

Jupiter's tropospheric composition and cloud structure from 5- μm spectroscopy



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A thesis submitted for the degree of
Doctor of Philosophy

Trinity 2016

Acknowledgements

I am very grateful to my two supervisors, Leigh Fletcher and Pat Irwin, for all of their support and encouragement. I couldn't have asked for better mentors. I am also fortunate to be part of a very welcoming wider academic community, with particular thanks going to Tommy Greathouse and Glenn Orton. AOPP has been a wonderful place to work over the last three years, and much of that is due to the people I shared an office with during that time: Charlotte, James, Ellie, Ashwin and Ryan (who wasn't technically an officemate, but might as well have been). And finally, huge thanks go to my parents and my sister, for always believing in me and for encouraging me to believe in myself.

Abstract

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This thesis uses infrared observations from spacecraft and ground-based telescopes to investigate the composition and cloud structure of the jovian atmosphere. It focuses on a single spectral region, known as the 5- μ m window, where Jupiter's upper atmosphere becomes optically thin. This allows us to probe down beneath the planet's thick cloud decks to the 4–8 bar region in the middle troposphere. Two different data sources are combined to build up a three-dimensional picture of Jupiter's troposphere. The first dataset is from the Cassini VIMS instrument, and was taken during the 2000–2001 Jupiter flyby. These observations cover a wide spectral range, provide global coverage and include both the nightside and the dayside of the planet, making them well suited to studying clouds. The VIMS spectra can be modelled using a single tropospheric cloud deck, subject to the following constraints: (i) the cloud base is located at pressures of 1.2 bar or lower; (ii) the cloud particles are highly scattering; and (iii) the cloud is sufficiently spectrally flat. The second dataset is from the CRIRES instrument at the Very Large Telescope in Chile. These observations have a very high spectral resolution, allowing the absorption lines of individual molecular species to be resolved. The CH₃D line shape varies between belts and zones, which can be interpreted as variations in the opacity of a deep cloud, located at around 5 bar. There is also evidence for spatial variability in two disequilibrium species, AsH₃ and PH₃, both of which show an enhancement at high latitudes. This is in contrast to a third disequilibrium species, GeH₄, which shows no evidence for spatial variability. The CRIRES dataset also includes several strong emission lines, which are identified as H₃⁺, an auroral species in Jupiter's ionosphere. The strengths of these lines were measured in order to determine the ionospheric temperatures. The work in this thesis contributes to our understanding of the dynamical, chemical and cloud-forming processes shaping Jupiter's troposphere and provides a reference point for future work, including observations made by NASA's Juno mission.

Contents

1	Introduction	1
1.1	Overview	1
1.2	5- μ m window	2
1.3	Jupiter’s troposphere	5
1.3.1	Composition	5
1.3.2	Clouds	7
1.4	Thesis structure	8
2	Radiative transfer and retrieval theory	11
2.1	Introduction	11
2.2	Radiative transfer	12
2.3	Molecular absorption	14
2.3.1	Ro-vibrational transitions	14
2.3.2	Line shapes	15
2.3.3	Line-by-line modelling	17
2.3.4	Correlated-k approximation	17
2.3.5	Sources of line data	18
2.4	Scattering	20
2.5	Retrieval theory	21
2.6	Reference atmosphere	24
2.7	Summary	25
3	Data	27
3.1	Introduction	27
3.2	Cassini VIMS	27
3.2.1	The Cassini mission	27
3.2.2	The VIMS instrument	28
3.2.3	Observations and data reduction	30
3.3	VLT CRIRES	32
3.3.1	The CRIRES instrument	32
3.3.2	Observations	33
3.3.3	Data reduction	35
3.4	Summary	38
4	Tropospheric clouds: part 1	41
4.1	Introduction	41
4.2	Observations	43
4.3	Sensitivity analysis	45
4.4	Basic retrievals	50
4.5	Cloud constraints	52

4.5.1	Vertical structure	52
4.5.2	Spectral features	58
4.5.3	Scattering properties	61
4.6	Global retrievals	64
4.6.1	Water vapour	64
4.6.2	Phosphine	67
4.6.3	Clouds	69
4.7	Reflected sunlight analysis	69
4.8	Conclusions	74
5	Tropospheric clouds: part 2	77
5.1	Introduction	77
5.2	Scattering properties of upper cloud	78
5.3	Deep cloud structure	82
5.3.1	Spatial variability	82
5.3.2	Latitudinal retrieval	85
5.3.3	Sensitivity to the scattering parameters	87
5.4	Conclusions	88
6	Disequilibrium species	91
6.1	Introduction	91
6.2	GeH ₄	92
6.2.1	Background	92
6.2.2	Identification of GeH ₄ features	94
6.2.3	Spatial variability	96
6.2.4	Latitudinal retrieval	98
6.2.5	Summary	100
6.3	AsH ₃	101
6.3.1	Background	101
6.3.2	Identification of AsH ₃ features	101
6.3.3	Spatial variability	102
6.3.4	Latitudinal retrieval	104
6.3.5	Summary	106
6.4	PH ₃	107
6.4.1	Background	107
6.4.2	Spatial variability	108
6.4.3	Latitudinal retrieval	110
6.4.4	Summary	111
6.5	Discussion	112
6.5.1	Abundances	112
6.5.2	Latitudinal profiles	113
6.6	Conclusions	116
7	Ionospheric H₃⁺ emission	117
7.1	Introduction	117
7.2	Line identification	119
7.3	Kinetic temperature	122
7.4	Rotational temperature	122
7.5	Vibrational temperature	124
7.6	Discussion and conclusions	125

8	Conclusions and future work	127
8.1	Introduction	127
8.2	Conclusions	128
8.3	Future work	132
8.3.1	Global mapping of disequilibrium species	132
8.3.2	Abundances of condensable species	132
8.4	Final comments	134
	Bibliography	137

