this
 225 value of n_{real} is typical of giant planet condensates, such as methane ice, but is not well
 226 constrained (other values were tested and gave similar overall results). We note that the
 227 Kramers-Kronig approach requires that we know $n_{imag}(\lambda)$ at all wavelengths to accu-
 228 rately reproduce $n_{real}(\lambda)$, whereas in this study $n_{imag}(\lambda)$ could only be estimated from
 229 0.3 to 2.4 μm . Hence, this reconstruction of $n_{real}(\lambda)$ is only an approximation. Mie the-
 230 ory was then used to compute the extinction cross-section and single-scattering albedo
 231 of the aerosols as a function of wavelength. Finally, the Mie-calculated phase functions
 232 were approximated with combined Henyey-Greenstein phase functions at each wavelength
 233 to average over features particular to spherical particles, namely the ‘rainbow’ peak and
 234 the back-scattered ‘glory’. Although containing several approximations, this approach,
 235 first presented by Irwin et al. (2015), has the merit of generating self-consistent values
 236 of the extinction cross-section, single-scattering albedo, and phase function spectra, and
 237 has considerable advantages, we believe, over approaches where the single-scattering albedo
 238 (or phase functions, or extinction coefficients) are modified directly and separately. The
 239 holistic IRW22 analysis found that the combined HST/STIS, IRTF/SpeX and Gemini/NIFS
 240 observations of both Uranus and Neptune were well approximated with an atmospheric
 241 model consisting of three main aerosol layers, outlined earlier: 1) ‘Aerosol-1’, a deep aerosol
 242 layer with a base pressure $\geq 5\text{--}7$ bar, assumed to be composed of a mixture of H_2S ice
 243 and photochemical haze of mean radius 0.05 μm ; 2) ‘Aerosol-2’, a layer of photochem-
 244 ical haze and methane ice of mean radius ~ 0.5 μm , confined within a layer of high static
 245 stability at the methane condensation level at 1–2 bar; and 3) ‘Aerosol-3’, an extended
 246 layer of small photochemical haze particles ($r \sim 0.05$ μm), probably of the same com-
 247 position as the haze in the 1–2-bar layer, extending from this level up through to the strato-
 248 sphere. An additional thin layer of micron-sized ($r \sim 3$ μm) methane ice particles at
 249 ~ 0.2 bar (‘Aerosol-4’) was required to explain enhanced reflection at methane-absorbing
 250 wavelengths in the Neptune data longer than 1.0 μm , caused probably by discrete clouds
 251 at this level being present along the central meridian of the IRTF/SpeX line-averaged
 252 data used, and unresolved clouds in the Gemini/NIFS observations. The region of high
 253 static stability at the methane condensation level has been noted by several previous au-
 254 thors (e.g., Hueso et al., 2020; Leconte et al., 2017).

255 We studied the VLT/MUSE data in the same way as IRW22 and concentrated on
 256 the best-resolved, deconvolved ‘Obs-6’ cube, described by Irwin et al. (2023). For this
 257 study we used the same simple ‘step’ model for the methane profile as IRW22, with vari-
 258 able deep abundance, a fixed relative humidity above the condensation level (discussed
 259 later) and limiting the mole fraction in the stratosphere to 1.5×10^{-3} (Lellouch et al.,
 260 2010). Sample profiles using this model are shown in Fig. 2. We first analysed the disc-
 261 average of the deconvolved ‘Obs-6’ cube, and then analysed the data in latitude bands
 262 of width 10° , spaced every 5° to achieve Nyquist sampling (masking out discrete clouds
 263 in both cases). The high spatial resolution of these observations, combined with a re-
 264 markably low level of cloud activity at this time (e.g., Chavez et al., 2023), meant that
 265 the few high-altitude discrete methane ice clouds that were visible were easily masked-
 266 out in our data. This meant that we did not need to include the upper troposphere (\sim
 267 0.2 bar) layer of micron-sized methane ice ‘Aerosol-4’ particles in our model, which only
 268 have a significant contribution at wavelengths greater than 1 μm anyway. Using a Min-
 269 naert analysis we extracted the disc-average nadir reflectance spectrum, $(I/F)_0(\lambda)$, and

limb-darkening spectrum, $k(\lambda)$, from the observations, where λ is wavelength. These spectra were used to reconstruct two synthetic observation spectra, calculated for back-scattering conditions, with the zenith angles (both viewing and solar) set to either 0° (i.e., nadir), or 61.45° , which were then fitted simultaneously with our NEMESIS radiative transfer and retrieval tool (Irwin et al., 2008). We fitted to these two angles simultaneously in order to fit both the mean reflectance and limb-darkening spectra, and these two particular zenith angles were chosen to coincide with two of the zenith angles in our matrix-operator multiple-scattering scheme (Plass et al., 1973), thus avoiding any interpolation errors. In this analysis (following Irwin et al., 2023) we increased the mean radius of the Aerosol-1 particles to $0.1 \mu\text{m}$ (standard Gamma distribution of sizes with variance $\sigma = 0.05$), consistent with expectation of fog-like particles, and fixed the mean radius of the Aerosol-2 particles to $0.7 \mu\text{m}$ ($\sigma = 0.3$), which was found by IRW22 to be the mean size of the Aerosol-2 particles that was most consistent with their observations. The size of the Aerosol-3 particles was left unchanged, with a mean radius $0.05 \mu\text{m}$ and variance $\sigma = 0.05$. When fitting to the disc-averaged MUSE data we found that the estimated random errors of the reconstructed spectra were smaller than the uncertainties in our radiative transfer model arising from parameterisation choices and the assumed gaseous absorption coefficients of Karkoschka and Tomasko (2010). Hence, following IRW22, the errors on these spectra were set to $1/50$ of the maximum disc-averaged nadir reflectance within 100 nm of each point in the spectrum. These errors were reduced by a factor of 2 in the $800 - 900 \text{ nm}$ range to ensure a good fit to the region best able to differentiate between cloud and methane abundance. In addition, the nadir reflectivity error was limited to not exceed 1% to ensure that NEMESIS fitted well to the wavelengths near the peak of reflection at $\sim 500 \text{ nm}$. With these errors (set to be the same for both the 0° and 61.45° spectra) we could fit the spectra to $\chi^2/n \sim 3$, giving retrieved parameters with meaningful error values. For the shorter wavelengths we also had to account for a small amount of Raman-scattering (where photons absorbed at shorter wavelengths can be scattered to longer wavelengths in hydrogen-dominated atmospheres) and polarisation effects, which we did following the procedures of Sromovsky (2005a) and Sromovsky (2005b), respectively, as outlined by IRW22. To account for the Raman scattering, we added 100 points below the MUSE spectrum in each pixel, covering $373 - 473 \text{ nm}$, which we reconstructed from a disc-averaged Minnaert analysis of the 2003 HST/STIS observations (Karkoschka & Tomasko, 2011), using the fitted Minnaert $(I/F)_0$ and k spectra to simulate the expected spectrum for the observing geometry of each pixel and scaling this to match the MUSE spectrum where they overlap. In our radiative transfer model we simulated the entire spectrum from 373 to 933 nm , including Raman-scattering. Hence, photons Raman-scattered from wavelengths as short as 373 nm were approximated for in the modelled spectra. However, to avoid the retrieval model trying to fit these synthetic $373 - 473 \text{ nm}$ data, the error on these points was set to 100%. Finally, for consistency with IRW22 and ease of comparison with their HST/STIS retrievals, the MUSE data were analysed at the same set of wavelengths as for the STIS data. The assumed gaseous absorption coefficients and the assumed vertical profiles of temperature and gaseous abundance were also those used by IRW22 and Irwin et al. (2023).

During the initial retrievals from the latitudinally-averaged data, we found that there was considerable degeneracy between the methane abundance and the opacities of the Aerosol-1 and Aerosol-2 layers, which led to these parameters not varying smoothly with latitude, but showing considerable degeneracy, or ‘cross-talk’. The reason for this can be understood from Fig. 2, which shows the best-fit cloud model of IRW22 determined from their combined Neptune data set. It can be seen that there is considerable overlap between the Aerosol-1 and Aerosol-2 profiles in the $2-3 \text{ bar}$ region. Since the modelled spectra were sensitive to both clouds in this pressure region, the two opacities are able to interfere with each other with, for example, one reducing and the other increasing leading to little change in the overall modelled reflectance. Hence, we revised the parameterisation of the Aerosol-1 layer to be like the Aerosol-2 layer, i.e., a single vertically thin cloud, but centred at a higher, fixed pressure of 5 bar (Fig. 2). This param-

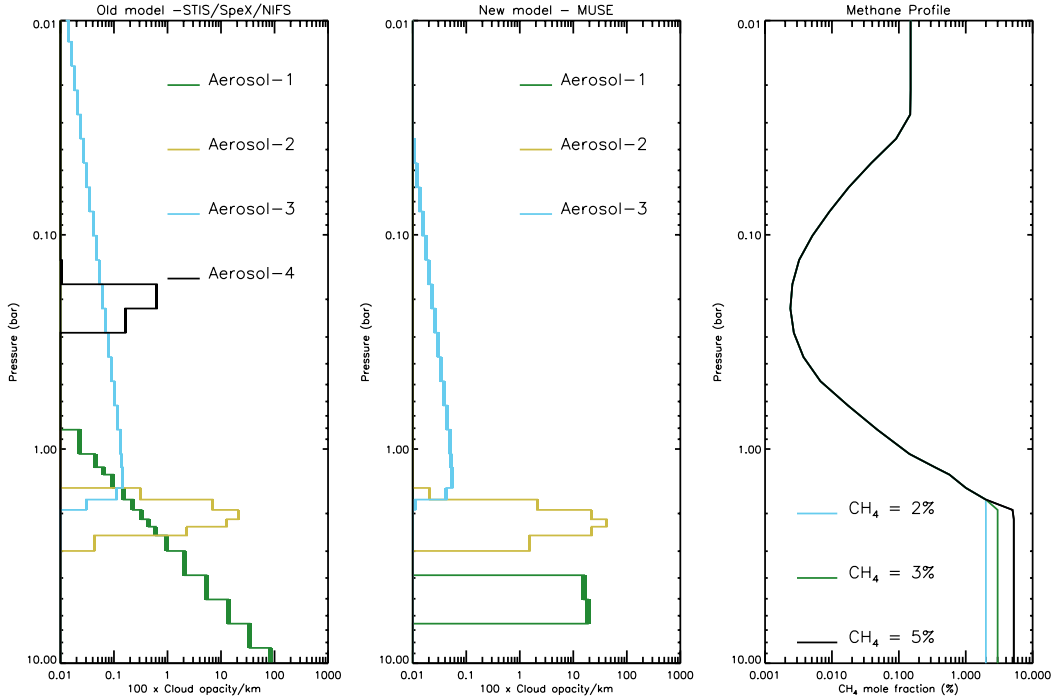


Figure 2. Cloud opacity profiles retrieved in this analysis from disc-averaged observations, and assumed example methane abundance profiles. Left: Cloud scheme used by IRW22 (Irwin, Teanby, Fletcher, et al., 2022) in their combined analysis of HST/STIS, IRTF/SpeX and Gemini/NIFS. Middle: Revised cloud scheme used in this analysis of VLT/MUSE observations. The horizontal thickness of the lines used to plot the profiles indicates the formal retrieved error ranges for the cloud profiles. Right: Assumed methane mole fraction profile for different values of the deep abundance, where the relative humidity above the condensation level is fixed to 50% and the stratospheric mole fraction is limited to 1.5×10^{-3} as described in the main text.

