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1 **Time variability of Neptune’s horizontal and vertical cloud**
2 **structure revealed by VLT/SINFONI and Gemini/NIFS from**
3 **2009 to 2013**

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ABSTRACT

18

19

New observations of Neptune’s clouds in the near infrared were acquired in October 2013 with SINFONI on ESO’s Very Large Telescope (VLT) in Chile. SINFONI is an Integral Field Unit spectrometer returning a 64×64 pixel image with 2048 wavelengths. Image cubes in the J-band ($1.09 - 1.41 \mu\text{m}$) and H-band ($1.43 - 1.87 \mu\text{m}$) were obtained at spatial resolutions of $0.1''$ and $0.025''$ per pixel, while SINFONI’s adaptive optics provided an effective resolution of approximately $0.1''$. Image cubes were obtained at the start and end of three successive nights to monitor the temporal development of discrete clouds both at short timescales (i.e. during a single night) as well as over the longer period of the three-day observing run. These observations were compared with similar H-band observations obtained in September 2009 with the NIFS Integral Field Unit spectrometer on the Gemini-North telescope in Hawaii, previously reported by Irwin et al., *Icarus* 216, 141-158, 2011, and previously unreported Gemini/NIFS observations at lower spatial resolution made in 2011.

We find both similarities and differences between these observations, spaced over four years. The same overall cloud structure is seen with high, bright clouds visible at mid-latitudes ($30 - 40^\circ\text{N,S}$), with slightly lower clouds observed at lower latitudes, together with small discrete clouds seen circling the pole at a latitude of approximately 60°S . However, while discrete clouds were visible at this latitude at both the main cloud deck level (at 2–3 bars) and in the upper troposphere (100–500mb) in 2009, no distinct deep (2–3 bar), discrete circumpolar clouds were visible in 2013, although some deep clouds were seen at the southern edge of the main cloud belt at $30-40^\circ\text{S}$, which have not been observed before. The nature of the deep sub-polar discrete clouds observed in 2009 is intriguing. While it is possible that in 2013 these deeper clouds were masked by faster moving, overlying

features, we consider that it is unlikely that this should have happened in 2013, but not in 2009 when the upper-cloud activity was generally similar. Meanwhile, the deep clouds seen at the southern edge of the main cloud belt at 30 – 40°S in 2013, should also have been detectable in 2009, but were not seen. Hence, these observations may have detected a real temporal variation in the occurrence of Neptune’s deep clouds, pointing to underlying variability in the convective activity at the pressure of the main cloud deck at 2–3 bars near Neptune’s south pole and also in the main observable cloud belt at 30 – 40°S.

20 *Subject headings:* planets and satellites: atmospheres — planets and satellites:
21 individual (Neptune)

1. Introduction

22

23 The highly dynamic clouds of Neptune have long fascinated planetary astronomers
 24 since Voyager 2’s flyby of that planet in 1989. Since that time, with the advent of techniques
 25 such as Adaptive Optics operating with larger and larger telescopes, ground-based
 26 observations of this most distant of the planets have improved beyond all recognition and
 27 the atmosphere of Neptune has been discovered to be even more active and dynamic than
 28 that seen by Voyager 2. In addition to larger telescopes and better imaging, a new class of
 29 instruments, Integral Field Unit (IFU) spectrometers, have been constructed, such as the
 30 NIFS instrument on Gemini-North and the SINFONI instrument at the European Southern
 31 Observatory’s (ESO) Very Large Telescope (VLT), which simultaneously map the entire
 32 FOV of the instrument at thousands of wavelengths with spectral resolving powers in excess
 33 of $R = \lambda/\Delta\lambda = 1000$.

34 Gemini/NIFS observations of Neptune recorded in the H-band in 2009 (1.48 – 1.80 μm)
 35 were presented by Irwin et al. (2011) and used to determine the vertical cloud structure of
 36 particular features at several locations on Neptune’s disc over a period of several days. High
 37 clouds were seen at mid latitudes (30–40°N,S) (with tops reaching to and in some cases
 38 above the expected tropopause level), slightly lower clouds observed at more equatorial
 39 latitudes near the morning terminator, and discrete clouds detected around the south pole
 40 at $\sim 60^\circ\text{S}$. These sub-polar clouds were seen to be of two types: one with very high cloud
 41 tops (extending to the tropopause again), and another which were apparently confined to
 42 the level of the main cloud deck at 2–3 bars. Since clouds at these two levels move with
 43 different wind speeds due to vertical wind shear, the upper clouds occasionally obscured
 44 the lower ones and so it was not possible to determine categorically whether the deeper
 45 clouds were long-lived or transient. It is possible that one of the deep clouds was present
 46 throughout the seven-day observing run, but one of the features seems to have appeared

47 and disappeared during a few days, which is remarkably fast for feature changes at the
48 main cloud deck level and would indicate vigorous convective activity.

49 Observing Neptune from the Earth is complicated by the fact that its rotational period
50 of 16.11 hours, as determined by the Voyager 2 Radio Science Experiment (e.g. Lacacheux
51 et al. (1993)), means that the hemisphere of Neptune observed on one night is almost
52 exactly the opposite to that which was observed on the previous night, and so we have to
53 wait two nights to see the same feature at the same place on Neptune’s disc. During this
54 long period the cloud features are distorted by Neptune’s zonal winds, which are extremely
55 strong and change enormously with latitude leading to huge latitudinal wind shears that
56 can tear apart newly formed cloud features on timescales of a few hours. To quantify this
57 level of shear, the winds at the equator are strongly retrograde (-400 m/s, Sromovsky et al.
58 (1993)) and thus the effective rotation period is 18.8 hours, while in the sub-polar jets the
59 effective rotation period is as small as 11 hours.

60 To counter these observational problems and also determine how Neptune’s cloud
61 activity is evolving, we proposed to use the SINFONI instrument on VLT in 2013 to
62 observe Neptune again, but this time making two observations per night, one near the
63 beginning of Neptune’s transit and one near the end so that we could observe the same
64 cloud features over a few hours as they transited the disc. The goals of our observations
65 were to: 1) determine how quickly Neptune’s cloud features evolve with time; 2) determine
66 if the equatorial clouds seen near Neptune’s morning terminator in 2009 survive as they
67 pass across Neptune’s disc; and 3) determine the spatial distribution of deep discrete cloud
68 features and monitor any changes that may have occurred since 2009.

2. VLT/SINFONI Observations

Observations of Neptune were made with the SINFONI instrument in October 2013 at the European Southern Observatory (ESO) Very Large Telescope (VLT) in La Paranal, Chile. At the time of observation, the diameter of Neptune’s disc was $2.3''$, while the sub-observer latitude was 27.73°S . SINFONI is an Integral Field Spectrograph that can make use of Adaptive Optics to yield a spatial resolution of typically $0.1''$. Each one of SINFONI’s 32 slitlets is imaged onto 64 pixels of the detector, giving 64×32 individual spectra, each with 2048 wavelengths, which are usually doubled in the cross-slit direction to give 64×64 pixel ‘cubes’. SINFONI has three pixel scale settings: $0.25''$, $0.1''$ and $0.025''$ giving Fields of View (FOV) of $8'' \times 8''$, $3'' \times 3''$ and $0.8'' \times 0.8''$, respectively. Neptune was observed on three nights from October 9th to 12th 2013 (UT) using the H- and J-grisms, which have spectral resolutions of $R = \lambda/\Delta\lambda \sim 2000$ and 3000, respectively. The individual observations are listed in Table 1. Since the disc size of Neptune comfortably fits in the FOV for the $0.1''$ plate scale, this was the default mode of operation. However, to increase the spatial resolution, observations were also made on the second and third nights with the $0.025''$ plate scale and stepping the FOV across the planet’s disc, building up 4×4 mosaics.

The data were reduced with the ESO VLT SINFONI pipeline, but correction for (i.e. removal of) the stellar absorption features of the telluric standard star was made using the Spextool (Cushing et al. 2004) *xtellcor-general* package, which uses the method of Vacca et al. (2003). Photometric correction was achieved by integrating the observations of the standard (A0V) star (HIP110963) across the entire FOV, using the quoted 2MASS (Cutri et al. 2003) J- and H-magnitudes of 8.603 and 8.601 and the 2MASS J- and H-filter profiles respectively. Geometric registration was done by visually aligning the images against a Neptune reference wire-grid, which was then used for determining the latitude, longitude, and emission angles; planetocentric latitudes were assumed throughout. We also

94 corrected for the airmass difference between each observed planet frame and the standard
95 star reference.

96 Figure 1 shows a typical reflectance spectrum of Neptune as measured by IRTF/Spex¹,
97 together with the pressure level at which the two-way transmission to space is 0.5 for
98 cloud-free conditions, assuming the standard atmospheric profile described in the next
99 section. The main absorption features seen in Neptune’s near-IR spectrum are formed by
100 gaseous methane. At wavelengths of strong methane absorption sunlight cannot penetrate
101 very far, and thus any light we see must have been reflected from hazes in the stratosphere.
102 Conversely in regions of weak absorption sunlight can penetrate to be reflected from clouds
103 at the deepest levels. Hence, such spectra allow us to probe the cloud density over a wide
104 pressure range. In this paper, we present many false colour plots that show the distribution
105 of deep, intermediate-level and high clouds/hazes. To map the deepest clouds we only use
106 wavelengths where the two-way transmission to space exceeds 0.5 at the 3-bar level. We
107 shall call this the ‘F3.0’ filter. To map the intermediate-level clouds we choose only those
108 wavelengths where the two-way transmission to space is **less** than 0.5 at the 1.25 bar level
109 (‘F1.25’ filter), and to map the highest clouds/hazes we choose only those wavelengths
110 where the two-way transmission to space is less than 0.5 at the 0.2 bar level (‘F0.2’ filter).
111 The wavelengths covered by these three ‘filters’ in the 0.9 – 1.87 μm range are shown in
112 Fig. 1. The mosaicked H-band appearance of Neptune recorded from 00:30 – 01:25UT
113 (observation ‘OB36’) on October 12th 2013, using the 0.025’’pixel scale in these three ‘filters’
114 is shown in the top row of Fig.2, while the appearance recorded slightly earlier (00:01 –
115 00:03UT, observation ‘OB34’) at the lower 0.1’’pixel resolution is shown in Fig. 3. The
116 bottom rows of Figs.2 and 3 show scaled differences between these images, highlighting the
117 cloud density at low and medium altitudes (the ‘F0.2’ map (panel (c)) already shows the

¹http://irtfweb.ifa.hawaii.edu/spex/IRTF_Spectral_Library/

118 cloud/haze density at high altitudes), and also shows the aspect of Neptune at the time
119 of observation. Figure 4 shows false-colour representations of all the useable H-grism and
120 J-grism data recorded in 2013, with red used to indicate the deep clouds (‘F3.0’), green for
121 the intermediate-level clouds (‘F1.25’), and blue for the highest clouds (‘F0.2’). As can be
122 seen, J- and H-band observations were taken at similar times and provide complementary
123 coverage, although it is apparent that the J-band observations are more affected by upper
124 tropospheric and stratospheric hazes, making them appear more yellowish in the false-colour
125 scheme chosen. This is understandable given their shorter wavelength ($1.3 \mu\text{m}$ compared
126 with $1.6 \mu\text{m}$) and the small estimated size of such haze particles ($\sim 0.1 - 1 \mu\text{m}$), which leads
127 to their cross-sectional area diminishing rapidly with increasing wavelength. Since Neptune
128 rotates so rapidly and latitudinal wind shear distorts clouds so quickly, it is difficult to
129 compare the raw images in Fig. 4, recorded over several days, with each other. To make
130 this easier, we have plotted the highest quality H-band observations (which are less affected
131 by haze and atmospheric seeing and thus clearer than the J-grism images) on a grid in
132 Fig.5, where the x-position is determined from the central meridian longitude at the time of
133 observation, while the y-position is the digital date (i.e. 00:00 on October 10th is 10.0, 06:00
134 on October 10th is 10.25, etc.). This plot allows us to compare observations taken with
135 notionally the same ‘face’ of Neptune pointed towards Earth and also to see the temporal
136 (if any) development of clouds as they traverse across the face of the planet. The ‘face’ of
137 Neptune seen from the Earth can be understood more clearly in Fig.6, which shows how
138 Neptune’s appearance would change on the time versus central meridian longitude grid of
139 Fig.5 if it were totally cloudy on one side and cloud-free on the other for two cases: 1) where
140 the rotation period is 16.11 hrs at all latitudes; and 2) where the rotation period varies with
141 latitude resulting from the zonal wind profile of Sromovsky et al. (1993). In the second
142 case, which is the real case on Neptune, it can be seen that the differential rotation quickly
143 leads to significant distortion of the the white ‘face’. We could have tried to ameliorate the

144 effects of this using ‘snakeskin’ plots (i.e. where the image is mapped on to a rectangular
145 latitude/longitude cylindrical projection) and applying latitudinally dependent corrections,
146 but Fig.6 makes it clear that such plots would themselves become quickly distorted and
147 hard to decipher. In addition, our spatial resolution is not sufficiently good for most of
148 our observations to produce accurate cylindrical (or ‘snakeskin’) maps, especially near the
149 South Pole. Hence, we settled on the method shown in Fig.5 of displaying our unreprojected
150 observations on a time versus central meridian longitude grid, which shows our observations
151 in their least processed form and gives some indication of which ‘face’ of Neptune is being
152 observed, although the latitudinally dependent distortions highlighted by Fig.6 must be
153 borne in mind.