Acronyms and abbreviations

AU	Astronomical Units
CIRS	Composite Infrared Spectrometer
CRIRES	Cryogenic High-Resolution Infrared Echelle Spectrograph
EqZ	Equatorial Zone
ESO	European Southern Observatory
EsoRex	ESO Recipe Execution Tool
FWHM	Full Width Half Maximum
GRS	Great Red Spot
IRIS	Infrared Interferometer Spectrometer
ISIS3	Integrated Software for Imagers and Spectrometers
JPL	Jet Propulsion Laboratory
KAO	Kuiper Airborne Observatory
LTE	Local Thermodynamic Equilibrium
MACAO	Multi-Applications Curvature Adaptive Optics
MWR	Microwave Radiometer
NEB	North Equatorial Belt
NEMESIS	Non-linear Optimal Estimator for Multivariate Spectral Analysis
NIMS	Near-Infrared Mapping Spectrometer
NTrZ	North Tropical Zone

SEB	South Equatorial Belt
STrZ	South Tropical Zone
TEXES	Texas Echelon Cross Echelle Spectrograph
VIMS	Visual and Infrared Mapping Spectrometer
VISIR	VLT Imager and Spectrometer for the Mid-Infrared
VLT	Very Large Telescope
VMR	Volume Mixing Ratio

Chapter 1

Introduction

1.1 Overview

As the archetypal giant planet, Jupiter has played an important role in shaping our understanding of the Solar System. With no obstructions to atmospheric flows, the entire planet can be treated as a laboratory for fundamental geophysical fluid dynamics. Observing and analysing the circulation patterns can provide more information about how atmospheres work than can be obtained from the Earth alone (Ingersoll et al., 2004). Furthermore, identifying the spectral signatures of different molecules in planetary atmospheres and calculating the bulk abundances and isotopic ratios helps us to understand how the planets in the Solar System were formed (Lunine et al., 2004). Jupiter is the largest planet in the Solar System and at its nearest point is only 4.2 AU from the Earth, making it more accessible than the other gas giants, Saturn, Neptune and Uranus. In addition, thousands of extra-solar planets now been discovered in orbit around other stars, and many of these are gas giants with a similar mass to Jupiter (e.g. Macintosh et al., 2015). Until improved observations allow for detailed atmospheric characterisation, Jupiter serves as an initial template for the dynamical, chemical and cloud-forming processes shaping the atmospheres of these distant planets.

Despite its significance, there are still many unanswered questions about Jupiter's atmosphere. This is particularly true of the middle troposphere, the region of the planet that lies below the thick upper cloud decks. The middle troposphere is the only part of the atmosphere where significant amounts of certain chemical species are found, including thermochemical disequilibrium species that can act as tracers for tropospheric

dynamics. It is also thought to be the location of several distinct cloud decks, but there is no clear consensus on their locations or composition.

Throughout much of the electromagnetic spectrum, Jupiter's middle troposphere is completely hidden from view. The presence of highly absorbing molecules at higher altitudes, as well as thick cloud decks, means that radiation from the troposphere does not reach the top of the atmosphere. One exception to this is a unique spectral region known as the 5- μm window, where the upper jovian atmosphere is optically thin. It provides a way to probe Jupiter's troposphere and this spectral region will therefore be the focus of this thesis. The aim of this thesis is to use 5- μm data from both spacecraft and ground-based telescopes to build up a three-dimensional picture of Jupiter's tropospheric cloud structure and composition.

1.2 5- μm window

The 5- μm atmospheric window is a unique part of the jovian infrared spectrum that allows us to probe deep into the planet's atmosphere. Between 4.5 and 5.2 μm , Jupiter's atmosphere becomes optically thin, allowing us to see radiation that originates below the main cloud decks. Figure 1.2.1 shows two images of Jupiter, taken at approximately the same time. On the right, is a visible-light image of the planet, taken by an amateur observer. This image shows Jupiter's typical banded structure, alternating between the pale, cloudy zones and the dark, cloud-free belts. On the left, is a 5- μm image of the planet, taken using the VISIR instrument (Lagage et al., 2004) at the Very Large Telescope. This image shows Jupiter's tropospheric clouds in silhouette against the bright tropospheric radiation from below. There is a strong contrast between the cloud-free belts, which offer an unobstructed view of the warm, deeper regions, and the zones, where some of this radiation is attenuated by the thick clouds.

In the cloud-free regions, the emission originates from the 4–8 bar region (Taylor et al., 2004), which corresponds to the middle troposphere (see Figure 1.2.2). This contrasts with the rest of the infrared region, which predominantly sounds pressures of less than 1 bar.

Figure 1.2.3 shows how the temperature functional derivative (i.e. dR/dT , where R is the top-of-atmosphere radiance and T is the atmospheric temperature) varies as

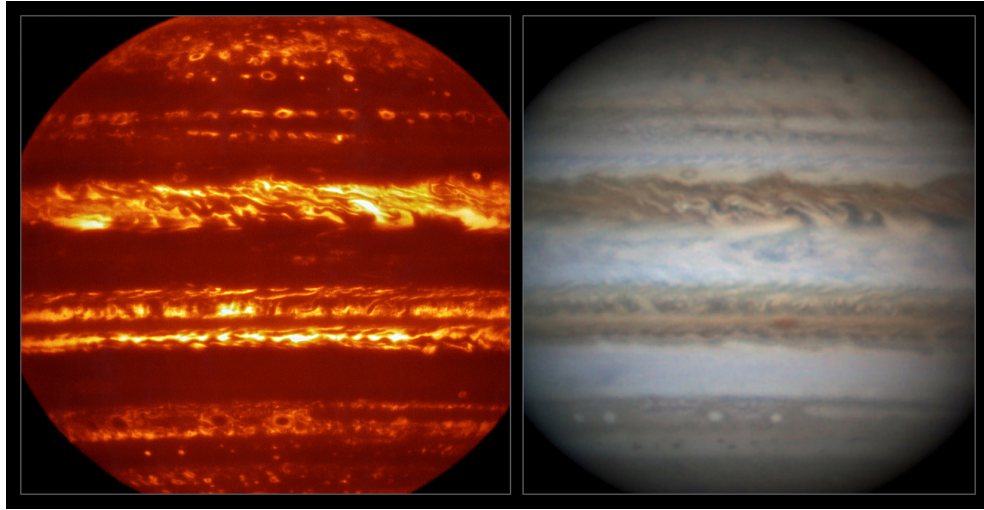


Figure 1.2.1: Images of Jupiter at two different wavelengths. On the left is a 5- μm image of Jupiter, made using VLT VISIR (Credit: ESO/L.N. Fletcher). On the right is a visible-light image of Jupiter, made by an amateur observer at nearly the same time (Credit: D. Peach).