325 eterisation was found to fit the MUSE data just as well as the original IRW22 param-
326 eterisation, but led to much less overlap between the aerosol distributions and thus much
327 less degeneracy between the Aerosol 1 and 2 opacities and methane abundances in the
328 latitudinal retrievals reported later. This revised Aerosol-1 cloud model was also used
329 by Irwin et al. (2023) in their analysis of the NDS-2018 and DBS-2019 features in this
330 data set. We note that our data do not allow us to determine whether there is a clear
331 gap between the Aerosol-1 and Aerosol-2 layers, only that this decoupling is necessary
332 for our retrieval approach to be reliable and stable.

333 4 Results

334 4.1 Disc-average retrievals and analysis

335 Using the deconvolved ‘Obs-6’ cube, we first determined the disc-averaged limb-
336 darkening properties for each wavelength (masking out discrete cloud features and in-
337 strument artefacts) to yield spectra of the fitted parameters $(I/F)_0(\lambda)$ and $k(\lambda)$. These
338 parameters were then used to compute the synthetic measurement spectra seen in Fig.
339 3, where the top panel compares two spectra reconstructed from the Minnaert coefficients
340 with both the solar and zenith angles set to either $\theta = 0^\circ$ or $\theta = 61.45^\circ$, using $(I/F)(\theta) =$
341 $(I/F)_0 \mu^{2k-1}$ where $\mu_0 = \mu = \cos \theta$. In this plot, the synthetic estimated error lim-

Table 2. Retrieval Models.

Parameter	Model 1	Model 2	Model 3
Aerosol-1:			
τ_1	variable	variable	variable
p_1	5 bar	5 bar	5 bar
Δp_1	0.1	0.1	0.1
$n_{imag}(\lambda)$ (8 wavelengths)	fixed	variable	variable
Aerosol-2:			
τ_2	variable	variable	1.75
p_2	variable	variable	2.1
Δp_1	0.1	0.1	0.1
$n_{imag}(\lambda)$ (8 wavelengths)	fixed	fixed	fixed
Aerosol-3:			
τ_3	variable	variable	variable
p_3	1.6 bar	1.6 bar	1.6 bar
FSH	2.0	2.0	2.0
$n_{imag}(\lambda)$ (8 wavelengths)	fixed	fixed	fixed
Methane:			
Deep VMR	variable	variable	variable
RH	50%	50%	50%
Number of variables n_x	5	13	11

N.B., n_{imag} tabulated from 0.3 to 1.0 μm in steps of 0.1 μm . Δp for Aerosol-1, Aerosol-2 is FWHM of Gaussian function in $\ln p$. Aerosol-3 is extended up from p_3 with a set fractional scale height (FSH). Note that where $n_{imag}(\lambda)$ is indicated as ‘fixed’, it means that the spectrum is fixed to that retrieved by the disc-average analysis.

342 its, discussed earlier, are shaded in grey for both spectra. Note that the spectrum recon-
343 structed at $\theta = 0^\circ$, is simply $(I/F)_0(\lambda)$. Our NEMESIS fits to these spectra, using our
344 modified aerosol model, are overplotted and show excellent agreement.

345 The bottom panel of Fig. 3 compares the limb-darkening coefficient spectrum of
346 the disc-averaged MUSE data with that extracted from our NEMESIS fits to the recon-
347 structed MUSE spectra at 0° and 61.45° , using

$$k_{fit} = 0.5 \times \left(1 + \frac{\log((I/F)_{fit}(61.45^\circ)) - \log((I/F)_{fit}(0^\circ))}{\log(\cos(61.45^\circ))} \right), \quad (2)$$

348 showing that NEMESIS correctly fits both the mean reflectivity and limb-darkening
349 spectra.

350 The spectra in both panels are compared with those obtained from an identical anal-
351 ysis by IRW22 of the HST/STIS observations of Neptune from 2003 (Karkoschka & Tomasko,
352 2011) and shows that the disc-average data are very similar, which is expected since the
353 MUSE data were scaled to match HST/STIS. However, when comparing the limb-darkening
354 spectra with HST/STIS it can be seen that the MUSE spectra show considerably more
355 limb-brightening at methane-absorbing wavelengths (i.e., $k(\lambda)$ is smaller than for STIS).
356 This may possibly result from the different deconvolution schemes used in the STIS and
357 MUSE datasets, but as we do not have the original undeconvolved STIS observations
358 we are unable to test this. However, this difference in the limb-darkening meant that the

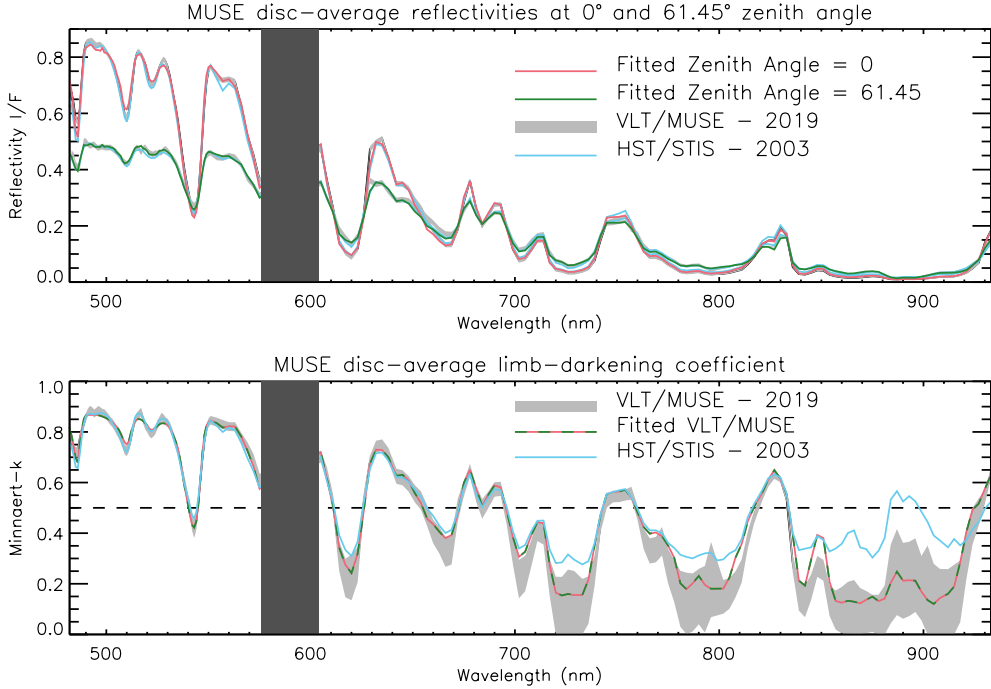


Figure 3. Top panel: MUSE spectra reconstructed at 0 and 61.45° zenith angles (viewing and solar) from our disc-average Minnaert analysis, and fits to them using our NEMESIS model. The errors on the synthetic measured spectra are indicated in grey. The spectrum calculated at $\theta = \theta_0 = 0^\circ$ is of course simply $(I/F)_0$. Bottom panel: MUSE disc-average limb-darkening spectrum $k(\lambda)$ and fitted limb-darkening spectrum, calculated from the NEMESIS fits to the reconstructed spectra at 0 and 61.45° zenith angle using Eq. 2. In both panels, the corresponding HST/STIS disc-average spectra from 2003, analysed by IRW22, are over-plotted. The wavelengths greyed-out from 585 – 602 nm, were reserved for the laser guide star adaptive optics system of MUSE. In the bottom panel, the dashed line at $k = 0.5$ is added to help differentiate between limb-darkened ($k > 0.5$) and limb-brightened ($k < 0.5$) wavelengths.

359 Mie-scattering properties calculated from the best-fitting n_{imag} spectra of the three aerosol
 360 layers derived by IRW22 did not achieve a very good fit to these MUSE data. Hence,
 361 the n_{imag} spectra had to be refitted to the MUSE observations, which we did using our
 362 modified ‘holistic’ aerosol model, also fitting the opacities of the three aerosol layers, the
 363 pressure of the Aerosol-2 layer, and the deep methane abundance. The methane relative
 364 humidity is difficult to constrain unambiguously from the MUSE wavelength range.
 365 IRW22 retrieved a methane relative humidity of 35% from their data set, which included
 366 longer-wavelength IRTF/SpeX observations, while Karkoschka and Tomasko (2011) used
 367 a fixed value of 60% to match their HST/STIS data. Here, we fix the methane relative
 368 humidity above the condensation level to 50%. Our final retrieved disc-averaged n_{imag}
 369 spectra for all three aerosols are compared with those derived by IRW22 for Neptune in
 370 Fig. 4. Here we can see that for VLT/MUSE the Aerosol-1 particles are determined to
 371 be more absorbing (i.e., n_{imag} is higher), the Aerosol-2 particles have similar n_{imag} spec-
 372 tra at short wavelengths, but have higher n_{imag} at $\lambda > 600$ nm, while the Aerosol-3 par-
 373 ticles (most visible at methane-absorbing wavelengths) are required to be considerably
 374 more scattering (i.e., n_{imag} is lower) in order to fit the MUSE observations.

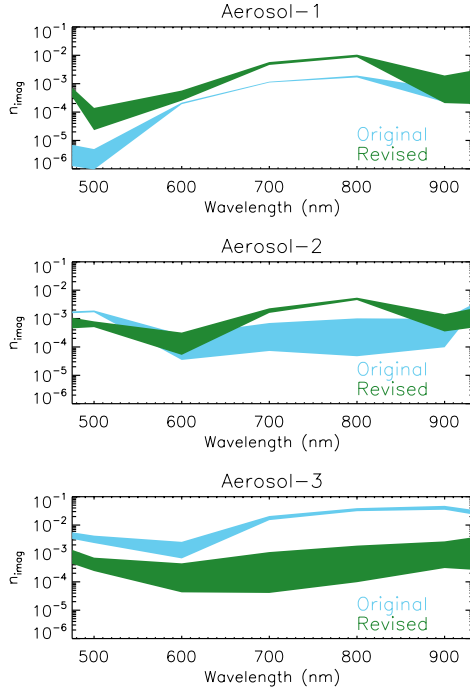


Figure 4. Imaginary refractive index (n_{imag}) spectra of Aerosol types 1, 2 and 3 derived from fitting the disc-averaged VLT/MUSE observations with our modified aerosol model, compared with the corresponding n_{imag} spectra from the IRW22 analysis of HST/STIS, IRTF/SpEx and Gemini/NIFS Neptune observations (Irwin, Teanby, Fletcher, et al., 2022).