154 **2.1. Observations of near-equatorial intermediate-level clouds and comparison** 155 **with Gemini/NIFS (2009)**

156 It can be seen that these VLT observations provide excellent coverage of Neptune’s
157 cloud structure, especially the 4×4 mosaicked observations taken with the $0.025''$ plate
158 scale, which show excellent spatial resolution. The $0.025''$ observations on the 2^{nd} night
159 unfortunately had the frame rotated with respect to the sample grid direction, leading to
160 small gaps between the ‘tiles’, but this error was recognised and corrected for the 3^{rd} night
161 of observations. We can see considerable temporal development of the clouds. Observations
162 on the 1^{st} and 3^{rd} nights were taken with similar central-meridian longitudes, assuming a
163 16.11 -hr rotation rate (as can be seen in Fig.5). Some similarities can be seen, including the
164 presence of a bright white cloud near Neptune’s south pole, which does not seem to have
165 evolved greatly over the elapsed 2 days. At equatorial and mid-latitudes, yellowish clouds
166 are visible in the false-colour plots, which are intermediate-level clouds that are visible in
167 the red (F3.0) and green channels (F1.25), but not so high as to be visible in the blue

168 channel (F0.2). Here we can see clear latitudinal wind shear in Neptune’s atmosphere as
169 the best correspondence between the 1st and 3rd nights is between the first image recorded
170 on the 1st night and the last image recorded on the 3rd night. Since the winds are strongly
171 retrograde at equatorial latitudes, the rotation period is effectively greater and so it takes
172 longer for the same clouds to appear in the central meridian, as can be seen here. However,
173 Fig.6 shows that small errors in the differential rotation rate can lead to large errors in the
174 observed East-West position of a feature, even after just a couple of days. Assuming that
175 the intermediate-level cloud features seen on the 3rd night are indeed the same as those seen
176 on the 1st night (which they appear to be given Neptune’s assumed latitudinal wind profile),
177 however, we can conclude these intermediate-level clouds survive for at least a couple of
178 rotations of the planet. In Gemini/NIFS observations of Neptune made in 2009, Irwin et
179 al. (2011) found that such clouds were only seen near the morning terminator. However,
180 there were not enough clouds or sufficiently well time-resolved observations to determine if
181 these clouds were simply local-time induced features or whether they were longer lived and
182 survived their transit across Neptune’s visible disc. This can be seen in Fig.7, which shows
183 the 2009 Gemini observations in the same format as in Fig.5, plotted as a function of time
184 versus central meridian longitude. Here we can see that observations were well spaced over
185 central meridian longitude, with little overlap, except after the passage of 5-6 days during
186 which time wind shear and general evolution have distorted the clouds so much as to make
187 them unrecognisable. The new VLT/SINFONI observations unequivocally confirm that
188 these equatorial intermediate-level clouds are not ephemeral and last for several days.

189 **2.2. Observations of south sub-polar deep clouds and comparison with**
 190 **Gemini/NIFS (2009 and 2011)**

191 One of the most interesting features of the 2009 Gemini/NIFS observing campaign
 192 was the presence of deep clouds near Neptune’s south pole (Irwin et al. 2011). In the
 193 Gemini/NIFS observations made on 1st, 5th and 6th September 2009 (Fig.7), we can see
 194 discrete red-coloured clouds near the south pole. The clouds are coloured red because
 195 they can only be seen at continuum wavelengths where the absorption of methane is least
 196 and must thus reside at pressures > 1.25 bar, presumably at the main cloud deck level,
 197 estimated to lie at around the 2–3 bar level. Irwin et al. (2011) showed that at least one
 198 of these clouds was long-lived, but was occasionally obscured by overhead clouds lying at
 199 levels of different wind speed. The new 2013 VLT/SINFONI observations show no evidence
 200 of such deep sub-polar clouds. However, several such clouds can be seen at the southern
 201 edge of the bright cloud belt at 30–40°S (most clearly seen in rows 4-6 of Fig.4), where
 202 they were not apparent in 2009. Later Gemini/NIFS observations of Neptune, previously
 203 unpublished, were also recorded on several nights between 30th August and 11th September
 204 2011 (Table 2). During this apparition, the Adaptive Optics module was non-functional
 205 and so spatial resolution was limited to the atmospheric seeing. Hence, these observations
 206 are much less spatially resolved than the 2009, and now 2013 observations. However, they
 207 were made with the I, J and H grisms and thus have greater spectral coverage. Fig.8 shows
 208 these 2011 observations in the same format as in Figs. 5 and 7, plotted as a function of
 209 time versus central meridian longitude; the grism used is indicated by each image. In this
 210 plot we can see that the images become progressively more ‘yellow’ as we move from H-
 211 (1.47 – 1.80 μm), through J- (1.14 – 1.36 μm) to the I-grism (0.94 – 1.16 μm), indicating
 212 rapidly increasing optical depth of tropospheric/stratospheric hazes as we move to shorter
 213 wavelengths. Although of poorer spatial resolution, a discrete deep sub-polar cloud as was
 214 seen in 2009 should have been discernible in 2011, but such features are absent as they

215 were also absent in the 2013 observations reported here. The fact that no such features are
216 apparent after 2009 suggests that such clouds may be short-lived.

217 **3. Radiative Transfer and Retrieval Analysis**

218 To quantitatively analyse the new VLT/SINFONI H-band spectra, they were first
219 smoothed to a lower spectral resolution using a a triangular-shaped instrument function with
220 Full Width Half Maximum (FWHM) = $0.002\mu\text{m}$ to simulate the IRTF-SpeX instrument,
221 giving a spectral resolution of $R \sim 775$. Although this sacrificed spectral resolution, it
222 greatly increased our computation speeds and improved the signal-to-noise (SNR) ratio.
223 This choice was justified by our previous high spectral resolution analysis of Neptune
224 spectra (Irwin et al. 2014). From this analysis we concluded that, for cloud parameter
225 retrievals, the lower IRTF-SpeX resolution was the best compromise between computational
226 efficiency, vertical resolution and SNR. Smoothing the spectra further would lower the noise
227 levels (i.e. increase the SNR), but degrade the vertical resolution, while a higher spectral
228 resolution greatly increases the computation time of the radiative transfer code (which uses
229 a Matrix-Operator multiple scattering model), while not greatly increasing the vertical
230 resolution due to the lower SNR.

231 **3.1. Temperature/Abundance Profiles**

232 The temperature and abundance profile assumed in this study was the same as
233 that used by Irwin et al. (2014). The temperature profile was based on the ‘N’ profile
234 determined by radio-occultation from Voyager 2 by Lindal (1992) and the He/H₂ ratio was
235 set to 0.177, which leads to a helium volume mixing ratio of 0.15 at altitudes of negligible
236 methane abundance, assuming 0.3% nitrogen, as favoured by Conrath et al. (1993) and

237 Burgdorf et al. (2003). Note that apart from at the south pole, Fletcher et al. (2014)
 238 found very little temporal evolution of Neptune’s temperature structure from the Voyager
 239 epoch to more recent times near the southern summer solstice, so using the Lindal (1992)
 240 profile is reasonable. The methane abundance profile was set with a deep CH₄ mole fraction
 241 of 4% and the volume mixing ratio limited to a maximum relative humidity of 60%, as
 242 recommended by Karkoschka and Tomasko (2011), but the stratospheric abundance was set
 243 to $(1.5 \pm 0.2) \times 10^{-3}$ as recommended by Lellouch et al. (2010). Although Karkoschka and
 244 Tomasko (2011) find that the deep abundance of CH₄ reduces at high southern latitudes,
 245 our analysis here is limited to latitudes where the assumption of latitude-invariance is a
 246 reasonable approximation.

247 **3.2. Gaseous Absorption data and Scattering Radiative Transfer Model**

248 These data were analyzed with the WKMC-80K line database (Campargue et al.
 249 2012) in the same method as described by Irwin et al. (2014). The spectra were fitted with
 250 the NEMESIS (Irwin et al. 2008) radiative transfer and retrieval code, using a correlated-k
 251 model (Lacis and Oinas 1991) and methane k-tables derived from the WKMC-80K line
 252 data, assuming the IRTF-SpeX triangular instrument function with FWHM = 0.002
 253 μm . These k-tables were computed using the hydrogen-broadened methane line shape
 254 of Hartmann et al. (2002) (suitable for atmospheres where H₂ is the main constituent)
 255 and have a line wing cut-off of 350 cm^{-1} , which we previously found to give good fits to
 256 our Uranus and Neptune Gemini/NIFS observations. Since the atmospheric composition
 257 and temperature of Uranus and Neptune are very similar in the upper troposphere/lower
 258 stratosphere it is reasonable to expect the methane lines of Neptune to be broadened in
 259 the same way as for Uranus. For this k-table, a CH₃D/CH₄ ratio of 3.6×10^{-4} determined
 260 from Uranus by de Bergh et al. (1986) was assumed. Although Irwin et al. (2014)

261 revised this value for Neptune downwards to 3.0×10^{-4} , the effect on cloud retrievals at
 262 IRTF/SpEx resolution is not significant and so there was no need to recompute the table.
 263 For $\text{H}_2 - \text{H}_2$ and $\text{H}_2 - \text{He}$ collision-induced absorption (CIA) we used the coefficients of
 264 Borysow (1991, 1992) and Zheng and Borysow (1995) and an equilibrium ortho/para- H_2
 265 ratio was assumed at all altitudes and latitudes, consistent with the latitudinal mean of
 266 results from the Voyager IRIS experiment at the tropopause or higher pressures (Conrath
 267 et al. 1998), although the effect of the para- H_2 fraction on the spectra in this wavelength
 268 band is insignificant. In addition to $\text{H}_2 - \text{H}_2$ and $\text{H}_2 - \text{He}$ CIA, $\text{H}_2 - \text{CH}_4$ and $\text{CH}_4 -$
 269 CH_4 collision-induced absorption was also included (Borysow and Frommhold 1986, 1987).
 270 The spectra were simulated using a Matrix Operator multiple scattering code, based on
 271 the method of Plass et al. (1973), including the Rayleigh scattering by the air molecules
 272 themselves, with 5 zenith angles (with Gauss-Lobatto calculated ordinates and weights)
 273 and N Fourier components to cover the azimuth variation, where N is set adaptively from
 274 the viewing zenith angle, θ , as $N = \text{int}(\theta/3)$. To perform this calculation the reference
 275 temperature, pressure and abundance profiles were split into 39 levels equally spaced in
 276 log pressure between 6.5 bar and 0.001 bar. The reference solar spectrum of Fiorenza and
 277 Formisano (2005) was used to simulate the solar flux.

278

3.3. Cloud Models

279 When modelling the 2009 Gemini/NIFS H-band observations, Irwin et al. (2011)
 280 favoured a simple two-cloud model, with a cloud in the 2–3 bar region, which we shall
 281 henceforth call the ‘Tropospheric Cloud’, and a second cloud near the tropopause at 0.1
 282 bar. At most locations the opacity of this second cloud was found to be very low and its
 283 low pressure suggests it is some form of haze. However, in Neptune’s mid-latitude cloudy
 284 zones at 30–40°N,S, the opacity of this ‘Haze’ becomes so large and spatially structured

285 that it more closely resembles a second, low-pressure condensation cloud, for which the
 286 existing haze particles act as condensation nuclei. However, for ease of identification we
 287 shall henceforth call this layer the ‘Haze’. Using the then best-available methane absorption
 288 data of Karkoschka and Tomasko (2010), Irwin et al. (2011) achieved a reasonably close
 289 fit by setting the extinction cross-section spectra of the particles in both layers to be as that
 290 calculated with Mie theory assuming a complex refractive index of $1.4 + 0i$ (with a standard
 291 Gamma distribution of sizes with mean radius $1.0 \mu\text{m}$ and variance 0.05), but adjusting
 292 the single scattering albedo manually, favouring, from limb-darkening considerations, a
 293 value of 0.75 for the lower, main cloud and values between 0.4 and 1 for the haze (varying
 294 between dark and bright, cloudy regions). Both particles were assumed to have a simple
 295 Henyey-Greenstein phase function, with asymmetry parameter $g = 0.6 - 0.7$. The analysis,
 296 and quality of fit to these Gemini/NIFS data, was greatly improved by Irwin et al. (2014)
 297 who made use of the newer WKMC-80K line database (Campargue et al. 2012) and who
 298 also applied an empirically derived single-scattering albedo spectrum for the Tropospheric
 299 Cloud, with the single-scattering albedo reducing with wavelength from 0.8 to 0.6 across the
 300 measured spectral range. With the new WKMC-80K generated k-tables and the modified
 301 scattering properties, Irwin et al. (2014) found that the simple 2-thin-cloud layer model
 302 still provided a very good fit to the observed spectra they analysed, even when compared to
 303 a model where a continuous vertical distribution of cloud particles was assumed, although
 304 the requirement for the Haze layer to be thin was found not to be strong.

305 Most recently, Irwin et al. (2015), analysing IRTF/SpeX observations of Neptune’s
 306 sister planet, Uranus, have developed a novel retrieval technique whereby, in addition to
 307 the cloud opacity and vertical position, the imaginary refractive index spectrum of a cloud
 308 is retrieved. This can then be used in a Kramers-Kronig analysis to estimate the real
 309 component of the refractive index and from this complex refractive index spectrum can
 310 be computed self-consistent extinction cross-section, single-scattering albedo and phase

311 function spectra using Mie scattering. To analyse these VLT Neptune spectra we adopted
 312 the same approach, assuming *a priori* particle sizes of $1.0 \mu\text{m}$ (with variance 0.05) for
 313 both the tropospheric cloud and haze, and *a priori* refractive indices of $1.4 + 0.001i$ at
 314 all wavelengths. As described by Irwin et al. (2015) the condensates in Uranus’ (and
 315 Neptune’s) atmospheres are very unlikely to be liquid and thus spherical, as is assumed for
 316 Mie theory. However, Mie theory provides a reasonable first approximation to the scattering
 317 characteristics of an array of randomly orientated non-spherical particles, provided that
 318 features such as the ‘rainbow’ and ‘glory’, which can only be produced by spherical
 319 particles, are removed from the phase function spectra. This was done by fitting double
 320 Henyey-Greenstein phase functions to the Mie-calculated phase functions, where the phase
 321 function is represented by the asymmetry factors of the forward and backward scattering
 322 peaks, g_1 and g_2 and the fraction of forward scattering, f .

323

3.4. Cloud Retrievals

324 To determine the effectiveness of this new retrieval technique for Neptune, in a case
 325 where deep discrete clouds are visible, we chose to analyse the ‘OB34’ H-band cube (Table
 326 1), recorded on October 12th 2013, which has a clearly visible deep cloud, just south of the
 327 main southern cloud belt (Fig. 4). Data from the pixels in a line passing through this cloud
 328 were extracted (Fig.9) and used as input to the retrieval model.