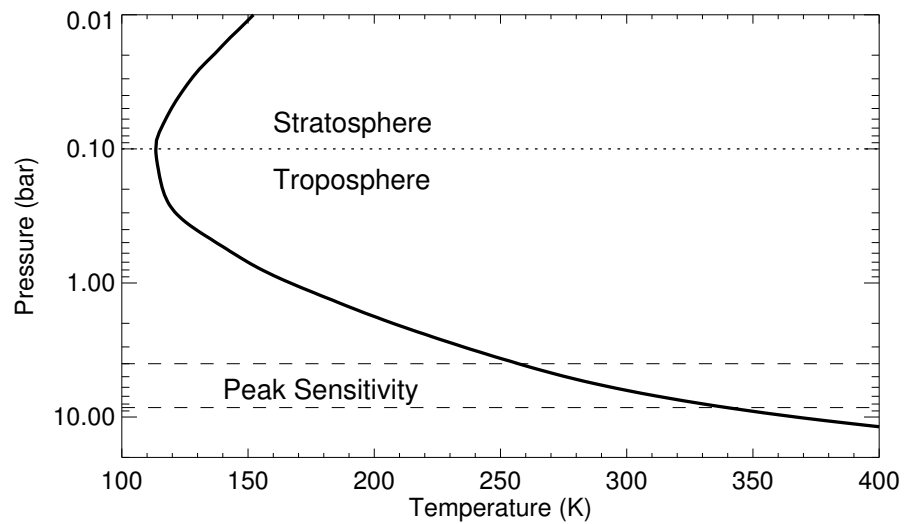


Figure 1.2.2: The temperature-pressure profile of Jupiter's lower atmosphere. The dotted line indicates the location of the tropopause, the boundary between the troposphere and the stratosphere. Sensitivity in the 5- μm window peaks at 4–8 bar, in the middle troposphere.

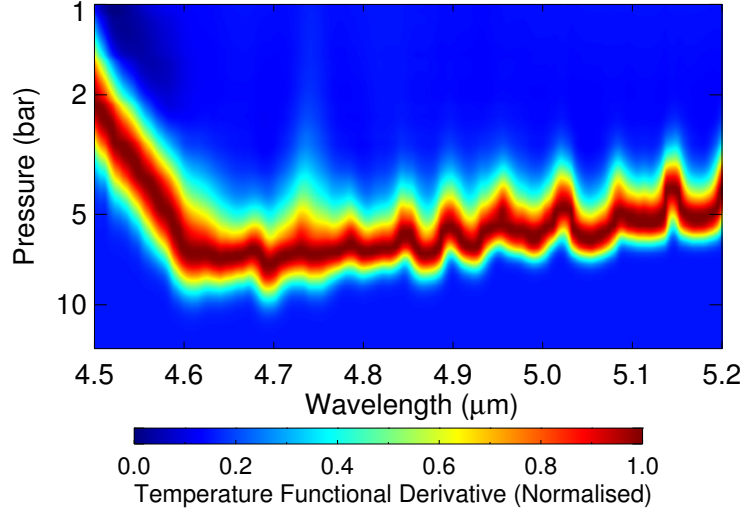


Figure 1.2.3: The temperature functional derivative for a cloud-free atmosphere, normalised at each wavelength.

a function of wavelength across the 5- μm band. This shows the extent to which each atmospheric layer contributes to the observed radiance; the pressure level where the temperature functional derivative is the highest is the pressure of maximum sensitivity. Within the window, the observed radiance is sensitive to changes deep in the atmosphere, at around 4–8 bar (if a thick tropospheric cloud is included in the model, there is additional sensitivity at the location of the cloud). The wavelengths on either side of the 5- μm window probe lower pressures, corresponding to the upper troposphere. The window exists because it is a point where the low opacities of the principal infrared absorbers, H_2 , CH_4 and NH_3 coincide. Its size is defined by the ν_1 and ν_2 PH_3 bands at the short-wavelength edge and the $2\nu_2$ and ν_4 CH_4 bands at long wavelengths (Encrenaz et al., 1996).

The atmospheric window was first described by Gillett et al. (1969) and the high degree of spatial variability was noted by Westphal (1969). Since then, the 5- μm region has been the focus of many studies from ground-based telescopes, airborne observatories and spacecraft. There have been several missions to Jupiter that carried instruments covering the 5- μm window range: the infrared interferometer spectrometer and radiometer (IRIS) on board Voyager 1 and 2 (Hanel et al., 1980), the near-infrared mapping spectrometer (NIMS) on Galileo (Carlson et al., 1992) and the visual and infrared mapping spectrometer (VIMS) on Cassini (Brown et al., 2004). 5- μm data from the first two missions have been used to determine the abundances of various species (Drossart

et al., 1982; Drossart and Encrenaz, 1982) and to shed light on the structure of the jovian cloud decks (Bézard et al., 1983; Irwin et al., 1998; Roos-Serote et al., 1999). The 5- μm part of the VIMS jovian spectrum has not yet been studied; an analysis of this data is carried out in Chapter 4.