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4.2 Latitudinally-resolved retrievals and analysis

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Having re-fitted the n_{imag} spectra (and thus all associated Mie-calculated scattering properties) of the three aerosol layers, we then determined latitudinal changes in opacity and methane abundance by analysing spectra reconstructed from the latitudinally-resolved Minnaert analysis of the MUSE dataset, where the data were averaged in latitudes bins of width 10° , spaced every 5° . A summary of the latitudinally-resolved Minnaert nadir reflectivity spectra, $(I/F)_0(\phi, \lambda)$, and limb-darkening spectra, $k(\phi, \lambda)$, where ϕ is the planetographic latitude, is shown in Fig. 5. For this latitudinal analysis there was not a sufficiently large range of zenith angles south of 70°S and north of 20°N to extract meaningful values of both $(I/F)_0(\phi, \lambda)$ and $k(\phi, \lambda)$. Hence for latitudes south of 70°S , the limb-darkening spectrum, $k(\lambda)$, was fixed to the average for all latitudes south of 70°S and only $(I/F)_0(\phi, \lambda)$ fitted. For latitudes north of 20°N , in the absence if any other information and since we believe the winds to be, to first order, hemispherically symmetric (Sromovsky et al., 1993), we assumed north-south symmetry in the cloud distribution also. Hence, the limb-darkening spectra were fixed to those found at the corresponding southern latitude, i.e., $k(\phi, \lambda) = k(-\phi, \lambda)$, and again only $(I/F)_0(\phi, \lambda)$ fitted. In Fig. 5 we can see the signature of the South Polar Wave (SPW) at $\sim 60^\circ\text{S}$ in the $(I/F)_0(\lambda)$ difference map at $\lambda < 650$ nm and also a slight increase in $(I/F)_0(\lambda)$ south of $\sim 25^\circ\text{S}$ at most wavelengths. Most of the features in the $k(\lambda)$ difference map are in regions of low k -values where the reflectances are low and are thus hard to interpret. However, we can see in the SPW at $\sim 60^\circ\text{S}$ that reflectances are more limb darkened, which is indicative of lower single-scattering albedo.

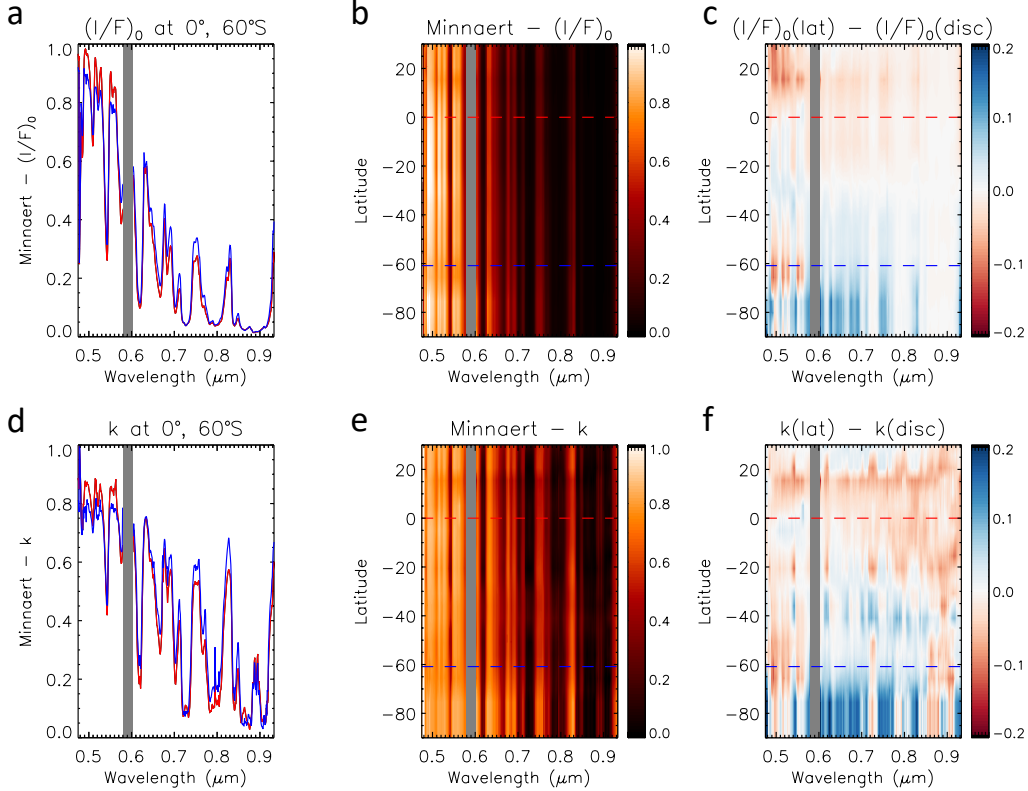


Figure 5. Latitudinally-resolved Minnaert limb-darkening analysis of MUSE data. Panel (a) compares the extracted $(I/F)_0$ spectra at the equator (red) and 60°S (blue). A contour plot of the extracted $(I/F)_0$ spectra as a function of wavelength and latitude (planetographic) is shown in Panel (b), while Panel (c) shows a contour plot of the difference of $(I/F)_0$ spectra compared with the disc-averaged $(I/F)_0$ spectrum (red regions are darker than disc-average, blue are brighter). In the contour plots the equator and 60°S are highlighted with coloured, dashed lines. Panels (d-f) show the same plots as Panels (a-c), but for the fitted Minnaert limb-darkening coefficients, k .

397 With our modified ‘holistic’ atmospheric model parameterisation we then used NEME-
 398 SIS to fit spectra reconstructed from these latitudinally-resolved Minnaert coefficients
 399 at the same two zenith angles used in the disc-average analysis (i.e., 0° and 61.45°) and
 400 retrieved latitudinal distributions of atmospheric properties using three different model
 401 setups. These are summarised in Table 2 and described in detail in the following sub-
 402 sections.

403 **4.2.1 Latitudinally-resolved retrievals with fixed Aerosol-1 n_{imag} spec-** 404 **trum (Model 1).**

405 Fixing the n_{imag} spectra of the three aerosol types to those determined from the
 406 disc-average analysis, we first fitted the latitudinally-resolved reconstructed spectra to
 407 retrieve latitudinal distributions of: 1) the opacity of all three aerosol layers (opacities
 408 are quoted at 800 nm); 2) the pressure of the Aerosol-2 layer; and 3) the deep methane
 409 abundance (Model 1 of Table 2), i.e., five parameters in total. The upper tropospheric
 410 methane relative humidity was again limited to 50%. In these retrievals the errors on

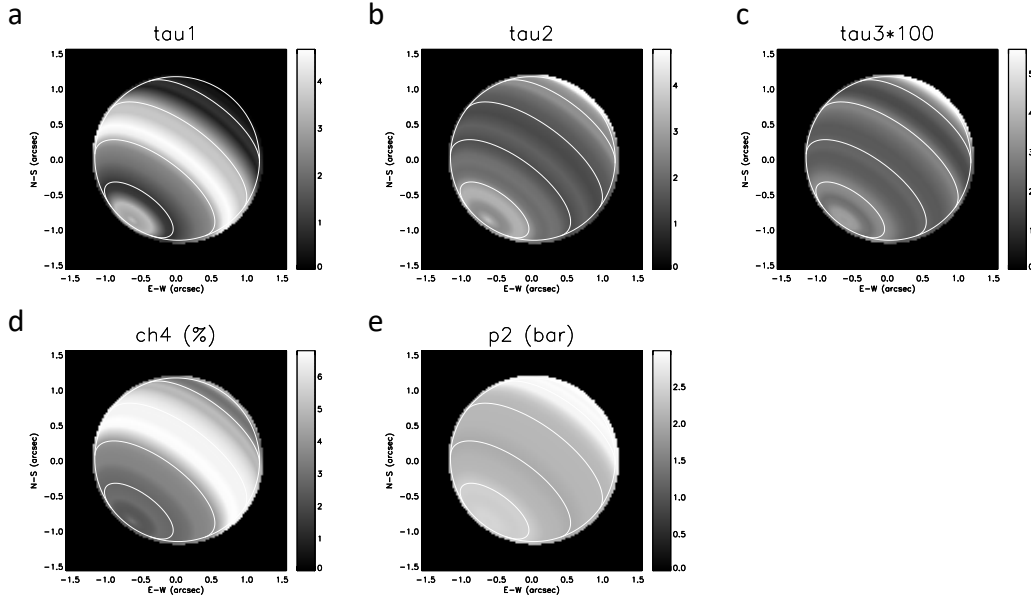


Figure 6. Zonally-averaged meridional profiles of atmospheric properties retrieved from the VLT/MUSE Neptune observations (Obs-6) using a model where the particle scattering properties were fixed to the disc-average for all aerosol layers (Model 1). The following fitted properties are projected on to Neptune’s disc: a) Aerosol-1 opacity; b) Aerosol-2 opacity; c) Aerosol-3 opacity ($\times 100$); d) deep methane mole fraction (%); and e) Pressure of base of Aerosol-2 layer (bar). The cloud opacities referred to here and elsewhere are those at 800 nm. Latitude circles are overplotted for ease of reference, with a spacing of 30° .

411 the Minnaert-reconstructed spectra were increased by a factor of 2 to reflect the fact that
 412 fewer points were averaged to determine the coefficients and to ensure that the fitted χ^2/n
 413 values were ~ 1 . The results of this analysis are shown in the form of projected images
 414 in Fig. 6 and as plots against latitude in Fig. 7. These retrievals show several very clear
 415 latitudinal dependencies of the fitted parameters, although the agreement between the
 416 observed and fitted reflectivities at 511 and 831 nm is not perfect (Fig. 7). The retrieved
 417 deep abundance of methane is seen to decrease from 6–7% at equator to $\sim 3\%$ at the
 418 pole, a very similar latitudinal dependence to that derived from previous analyses of Nep-
 419 ture’s visible/near-IR spectra (Karkoschka & Tomasko, 2011; Luszcz-Cook et al., 2016;
 420 Irwin et al., 2021), with a boundary at $\sim 20 - 30^\circ\text{S}$. Although the latitudinal depen-
 421 dence of methane abundance is similar to previous estimates, however, the absolute abun-
 422 dances are slightly higher and we note in the Discussion that these depend also on the
 423 assumed vertical profile of methane and the modelled aerosol structure, which are here
 424 different. In addition to methane, we also find significant latitudinal variation in the opac-
 425 ity of all three aerosol layers, with variations in the opacity of the Aerosol-2 layer ap-
 426 pearing to match the latitudinal variation seen in the 831-nm image (Fig. 1), with opac-
 427 ity peaks at 75°S , 45°S and 20°S . Higher in the atmosphere, similar, but offset, peaks
 428 in opacity can be seen in the retrieved Aerosol-3 opacity, with notable peaks at 80°S , 60°S
 429 and at the equator. The Aerosol-3 opacity shows some similarity with features in the 848-
 430 nm image, which is a wavelength of medium methane absorption, but is better correlated
 431 with the features in the 860-nm image, where methane absorption is strong. In particu-
 432 lar, the peak in Aerosol-3 opacity at 80°S corresponds well with a faint ring of bright-
 433 ness near Neptune’s south pole at 860 nm and similarly with a brighter zone at the equa-
 434 tor. Finally, there is a good correlation between the Aerosol-1 opacity and variations seen