329 Following our previous Neptune modelling work, we initially attempted to fit the
 330 observations with a simple two-cloud model, with variable imaginary refractive index
 331 spectra for both layers. The *a priori* Tropospheric Cloud and Haze were based at 2 bar
 332 and 0.08 bar respectively and both had a fixed fractional scale height of 0.1. Although
 333 this model fitted most observed spectra very well, we were unable to fit the data to within
 334 the predicted random error of the VLT reductions and so additional noise was added to

335 account for ‘forward-modelling error’ to bring the final $\chi^2/n \sim 1$. Such forward-modelling
 336 error may arise from uncertainties in the spectral absorption data, and the various other
 337 assumptions that go into constructing a radiative-transfer model. Figure 10 shows the fit
 338 we can achieve with this model at near-equatorial latitudes (in this case 24.1°S), away
 339 from the main cloud belts, where the Haze opacity is low. As can be seen the fit is at
 340 most wavelengths extremely good. For reference Fig.10 also shows the spectrum calculated
 341 with: 1) the Haze removed; 2) the Tropospheric Cloud removed; and 3) when both Haze
 342 and Tropospheric Cloud are removed, leaving only the reflectivity resulting from Rayleigh
 343 scattering from the air itself. As expected, it can be seen that reflection from the Haze is
 344 mostly responsible for the modelled reflectivity at methane-absorbing wavelengths, while
 345 the reflection from the deeper Tropospheric Cloud (TC) is important in regions where
 346 methane is more transparent. The retrieved cloud/haze opacity profiles and imaginary
 347 refractive index spectra of the cloud and haze particles are shown in Fig.11. The imaginary
 348 refractive index spectrum of the TC particles is almost identical to that derived for Uranus’
 349 tropospheric clouds by Irwin et al. (2015) and is reasonably well constrained (i.e. the
 350 retrieved errors are significantly smaller than the *a priori* errors). However, the Haze
 351 imaginary refractive index spectrum has barely moved from its *a priori* and the retrieved
 352 errors are not significantly smaller than the *a priori* errors, indicating that the spectral
 353 properties of the Haze are not well determined. Hence, the Haze refractive index spectrum
 354 was fixed to $1.4 + 0.001i$ at all wavelengths in subsequent retrievals, unless stated otherwise.

355 While the fit at 24.1°S is very good, at other latitudes the fit with a two-cloud model
 356 is significantly worse. This can be seen in Fig.12, which shows the fit at 38.5°S , where
 357 χ^2/n is greatest. Clearly there is something missing in our assumed two-cloud model at
 358 this location. Looking at the differences between the measured and modelled spectra, the
 359 difference seemed to be caused by missing reflection from a level between 2 – 3 bar and 1
 360 bar. We surmised that this might be due to a missing methane cloud. Using the assumed

361 temperature/abundance profile, we determined that such a cloud must be based at 1.44 bar
 362 and we added a thin cloud, based at this pressure level, assumed to be composed of methane
 363 ice particles. To model the reflectivity of these particles we used the complex refractive
 364 indices of Martonchik et al. (1994) and assumed a standard Gamma size distribution with
 365 mean radius $r = 1.2\mu\text{m}$ and variance = 0.1. This size distribution was the same as that
 366 chosen to model the Upper Tropospheric Cloud of Uranus by Sromovsky et al. (2011),
 367 which was also used by Irwin et al. (2015) and Irwin et al. (2016) to model Uranus’
 368 methane cloud. We assume here that the methane clouds of Uranus and Neptune have
 369 similar size distributions. Figs.10 and Figs.12 show the result of adding this extra cloud to
 370 our best and worst fitting cases. As can be seen the improvement in our best test case at
 371 24.1°S (Fig.10) is minimal (in fact it is very slightly worse), but the improvement at 38.5°S
 372 is very significant and clearly indicates that in the region where deep discrete clouds are
 373 visible, additional opacity is required that would appear to be consistent with the presence
 374 of a methane ice cloud.

375 Having considered two test cases, we then applied our retrieval model to all the pixels
 376 in the line passing through the deep cloud feature. Figure 13 shows the variation in
 377 the retrieved cloud/haze opacities and base pressures as a function of latitude along the
 378 sampled line of observation ‘OB34’ (Fig.9), plotted as a function of latitude using either
 379 the two-cloud or three-cloud models, depending on which fits better. Only retrievals at
 380 latitudes between the northernmost latitude observable (40°N) and 55°S have been plotted;
 381 accurate assignment of viewing geometries is difficult for pixels south of 55°S at this spatial
 382 resolution. The χ^2/n of the fit of both models is shown in the bottom right panel of Fig.13.
 383 In these retrievals the *a priori* tropospheric cloud particles’ complex refractive index was
 384 set to $1.4 + 0.001i$ at all wavelengths. The haze particles’ complex refractive indices were
 385 also set to $1.4 + 0.001i$ at all wavelengths, but fixed since Fig.11 showed we have very little
 386 sensitivity to spectral properties of these particles, assuming that they are highly scattering.

387 We can see that the two-cloud model gives a better fit at most latitudes, and the addition
 388 of methane clouds is only necessary at certain locations, here at the southern edge of the
 389 main southern mid-latitude cloud belt as we saw in Fig.12. At most other locations, adding
 390 a methane cloud actually worsens χ^2/n , suggesting that cross-correlation errors within the
 391 scheme, probably arising from the fact that the methane cloud becomes indistinguishable
 392 from the main cloud when its opacity is low, prevents the model from reaching as good
 393 a solution as if the methane cloud were omitted altogether. Apart from at 38.5°S, where
 394 the fit is significantly improved by adding a methane cloud, the only other locations where
 395 adding a methane cloud improves the fit is near 5°S and 15°S. However, at these locations
 396 the improvement in χ^2/n is actually very small and the presence of a methane cloud can be
 397 discounted. For reference, the errors on the retrieved opacities in Fig. 13 are of the order
 398 of 5% and 2% for the cloud and haze, respectively, at all latitudes, while the error on the
 399 retrieved opacity of the methane cloud, where it is detected, is approximately 5%. For the
 400 pressure levels shown in the panel B) of Fig.13, the base pressure of the Tropospheric Cloud
 401 at ~ 2 bars is well constrained to within ~ 0.1 bar, while the pressure of the methane
 402 cloud is fixed at 1.44 bar. The error on the Haze pressure is more problematic since with
 403 the *a priori* base pressure at ~ 0.1 bar the transmission to space is effectively unity at all
 404 wavelengths and thus there is very little sensitivity to the precise pressure level (Fig.1).
 405 Hence, the retrieval does not stray far from the *a priori* except towards the northern edge
 406 of the line, where the longer path lengths corresponding to the higher zenith angles leads
 407 to some discrimination, and we find that the Haze base pressure needs to decrease. It is
 408 possible that the Haze lies at pressures less than ~ 0.1 bar at all latitudes, but without
 409 observations at wavelengths where methane is more absorbing (for instance in the K-band)
 410 this cannot be determined. Finally, it should be noted that refractive index spectra very
 411 similar to those shown in Fig.11 were retrieved for the Tropospheric Cloud particles at all
 412 locations and in all subsequent cases reported in this paper.

3.5. Limb-darkening Considerations

413
 414 Figure 13 shows a steady decrease of the opacity of the main Tropospheric Cloud deck
 415 from the cloudy southern zone (at 30 – 40°S) towards the northern edge of the sampled line.
 416 We wondered whether this effect was physical or perhaps a geometrical effect of looking at
 417 increasingly high zenith angle. We thus revisited the required scattering properties of the
 418 Haze. From the same ‘cube’ we extracted all observations between 5 – 15°S, plotted them as
 419 a function of zenith angle and extracted the general limb-darkening/limb-brightening curves
 420 at all wavelengths. We found these curves to be essentially identical to those determined
 421 from our previous study of Gemini/NIFS near-equator observations made in 2009 (Irwin
 422 et al. 2011), in which we showed that the Haze must be considerably non-scattering
 423 to avoid limb brightening at all wavelengths. In this previous Gemini/NIFS study we
 424 compared observed and modelled limb-darkening curves at just a few wavelengths and a
 425 limited set of assumed Haze scattering properties. We found that Haze particles having a
 426 single-scattering albedo, $\varpi \sim 0.4$ and Henyey-Greenstein phase function with asymmetry
 427 parameter, g , in the range 0.6 to 0.7 were most consistent with observations. With our new
 428 self-consistent cloud retrieval scheme we reanalysed the limb-darkening at 20 wavelengths
 429 spread evenly across the 1.47 to 1.71 μm range. At each wavelength, the limb-darkening
 430 observations were averaged and fitted with smooth reflectance versus emission angle curves,
 431 and sampled at four zenith angles between 0 and 65° (Fig.14), corresponding to the first
 432 four zenith angles of our five-zenith angle quadrature scheme. Since we had previously
 433 (Irwin et al. 2011) found that we need the Haze particles to be quite dark at equatorial
 434 latitudes, we revised the Haze *a priori* refractive indices from $1.4 + 0.001i$ to $1.4 + 0.3i$
 435 (at all wavelengths) to lower the *a priori* single-scattering albedo to the low values found
 436 by Irwin et al. (2011), and fitted the opacity of the Tropospheric Cloud (TC) and Haze,
 437 the imaginary refractive index spectra of both the TC and Haze, and the TC and Haze
 438 particle sizes. For this limb-darkening analysis, starting with a less scattering Haze *a priori*

439 we found that we were sensitive to the refractive indices of both Haze and Tropospheric
440 Cloud and we retrieved good limb-darkening curves at all wavelengths as can be seen
441 in Fig.14. The particle scattering properties deriving from the retrieved refractive index
442 spectra and particle sizes can be seen in Fig.15. As we can see the Tropospheric Cloud is
443 found to be almost entirely forward scattering, with scattering asymmetry $g_1 \sim 0.7$, and
444 has a single scattering albedo varying between 1 and 0.5 across the range. For the Haze
445 we find that the particles are considerably darker than determined by Irwin et al. (2011),
446 but the phase function is also substantially different, approaching Rayleigh-scattering.
447 However, we believe this solution to be more reliable than our previous conclusions since
448 this combination of properties was derived in a self-consistent manner instead of being
449 chosen from a limited set of self-inconsistent properties in our previous work (Irwin et al.
450 2011), where this combination was never explored. Fig.16 shows the retrieved cloud/haze
451 opacity profiles and retrieved imaginary refractive indices from this analysis at $5 - 15^\circ\text{S}$.
452 In this case we can see that the imaginary refractive index spectra of both the TC and
453 Haze are well constrained at the wavelengths sensitive to these particles since the retrieved
454 errors are significantly smaller than *a priori*. The position of the cloud/haze decks is
455 almost indistinguishable from our retrievals with a highly scattering Haze *a priori* as is
456 the retrieved refractive index spectrum of the Tropospheric Cloud. To determine whether
457 we would find substantially different latitudinal variations using a low-scattering Haze *a*
458 *priori*, we repeated our retrievals of the N/S strip through the deep cloud seen in ‘OB34’,
459 but instead set the *a priori* Haze refractive indices to be $1.4 + 0.3i$ at all wavelengths and
460 retrieved the cloud opacities for the Tropospheric Cloud, methane cloud (for the three-cloud
461 model only) and Haze, and the refractive index spectra of both the TC and Haze for the
462 two-cloud and three-cloud models. We found these models (Fig.17) had similar fitting
463 accuracies at most locations to the retrievals shown in Fig.13 for the highly scattering *a*
464 *priori* Haze, but that a poor fit was achieved in the cloudy zones at $30-40^\circ\text{N}$ and $30-40^\circ\text{S}$

465 for the two-cloud model. Note that in Fig.17 we again plot the fitted parameters of model
466 that has the lowest χ^2/n at each latitude. The three-cloud model was able to significantly
467 improve the fit at at 30–40° N, but had a similarly poor fit at 30–40° S. For both two-cloud
468 and three-cloud models we found that we needed a far greater opacity of Haze to achieve
469 the same levels of reflectivity from the upper troposphere/lower stratosphere, as might be
470 expected, but that the increased absorption of this Haze worsened the model’s ability to
471 fit the reflected spectrum from the lower clouds. Although the retrieval model lowered
472 the imaginary refractive indices of the Haze (as is indicated in Fig.17), and thus increased
473 the single-scattering albedos of these particles over the mid-latitude cloudy zones, the
474 relative weights in our retrieval set-up (and absence of limb-scattering constraints at higher
475 latitudes) meant that the Haze particles did not become scattering enough to avoid having
476 extremely large opacities at mid-latitudes. The marked increase of haze single-scattering
477 albedo in the cloudy zones was also a conclusion of our previous Gemini/NIFS study
478 (Irwin et al. 2011). Comparing the latitudinal variation in the retrieved opacity of the
479 Tropospheric Cloud with that obtained for a highly scattering Haze *a priori* (Fig.13), we
480 found a very similar decrease of opacity running north from the southern cloudy zone at
481 30 – 40° until 20°N, but a divergence from the highly scattering *a priori* Haze case at
482 higher latitudes. At these higher latitudes, (and high zenith angles) the increased opacity
483 of the overlying less-scattering Haze made it necessary to greatly increase the TC opacity
484 in order to match the observed spectra. The results suggest that a reliable estimate of
485 how the opacity of the Tropospheric Cloud varies with latitude depends greatly on the
486 assumed and/or modelled scattering characteristics of the overlying Haze. Alternatively,
487 if we assume the Tropospheric Cloud has a similar opacity at all latitudes, this could in
488 future be used to provide a constraint on the single-scattering albedo of the overlying Haze.

3.6. Intermediate-Level Equatorial Clouds

489 To examine the cloud structure of regions with ‘intermediate-level’ clouds (the yellowish
 490 regions in the false colour plots, where the cloud is visible in the F3.0 and F1.25 filters, but
 491 not in the F0.2 filter, indicating these clouds to lie at pressures between 1.25 and 0.2 bar),
 492 the observation ‘OB37’ (Table 1) was selected and a north/south strip selected through
 493 the centre of such a cloud (Fig.9). We ran NEMESIS on the selected spectra using the
 494 two-cloud and three-cloud models with fixed Haze refractive indices of $1.4 + 0.001i$; the
 495 results are shown in Fig.18. Here we see that our fitting accuracy, except in the centre
 496 of the intermediate-level level cloud at 10.7°N , is similar to the ‘OB34’ retrievals. The
 497 addition of a methane cloud only improves the fit near 15°S , but only insignificantly and in
 498 most cases significantly worsens the fit. In the centre of the ‘intermediate-level’ feature, the
 499 two-cloud model is clearly preferred. Here the opacity of the Tropospheric Cloud is seen
 500 to decrease (relative to the overall decrease from south to north) while the Haze opacity
 501 (which here accounts for the reflection from the ‘intermediate cloud’) increases and its base
 502 pressure increases from ~ 80 mb to 400-500mb. This worst fitting spectrum at 10.7°N is
 503 shown in Fig.19 and we can see that there is a systematic difference between the modelled
 504 and measured spectra. We performed a number of retrieval tests, for example, adding a
 505 CH_4 cloud and allowing both it and the Haze to be vertically extended. While allowing
 506 a CH_4 cloud, based at 1.44 bar, to be extended produced no noticeable improvement in
 507 the fit, allowing the Haze to be extended (giving it an *a priori* fractional scale height of
 508 0.5 ± 0.1 with a higher *a priori* base pressure of 0.25 bar (compared with 0.08 bar before)
 509 produced a noticeably improved fit, which is also shown in Fig. 19. The results for the
 510 two-cloud and three-cloud models with the vertically extended Haze applied to all latitudes
 511 in the ‘OB37’ north/south strip are shown in Fig.20. For the two-cloud model, extending
 512 the Haze leads to a significantly improved fit near the ‘intermediate-level’ feature at 10.7°N ,
 513 but the improvement at other latitudes is marginal. However, the ‘intermediate-level’

515 feature at 10.7°N is the only one where the Haze base pressure is required to be increased to
 516 pressures ($\sim 300 - 400$ mbar, similar to the case where the Haze is assumed to be vertically
 517 thin) where the observations are actually sensitive to the fractional scale height of the Haze
 518 (Fig.1). The three-cloud model is again generally found to be less successful, especially near
 519 the south pole, where considerable cross-correlation between different cloud parameters
 520 made the retrieval unstable, leading to the solution not varying far from *a priori* resulting
 521 in high χ^2/n values, as can be seen. Adding a methane cloud only marginally improves the
 522 fit near 15°S and 25°N as can also be seen.