There have also been several ground-based and airborne studies using Jupiter’s 5- μm spectrum. Bjoraker et al. (1986) used data from the Kuiper Airborne Observatory to determine the tropospheric gaseous composition and a similar analysis was carried out by Encrenaz et al. (1996) using the Infrared Space Observatory. The Canada-France-Hawaii Telescope and the United Kingdom Infrared Telescope have been used by several groups to make high spectral resolution observations of the bright ‘hot-spots’ in Jupiter’s North Equatorial Belt in order to target individual gaseous species (Bézard et al., 2002; Noll et al., 1989). This thesis uses data from the CRIRES instrument at the Very Large Telescope in Chile to study the tropospheric cloud structure and composition. These observations provide higher spectral resolution and better spatial coverage than previous ground-based observations and are analysed in Chapters 5–7.

1.3 Jupiter’s troposphere

1.3.1 Composition

Jupiter’s atmosphere is predominantly made up of hydrogen and helium, but there are also many other species present. Table 1.3.1 provides a summary of Jupiter’s tropospheric composition. The global abundances of the minor chemical species are important pieces of evidence in determining the origin of the planet (Lunine et al., 2004). Jupiter has an observed enrichment in heavy elements relative to the Sun, which suggests that it cannot have formed by a direct collapse of gases via gravitational instability. Instead, it is thought that accretion of icy planetesimals brought these heavy elements to the planet. Precise measurements of Jupiter’s deep abundances are required to understand the origin of these planetesimals and how they reached Jupiter’s orbit. However, these are difficult values to determine accurately, since the vertical profiles of the gaseous species are governed by a range of physical processes, including non-equilibrium chemistry, vertical transport in the atmosphere, condensation to form clouds, and photochemistry in

Species	VMR	Source
Hydrogen, H ₂	0.86	Niemann et al. (1998)
Helium, He	0.136	Niemann et al. (1998)
Methane, CH ₄	0.002	Niemann et al. (1998)
Ammonia, NH ₃	0.0002	Fletcher et al. (2009)
Water, H ₂ O	>0.0005	Taylor et al. (2004)
Hydrogen Sulphide, H ₂ S	77 ppm	Niemann et al. (1998)
Neon, Ne	23 ppm	Mahaffy et al. (2000)
Argon, Ar	15 ppm	Mahaffy et al. (2000)
Hydrogen Deuteride, HD	15 ppm	Taylor et al. (2004)
Phosphine, PH ₃	0.2 ppm	Fletcher et al. (2009)
Deuterated Methane, CH ₃ D	0.3 ppm	Lellouch et al. (2001)
Krypton, Kr	9.2 ppb	Mahaffy et al. (2000)
Carbon Monoxide, CO	1.0 ppb	Bézard et al. (2002)
Xenon, Xe	0.88 ppb	Mahaffy et al. (2000)
Germane, GeH ₄	0.45 ppb	Bézard et al. (2002)
Arsine, AsH ₃	0.24 ppb	Bézard et al. (2002)

Table 1.3.1: The mean volume mixing ratios of the various species present in Jupiter’s troposphere. The species with prominent spectral signatures in the 5- μ m region are NH₃, H₂O, PH₃, CH₃D, CO, GeH₄ and AsH₃.

the upper atmosphere (Taylor et al., 2004). The species with prominent spectral signatures in the 5- μ m region are ammonia (NH₃), water vapour (H₂O), deuterated methane (CH₃D), carbon monoxide (CO), phosphine (PH₃), germane (GeH₄) and arsine (AsH₃).

The temperature profile of the jovian troposphere means that both H₂O and NH₃ condense to form clouds, which makes determining their vertical profiles complex. Water is expected to condense first to produce the deepest cloud deck at ~ 7 bar, while ammonia condenses out at two different levels, producing a cloud made of solid NH₄SH particles at ~ 3 bar and a higher cloud of ammonia ice at ~ 0.8 bar (see Section 1.3.2). Analysis of these gaseous species does not form a major part of this thesis (although H₂O is briefly discussed in Chapter 4), but Chapter 8 describes how they could be studied in future work.

CH₄ and its isotopologue CH₃D both have a relatively simple profile on Jupiter, with a constant volume mixing ratio throughout most of the atmosphere. The temperatures are high enough that they do not condense, and they are chemically stable up until the upper stratosphere where they are dissociated by UV radiation and energetic particles (Moses et al., 2004). CH₃D has strong spectral signatures in the 5- μ m window and, due to its constant volume mixing ratio, measurements of the CH₃D line width have been used as a proxy for measuring cloud structure in the deep troposphere (Bjo-

raker et al., 2015). Chapter 5 provides further background on CH_3D and analyses the latitudinal variability observed in the CH_3D line shapes.

The remaining four gases are all disequilibrium species in Jupiter’s troposphere. This thesis focusses on PH_3 , GeH_4 and AsH_3 ; CO is not included, as the strong telluric absorption lines from CO at $5\text{ }\mu\text{m}$ make the jovian abundance difficult to determine from ground-based observations (unless there is a strong Doppler shift). The abundances of disequilibrium species are determined by temperature-dependent equilibrium reactions. Deep in the atmosphere, the temperature is high enough that they have stable mixing ratios. However, the temperature in the observable troposphere is much lower and so the abundances of these species should rapidly drop to very low amounts. The fact that GeH_4 , AsH_3 and PH_3 have all been detected in Jupiter’s atmosphere with higher than expected abundances (Fink et al., 1978; Noll et al., 1989; Ridgway et al., 1976) is therefore indicative of rapid vertical motion forcing the disequilibrium species upwards at a rate faster than they can decompose. This means that detection of an enhanced abundance of a disequilibrium species can indicate a region of strong vertical mixing, while a lower value can suggest a region of weak vertical mixing. Mapping the abundances of species such as PH_3 , GeH_4 and AsH_3 can therefore provide information about atmospheric dynamics in Jupiter’s troposphere (Taylor et al., 2004). One of the objectives of this thesis is to measure the latitudinal distribution of these disequilibrium species and this is addressed in Chapter 6 (PH_3 is also briefly discussed in Chapter 4). Chapter 6 also provides further background information about these species.