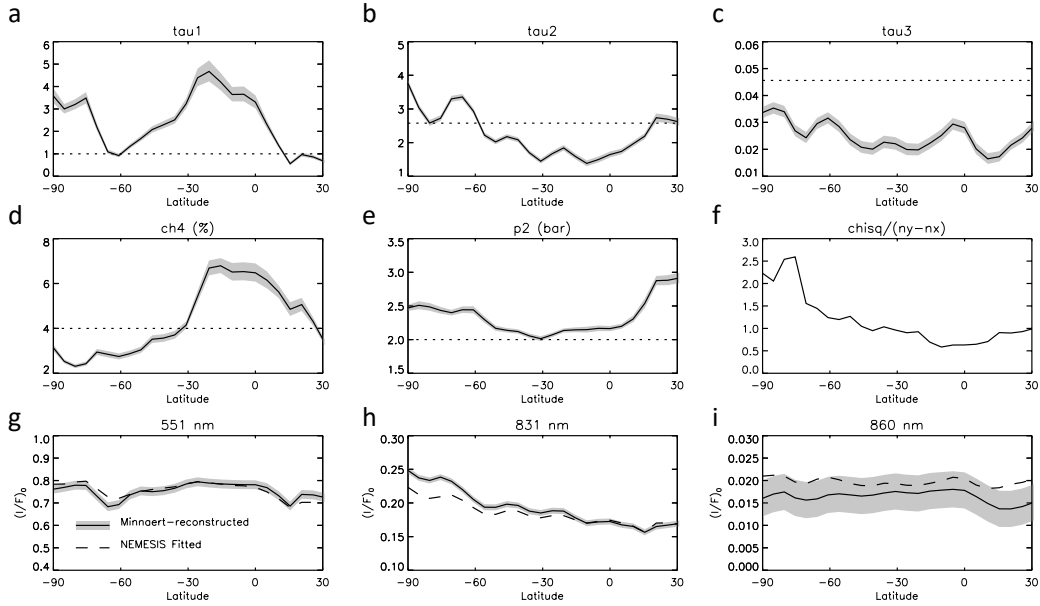


Figure 7. Zonally-averaged atmospheric properties retrieved from the VLT/MUSE Neptune observations (Obs-6), using a model (Model 1 of Table 2) where the particle scattering properties were fixed to the disc-average for all aerosol layers, showing: a) Aerosol-1 opacity; b) Aerosol-2 opacity; c) Aerosol-3 opacity ($\times 100$); d) deep methane mole fraction (%); e) Pressure at the base of Aerosol-2 layer (bar), and f) $\chi^2/(n_y - n_x)$ of the fits at different latitudes. The dotted lines in these panels show the *a priori* values assumed. Note that the error bars shown are the formal errors on the retrievals and do not include cross-correlation effects (see Discussion). The bottom row (panels g – i) shows the latitudinal variations of $(I/F)_0$ at 551, 831 and 860 nm, respectively, where the solid lines show the Minnaert-fitted values (and uncertainties) and the dashed lines show the best-fit values from our retrieval.

435 at 551 nm, and a sharp reduction in the retrieved opacity near 60°S , which coincides with
 436 the SPW, with the opacity increasing south of this, corresponding to the brighter reflectivities
 437 seen south of the SPW at 511 and 831 nm. Otherwise, the Aerosol-1 opacity and
 438 deep methane abundance seem moderately correlated between the equator and 60°S , which
 439 might suggest upwelling in the $25^\circ\text{S} - 25^\circ\text{N}$ region, condensing more H_2S cloud at ~ 5
 440 bar, and downwelling elsewhere. It should also be noted that the spatial variation in the
 441 pressure of the Aerosol-2 layer is very small, as expected from previous cloud and methane
 442 retrievals for Uranus and Neptune (Karkoschka & Tomasko, 2009, 2011). Finally the χ^2/n
 443 of the fits is good and generally less than ~ 2 , although there are clearly deficiencies at
 444 some wavelengths such as 511 and 831 nm. The *a priori* values assumed for the retrieved
 445 parameters are overplotted in Fig. 7 for reference. The *a priori* error for all parameters
 446 was set to 100%, except deep methane abundance, for which a more constrained error
 447 of 50% was assumed.

448 **4.2.2 Latitudinally-resolved retrievals with variable Aerosol-1 n_{imag} spectrum (Models 2 and 3).**

450 Although the spectral properties of the SPW at $\sim 60^\circ\text{S}$ can, to first order, be ex-
 451 plained by a thinning of the Aerosol-1 layer, the agreement with the observed reflectivities
 452 at 511 nm in Fig. 7 is not perfect, and the model does not match the 831 nm re-
 453 flectivities very well. Instead, we wondered if the SPW might be caused by a spectrally-

454 dependent darkening of the Aerosol-1 layer. The values of many properties are varying
455 with latitude towards the pole, but by subtracting from the $(I/F)_0$ spectrum at $\sim 60^\circ\text{S}$
456 the average of the $(I/F)_0$ spectra either side (i.e., those at 50°S and 70°S) we can at-
457 tempt to isolate the signature of the SPW itself, obtaining the difference spectrum shown
458 in Fig. 8. This spectrum is compared with the difference spectrum of the NDS-2018 dark
459 spot (Irwin et al., 2023), which is the difference between the observed I/F spectrum in
460 the dark spot and that expected at the same location from the average limb-darkening
461 at this latitude (15°N). Figure 8 shows that there is considerable similarity between the
462 SPW and NDS-2018 difference spectra, both showing a darkening at wavelengths < 650
463 nm, with strong methane absorption features indicating the features lie at considerable
464 depth in Neptune’s atmosphere. Irwin et al. (2023) found that the darkness of the NDS-
465 2018 dark spot could only be explained by a spectrally-dependent darkening of the Aerosol-
466 1 particles, reducing their single-scattering albedo at $\lambda < 650$ nm, and it is quite pos-
467 sible, and indeed arguably probable, that the SPW darkening is caused by the same mech-
468 anism. To test this we repeated our latitudinally-resolved retrievals, but this time we also
469 retrieved the imaginary refractive index spectrum of the Aerosol-1 particles, retrieving
470 this at eight equally-spaced wavelengths from 300 – 1000 nm, spaced by 100 nm, and with
471 a correlation length of 100 nm (Model 2 of Table 2). As noted earlier, the real part of
472 the complex refractive index spectrum was approximately reconstructed with a Kramers-
473 Kronig analysis and the particle scattering properties calculated with Mie theory, smooth-
474 ing the phase functions with combined Henyey-Greenstein functions.

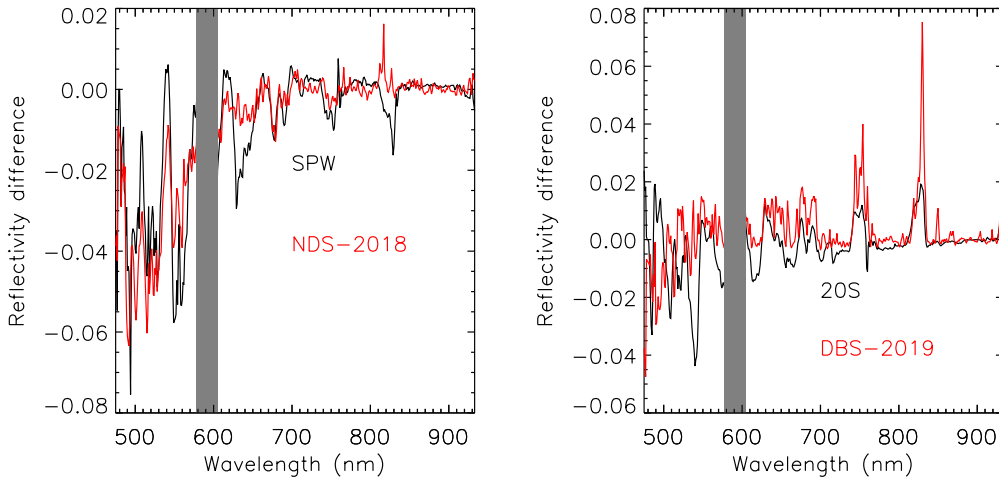


Figure 8. Difference spectrum of the South Polar Wave (SPW) at 60°S (left panel) and the bright deep ‘zone’ at 20°S (right panel) compared with the difference spectra of the NDS-2018 and DBS-2019 features, respectively, reported by Irwin et al. (2023). The difference spectra of these latitudinal bands is computed as $D(\phi, \lambda) = (I/F)_0(\phi, \lambda) - 0.5 \times ((I/F)_0(\phi - 10^\circ, \lambda) + (I/F)_0(\phi + 10^\circ, \lambda))$. The difference spectra of the NDS-2018 and DBS-2019 features are computed as the observed spectra at these locations minus the expected spectra at the same locations calculated from the centre-to-limb reflectivity functions at these latitudes of 15°N and 10°N , respectively. The comparability between the two sets of difference spectra points to a likely common origin for all these features, namely spectrally-dependent single-scattering albedo spectrum perturbations of the Aerosol-1 layer at ~ 5 bar.

475 Allowing the Aerosol-1 n_{imag} spectrum to vary leads to greatly improved fits to
476 the Minnaert-reconstructed reflectivities as shown in Fig. 9, with closer fits to the re-

477 constructed nadir reflectances at all wavelengths, including those at 551, 831 and 860
 478 nm, shown here. Although the overall retrievals of Aerosol-3 opacity and methane abun-
 479 dance are similar to those previously determined (Fig. 7) the variation in Aerosol-1 opac-
 480 ity shows a less pronounced minimum at 60°S and the Aerosol-2 opacity and base pres-
 481 sure both seem less correlated with observable variations, although a reduction in Aerosol-
 482 2 opacity and pressure level are retrieved at 80°S. However, the goodness of fit is greatly
 483 improved at all latitudes and especially south of 50°S, with χ^2/n at 60°S reducing from
 484 1.23 to 0.99. Of course, the retrieval including n_{imag} has additional model parameters,
 485 so we should use the reduced $\chi^2/(n_y - n_x)$ statistic here, where n_y is the number of spec-
 486 tral points and n_x is the number of model parameters. In these retrievals, the total num-
 487 ber of fitted spectral points is 374, while the total number of fitted parameters is 5 for
 488 the fixed n_{imag} case, and 13 for the variable Aerosol-1 n_{imag} case. This leads to $\chi^2/(n_y -$
 489 $n_x)=1.24$ at 60°S for the retrieval with fixed n_{imag} and 1.02 for the variable case, which
 490 is still clearly a very significant improvement. Fig. 10 shows the latitudinal dependence
 491 of the computed single-scattering albedo (SSA) of the Aerosol-1 particles at 500 nm, and
 492 we can see a very good correspondence between this and the location of the SPW in the
 493 551-nm image (Fig. 1), with the single-scattering albedo, ϖ , of the Aerosol-1 particles
 494 significantly reduced at 60°S. The reduction of ϖ north of 10°N probably arises due to
 495 contamination with the NDS-2018 dark spot not being wholly deconvolved, combined
 496 with the fact that the zenith angles are less well sampled at these latitudes and so Minnaert-
 497 reconstructed spectra are less reliable. We note here that Karkoschka (2011a) also in-
 498 ferred a very similar latitudinal variation in the IR single scattering albedo (their Fig.
 499 20).