523 4. Discussion

524 The clouds of Neptune can be seen from our observations to be comprised of four main
 525 types: 1) the main deep Tropospheric Cloud (TC) at 2–3 bars composed, probably, of H₂S
 526 (e.g. de Pater et al. (2014)), 2) the high altitude, highly reflective, high opacity clouds
 527 seen in the mid-latitude bands at 30 – 40° N,S; 3) small, bright ‘deep’ clouds seen near
 528 the south pole in 2009 and along the southern edge of the main 30 – 40°S in 2013; and 4)
 529 ‘intermediate-level’, vertically extended clouds, with base pressures of $\sim 300 - 400$ mb at
 530 low latitudes. These four cloud types are clearly distinguishable in our false colour plots
 531 and also in our retrieved vertical cloud profiles.

532 The distribution and appearance of the high altitude, mid-latitude clouds at 30 – 40°
 533 N,S in 2013 (cloud type 2) seems very similar to that observed in 2009, but the distribution
 534 of the small, bright ‘deep’ clouds (cloud type 3) is completely different. In 2013 no such
 535 clouds were seen near the south pole, but instead such clouds appeared at the southern
 536 edge of the 30 – 40°S region. Irwin et al. (2011) postulated that the deep sub-polar clouds
 537 seen in 2009 might be linked to the offset sub-polar hotspots observed near 70°S at mid-IR
 538 wavelengths (8.6 μ m) by VLT/VISIR in September 2006 (Orton et al. 2007, 2012). These

539 hotspots, observed via stratospheric CH₄ emission, were ephemeral in nature between 2003
540 and 2010. This is in contrast to the general warming trend towards Neptune’s summer pole
541 that has been observed consistently in ground-based mid-IR datasets since 2003 (Fletcher
542 et al. 2014), and which could be explained by subsidence and adiabatic-warming of the air
543 within a summer stratospheric vortex. Orton et al. (2012) suggested that a high-latitude
544 wave, excited by powerful dynamics at deeper tropospheric levels (e.g., convective activity),
545 could be interacting with and perturbing the polar stratospheric vortex. Generation
546 of warm stratospheric airmasses by vertically-propagating waves was also a suggested
547 mechanism for the formation of Saturn’s stratospheric anticyclone following its 2010-2011
548 tropospheric storm (Fletcher et al. 2012), hinting at a coupling between the sporadic
549 sub-polar clouds observed in the troposphere and the offset polar hotspots observed in
550 the stratosphere. However, simultaneous observations in the near- and mid-IR were only
551 attempted once for Neptune, at southern summer solstice in 2005 (Hammel et al. 2007),
552 and did not reveal a direct correlation between the two (although no sub-polar clouds
553 were visible in the near-IR at the time). A simultaneous campaign of near-IR and mid-IR
554 imaging at comparable spatial resolutions will be required to confirm this coupling between
555 tropospheric and stratospheric activity. Whether such clouds are linked to mid-IR features
556 or not, the more precise radiative transfer modelling enabled by improved methane line
557 data and retrieval methods reveals that such discrete deep clouds (i.e. cloud type 3) are
558 possibly methane ice condensation clouds, formed presumably in regions of rapid upwelling.
559 The global meridional circulation of Neptune can be inferred through observations of upper
560 tropospheric temperature (de Pater et al. 2014; Fletcher et al. 2014) and is believed to
561 be rising at mid-latitudes and sinking at the equator and poles. The appearance of deep
562 convection methane clouds at the edges of the main cloudy zones at 30 – 40° N,S is then
563 perhaps only to be expected. How such clouds might appear near the south pole as they did
564 in 2009, however, remains a mystery, as is their apparent absence along the southern edge of

565 the main 30 – 40°S cloud belt at that time. However, we have found in this study that the
566 spectra of such ‘deep’ clouds is well-modelled by the addition of a methane condensation
567 cloud at 1.44 bar (the expected condensation level for the assumed temperature-pressure
568 profile). It should be noted, though, that the addition of such a methane condensation cloud
569 does not generally improve the fit to Neptunian near-IR spectra and in many locations
570 significantly worsens it.

571 For the high-altitude clouds themselves (i.e. cloud type 2), these appear to be based
572 at around the 100 – 200 mb level and vary in single-scattering albedo with latitude. We
573 find that they have higher albedo in the main cloud belts, but very low albedo and low
574 opacity elsewhere, as previously determined by Irwin et al. (2011). We suggest that
575 Neptune is generally covered by a dark ‘sooty’ haze layer at these pressure levels, which
576 only become highly scattering and optically thick in regions of upwelling, where they are
577 coated with freshly condensed material - again, presumably methane ice. As mentioned
578 earlier, if we assume the Tropospheric Cloud has a similar opacity at all latitudes, then this
579 assumption could be used to better constrain the single-scattering albedo of the overlying
580 Haze. However, we shall leave such an analysis to a future study.

581 The ‘intermediate-level’ equatorial clouds (cloud type 4) were only seen by Gemini/NIFS
582 (Irwin et al. 2011) near the morning terminator, and it was thus possible from these
583 observations that these might be ephemeral features linked in position to the diurnal cycle.
584 Our new observations show that these ‘intermediate-level’ clouds are uniformly distributed
585 with local time at equatorial latitudes. In addition, because our observations were made
586 near the start and end of Neptune’s transit on three consecutive nights we can see that
587 these features, and indeed all the others seen, do not evolve significantly during a single
588 night. Indeed, the ‘intermediate-level’ clouds seem to last for several days, which since they
589 reside at latitudes of relatively low latitudinal wind shear is perhaps not surprising. In

590 terms of the overall meridional circulation it is curious that the ‘intermediate-level’ clouds
591 at 300 – 400mb should appear at near-equatorial latitudes – a region generally thought to
592 be one of subsiding air in the upper troposphere (de Pater et al. 2014). The situation
593 is analogous to the appearance of convective plumes in Jupiter’s North Equatorial Belt,
594 an area similarly thought to be a region of generally subsiding air. It may be that in
595 both ‘belt’ locations, conditional instabilities mean that small convective events can occur
596 amongst otherwise descending air and, in the case of Neptune, lead to condensation that
597 perhaps becomes vertically extended. The most obvious candidate for such condensation
598 is, again, methane and it is thus puzzling that no ‘deep’ methane clouds have been seen
599 at the equator, but instead only ‘intermediate-level’ level ones. Alternatively, it may be
600 that the ‘intermediate-level’ clouds are caused by material descending and freezing out
601 through the tropopause cold-trap as part of the overall upper tropospheric meridional
602 circulation scheme indicated from mid-IR observations with air rising at mid-latitudes and
603 sinking at the poles and equator (de Pater et al. 2014). This descending branch at the
604 equator presumably weakens at pressures greater than 1 bar since Karkoschka and Tomasko
605 (2011) find that methane is enriched at all latitudes equatorwards of $\sim 45^\circ\text{N,S}$, indicating
606 upwelling, and only decreases at more polar latitudes, possibly indicating subsidence, or
607 decrease in convective overturning. This picture is also mirrored in radio images of Neptune
608 (de Pater et al. 2014), which shows increased emission at the south pole, indicating dryer
609 air at pressures greater than 10 bar, but do not show increased emission at the equator.

610 5. Conclusions

611 We have compared Integral Field Unit Spectrometer observations of Neptune made in
612 2013 with VLT/SINFONI, with Gemini/NIFS observations made in 2009 and 2011. We
613 have shown that the small, deep, discrete clouds seen near Neptune’s south pole in 2009

614 by Gemini/NIFS were absent in 2011 and 2013, but similar deep clouds appeared at the
 615 southern edge of the southern mid-latitude cloudy zone at 30–40°S region in 2013, which
 616 were not apparent in 2009 and 2011. Our observations, taken at the beginning and end
 617 of three consecutive nights show that the cloud features are not significantly deformed by
 618 latitudinal windshear during a single night. In particular, the ‘intermediate-level’ level
 619 clouds observed by Gemini/NIFS in 2009 (Irwin et al. 2011) do not appear to be limited in
 620 their distribution to be near the morning terminator, a possibility that could not be ruled
 621 out by the Gemini/NIFS observations, but instead can be seen to survive several transits
 622 across Neptune’s disc.

623 We have analysed our new VLT/SINFONI H-band observations using a self-consistent
 624 cloud-retrieval model, previously applied to Uranus IRTF/SpeX observations by Irwin et
 625 al. (2015). This improvement in our retrieval technique, coupled with the use of greatly
 626 improved methane absorption data from Campargue et al. (2012) means that we can fit
 627 the observations to much higher precision and at greater spectral resolution, allowing us to
 628 extract more precise information from these data than was possible in our previous study of
 629 Gemini/NIFS H-band observations made in 2009 (Irwin et al. 2011). We find that a simple
 630 two-cloud model (a ‘Tropospheric Cloud’ near the 2–3 bar level and a ‘Haze’ based near 0.1
 631 bar) recommended by Irwin et al. (2011) is sufficient to model the bulk of spectra across
 632 Neptune’s disc at these wavelengths. The opacity of the Tropospheric Cloud is seen to vary
 633 slowly with latitude, while the ‘Haze’ optical depth varies greatly from being optically thin
 634 (and poorly scattering with low single scattering albedo) at most latitudes, to becoming
 635 optically thick and highly scattering in the bright mid-latitude belts at 30–40°N,S. At these
 636 locations we suggest that cloud (methane ice) is condensing on the background dark haze
 637 particles. However, we find that the discrete, bright, ‘deep’ clouds seen at the southern
 638 edge of the southern mid-latitude cloudy zone at 30–40°S are much better modelled by
 639 adding a methane cloud layer, based at the condensation level of 1.44 bar expected from

640 the assumed temperature-pressure-abundance profile. Hence, these features appear to be
641 localised methane clouds, caused by rapid convection and condensation of material in the
642 1–1.5 bar region. For the ‘intermediate-level’ level clouds seen at more equatorial latitudes,
643 we have shown that these clouds can again be modelled (as done in our previous analysis)
644 with the two-cloud model, by lowering the base of the ‘Haze’ layer to the 300 – 400mb
645 pressure level, but that we are unable to achieve as good a fit unless we allow the Haze
646 layer to become vertically extended.

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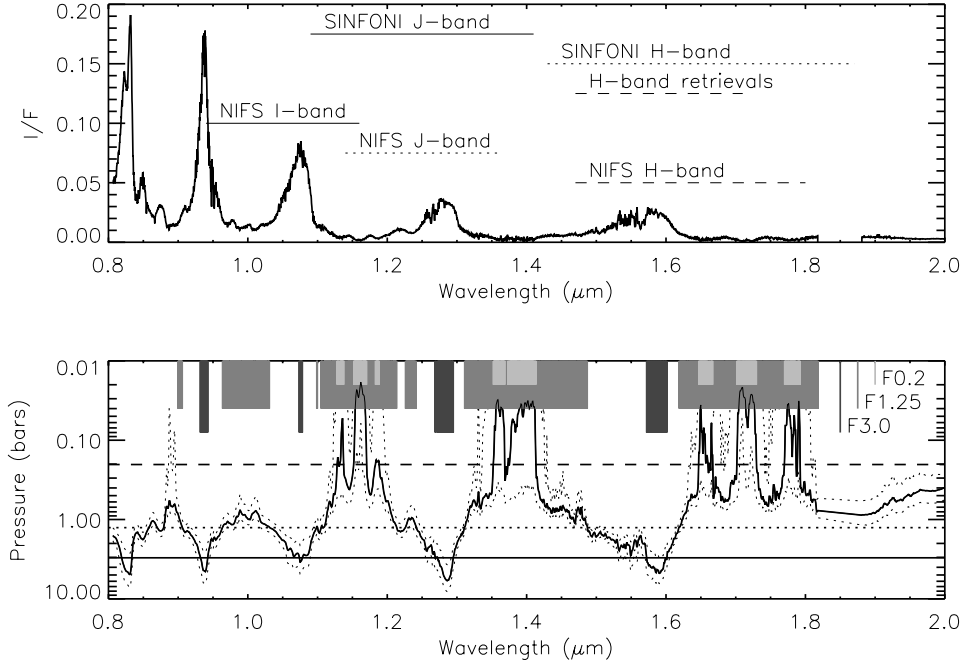


Fig. 1.— Top panel shows a typical I/F spectrum of Neptune as observed by IRTF/SpeX, together with the wavelengths spanned by selected VLT/SINFONI and Gemini/NIFS grisms. Bottom panel shows the pressure level in Neptune’s atmosphere at which two-way transmission to space is 0.5 for a cloud-free atmosphere. Overplotted in the bottom panel are the pressure levels (dotted lines) for which the two-way transmission to space is 0.25 and 0.75, giving an indication of the vertical resolution of the observations at a single wavelength. Also overplotted in the bottom panel are the chosen cut-off pressures of 3, 1.25 and 0.2 bar. Continuum images (‘F3.0’) are averaged over all wavelengths where the transmission to 3 bars exceeds 0.5. Medium-absorption and high-absorption images are averaged over all wavelengths where the transmission at 1.25 and 0.2 bars is respectively less than 0.5, labelled respectively as ‘F1.25’ and ‘F0.2’. The wavelengths selected by these filters in the wavelength range (0.9 – 1.87 μm) are indicated by the grey regions in the bottom panel of differing length and greyness, indicated by the vertical bars in the top right of the bottom panel.