1.3.2 Clouds

Jupiter’s troposphere is also home to several cloud decks. Theory predicts that there should be three main layers: a deep cloud composed of solid H_2O , a middle cloud composed of solid NH_4SH (ammonium hydrosulphide) and an upper cloud composed of NH_3 ice (West et al., 2004). The results of a typical cloud condensation model from Atreya et al. (1999) are shown in Figure 1.3.1. If solar abundances of N, S and O are assumed, this model places the NH_3 cloud base at 0.7 bar, the NH_4SH cloud base at 2.2 bar and the H_2O cloud base at 5.4 bar, with an additional thin layer of aqueous-ammonia solution just below the H_2O cloud. If the abundances are increased

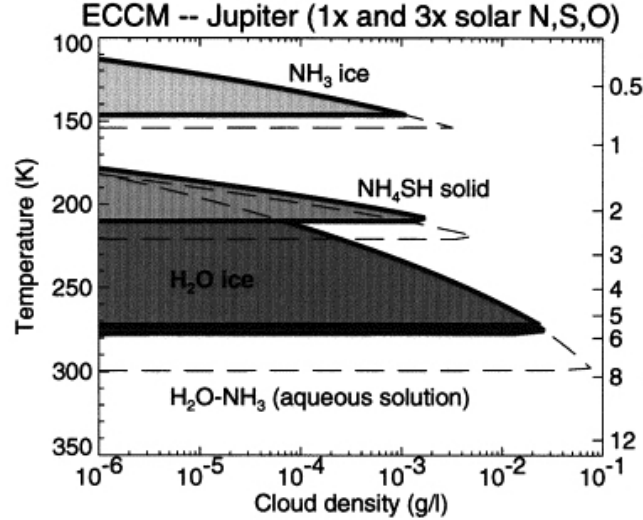


Figure 1.3.1: Equilibrium cloud condensation profiles from Atreya et al. (1999). The case where the abundances of N, S and O are solar is depicted by the solid area and the case where the abundances are three times solar is depicted by the dashed lines.

to three times solar, which is more accurate for Jupiter (Taylor et al., 2004), the clouds move downwards to 0.8 bar, 2.6 bar and 7.2 bar respectively. In addition to these main tropospheric cloud layers, there are also small-particle hazes in the upper troposphere and stratosphere, but these are mostly transparent to 5- μm radiation (West et al., 2004).

The huge spatial variations in brightness temperatures that are found in the 5- μm spectra clearly indicate the presence of clouds of variable optical thickness in the atmosphere, as the changes cannot be accounted for by variations in chemical composition. However separating the effects of the discrete cloud decks and conclusively identifying the components of the clouds has proved difficult. Further information about previous observational studies of Jupiter’s tropospheric clouds is provided in Chapter 4. In this thesis, 5- μm observations of Jupiter are used to constrain the tropospheric cloud properties. This makes use of both spacecraft data from Cassini VIMS (Chapter 4) and ground-based observations from VLT CRIRES (Chapter 5).

1.4 Thesis structure

In this thesis, 5- μm spectroscopy is used to study the composition and cloud structure of Jupiter’s troposphere. These two subjects are closely entwined. Firstly, they are physically connected, as the formation of clouds both depends on, and alters, the chem-

ical composition. Secondly, both clouds and chemical species can have similar effects on the observed spectrum, leading to degeneracies in retrieving abundances or opacities. An analysis of one therefore requires good knowledge of the other. Chapter 4 shows that in order to study Jupiter’s cloud structure, one must have an understanding of the effects of chemical species on the planet’s spectrum. Similarly, Chapters 5 and 6 show that an accurate measurement of gaseous abundances is heavily dependent on the cloud properties.

In addition to studying these two main topics, this thesis also makes use of two different datasets: observations from the VIMS instrument on the Cassini spacecraft and observations from the CRIRES instrument at the Very Large Telescope. These two instruments provide complementary views of Jupiter’s troposphere. The VIMS instrument is relatively low spectral resolution ($R=300$), but can simultaneously observe the entire $5\text{-}\mu\text{m}$ spectral window. This is ideal for studying broad-band features, such as the cloud structure or gases with very strong spectral features in the $5\text{-}\mu\text{m}$ window. VIMS also provides observations of Jupiter’s nightside, which removes the complication of reflected sunlight, and provides full global coverage, allowing us to map the cloud structure and composition. In contrast, CRIRES has very high spectral resolution ($R=96,000$), which allows us to resolve the individual absorption features of gases in Jupiter’s troposphere. This is well-suited to studying minor chemical species with relatively weak spectral signatures. CRIRES provides latitudinal coverage, so this data can be used to search for latitudinal variability in composition.

The scientific results reported in this thesis will be structured according to these two datasets. In Chapter 2, the concepts of radiative transfer and retrieval theory are introduced, and NEMESIS, the retrieval algorithm that is used to analyse the data, is discussed. Chapter 3 describes the two instruments used to make observations and the methods used to reduce and calibrate the data. Chapter 4 analyses the data from Cassini VIMS. This is primarily focussed on Jupiter’s tropospheric clouds, but the tropospheric composition is also briefly discussed. This chapter is heavily based on Giles et al. (2015), which has been published in *Icarus*. Chapters 5 and 6 analyse the data obtained from VLT CRIRES. Chapter 5 explores how the CRIRES data is able to further constrain the tropospheric cloud properties and discusses how the cloud structure can affect the

analysis of gaseous species. In Chapter 6, Jupiter’s disequilibrium chemical species are measured, and their latitudinal profiles are determined. These two chapters are based on Giles et al. (2016a), a paper which has published in *Icarus*.

The final results chapter, Chapter 7, also makes use of the CRIRES data, but unlike the previous chapters, it is not focussed on Jupiter’s troposphere. Instead, it makes use of the serendipitous detection of H_3^+ emission lines in Jupiter’s 5- μm spectrum. H_3^+ is an auroral species in Jupiter’s ionosphere, which provides an insight into the energy budget of the upper atmosphere. This chapter is taken from Giles et al. (2016b), a paper published in *Astronomy & Astrophysics*.