500 If the dark SPW at $\sim 60^\circ\text{S}$ can be explained by a darkening of the Aerosol-1 par-
 501 ticles, might a similar process help explain the banded structure seen at 831 nm? The
 502 right hand panel of Fig. 10 shows the latitudinal variation of the Aerosol-1 particles' single-
 503 scattering albedo at 800 nm. Here, apart from a general trend of increasing ϖ towards
 504 the south pole, we see local peaks of ϖ at 0°, 20°, 45° and 70°, which correspond to peaks
 505 in the observed reflectivity at 831 nm (Fig. 1). This suggests that these 831-nm features
 506 (and similar banding seen at other longer wavelengths of minimal methane absorption
 507 near 680, 750 and 930nm) are caused by a brightening/darkening of the Aerosol-1 par-
 508 ticles at these wavelengths. If this is the case, then the effect is very similar to that in-
 509 ferred for the spectral properties of the 'Deep Bright Spot', DBS-2019, seen near NDS-
 510 2018 and reported by Irwin et al. (2023). To test this hypothesis, Fig. 8 compares the
 511 difference spectrum at 20°S (i.e., the difference between the $(I/F)_0$ spectrum at 20°S
 512 and the average of those at 10°S and 30°S) with the difference spectrum of DBS-2019.
 513 As can be seen there is again a considerable degree of similarity between these difference
 514 spectra, with the difference peaks longer than 650 nm being mostly restricted to narrow
 515 bands centred on 750 and 830 nm. These spectra are not identical, however, with some
 516 differences seen at shorter wavelengths. The DBS-2019 feature is small and it is possi-
 517 ble that there remains some spatial mixing of the light from nearby locations left over
 518 from incomplete deconvolution. However, the similarity of these spectra at longer wave-
 519 lengths, especially the narrowness of the reflectivity peaks, leads us to the conclusion that
 520 these features likely have a similar cause. Hence, we conclude that the SPW and NDS-
 521 2018 features are both caused by a spectrally-dependent darkening of the Aerosol-1 par-
 522 ticles at $\lambda < 650$ nm, while the bright 'zones' seen at 831 nm and DBS-2019 are both
 523 caused by a spectrally-dependent brightening of the same Aerosol-1 particles at $\lambda > 650$
 524 nm. Furthermore, the 511 nm and 831 nm features **must** be caused by spectrally-dependent
 525 perturbations of the Aerosol-1 layer, rather than changes in its opacity, since opacity changes
 526 would affect both wavelengths while it is clear that the latitude dependence of these fea-
 527 tures are very different.

528 Although the formal retrieval errors of the opacity and pressure level of Aerosol-
 529 2 shown in Fig. 9 are smaller than the latitudinal variations, we note that these errors
 530 do not include cross-correlation with other model parameters and so may be underes-

531 timates. Given that we find that the bright and dark features in our data can mostly be
 532 explained by variations in the reflectivity of the Aerosol-1 particles only, we explored whether
 533 we could fit the observations by fixing the properties of Aerosol-2 and varying only the
 534 other remaining parameters (τ_1 , $n_{imag}(\lambda)$, τ_3 , and deep methane abundance). Fixing
 535 τ_2 and p_2 at all latitudes to the mean values found of 1.75 (at 800 nm) and 2.1 bar, re-
 536 spectively (Model 3 of Table 2), we re-ran our retrieval model, whose results are over-
 537 plotted in red in Fig. 9. The quality of the fits can be seen to be only very slightly re-
 538 duced, with the same trends seen in all the other parameters. Hence, we conclude that
 539 to first order the Aerosol-2 layer is latitudinally invariant and that the vast majority of
 540 the latitudinal changes seen in the MUSE data can be attributed to changes in: 1) the
 541 methane abundance; 2) the opacity and scattering properties of the Aerosol-1 layer at
 542 ~ 5 bar; and 3) the opacity of the upper tropospheric Aerosol-3 layer. We will return to
 543 discuss retrieval errors in the discussion section.

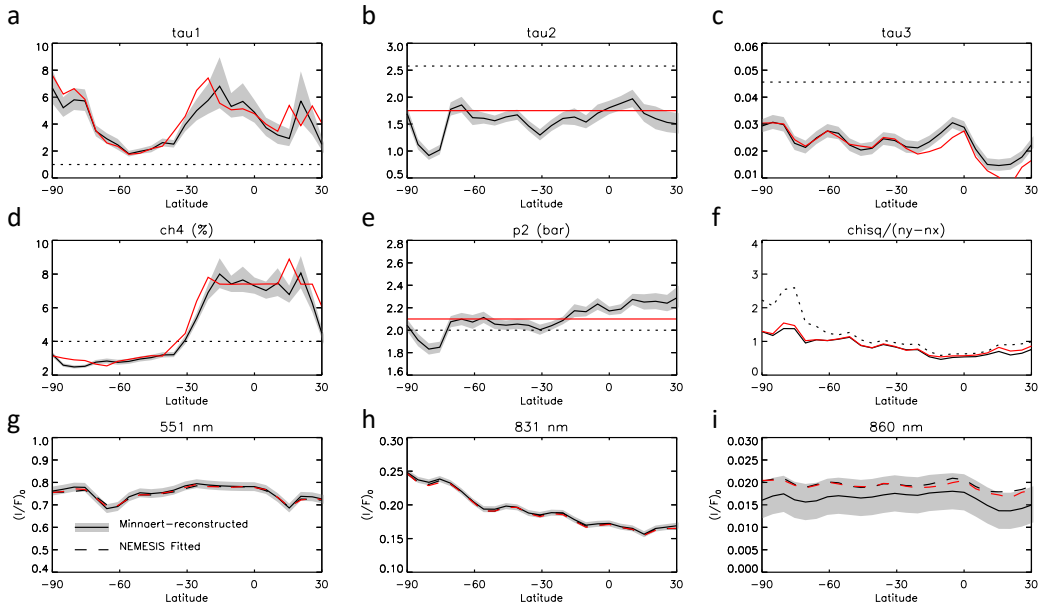


Figure 9. As Fig. 7, but showing atmospheric properties retrieved from the VLT/MUSE Nep-
 ture observations (Obs-6), when the n_{imag} spectrum of the Aerosol-1 particles is also allowed to
 vary (Model 2). In the middle-right panel, showing the latitudinal (planetographic) variation of
 $\chi^2/(n_y - n_x)$, the reduced χ^2 for the case where n_{imag} for Aerosol-1 is fixed is overplotted as a
 dotted line for comparison. Overplotted in red in all panels are the results when the opacity (at
 800 nm) of Aerosol-2 is fixed to 1.75 and its pressure fixed to 2.1 bar (Model 3), showing very
 similar fits to the data and similar reduced χ^2 . Note that the error bars shown in panels a – e
 are the formal errors on the retrievals and do not include cross-correlation effects (see Discussion).

544 5 Discussion

545 The presence of bright ‘zones’ and dark ‘belts’ in the Aerosol-1 layer at wavelengths
 546 of minimal methane absorption near 830 nm (and also less clearly at 670, 750, and 930
 547 nm), with the zones separated by $\sim 25^\circ$ is not something that has specifically been noted
 548 before and is very curious given that the measured wind speeds show a much more slowly
 549 varying latitudinal dependence. These finer-scale latitudinal variations in reflectivity are,
 550 in fact, just visible in HST/WFC3 F845M images (e.g., Chavez et al., 2023), although

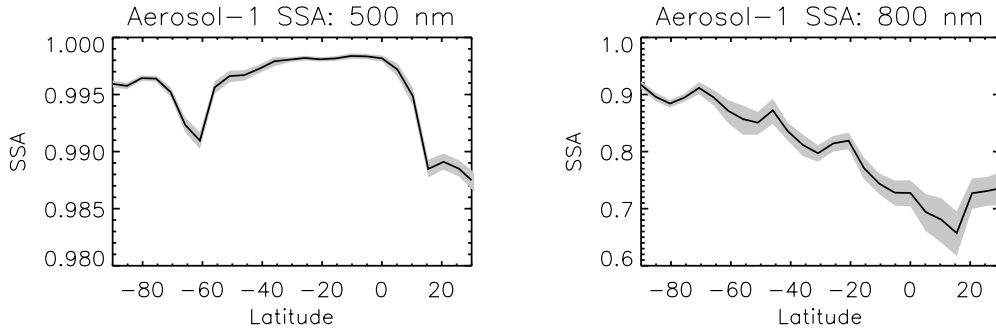


Figure 10. Estimated single-scattering albedoes of the best fit Aerosol-1 particles at 500 nm and 800 nm as a function of latitude (planetographic), derived from our fitted n_{imag} spectra using Mie theory. The formal confidence limits are indicated by the grey shading and do not include cross-correlation effects.

551 much less clear, and at longer wavelengths Sromovsky et al. (2014) note finer-scale struc-
 552 ture in H-band ($1.5 \mu\text{m}$) Keck observations of Uranus. Although just visible in broader
 553 wavelength filters, the banding seen here is only very clear at wavelengths of extremely
 554 weak methane absorption and is not seen at wavelengths of stronger methane absorp-
 555 tion. Only high-resolution IFU spectrometers such as MUSE can discriminate between
 556 these deep features and shallower cloud structures that would appear identical in broader
 557 filters, and thus isolate their surprising origin. Such very different structure below the
 558 visible haze layers is reminiscent of Cassini/VIMS observations of the $5\text{-}\mu\text{m}$ emission from
 559 Saturn’s atmosphere, which revealed finely detailed bright and dark bands that bore lit-
 560 tle resemblance with the broader structures seen at lower pressures (e.g., Baines et al.,
 561 2006). This suggests that the circulation of Neptune’s atmosphere below the 2–3-bar Aerosol-
 562 2 ice/haze layer is likely very different from that seen above that layer.

563 Analysing these VLT/MUSE data we have shown that Neptune’s SPW and dark
 564 spots are likely to be caused by the same process, i.e., a darkening of the particles in the
 565 $\sim 5\text{-bar}$ -Aerosol-1 layer at wavelengths less than 650 nm . Our analysis of the SPW sig-
 566 nature is helped by the zenith-angle coverage allowing us to constrain the limb-darkening
 567 properties, but is complicated by the difficulty in extracting the SPW signature from the
 568 latitudinal variations in other parameters. In contrast, the NDS-2018 difference spec-
 569 trum is relative to other locations in the same latitude band, so extracting its signature
 570 is not complicated by latitudinal variations. However, during the time that NDS-2018
 571 was visible in our data set, from ‘Obs-6’ and ‘Obs-10’, the observed zenith angle only
 572 increased from 42.36° to 45.37° and thus we do not have sufficient zenith angle data to
 573 constrain its limb-darkening characteristics. However, IRW22 analysed the limb-darkening
 574 properties of both the SPW and GDS from Voyager-2 imaging observations of Neptune
 575 in 1989 at several wavelengths and found they had similar limb-darkening, which again
 576 points to a likely common cause of these short-wavelength dark features. At longer wave-
 577 lengths, the signature of the deep bright spot DBS-2019 is similar to the differences be-
 578 tween the bright ‘zones’ and dark ‘belts’ seen at longer continuum wavelengths, such as
 579 831 nm , and both seem to be caused by a brightening of the particles in the $\sim 5 \text{ bar}$ -Aerosol-
 580 1 layer at wavelengths $> 650 \text{ nm}$. It is intriguing that two very distinctive and very dif-
 581 ferent features are caused by modifications of the same Aerosol-1 layer at wavelengths
 582 either less than or greater than 650 nm , and we thus looked to determine whether there
 583 might be a simple explanation for these very different reflectance signatures.