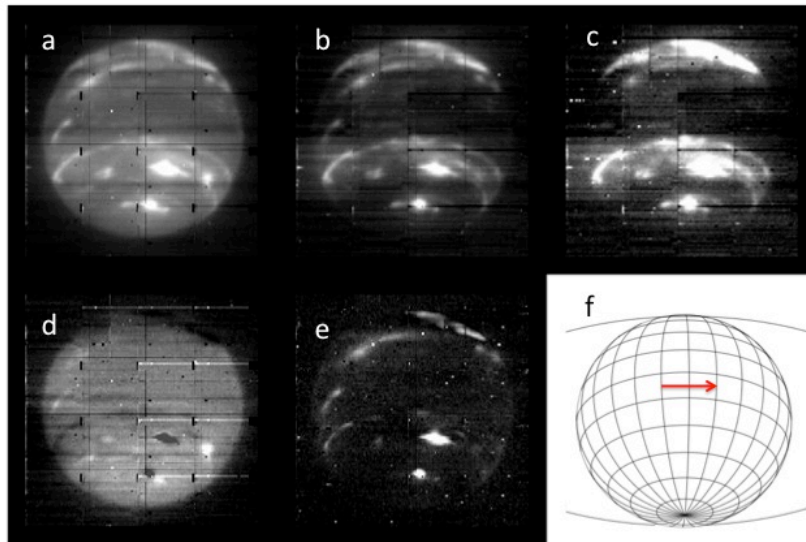


Fig. 2.— One of the best H-band observations of 12th October 2013 (OB36), formed of a 4×4 mosaic of $0.025''$ plate scale cubes. The top row shows the appearance of Neptune in the different wavelength ‘filters’. Panel (a) shows the planet at wavelengths where the two-way transmission to space, for a cloud-free atmosphere, exceeds 0.5 at the 3-bar level (i.e. the ‘F3.0’ filter). Panel (b) shows the planet at wavelengths where the two-way transmission to the 1.25 bar level is less than 0.5 (‘F1.25’), while panel (c) shows the planet at wavelengths where the two-way transmission to the 0.2-bar level is less than 0.5 (‘F0.2’), which is only sensitive to the hazes at pressures less than 0.2 bar. The bottom row shows differences between the images to highlight the clouds at different levels. Panel (d) shows the F3.0 image (Panel (a)) minus the F1.25 image (Panel (b)) and shows the distribution of cloud reflectivity at the main cloud deck at $\sim 1 - 3$ bars, while panel (e) shows the F1.25 image (Panel(b)) minus the F0.2 image (Panel (c)), showing the distribution of clouds between roughly 1.25 and 0.2 bar. Panel (f) shows a projection of Neptune’s disc and ring for reference. The dark patches in Panel (d) do not indicate holes in the deep cloud, only that the scaling chosen to eliminate the reflectivity of overlying clouds leads to slightly too much reflectivity being subtracted in cases where the overlying clouds are very thick. Neptune’s sense of rotation is indicated by the arrow in panel (f).

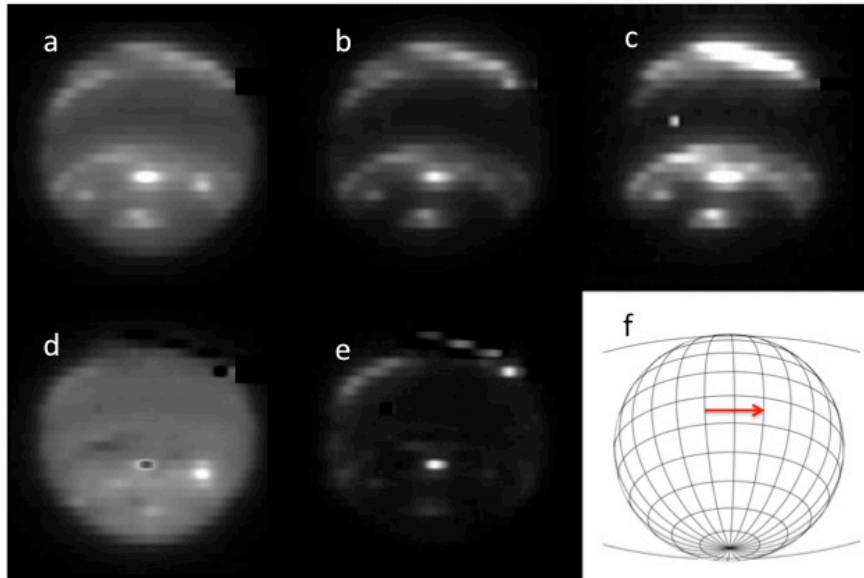


Fig. 3.— Lower-resolution H-band observation ($0.1''$ plate scale) made on 12^{th} October 2013 (OB34), recorded shortly before OB36 (Fig.2). The layout of the figure is identical to Fig.2.

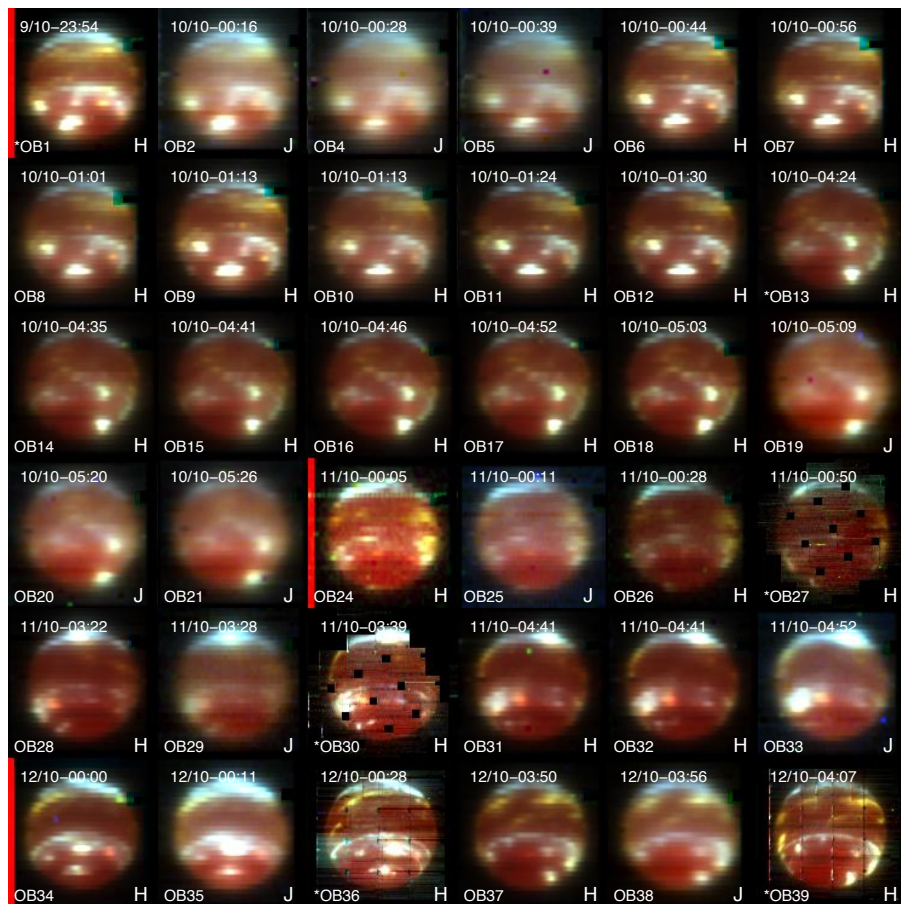


Fig. 4.— Summary of October 2013 VLT/SINFONI H- and J-band Neptune observations, gridded with time running from top left to bottom right. The observation date/time and grism are indicated by each image. The starting image from each night is indicated by the red bar on the left hand edge side of the image. Images are plotted in false colour, where red is the continuum F3.0 image (i.e. panel (a) in Figs. 2, 3), green is the F1.25 image where $\text{Trans}_{1.25\text{bar}} < 0.5$ (i.e. panel (b) in Figs. 2, 3) and blue is the F0.2 image where $\text{Trans}_{0.2\text{bar}} < 0.5$ (i.e. panel (c) in Figs. 2, 3). In this scheme, deep clouds appear red, intermediate-level clouds appear yellow and high hazes appear bluish. Bright ‘white’ (and thus high) clouds are seen at mid-latitudes in both hemispheres, with lower altitude clouds seen at more equatorial latitudes. No discrete deep (indicated as ‘red’) clouds are seen near the south pole, but such clouds are visible near the southern edge of the southern equatorial cloudy zone. Images selected for the ‘scan’ plot, shown in Fig.5 are indicated by the ‘*’ symbol to the left of the observation name.

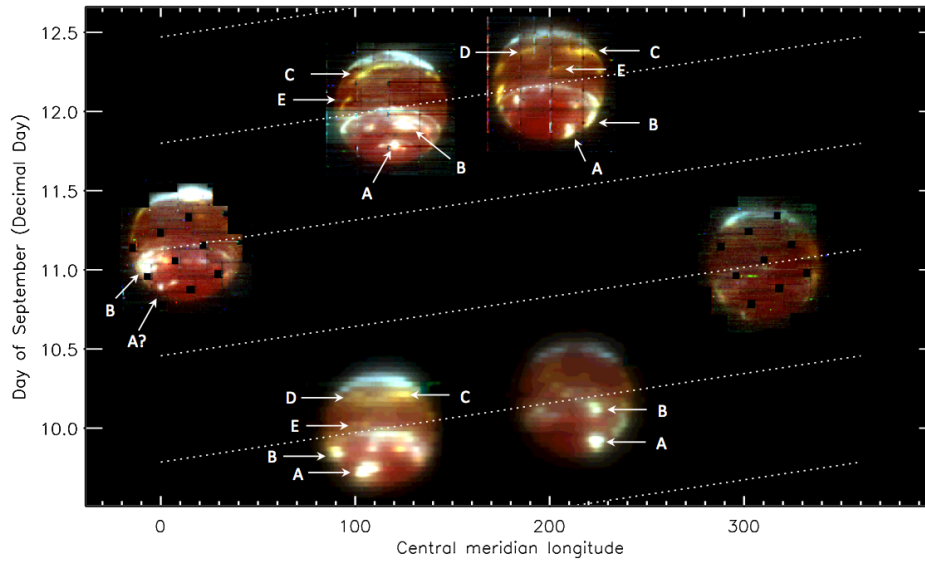


Fig. 5.— Scan plot of selected 2013 VLT/SINFONI H-band observations made in October 2013. The images are plotted at a point depending on the central meridian longitude of the observation (assuming a rotation rate of 16.11 hours) and the observation time plotted as the digital day of the month i.e. October 10th at 00:00 UT is 10.0). Images with best resolution have been chosen covering as great a time period as possible. The diagonal dotted lines indicate how the central meridian longitude varies with time. Distinct identifiable features have been labelled: A) sub-polar discrete cloud; B) mid-latitude bright cloud; C, D and E) near-equatorial intermediate level clouds. The approximate planetocentric latitudes of the five labelled features are 67°S , 42°S , 12°N , 12°N , and 9°S , respectively.

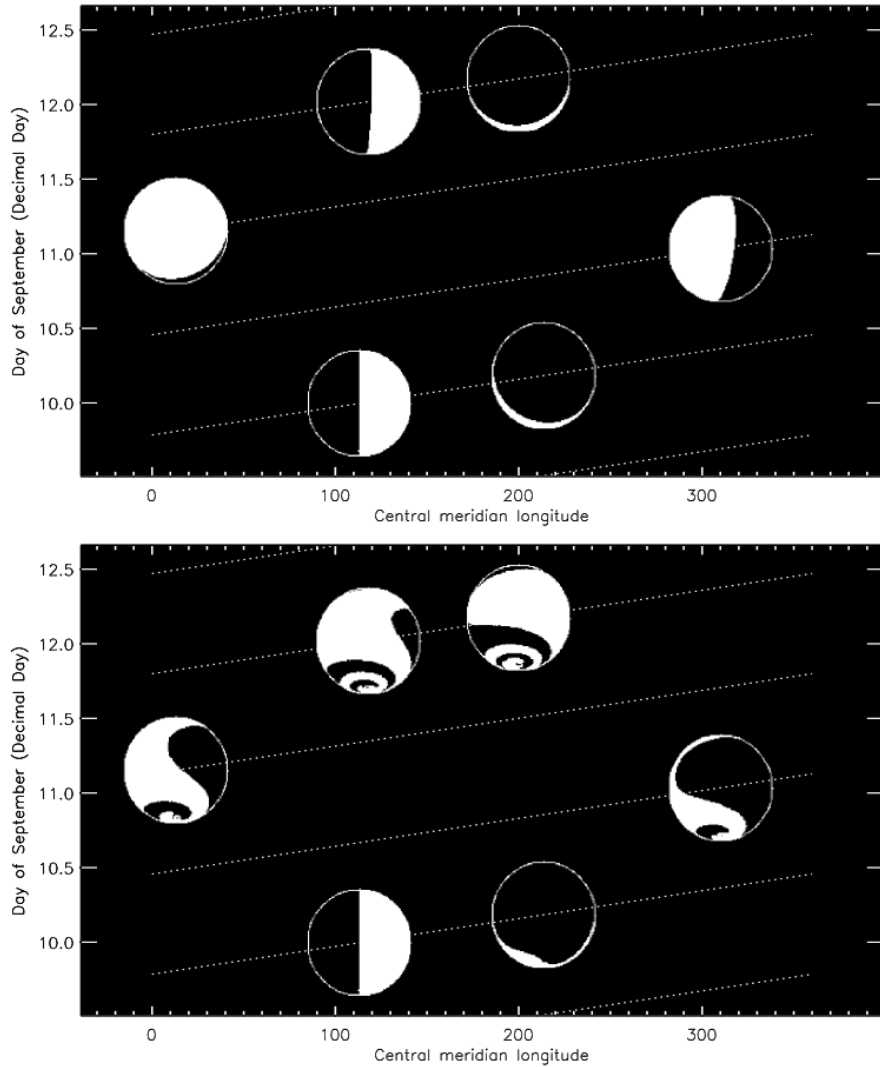


Fig. 6.— For the same time period as is presented in Fig.5, the top plot shows how Neptune would appear with a cloud distribution that is white in one hemisphere and dark in the other if the latitudinally different rotation rates are neglected. The terminator is set to be at a longitude of 0° with white on the right hand side at the time of the first observation plotted. The bottom plot shows how the appearance of such a distribution would be distorted by the very different rotation rates seen at different latitudes in Neptune’s atmosphere due to the extreme latitudinal variation of zonal wind speed.