The overall aim of this thesis is to answer the following questions:

1. When studying Jupiter’s troposphere using 5- μm spectroscopy, what are the advantages and disadvantages of low-resolution spacecraft data, compared with high-resolution ground-based data?
2. What degeneracies exist between different atmospheric parameters, and what limitations do these place on atmospheric retrievals?
3. What is the vertical structure of Jupiter’s tropospheric cloud decks, and how do they vary across the planet?
4. What are the scattering properties of Jupiter’s cloud particles, and are there any spectral features?
5. How do the abundances of disequilibrium species vary across the planet, and what can be inferred about the tropospheric dynamics?
6. What information can measurements of H_3^+ emission provide about Jupiter’s upper atmosphere?

In Chapter 8, these questions are revisited, and the conclusions from the previous four chapters are drawn together. Chapter 8 then discusses opportunities for future work.

Chapter 2

Radiative transfer and retrieval theory

2.1 Introduction

Remote sensing involves analysing the radiation that is reflected, absorbed and emitted by a planetary atmosphere in order to draw conclusions about the atmospheric properties. This can include the temperature profile, the mixing ratios of chemical species and the cloud cover. It differs from in situ methods, such as atmospheric probes, in that the properties of interest are not directly measured and must be retrieved from the data. This is a complex process, but remote sensing has the advantage of being able to study phenomena from a distance and over planetary-wide scales.

In this thesis, Jupiter's 5- μm spectrum is modelled using the NEMESIS retrieval algorithm developed by Irwin et al. (2008). NEMESIS is made up of two parts: a radiative transfer code that computes the emergent radiation for a given atmospheric profile and an optimal estimation retrieval algorithm, which iteratively determines the best-fit atmospheric parameters for an observed spectrum. This chapter provides an introduction to NEMESIS and how it is used in this thesis. Section 2.2 describes the theory behind radiative transfer models, and Sections 2.3 and 2.4 describe the roles of gaseous species and cloud particles respectively. Section 2.5 describes the retrieval algorithm and Section 2.6 describes the reference atmosphere used in this work.

2.2 Radiative transfer

When light passes through a non-scattering material, there is a change in radiance due to absorption and emission. For light with a radiance L , the change in radiance, dL , over a vertical distance dz , at an angle θ to the zenith, is given by Schwarzschild's equation (e.g. Andrews, 2010):

$$dL = \frac{kn}{\mu}(B(T) - L)dz \quad (2.1)$$

where k is the absorption coefficient of the material, n is the number density of the material, $\mu = \cos \theta$, and $B(T)$ is the Planck function at the temperature of the material (which describes the radiation emitted by a blackbody of temperature T). k depends on the composition of the material (see Section 2.3). L , k and $B(T)$ are all wavelength specific.

The first term on the right-hand side of Equation 2.1 shows the increase in radiation due to emission from the material, while the second term shows the decrease in radiation due to absorption by the material. Depending on the relative sizes of the two terms, the presence of an absorbing material can lead to either an absorption line (decrease in radiance) or an emission line (increase in radiance) in the spectrum.

An observer making remote sensing observations measures the top-of-atmosphere radiation from a planet. The optical depth $\chi(z)$ is the optical path between an altitude z and the top of the atmosphere and is defined by:

$$\chi(z) = - \int_{s'=\infty}^{z'=z} \frac{k(z')n(z')}{\mu} dz' \quad (2.2)$$

The optical depth at the top of the atmosphere is zero, and it increases as the altitude decreases. Rewriting Equation 2.1 in terms of Equation 2.2 gives

$$\frac{dL}{d\chi} - L = -B(T) \quad (2.3)$$

Using an integrating factor $I = e^{-\chi}$, this can be integrated between $z = z_0$ ($\chi = \chi(z_0)$) and $z = \infty$ ($\chi = 0$) to give

$$L_{\infty} = L_{z_0} e^{-\chi(z_0)} + \int_0^{\chi(z_0)} B(T(\chi)) e^{-\chi} d\chi \quad (2.4)$$

where L_{∞} is the radiance at the top of the atmosphere and L_{z_0} is the radiance at an altitude z_0 . This can be further rewritten in terms of the transmission $\tau(z, \infty) = e^{-\chi(z)}$ as

$$L_{\infty} = L_{z_0} \tau(z_0, \infty) + \int_{z_0}^{\infty} B(T(z)) \frac{d\tau}{dz} dz \quad (2.5)$$

In the case of a rocky planet, z_0 refers to the altitude of the surface. The first term on the right hand side of Equation 2.5 therefore describes the radiation emitted by the surface of the planet, attenuated by the total optical depth of the atmosphere and the second term represents the radiation emitted by the atmosphere itself. $d\tau/dz$ is known as the transmission weighting function, and it describes how much each layer of the atmosphere contributes to the observed top-of-atmosphere radiation. This weighting function generally varies smoothly as a function of altitude, peaking in a region of the atmosphere where the optical depth is approximately unity.

In the case of a gas giant, such as Jupiter, the first term becomes redundant, as the entirety of the observed radiation comes from the atmosphere. As long as the base of the model atmosphere is set at an altitude much deeper than where the weighting functions peak, the precise location is not important. In this thesis, the base of the atmosphere is set to 30 bar (see Section 2.6); at these high pressures, the optical depth is high enough that no contribution is provided to the observed top-of-atmosphere radiation.

Equation 2.5 forms the basis of the radiative transfer code in NEMESIS. Given an atmospheric temperature profile, abundances of gaseous species and information about the absorption coefficients, it allows the top-of-atmosphere radiation to be calculated as a function of wavelength. This produces a ‘synthetic spectrum’ of the atmosphere, which can then be compared to observations.

2.3 Molecular absorption

2.3.1 Ro-vibrational transitions

Section 2.2 makes use of the absorption coefficient, k , which varies as a function of wavelength. The value of k depends on the gaseous composition of the atmosphere, as well the temperature and pressure. In the infrared, the gaseous absorption is primarily dependent on the vibrational and rotational modes of the molecules in the atmosphere. This section is based on content from Andrews (2010) and gives a simplified overview in the case of a diatomic molecule. For polyatomic molecules, the equations in this section become significantly more complex and further details can be found in Herzberg (1989).