584 In Fig. 11 we have again plotted the n_{imag} spectrum of the Aerosol-1 particles re-
 585 trieved from our disc-average Minnaert limb-darkening analysis. As can be seen we re-

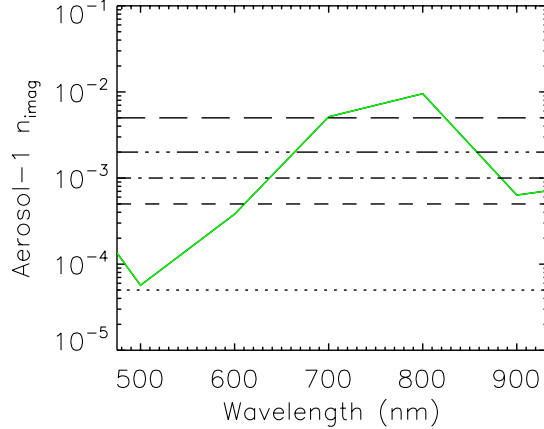


Figure 11. Imaginary refractive index spectrum of the fitted disc-averaged Aerosol-1 particles (green), compared with the n_{imag} spectra of the additional particles added to the Aerosol-1 layer in the reflectivity calculations shown in Fig. 12.

586 retrieve low values near 500 nm, making the particles highly scattering ($\varpi = 0.999$), ris-
 587 ing to large values at 800 nm, making the particles poorly scattering ($\varpi = 0.777$). The
 588 increase of n_{imag} with wavelength is commonly seen with photochemically-produced par-
 589 ticles as discussed by IRW22, who conclude that the particles in Aerosol-1 layer are likely
 590 composed of a mixture of bright H_2S ice particles and photochemical products mixed
 591 down from above. The large single-scattering albedo difference on either side of 650 nm
 592 is reminiscent of the difference between deep dark and bright spots and we wondered what
 593 would happen to the computed disc-averaged $(I/F)_0$ spectrum if we added an additional
 594 component of particles to the Aerosol-1 layer with different, fixed n_{imag} spectra. The
 595 n_{imag} values we chose were 5×10^{-5} , 5×10^{-4} , 5×10^{-3} , 2×10^{-3} and 1×10^{-3} (over-
 596 plotted in Fig. 11), roughly spanning the range of n_{imag} seen in the retrieved Aerosol-
 597 1 spectrum. We also tested whether the particle size might be important and used a size
 598 distribution for the additional particles either equal to that assumed for the Aerosol-1
 599 layer with mean radius $r_0 = 0.1 \mu\text{m}$, or increased to $r_0 = 1.0 \mu\text{m}$. The particle size vari-
 600 ance in both cases was set to $\sigma = 0.05$. The additional particles had the same verti-
 601 cal distribution as the existing Aerosol-1 layer and had an opacity (at 800 nm) relative
 602 to the existing Aerosol-1 layer of 10% for the $0.1\text{-}\mu\text{m}$ particles or 30% for the less back-
 603 scattering $1.0\text{-}\mu\text{m}$ particles.

604 The differences in the computed $(I/F)_0$ spectra are shown in Fig. 12, which demon-
 605 strates that when the additional particles have larger n_{imag} values, and thus lower single-
 606 scattering albedo, we see a darkening at shorter wavelengths, but little effect at longer
 607 wavelengths. This is understandable given that the additional darker particles will have
 608 a large effect at wavelengths where the surrounding Aerosol-1 particles are highly scatter-
 609 ing, but will have a minimal impact at wavelengths where the surrounding particles
 610 are already rather absorbing. Similarly, adding highly-scattering particles to the Aerosol-
 611 1 layer, with low n_{imag} , will not make much difference at wavelengths where the parti-
 612 cles are already highly scattering, but will significantly affect wavelengths where the sur-
 613 rounding particles have lower albedo. For either additional particle size, when n_{imag} is
 614 high (e.g., 1×10^{-3}), the difference in the computed $(I/F)_0$ spectrum matches closely
 615 to the SPW and NDS-2018 difference spectra (Fig. 8). On the other hand, for highly scatter-
 616 ing particles (e.g., $n_{imag} = 5 \times 10^{-5}$) we see not only narrow reflectance peaks at
 617 longer wavelengths, but also an increase in reflectance at shorter wavelengths. However,

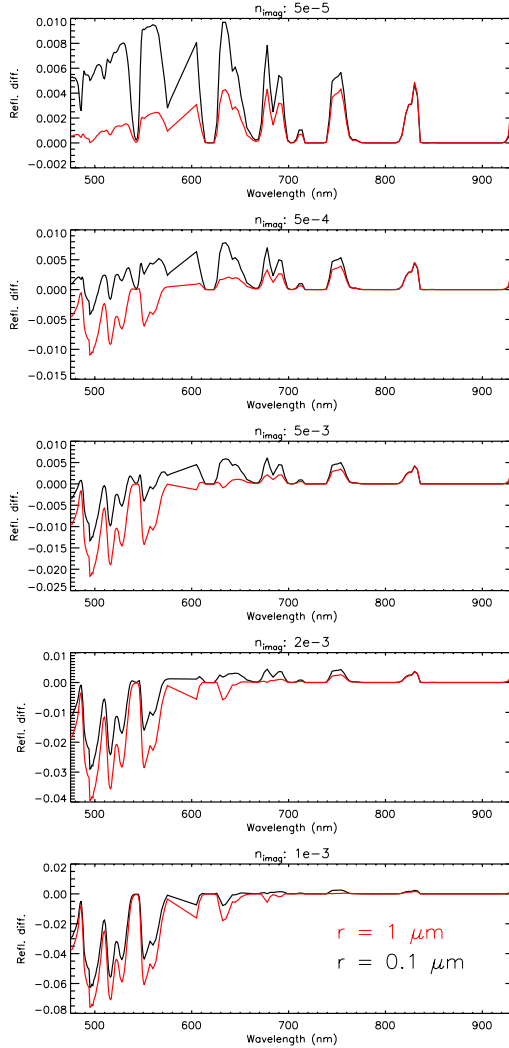


Figure 12. Change in calculated disc-averaged $(I/F)_0$ spectrum when an additional opacity of particles with specified n_{imag} is added to the particles in the Aerosol-1 layer. The additional particles either have a mean radius of $0.1 \mu\text{m}$ (black) or $1.0 \mu\text{m}$ (red) and the additional opacity (at 800 nm) is 10% or 30% of the existing Aerosol-1 opacity, respectively. In both cases the variance of the additional size distribution is 0.05.

618 for small particles and n_{imag} in the range of 5×10^{-4} to 5×10^{-3} the effect on the $(I/F)_0$
 619 spectra is very similar to the 20°S and DBS-2019 difference spectra of Fig. 8, with nar-
 620 row reflectance peaks at longer wavelengths and little clear signature at shorter wave-
 621 lengths. Hence, rather than trying to explain why the average scattering spectra of the
 622 Aerosol-1 particles in the 831-nm bright regions and 500-nm dark spots are so different
 623 from the background, we could instead consider simpler solutions where dark spots are
 624 caused by the addition of dark particles, or ‘chromophores’, that are generally absorb-
 625 ing across the MUSE range and 831-nm bright regions are caused by the addition of other
 626 particles that are generally scattering, such as fresh H_2S ice.

627 Although this chromophore model is simple and easy to understand, it does then
 628 leave the question of what the additional chromophores might be. An alternative expla-
 629 nation for dark regions in Neptune’s atmosphere, noted by IRW22, is that warm regions

630 at the Aerosol-1 pressure level might lead to H₂S ice sublimating off from the particles,
 631 revealing their darker photochemically-produced cores. This would have more of an ef-
 632 fect at wavelengths where the reference Aerosol-1 particles are bright. Equally, cooler
 633 regions might result in more H₂S ice condensing on to the particles, which would not make
 634 them more reflective at wavelengths where they are already bright, but could lead to them
 635 becoming brighter at the longer wavelengths where we see belts and zones. Observations
 636 of Neptune with VLA and ALMA (Tollefson et al., 2021) note latitudinal variations in
 637 brightness temperature at wavelengths sounding 4 – 8 bar that are similar to the reflect-
 638 ivity variations seen in our longer wavelength (> 600 nm) methane windows, which sug-
 639 gests there might indeed be a link. However, the direction of such a link is arguable: warmer
 640 regions might lead to H₂S ice sublimating off the haze cores to make the particles darker,
 641 but it might also be that darker aerosol particles will absorb more sunlight and thus in-
 642 crease the local atmospheric warming.

643 While the ‘zones’ at 831 nm are consistent with a brightening of the Aerosol-1 par-
 644 ticles, we can see from Fig. 10 that there is general increase of the 800-nm single-scattering
 645 albedo (SSA) towards the south pole. In Fig. 13 selected retrieved parameters (for Model
 646 3, where the Aerosol-1 n_{imag} spectrum was allowed to vary, but the opacity and pres-
 647 sure level of the Aerosol-2 layer fixed) are plotted as disc images. The SSA image at 500
 648 nm shows generally good correspondence with the brightness of the 511-nm image in Fig.
 649 1 and the SSA image at 800 nm shows good correspondence with the 831-nm image south
 650 of the equator, except north of the equator, where the Minnaert-reconstructed spectra
 651 are less reliable. In particular, the bright 831-nm zone seen at 75°S (which was noted
 652 by Irwin et al. (2023)), just south of the SPW corresponds with a region of high 800-nm
 653 SSA, and another such region lies on the SPW’s northern edge. Might it be that fresh
 654 material is upwelling on either side of the SPW, then moves into the SPW, suffering pho-
 655 tochemical alteration along the way and darkening it to form the 500-nm-dark SPW fea-
 656 ture? A possible candidate for the fresh material might be H₂S ice, and gaseous H₂S was
 657 detected in Gemini/NIFS observations of Neptune in 2010 (Irwin, Toledo, Garland, et
 658 al., 2019), who found its signature (near 1.5 μ m) to be stronger towards Neptune’s south
 659 pole, indicating higher abundance at 2–3 bar. However, deeper in the atmosphere, ob-
 660 servations with the Very Large Array and ALMA (Tollefson et al., 2021) find low ther-
 661 mal emission at equatorial latitudes and high thermal emission at polar latitudes. At the
 662 depths sounded at these microwave wavelengths (\sim 10 bar), low thermal emission regions
 663 are interpreted as being regions of high H₂S abundance and thus Tollefson et al. (2021)
 664 concluded that H₂S is enriched at equatorial latitudes, just as we see in our CH₄ retrievals,
 665 indicative of strong and persistent upwelling. It may be that the H₂S signature seen by
 666 Gemini/NIFS was obscured near the equator by the enhanced Aerosol-1 opacity we re-
 667 trieve in this study, making it easier to detect at more polar latitudes near 1.5 μ m. Or
 668 is it the case that the latitudinal distribution of H₂S is different at different pressure lev-
 669 els?