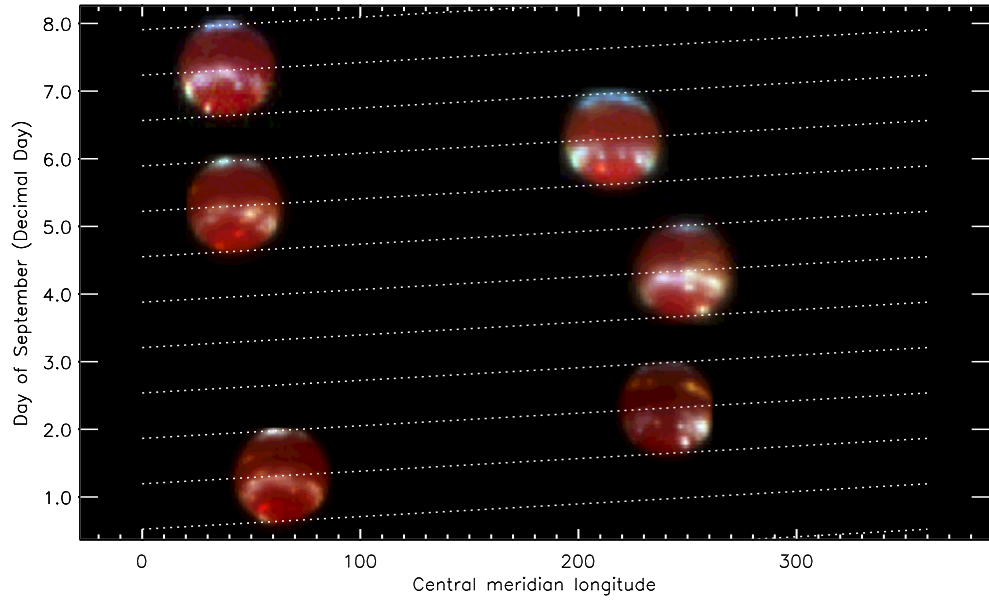


Fig. 7.— As Fig.5, but showing a scan plot of the 2009 Gemini/NIFS H-band observations during September 2009.

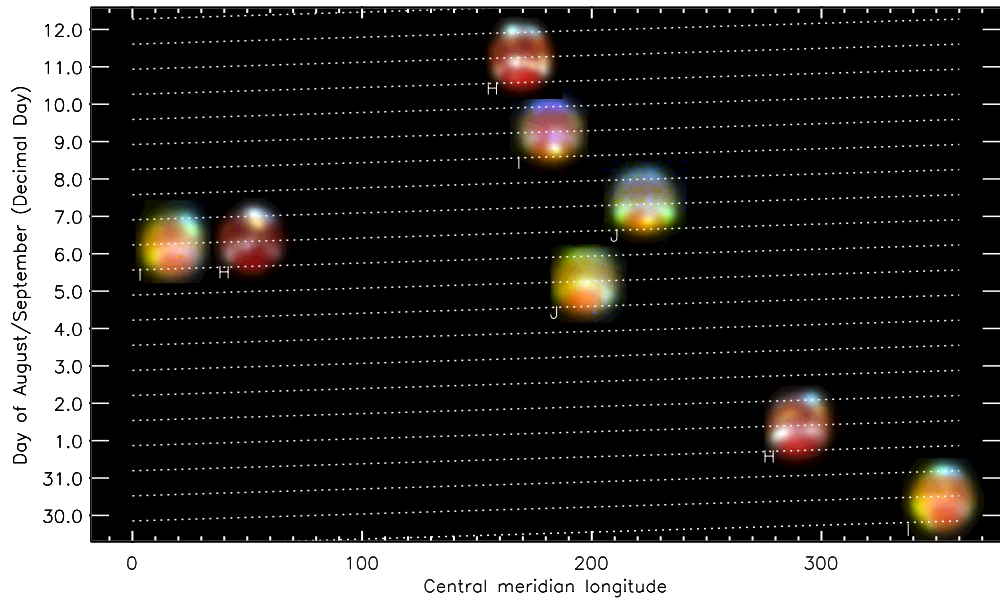


Fig. 8.— As Fig.5, but showing a scan plot of the 2011 Gemini/NIFS observations in the I-, J-, and H-bands during August/September 2011. The grism used for the observation is indicated by each image.

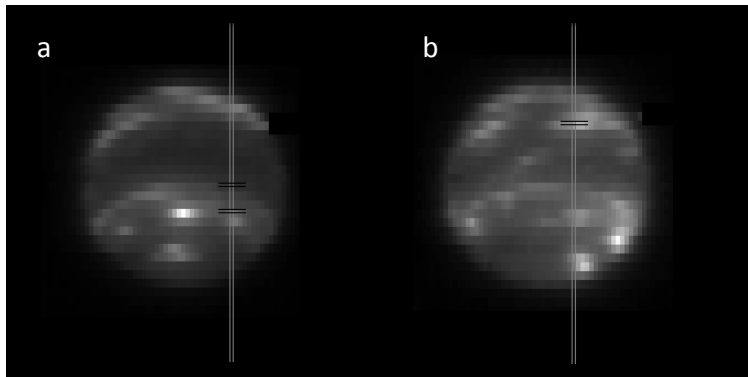


Fig. 9.— The two lines of pixels selected for H-band retrieval analysis. Panel (a) shows observation ‘OB34’ with line cutting through the deep cloud feature in southern hemisphere. Panel (b) shows observation ‘OB37’ with line cutting through the intermediate-level cloud just north of the equator. Figs. 10–13 and Fig.17 show the line retrievals for ‘OB34’, while Figs.18 – 20 show the line retrievals for ‘OB37’. The locations highlighted by the black horizontal lines are those for which the retrieved spectra are presented in detail in Figs. 10, 12, and 19.

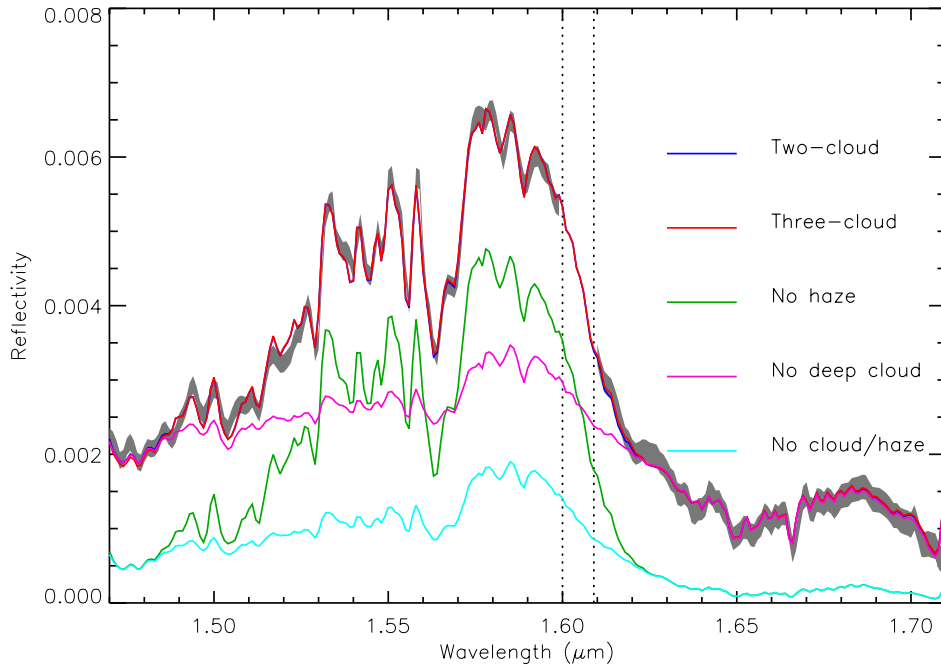


Fig. 10.— Retrieved spectra for a typical sample pixel at 24.1°S (the topmost indicated pixel in panel (a) of Fig.9), just north of the southern mid-latitude cloud belt, using a two-cloud and three-cloud model. The grey shaded region is the measured spectrum and errors, while the solid coloured lines are the retrieved spectra. At this latitude the two-cloud (blue) and three-cloud (red) models are effectively indistinguishable. For reference, also plotted are the spectra calculated for the two-cloud haze case: 1) when just the Haze is removed (green); 2) when just the Tropospheric Cloud is removed (mauve); and 3) when both cloud layers are removed (cyan), in which case the reflectivity calculated is due entirely to Rayleigh scattering. NB in this case data were missing between 1.6 and 1.61 μm .

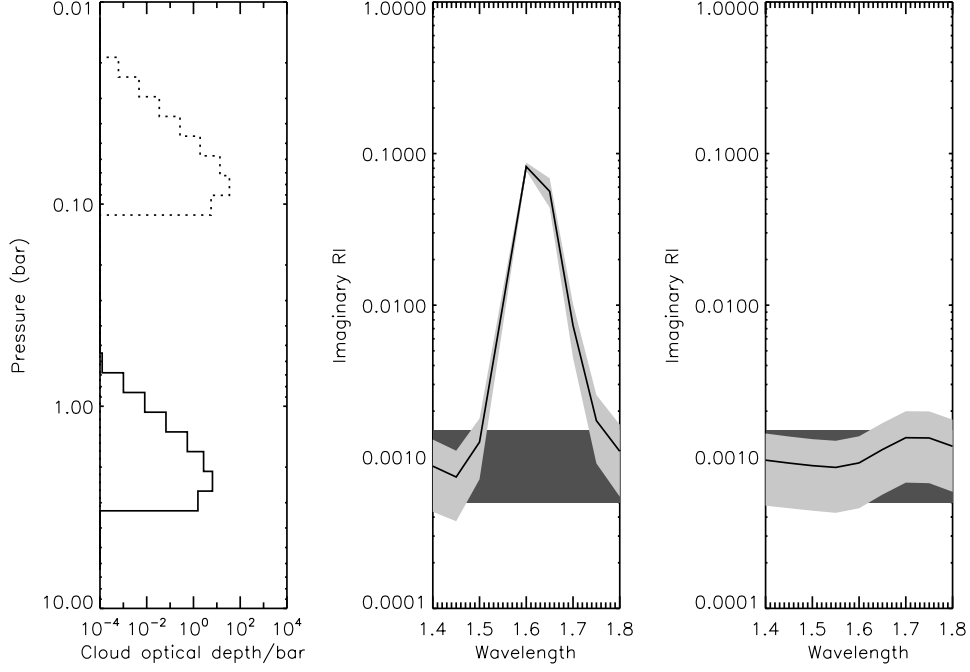


Fig. 11.— Retrieved cloud/haze opacity profile and imaginary refractive index spectra for the Tropospheric Cloud (TC) and Haze from the sample pixel at 24.1°S . Left hand panel shows the retrieved TC/Haze opacity profiles (solid line - TC, dotted line - Haze), while the middle and right hand panels show the retrieved imaginary refractive index spectra for the Tropospheric Cloud (middle panel) and Haze (right hand panel) respectively. For the imaginary refractive indices, the *a priori* value and range is indicated by the darker shaded region, while the retrieved spectra are indicated with the solid line and errors indicated by the lighter shaded region. The imaginary refractive index spectrum of the Tropospheric Cloud can be seen to be generally well-retrieved. However, the imaginary refractive index spectrum of the Haze has barely moved from the *a priori* and the retrieved errors are no smaller than *a priori*. Hence, we conclude that the imaginary refractive index spectrum of the Haze particles are not retrievable in this case.

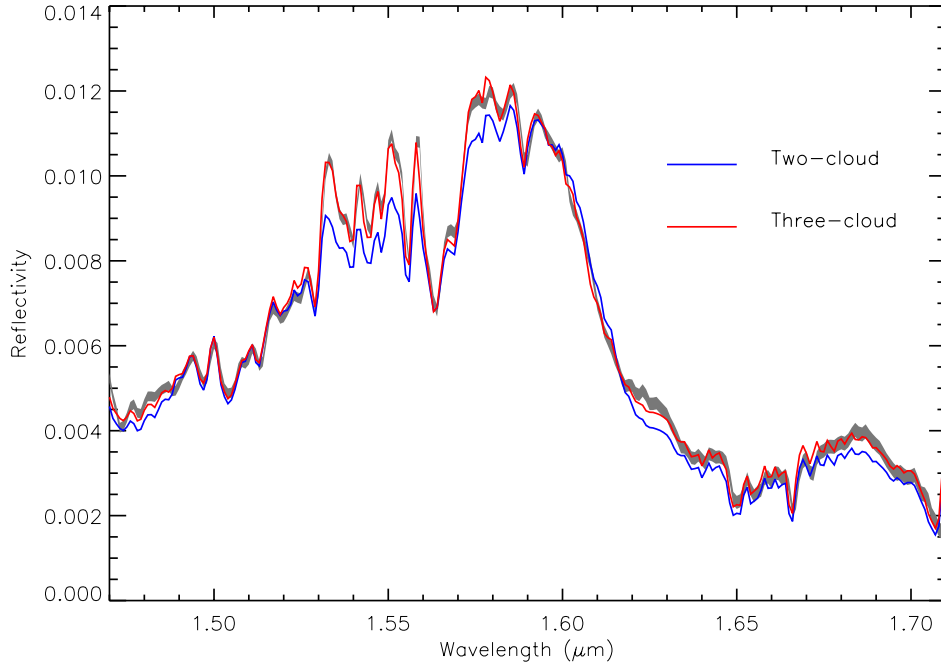


Fig. 12.— Retrieved spectra for sample pixel at 38.5°S (the lowermost indicated pixel in panel (a) of Fig.9, in the centre of the discrete ‘deep’ cloud feature), using a two-cloud (blue), and three-cloud (red) models, where our fit with the two-cloud model is worst. The form of the figure is identical to Fig. 10 and again the grey shaded region is the measured spectrum and errors, while the solid coloured lines are the retrieved spectra. The two-cloud model clearly gives a worse fit at this location, but the addition of a methane cloud based at 1.44 bar greatly improves the fit to the observed spectrum.