Vibrational modes of a diatomic molecules are classically compared to the oscillation of a spring. By inserting the potential function of a harmonic oscillator into Schrödinger's equation, the infinite set of vibrational energy levels, E_v , can be obtained:

$$E_v = h\nu_0 \left(v + \frac{1}{2} \right) \quad \text{where} \quad v = 0, 1, 2, \dots \quad (2.6)$$

where h is the Planck constant, ν_0 is the frequency of oscillation of the system (dependent on the spring constant and the reduced mass of the molecule), and v the vibrational quantum number, which takes integer values. The dipole selection rule states that in a transition from one vibrational mode to another, $\Delta v = \pm 1$, so $|\Delta E_v| = h\nu_0$.

In addition to different vibrational modes, a molecule may also have different rotational modes. For diatomic molecules, it can be shown that there is an infinite set of rotational energy levels,

$$E_J = \frac{h^2}{8\pi^2 I} J(J+1) \quad \text{where} \quad J = 0, 1, 2, \dots \quad (2.7)$$

where I is the moment of inertia of the molecule (dependent on the molecular geometry and reduced mass), and J is the rotational quantum number, which also takes only integer values. As with vibrational transitions, the ordinary selection rule for transitions between rotational states is $\Delta J = \pm 1$, leading to $|\Delta E_J| = (h^2/4\pi^2 I) (J+1)$, where J is the quantum number of the lower state.

The energy differences between vibrational modes of a molecule are typically equiv-

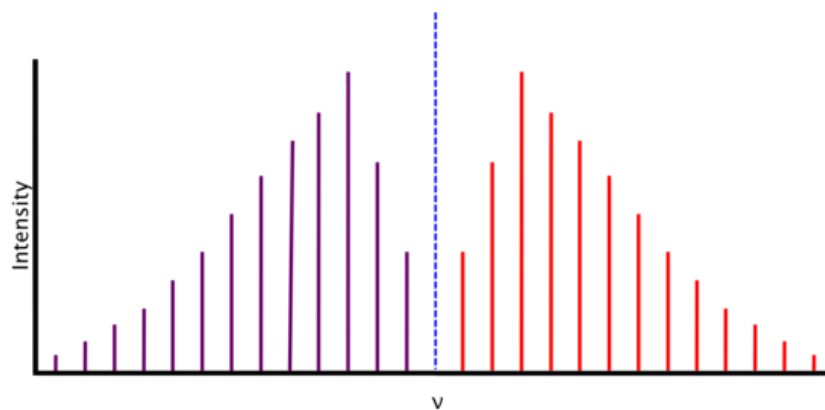


Figure 2.3.1: A cartoon depiction of an ideal ro-vibrational spectrum. Credit: Joya Cooley, UC Davis

alent to photons with wavelengths 1–20 μm (thermal infrared). The energy differences between rotational modes are much smaller and are associated with wavelengths of 10^2 – 10^4 μm (far infrared and microwave). Generally, vibrational and rotational transitions occur together, leading to a joint ro-vibrational spectrum. A cartoon of an ideal ro-vibration spectrum is shown in Figure 2.3.1. The band is centred (dashed blue line) around the vibrational energy transition, with the lower energy rotational transition providing fine structure on either side. The purple lines correspond to transitions where $\Delta J = -1$ and these lines are known as the ‘P-branch’. The red lines correspond to transitions where $\Delta J = +1$ and these lines are known as the ‘R-branch’. In some cases, the ‘Q-branch’, which corresponds to $\Delta J = 0$ can also be observed; its location is marked by the blue dashed line.

2.3.2 Line shapes

Figure 2.3.1 shows infinitely narrow absorption lines, but in reality, the lines in the ro-vibrational spectrum are broadened due to physical effects. This section is based on content from Irwin (2008).

The first effect is due to the thermal motion of the molecules and is known as Doppler broadening. The molecules of the gas have a random motion, and at any given moment, some will be travelling towards the observer (giving a blue-shift to the spectral line) and some will be travelling away from the observer (giving a red-shift to the spectral line). The hotter the temperature of the gas, the faster the motion of the molecules, leading to more pronounced Doppler broadening. If the line centre is located at frequency ν_0 ,

then the Doppler-broadened absorption coefficient k_ν at frequency ν is given by:

$$k_\nu = \frac{S}{\pi^{1/2}} \frac{1}{\gamma_D} \exp\left(-\frac{(\nu - \nu_0)^2}{\gamma_D^2}\right) \quad \text{where} \quad \gamma_D = \gamma_{D_0} \left(\frac{T}{T_0}\right)^{1/2} \quad (2.8)$$

where S is the integrated line strength and γ_{D_0} is the Doppler line width at a reference temperature, T_0 . This is a Gaussian line shape, where the line width is proportional to $T^{1/2}$.

The second line broadening effect is due to the collisions between molecules and is known as pressure broadening. Energy levels have finite lifetimes due to collisions between molecules, and this leads to a broadening of the lines. The greater the pressure of the gas, the higher the rate of collisions, leading to more pronounced pressure broadening. If the line centre is located at frequency ν_0 , then the pressure-broadened absorption coefficient k_ν at frequency ν is given by:

$$k_\nu = \frac{S}{\pi} \frac{\gamma_L}{(\nu - \nu_0)^2 + \gamma_L^2} \quad \text{where} \quad \gamma_L = \gamma_{L_0} \frac{p}{p_0} \left(\frac{T_0}{T}\right)^n \quad (2.9)$$

where γ_{L_0} is the pressure-broadened line width at a reference temperature T_0 and pressure p_0 , and n is the temperature coefficient. According to simple theory, $n = 0.5$, but the value varies between different molecules. This is a Lorentz line shape, where the line width is proportional to pT^{-n} . A similar finite-lifetime effect also exists due to the spontaneous decay from excited states, but this is negligible (in the infrared) compared to pressure broadening.