670 Much higher in the atmosphere the ring of stratospheric haze (Aerosol-3) surround-
 671 ing the south pole at 80°S is intriguing. We can see the feature in our MUSE data at
 672 all methane-absorbing wavelengths longer than 650 nm and also searched for it in HST/WFC
 673 observations made during the same apparition, detecting it in the FQ619N and FQ727N
 674 filters (both narrow-band filters, centred on wavelengths of strong methane absorption),
 675 but not finding it in the broadband HST/WFC3 filters. The ring of material clearly re-
 676 sides high in the atmosphere, near the tropopause and perhaps marks the edge of the
 677 south polar vortex in Neptune’s upper atmospheric circulation, although Fletcher et al.
 678 (2014) and de Pater et al. (2014) put the edge of the south polar vortex, or the prograde
 679 polar jet, at \sim 70°S. In addition to the bright ring at 80°S at methane-absorbing wave-
 680 lengths we also see a slight increase of reflectivity at the equator. Such a distribution in-
 681 dicates upwelling of air at mid-latitudes followed by photodissociation of methane lead-
 682 ing to haze production, with the air then moving the haze north and south and concen-

683 trating the aerosols at the equator and poles before it downwells (de Pater et al., 2014;
684 Tollefson et al., 2021).

685 The retrieved ‘deep’ abundance of methane varies from 6–7% at the equator to ~3%
686 at polar latitudes, with a boundary at 20–30°S. These abundances are slightly higher
687 than previously retrieved (Karkoschka & Tomasko, 2010; Irwin, Toledo, Braude, et al.,
688 2019; Irwin et al., 2021), but we know that the absolute abundance depends both on the
689 assumed vertical profile of methane and also the assumed vertical profile of clouds/hazes.
690 Our scheme has a methane profile that is fixed up to the condensation level, falling with
691 a prescribed relative humidity above, and is combined with three simply vertically pa-
692 rameterised aerosol layers. Although we show that this model matches the data very well,
693 alternative parameterisations could fit similarly well, but return different absolute methane
694 abundances. In particular, Karkoschka and Tomasko (2009, 2011) and Sromovsky et al.
695 (2011) favour a ‘descended’ methane profile that is depleted in the lower troposphere
696 (at 1–5 bar), but returns to the same value at all latitudes at great depth. Indeed, if lat-
697 itudinal variations in methane really extended to great depth, then Sromovsky et al. (2011)
698 notes that there would be a significant gradient of mean molecular weight with latitude
699 and significant gradients of vertical wind shear (e.g., Sun et al., 1991; Tollefson et al.,
700 2018) that are inconsistent with the observed winds. Hence, while we are confident of
701 the shape of the retrieved latitudinal abundance of methane, we can be less sure of the
702 absolute profile and abundances. However, the general increase at longer MUSE wave-
703 lengths of the latitudinally-resolved Minnaert nadir reflectivity $(I/F)_0$ south of 20–30°S,
704 relative to the disc-averaged $(I/F)_0$ spectrum, noted in Fig. 5, can now simply be in-
705 terpreted as being caused by the lower methane abundances determined at these lati-
706 tudes.

707 The circulation of Neptune’s atmosphere has been studied for decades, but is still
708 puzzling. In the horizontal direction, we know that the winds are predominantly zonal
709 (i.e., east-west), with strongly retrograde winds at the equator with speeds of nearly 400
710 m/s, becoming prograde polewards of ~45°N and ~45°S, reaching peak prograde speeds
711 of over 200 m/s at ~75°N and ~75°S, before falling to zero at the poles (Sromovsky et
712 al., 1993; Sánchez-Lavega et al., 2019). In the vertical direction, the presence of thick
713 upper-tropospheric methane ice clouds and cooler retrieved upper-tropospheric temper-
714 atures at mid-latitudes (20–40°N and 20–40°S) indicates upwelling at these latitudes and
715 cooling as the air adiabatically expands at the tropopause and moves towards the equa-
716 tor and poles (Conrath et al., 1991; Bezaud et al., 1991; Fletcher et al., 2014; de Pater
717 et al., 2014; Roman et al., 2022). This circulation is consistent with the distribution of
718 Aerosol-3 particles we detect in our MUSE observations. Deeper down, the significant
719 vertical reduction of the mean molecular weight at the CH₄ condensation level at ~ 2
720 bar seems to act as a barrier to vertical motion (e.g., IRW22) and below this the ver-
721 tical circulation appears to switch with air rising at equatorial latitudes and falling nearer
722 the pole, leading to higher equatorial abundances of CH₄, seen here, and also high deep
723 H₂S abundances seen at microwave wavelengths (Tollefson et al., 2021). This simple ‘stacked
724 circulation’ view (e.g., Tollefson et al., 2019; Fletcher et al., 2020), however, is inconsis-
725 tent with the banded structure seen here at methane continuum wavelengths longer than
726 650 nm and may also be inconsistent with retrieved H₂S abundances (at 1.5 μm) at pres-
727 sures less than 5 bar (Irwin, Toledo, Garland, et al., 2019), suggesting that the circula-
728 tion may actually be even more complicated than previously thought. An interesting anal-
729 ogy is the distribution of NH₃ seen in Jupiter’s deep atmosphere by the Juno spacecraft
730 (Li et al., 2017), which appears to rise in an equatorial plume, with much greater abun-
731 dances than at mid-latitudes, although a more zonal NH₃ structure is seen at the 0.5–
732 2-bar level by both Juno (Fletcher et al., 2021), and also in mid-infrared observations
733 (e.g., Fletcher et al., 2016); this is believed to arise and be maintained by a similar dou-
734 ble ‘stacked cell’ system (e.g., Ingersoll et al., 2000; Showman & de Pater, 2005; Duer
735 et al., 2021; Fletcher et al., 2021; Moeckel et al., 2023; de Pater et al., 2023).

736 The data analysed in this study have been fitted by varying just the following few
 737 variables: the opacity of the three aerosol layers, the pressure level of Aerosol-2 layer,
 738 the ‘deep’ methane abundance, and the imaginary refractive index spectra of the three
 739 aerosol types. The scattering properties of a distribution of particles in a planetary at-
 740 mosphere depend on very many factors, including the size distribution, composition, and
 741 mix of different particle species. Regrettably, for Neptune we have very little *a priori* con-
 742 straint on what the expected particles and their size distributions should be, nor any re-
 743 liable information on their complex refractive index spectra. As a result, have to try and
 744 retrieve these from the data themselves if we are fit the observations satisfactorily. We
 745 do this using optimal estimation method (Rodgers, 2000), expanded upon below, which
 746 is a deepest descent approach and thus is most stable when the rate of change of radi-
 747 ance with model parameter is a smoothly varying function of the value of that param-
 748 eter. Unfortunately, we find that the rate of change of radiance does not vary smoothly
 749 enough with the mean radius of the size distribution to reliably retrieve this with our
 750 model. Hence, instead we conduct separate retrievals for a range of fixed particle size
 751 distributions and choose those that match the data best (e.g., IRW22). We do find, how-
 752 ever, that the model behaves well when varying the n_{imag} spectra, and we do find that
 753 we need for the scattering properties of the particles to vary more rapidly with wavelength
 754 than simple changes in r_{mean} or the variance of the size distribution can achieve. We at-
 755 tribute this to the fact that the particles are likely made up of unknown photochemical
 756 products that have strong absorption bands. We thus fix the size distribution of the par-
 757 ticles in a certain aerosol layer to an acceptable shape (compared with a range of obser-
 758 vations) and then retrieve the n_{imag} spectra, reconstructing the n_{real} spectra using the
 759 Kramers-Kronig approach. In reality the particles may be a mix of separate ice and pho-
 760 tochemical particles, but they could also be combined together by ‘riming’ or some other
 761 combination mechanism. Unfortunately, we just do not have this information and so we
 762 instead retrieve the mean scattering properties of the layer by fitting n_{imag} as we describe.
 763 Although an approximation, we find that our approach allows us to fit the data well and
 764 we can then work on interpreting what we have found. This is precisely the point of the
 765 first part of this discussion, where we show that the variation of the n_{imag} spectra with
 766 latitude of the ‘mean’ particles in the Aerosol-1 layer can be interpreted as being due
 767 by the presence of a background distribution of particles, presumably rich in photochem-
 768 ical haze, combined with a latitudinally-varying opacity of fresh ice crystals. It may be
 769 that these two particle distributions have different mean radii and variances, but these
 770 cannot be uniquely separated using these data and so we assume a single mean size dis-
 771 tribution. Ideally, we would have the complex refractive index spectra of a set of laboratory-
 772 measured possible condensates to test against the measured spectra, but this data set
 773 does not yet exist. In the meantime, we believe that we have developed a method that
 774 reliably retrieves the mean scattering properties of the aerosol layers, which can then be
 775 interpreted to gain novel insights into the hazes and clouds in Neptune’s atmosphere.

776 Finally, we note that the retrievals presented here were conducted using the op-
 777 timal estimation (Rodgers, 2000) framework of the NEMESIS model (Irwin et al., 2008).
 778 To model these spectral observations requires a full multiple-scattering radiative trans-
 779 fer model, which must be run at many latitudes, many wavelengths, and with many tun-
 780 able parameters. Hence, this model is computationally very expensive and to combine
 781 it with a Bayesian retrieval framework (such as Monte-Carlo Markov Chain or Nested
 782 Sampling, e.g., Skilling (2006)), which require tens of thousands of iterations, would be
 783 prohibitively computationally expensive. However, although cross-correlation between
 784 parameters can to be interpreted in optimal estimation method from the computed co-
 785 variance matrix, this is much less easy to comprehend and present than the ‘corner plots’
 786 of Bayesian approaches. The retrieval errors we present in this work are derived from
 787 diagonal elements of the retrieved covariance matrix and corrected for the original *a pri-*
 788 *ori* errors, since if the retrieved error is the same as the *a priori* error we have not learnt
 789 anything new. Hence, the errors shown are $\sigma = 1/\sqrt{(1/\sigma_{ret}^2 - 1/\sigma_{apr}^2)}$, where σ_{ret}^2 are
 790 diagonal components of the retrieved covariance matrix and σ_{apr}^2 are diagonal compo-

791 nents of the *a priori* covariance matrix. However, these errors still do not wholly account
 792 for cross-correlation effects. Instead, we estimate the magnitude of these effects by fix-
 793 ing some parameters and observing the effect on the other retrieved variables. For ex-
 794 ample, going from Model 2 to Model 3 we show that by fixing the pressure and opac-
 795 ity of the Aerosol-2 layer we can achieve very similar quality fits and very similar vari-
 796 ations with latitude of the other retrieved parameters. How, then, can we be sure that
 797 the retrieved latitudinal variations of the remaining parameters in Model 3 are reliable?
 798 Figure 8 shows that dark spots, the SPW and the zone/belt signature at longer contin-
 799 uum wavelengths are all most consistent with changes in the Aerosol-1 layer, and that
 800 this study and previous analyses (Irwin, Teanby, Fletcher, et al., 2022; Irwin et al., 2023)
 801 find that spectrally-dependent changes in the reflectivity of the Aerosol-1 particles pro-
 802 vide the best solution, which is what Model 3 achieves (Fig. 10). There are no clear lat-
 803 itudinal variations that have a spectral signature consistent with significant latitude vari-
 804 ations in the Aerosol-2 layer, but variations seen at methane-absorbing wavelengths, and
 805 which thus must be high in the atmosphere, are matched by latitude variations in the
 806 opacity of the Aerosol-3 layer and there is a clear methane variation signature in the ob-
 807 servations, noted earlier. Hence, our Model 3 is consistent with all the evidence in the
 808 MUSE data set and is, we believe, reliable. We also note that as this model was based
 809 on the ‘holistic’ model of IRW22, which was originally derived from a much wider wave-
 810 length range data set, it is likely to be applicable to longer wavelengths also. Observa-
 811 tions of Neptune have recently been made with the NIRSpc and MIRI instruments on
 812 the James Webb Space Telescope (JWST) and we look forward to seeing if our param-
 813 eterisation can be extended successfully to the longer wavelengths sampled by JWST also.