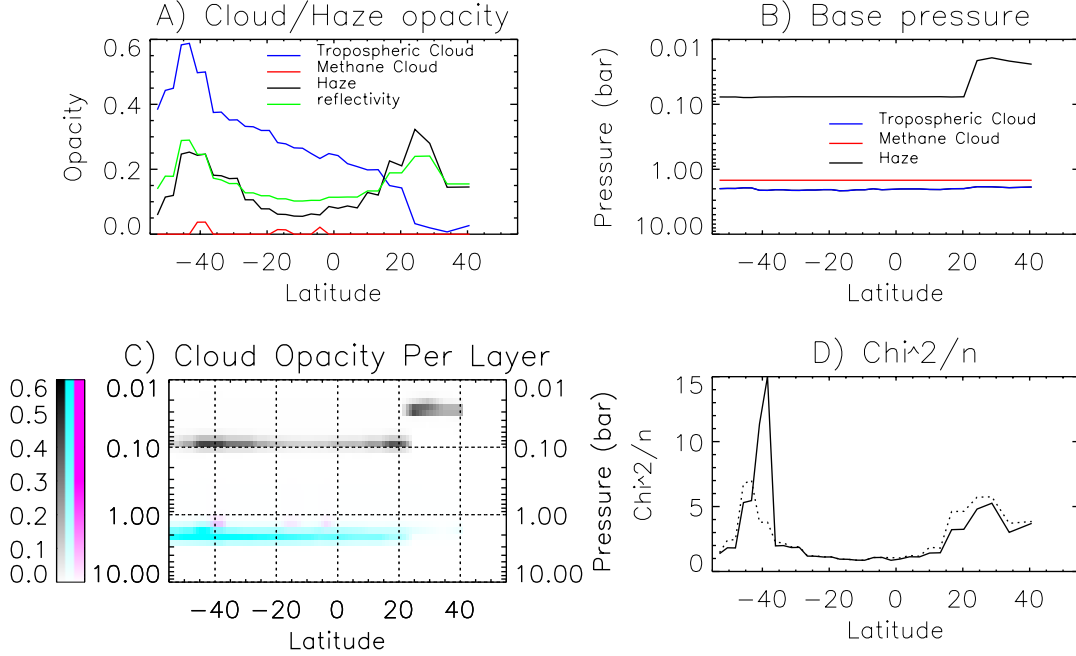


Fig. 13.— Retrieved cloud/haze opacities (Panel A) and base pressures (Panel B) as a function of latitude along the selected line for observation ‘OB34’ (Fig.9), together with an image representation of the resulting opacity per layer in the atmospheric model of each cloud type (Panel C) and the estimated χ^2/n of the fit for the three-cloud (dotted) and two-cloud (solid) models (Panel D). Where the three-cloud model fits better than the two-cloud model, its retrieved quantities have been plotted, otherwise the two-cloud model results are shown. Panel A also shows the observed reflectivity averaged between 1.57 and 1.6 μm to help identify the cloud features. In the cloud opacity per layer plot (Panel C), the opacity of the Haze is represented in grey, the opacity of the Tropospheric Cloud is coloured in cyan, and the methane cloud (where its addition is found to improve the fit) is coloured magenta. The χ^2/n for the two-cloud (solid line) and three-cloud (dotted line) models shown as the dotted line in Panel C, indicates that adding a methane cloud layer only improves the fit at certain locations. In these retrievals the *a priori* tropospheric cloud particles’ complex refractive index was set to $1.4 + 0.001i$ at all wavelengths. The complex refractive index of the haze particles was also set to $1.4 + 0.001i$ at all wavelengths, but fixed since Fig.11 shows we have little sensitivity to the haze refractive index spectrum, assuming the *a priori* particles are highly scattering.

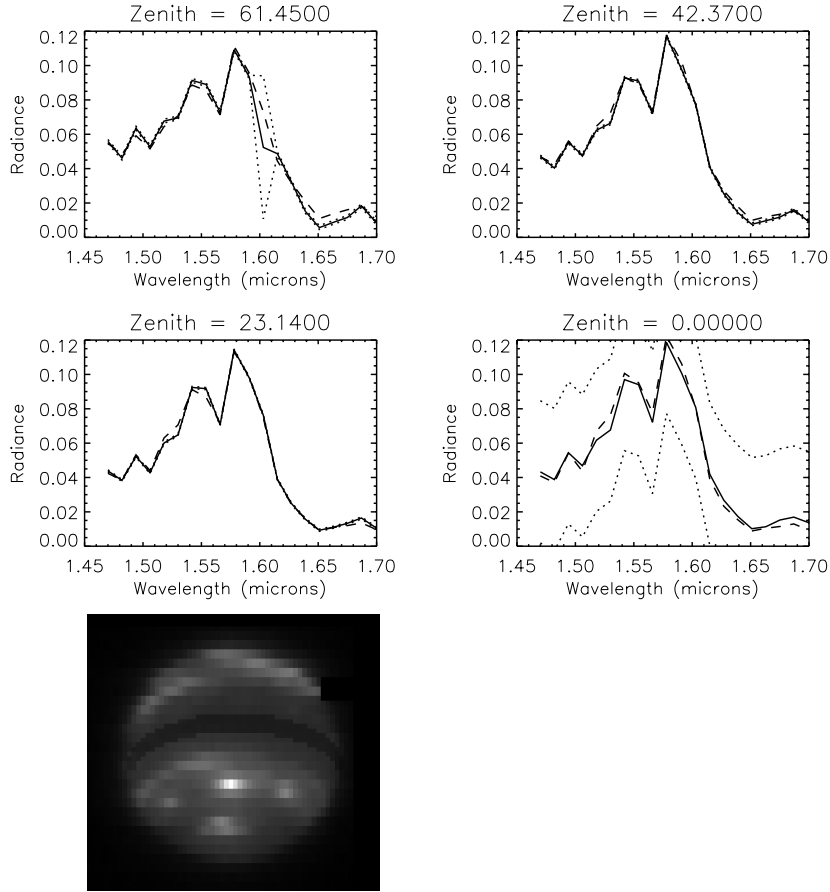


Fig. 14.— Latitude band ($5 - 15^{\circ}\text{S}$) selected for limb-darkening analysis (bottom left) and extracted averaged spectra at the first four angles of the zenith-angle quadrature scheme. Solid lines and dotted lines indicate measured spectra and errors, while the dashed lines are the fitted spectra. Just twenty wavelengths were selected for this analysis. The larger measurement errors for zenith angle = 0 are to account for the fact that we have extrapolated the data beyond the range of measured zenith angles. However, the fit remains good. The larger measurement errors near $1.6 \mu\text{m}$ for zenith angle = 61.45° indicate missing data.

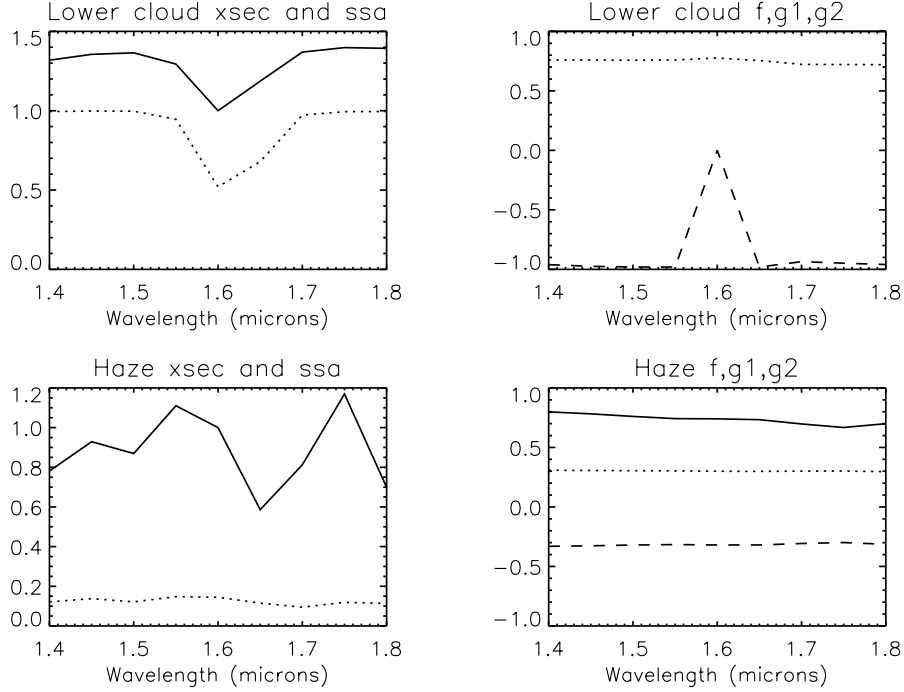


Fig. 15.— Derived scattering properties of the Tropospheric Cloud and Haze from the limb-scattering analysis at $5 - 15^\circ\text{S}$ – extinction cross-section, single-scattering albedo, and Henyey-Greenstein phase function coefficients f , g_1 and g_2 . For cross-section and single scattering albedo, the solid lines are the cross-sections, while dotted lines are single scattering albedoes. For the phase function parameters, f is indicated by the solid lines, g_1 is indicated by the dotted lines and g_2 are indicated by the dashed lines. These properties were derived from the fitted imaginary refractive index spectra and particle sizes. For the Tropospheric Cloud (TC) phase function parameters, f is effectively unity at all wavelengths. Mean particle sizes of 1.1 and $0.2 \mu\text{m}$ were retrieved for the TC and Haze particles respectively.

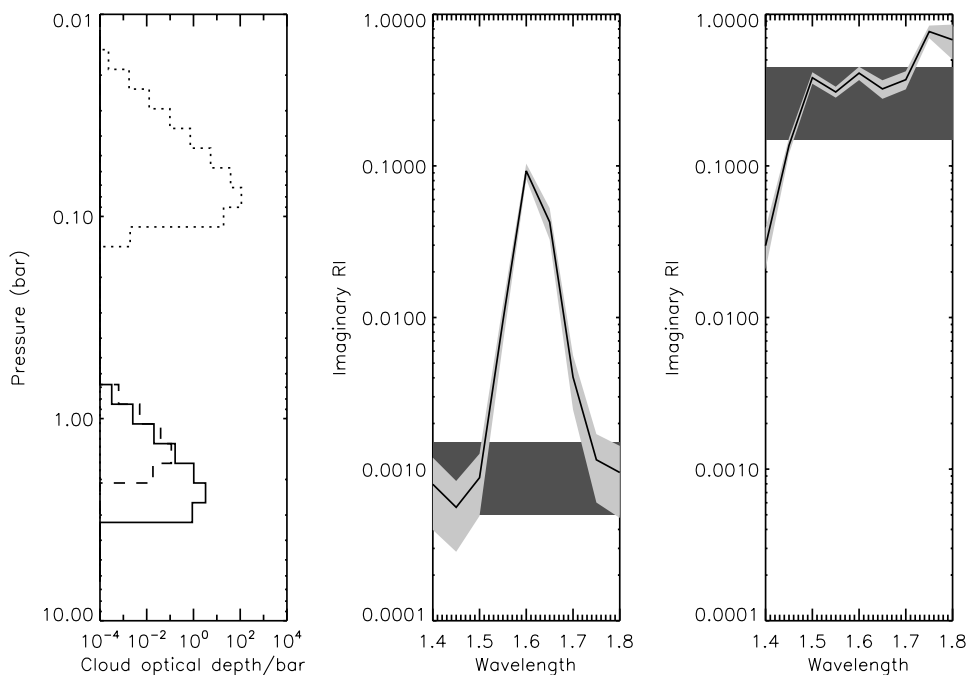


Fig. 16.— Retrieved cloud/haze opacity profile and imaginary refractive index spectra for the Tropospheric Cloud (TC) and Haze from the limb-scattering analysis in the latitude band $5 - 15^{\circ}\text{S}$. As in Fig.11, the *a priori* value and range is indicated by the darker shaded region, while the retrieved spectra and errors are indicated with the solid line and lighter shaded region, respectively. The cloud/haze positions are well-constrained as are the imaginary refractive index spectra of both the TC and the Haze in this case. NB In these retrievals the *a priori* haze complex refractive index was set to $1.4 + 0.3i$ at all wavelengths, as described in the text, while the *a priori* tropospheric cloud complex refractive index was set again to $1.4 + 0.001i$.

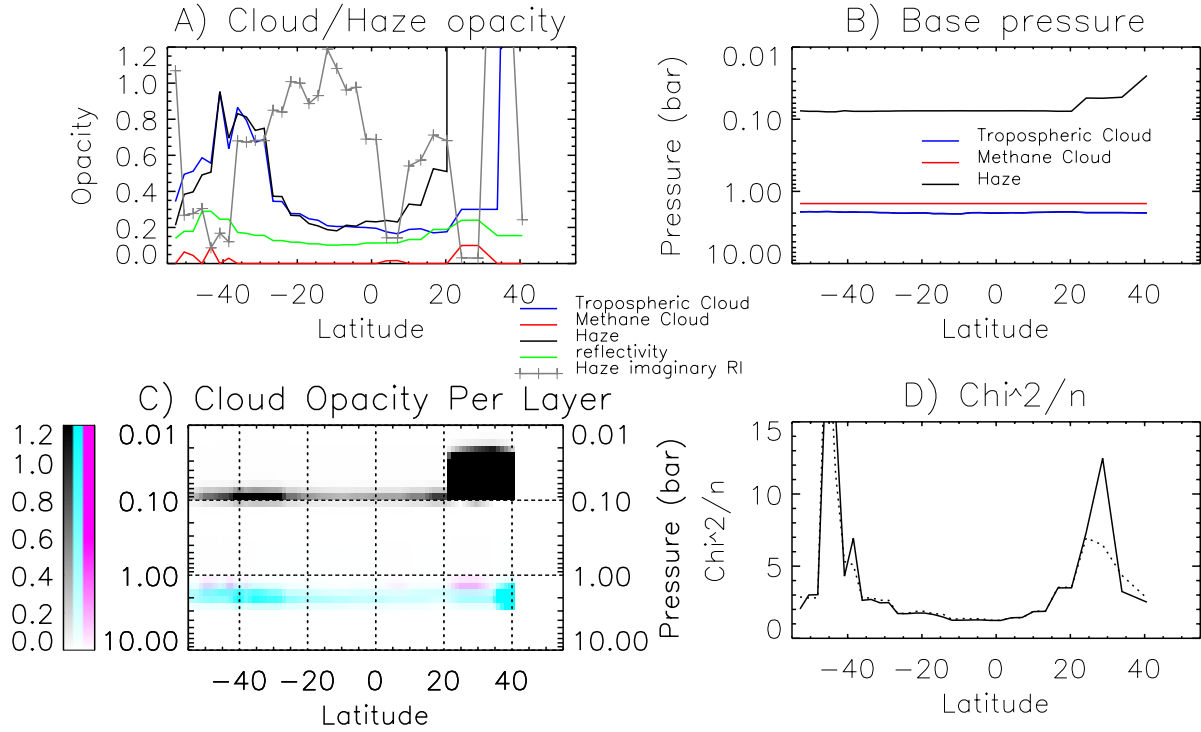


Fig. 17.— As Fig.13, but showing the retrieved cloud/haze opacities and base pressures as a function of latitude along the selected line for observation ‘OB34’ (Fig.9). Here the Haze particles are assumed to be less scattering with an *a priori* complex refractive index set to $1.4 + 0.3i$ at all wavelengths and allowed to vary. The tropospheric cloud particles have the same *a priori* complex refractive index of $1.4 + 0.001i$. Panel A additionally plots the retrieved imaginary refractive index of the Haze particles at $1.65 \mu\text{m}$. As before, the χ^2/n for the two-cloud (solid line) and three-cloud (dotted line) models is shown in Panel D. NB the labels for Panel A have been moved to the centre for clarity.

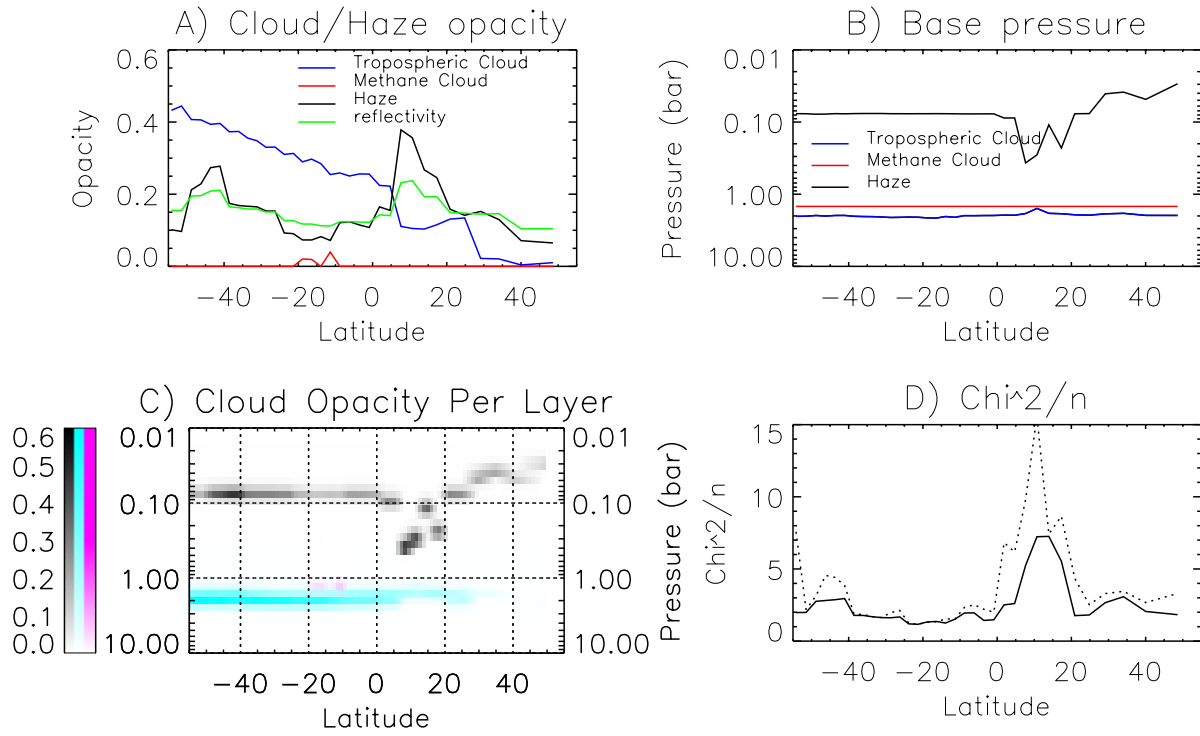


Fig. 18.— As Fig.13, but showing retrieved cloud/haze opacities and base pressures as a function of latitude along the selected line for ‘OB37’, which runs through the ‘intermediate-level’ cloud at $\sim 10^\circ\text{N}$, together with the estimated χ^2/n of the two-cloud (solid line) and three-cloud (dotted line) model fits. In these retrievals the *a priori* Haze complex refractive index was again set to $1.4 + 0.001i$ at all wavelengths and fixed, while the complex refractive index spectra of the tropospheric cloud particles was allowed to vary. A poor fit is obtained in the region of the near-equatorial ‘intermediate-level’ cloud for both models.

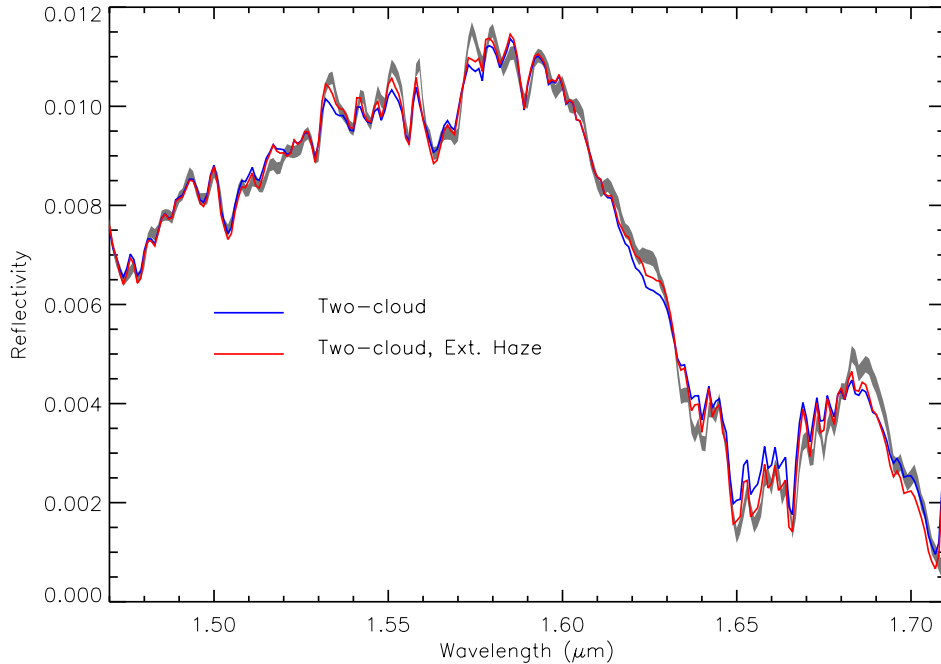


Fig. 19.— Retrieved spectrum for sample pixel at 10.7°S in the ‘intermediate-level’ cloud (indicated in panel (b) of Fig.9) where our fit is worst for the ‘OB37’ line sample. The form of the figure is identical to Fig. 10 and again the grey shaded region is the measured spectrum and errors. The fit with our original two-cloud model is shown in blue, while that in which the Haze is allowed to be vertically extended (i.e. not made to be physically thin), with an *a priori* fractional scale height of 0.5 ± 0.1 and with a higher *a priori* base pressure of 0.25 bar (compared with 0.08 bar before) is shown in red and can be seen to significantly improve the fit.

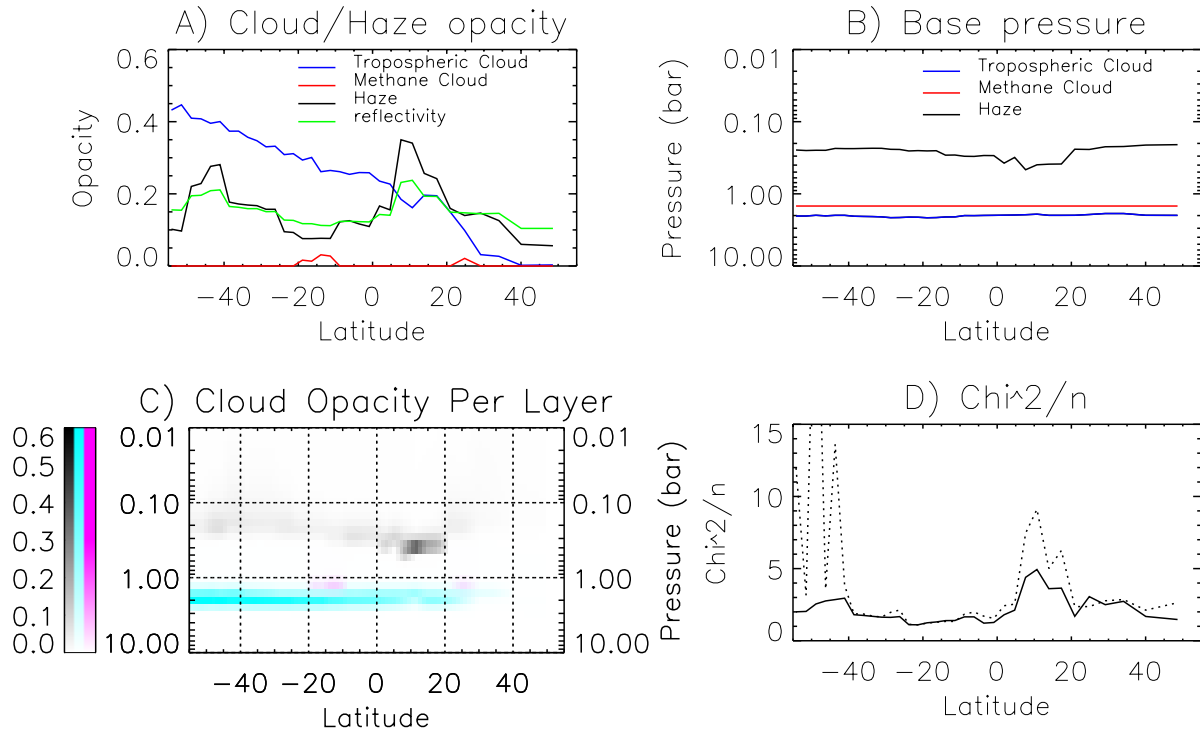


Fig. 20.— As Fig.18, but showing retrieved cloud/haze opacities and base pressures as a function of latitude along the selected line for ‘OB37’ where the Haze *a priori* base pressure was increased to 0.25 bar and the distribution allowed to be vertically extended with an *a priori* fractional scale height of 0.5 ± 0.1 . The fit in the region of the ‘intermediate-level’ cloud can be seen to be significantly improved as compared with Fig.18.

Table 1. 2013 VLT Observations.

Name	Date	T_1^a	T_2^a	Grism	T_{exp}^a	NDIT ^a	Plate Scale
OB1 ^b	9th October 2013	23:52	23:55	H	180s	1	0.1''
OB2	10th October 2013	00:14	00:18	J	180s	1	0.1''
OB3 ^c	10th October 2013	00:21	00:25	J	180s	1	0.1''
OB4	10th October 2013	00:28	00:32	J	180s	1	0.1''
OB5	10th October 2013	00:36	00:39	J	180s	1	0.1''
OB6	10th October 2013	00:46	00:49	H	180s	1	0.1''
OB7	10th October 2013	00:53	00:56	H	180s	1	0.1''
OB8	10th October 2013	01:00	01:04	H	180s	1	0.1''
OB9	10th October 2013	01:07	01:11	H	180s	1	0.1''
OB10	10th October 2013	01:15	01:19	H	180s	1	0.1''
OB11	10th October 2013	01:23	01:26	H	180s	1	0.1''
OB12	10th October 2013	01:30	01:34	H	180s	1	0.1''
OB13 ^b	10th October 2013	04:24	04:28	H	180s	1	0.1''
OB14	10th October 2013	04:31	04:35	H	180s	1	0.1''
OB15	10th October 2013	04:38	04:42	H	180s	1	0.1''
OB16	10th October 2013	04:45	04:49	H	180s	1	0.1''
OB17	10th October 2013	04:53	04:56	H	180s	1	0.1''
OB18	10th October 2013	05:00	05:03	H	180s	1	0.1''
OB19	10th October 2013	05:10	05:14	J	180s	1	0.1''
OB20	10th October 2013	05:17	05:21	J	180s	1	0.1''
OB21	10th October 2013	05:24	05:28	J	180s	1	0.1''

Table 1—Continued

Name	Date	T_1^a	T_2^a	Grism	T_{exp}^a	NDIT ^a	Plate Scale
OB22 ^c	10th October 2013	05:32	05:35	J	180s	1	0.1''
OB23 ^c	10th October 2013	05:40	05:43	J	180s	1	0.1''
OB24	11th October 2013	00:03	00:05	H	60s	1	0.1''
OB25	11th October 2013	00:08	00:10	J	60s	1	0.1''
OB26	11th October 2013	00:29	00:30	H	60s	1	0.1''
OB27 ^b	11th October 2013	00:48	01:48	H	70s	2	0.025''
OB28	11th October 2013	03:20	03:22	H	60s	1	0.1''
OB29	11th October 2013	03:26	03:27	J	60s	1	0.1''
OB30 ^b	11th October 2013	03:40	04:40	H	70s	2	0.025''
OB31	11th October 2013	04:40	04:42	H	60s	1	0.1''
OB32	11th October 2013	04:44	04:45	H	60s	1	0.1''
OB33	11th October 2013	04:50	04:51	J	60s	1	0.1''
OB34	12th October 2013	00:01	00:03	H	60s	1	0.1''
OB35	12th October 2013	00:08	00:10	J	60s	1	0.1''
OB36 ^b	12th October 2013	00:30	01:25	H	70s	2	0.025''
OB37	12th October 2013	03:48	03:50	H	60s	1	0.1''
OB38	12th October 2013	03:55	03:56	J	60s	1	0.1''
OB39 ^b	12th October 2013	04:08	05:02	H	70s	2	0.025''

^a T_1 and T_2 are the start and end time (UT) of each observation (hh:mm), T_{exp} is the exposure time, NDIT is the number of exposures per observation.

^bData included in scan image in Fig. 5

^cPoor image quality and not included in analysis

Table 2: Summary of Gemini-N/NIFS Neptune observations in the 2011 campaign.

Date	T_1	T_2	Grism	Integration Time
30th August 2011	10:35	11:28	I	20min (2min \times 10 frames)
1st September 2011	09:44	10:38	H	20min (2min \times 10 frames)
1st September 2011	10:49	11:54	J	20min (2min \times 10 frames) ^a
5th September 2011	06:15	06:42	J	10min (2min \times 5 frames)
6th September 2011	06:27	07:20	I	20min (2min \times 10 frames)
6th September 2011	08:00	08:54	H	20min (2min \times 10 frames)
7th September 2011	07:45	08:38	J	20min (2min \times 10 frames)
9th September 2011	06:16	07:09	I	20min (2min \times 10 frames)
11th September 2011	06:00	06:53	H	20min (2min \times 10 frames)

^aNot shown in Fig.8 as it overlaps on figure with previous observation.