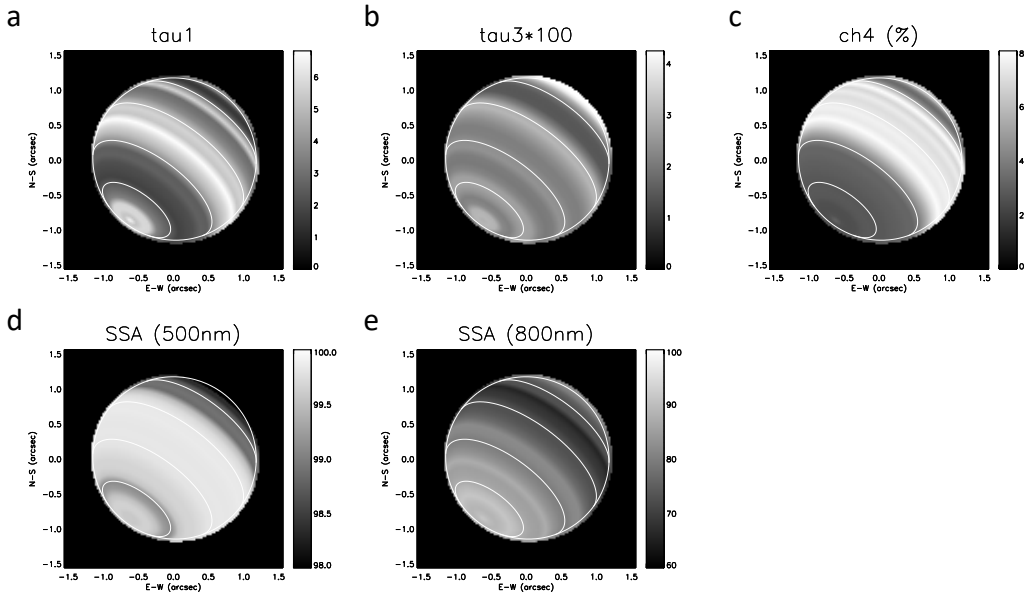


Figure 13. Similar to Fig. 6, but showing atmospheric properties retrieved and projected on to Neptune’s disc when the Aerosol-1 *n_{imag}* spectrum is allowed to vary, and the Aerosol-2 properties are fixed (Model 3), mapping: a) Aerosol-1 opacity; b) Aerosol-3 opacity ($\times 100$); c) deep methane mole fraction (%); d) Aerosol-1 single-scattering albedo (%) at 500 nm; and e) Aerosol-1 single-scattering albedo (%) at 800 nm. The cloud opacities are those at calculated at 800 nm. Latitude circles are overplotted for ease of reference, with a spacing of 30° .

6 Conclusions

Our analysis of latitudinally-resolved centre-to-limb spectra of Neptune, observed with VLT/MUSE in October 2019, has revealed new constraints on the mechanism that causes the darkening of the South Polar Wave at $\sim 60^\circ\text{S}$, which we conclude is common with the darkening mechanism for discrete dark spots such as Voyager-2’s Great Dark Spot and the more recent NDS-2018 feature. Using a modified version of the ‘holistic’ aerosol model of IRW22 we find that both these features are consistent with being caused by the darkening (at wavelengths less than 650 nm) of particles in an Aerosol-1 layer, based at ~ 5 bar, which we suggest is composed of a mixture of photochemically-produced haze, mixed down from their production level above, and H_2S ice. In addition, we note a latitudinal brightening and darkening (with a scale of $\sim 25^\circ$) in Neptune’s aerosols, which is only visible at regions of very low methane absorption longer than 650 nm. We show that this feature is caused by a brightening (at wavelengths greater than 650 nm) of the particles in the same Aerosol-1 layer at ~ 5 bar. We conclude that these different features seen at $\lambda < 650$ nm and $\lambda > 650$ nm must be caused by spectrally-dependent perturbations of the Aerosol-1 scattering properties, rather than opacity changes of this layer, since they have very different spatial structures and changes in the opacity would affect both wavelength ranges similarly.

Above the Aerosol-1 layer at ~ 5 bar, we find the properties of the main Aerosol-2 layer, confined to a region of high static stability at the CH_4 -condensation level at ~ 2 bar, is to first order invariant with latitude, although the base pressure and opacity may be slightly reduced at 80°S . Even higher in the atmosphere, variations in the opacity of the Aerosol-3 tropospheric haze are able to reproduce the observed variation in reflectivity at methane-absorbing wavelengths. We find the abundance of this aerosol to be higher at the equator and also in a narrow ‘zone’ at 80°S , which supports the hypothesis that at the tropopause air is rising at mid-latitudes and then moving polewards and equatorwards, concentrating the photochemical haze products there. The bright ring at 80°S would appear to define the edge of a polar cyclone that has been observed on Neptune for the past two decades, and which are often seen in giant planet atmospheres.

The detailed conclusions of this paper are:

1. We find a slightly modified version of the ‘holistic’ aerosol model of IRW22, consisting of three distinct layers, fits the VLT/MUSE data very well, with the Aerosol-1 parameterisation revised to be a single, vertically-confined layer;
2. We find similar, but revised n_{imag} wavelength dependences to those derived by IRW22 for the three aerosol types. This revision was necessary to match centre-to-limb functions of the deconvolved MUSE observations. In particular, we see more limb-brightening at methane-absorbing wavelengths in our deconvolved MUSE data, requiring a higher single-scattering albedo for the upper atmospheric haze particles in the Aerosol-3 layer;
3. We find that the Aerosol-1 opacity varies strongly with latitude, with high values retrieved at the equator and south pole and lower values retrieved at mid latitudes;
4. We find that to a first approximation, the opacity and pressure of the Aerosol-2 layer remains constant, although there are indications that both may be slightly reduced near 80°S ;
5. We find that the retrieved opacity of the Aerosol-3 layer matches well the distribution of upper atmospheric haze seen at methane-absorbing wavelengths, and has highest abundance at the equator and 80°S , with secondary maxima at 30°S and 60°S ;
6. We find a smooth latitudinal distribution of ‘deep’ mole fraction of methane (i.e., below the condensation level) in our simple ‘step’ methane profile, with values of 6–7% at the equator reducing to $\sim 3\%$ south of 25°S ;

- 866 7. We find the South Polar Wave at $\sim 60^\circ\text{S}$ to be caused by a perturbation of the par-
867 ticles in the Aerosol-1 layer that reduces the single-scattering albedo at shorter
868 wavelengths. The darkening appears identical to that which darkens the dark spot
869 NDS-2018 and could be caused either by the addition of a chromophore in this
870 layer that is darker than the background particles at all MUSE wavelengths, or
871 by local heating at the ~ 5 -bar level that sublimates H_2S ice off to reveal the dark
872 haze cores of the Aerosol-1 particles;
- 873 8. We note a newly apprehended latitudinal variation in the reflectivity in narrow
874 continuum wavelengths longer than 650 nm. We interpret these changes as being
875 caused by variations in the scattering properties of the Aerosol-1 layer at wave-
876 lengths longer than 650 nm, with bright ‘zones’ at the equator, 20°S , 45°S , and
877 75°S , interspersed with darker ‘belts’ at 5°S , 30°S , 50°S , and 80°S . This coloration
878 could be caused by the addition of brighter, more scattering particles at the zone
879 latitudes, or by local cooling at the ~ 5 -bar level that condenses more bright H_2S
880 ice on to the Aerosol-1 particles.

881 The banding seen in the narrow windows of very low methane absorption longer
882 than 650 nm is suggestive of a finer latitudinal scale variation than has been noted be-
883 fore on Neptune at the level of the H_2S /haze Aerosol-1 layer at ~ 5 bar and indicates that
884 the circulation of Neptune’s atmosphere is even more complicated than that suggested
885 by the ‘stacked circulation’ models of Tollefson et al. (2019) and Fletcher et al. (2020).
886 Further work is needed to explore such changes more fully and recent observations with
887 the James Webb Space Telescope should provide strong new constraints. In addition,
888 the spatial deconvolution techniques developed for this study could be applied to exist-
889 ing ground-based IFU measurements from instruments such as Gemini/NIFS (Irwin et
890 al., 2011) and VLT/SINFONI (Irwin et al., 2016) to extend the high spatial resolution
891 analysis to longer wavelengths, and perhaps better constrain the latitudinal dependence
892 of H_2S abundance. Further in the future, a space mission to one of the Ice Giants, com-
893 prising an orbiter and an entry probe would be invaluable in providing some measure
894 of ‘ground truth’ for aerosol models. In particular, it would be useful to observe with a
895 probe the penetration depth of the UV flux to establish the pressure levels at which we
896 may expect the photolysis of methane and other components. Meanwhile, an orbiter would
897 provide reflectivity observations at higher phase angle, which will help to further con-
898 strain the allowable range of solutions that are consistent with observations.

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910 7 Open Research

911 The raw VLT/MUSE datasets studied in this paper (under ESO/VLT program:
912 0104.C-0187) are available from the ESO Portal at https://archive.eso.org/eso/eso_archive_main.html. The reduced raw and deconvolved ‘cubes’ for the observation IDs:
913 6 – 10, discussed in this paper, are available at Irwin (2023b). Data files associated with
914 this analysis are available at Irwin (2023a). The spectral fitting and retrievals were per-
915

916 formed using the NEMESIS radiative transfer and retrieval algorithm Irwin et al. (2008)
 917 and can be downloaded from Irwin, Teanby, de Kok, et al. (2022a) (or [https://github](https://github.com/nemesiscode/radtrancode)
 918 [.com/nemesiscode/radtrancode](https://github.com/nemesiscode/radtrancode)), with supporting website information at Irwin, Teanby,
 919 de Kok, et al. (2022b) (or <https://github.com/nemesiscode/nemesiscode.github.io>).

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