At very high pressures, the overall line shape tends towards the pressure-broadened Lorentz line shape, and at very low pressures, the line shape tends towards the Doppler-broadened Gaussian line shape. If the Doppler line width and the pressure-broadened line width are comparable in size, then the Voigt profile can be used, which combines both line shapes. The Voigt profile is given by:

$$k_\nu = \frac{S}{\pi^{3/2}} \frac{\gamma_L}{\gamma_D^2} \int_{-\infty}^{\infty} \frac{\exp(-t^2)}{(x - t)^2 + (\gamma_L/\gamma_D)^2} dt \quad (2.10)$$

where $x = (\nu - \nu_0)/\gamma_D$. Equation 2.10 does not have an analytical solution and must be integrated numerically.

2.3.3 Line-by-line modelling

Sections 2.3.1 and 2.3.2 describe the positions and line shapes of individual spectral lines. A planetary atmosphere contains many different molecular species, with overlapping spectral lines; in order to calculate the overall opacity of a given spectral interval, all of the spectral lines that contribute to the opacity at those wavelengths must be taken into account. The most accurate way to do this is using a line-by-line calculation, where the contributions from each individual line are summed together (Irwin, 2008).

For each individual line, the line strength and line shape are functions of temperature and pressure, both of which vary throughout the atmosphere. For observations with a moderately low spectral resolution, tens of thousands of these lines must then be added together, making this line-by-line method very computationally expensive. This is especially true when several such models need to be run in order to retrieve an iterative best-fit solution (see Section 2.5).

The analysis of the very high spectral resolution CRIRES observations in Chapters 5 and 6 uses the line-by-line method. In this case, only small wavelength ranges ($\sim 0.02\mu\text{m}$) are studied simultaneously, and so the line-by-line method is relatively fast. In contrast, the Cassini VIMS observations (analysed in Chapter 4) have a much lower spectral resolution and the entire $4.5\text{--}5.2\mu\text{m}$ wavelength range is simultaneously analysed. This includes an enormous number of spectral lines, making the line-by-line method prohibitively time-consuming. In this case, the correlated-k approximation (Section 2.3.4) is used, in order to speed up the radiative transfer calculation.

2.3.4 Correlated-k approximation

The correlated-k approximation (Goody et al., 1989) is a method for rapidly calculating finite resolution spectra. This approximation takes advantage of the fact that when calculating the mean transmission of small spectral interval $\lambda_0 \rightarrow \lambda_0 + \Delta\lambda$, the precise location of the absorption lines is not important; knowing the fraction of the spectral interval that is occupied by different absorption coefficients is sufficient. This means that the absorption coefficients in the spectral interval can be reshuffled into ascending order, producing a smoothly varying function known as a k-distribution (Irwin et al., 2008).

In order to calculate the mean transmission across a spectral interval, the absorption coefficient must be integrated across the spectral range. The advantage of using the smoothly-varying k-distribution is that it can be integrated with far fewer quadrature points (NEMESIS typically uses 20), vastly reducing the computational time (Irwin et al., 2008). k-distributions can be pre-calculated for each gas, for a range of temperatures and pressures, and stored in ‘k-tables’ which are then used in the radiative transfer calculation.

The correlated-k method can be used for inhomogeneous paths through the atmosphere, as it is found that there is a correlation between the k-distributions for adjacent layers (Goody et al., 1989; Lacis and Oinas, 1991). This means that an inhomogeneous atmosphere can be modelled by summing together homogeneous layers. This has been shown to be a valid assumption for most physically realistic atmospheres by Fu and Liou (1992). A particular advantage of the correlated-k approximation over other radiative transfer approximations is that scattering calculations can be summed in the same way (Irwin, 2008). The importance of scattered light is described in Section 2.4 and forms a large part of the analysis in Chapters 4 and 5.

2.3.5 Sources of line data

The sources of the molecular line data are provided in Table 2.3.1. The line positions and intensities are primarily obtained from the HITRAN2004 (Rothman et al., 2009) and the GEISA2009 (Jacquinet-Husson et al., 2011) molecular databases. The two exceptions are AsH₃, where laboratory data from Dana et al. (1993) and Mandin et al. (1995) are used, and GeH₄, where the Spherical Top Data System (Wenger and Champion, 1998) is used and these theoretical intensities are scaled to match the more limited data from GEISA2009 (see Section 6.2 for more details).

In addition to providing information about the line centres and intensities, these molecular databases also provide data for the air-broadened line widths and temperature coefficients (see Equation 2.9). These air-broadened values are measured/calculated using a mixture of nitrogen and oxygen, as found on Earth. Jupiter’s atmosphere is primarily H₂, so H₂-broadening coefficients are more useful. Where possible, estimates of H₂-broadening are used instead (CH₄, CH₃D, NH₃, PH₃, AsH₃). For those gases

Gas	Line intensities	H ₂ pressure-broadening half-width	Temperature dependence
CH ₄ , CH ₃ D	Rothman et al. (2009)	0.061 cm ⁻¹ atm ⁻¹ (B. Bézard, pers. comm.)	Margolis (1993)
NH ₃	Rothman et al. (2009)	Brown and Peterson (1994)	Brown and Peterson (1994)
H ₂ O	Rothman et al. (2009)	-	-
PH ₃	Jacquinet-Husson et al. (2011)	Levy et al. (1993)	Salem et al. (2004)
CO	Rothman et al. (2009)	-	-
GeH ₄	Wenger and Champion (1998), Jacquinet-Husson et al. (2011)	-	-
AsH ₃	Dana et al. (1993), Mandin et al. (1995)	0.075 cm ⁻¹ atm ⁻¹ (L. Brown, pers. comm.)	$n = 0.50$ (L. Brown, pers. comm.)

Table 2.3.1: The sources of line data used in the radiative transfer model of Jupiter’s atmosphere.

with gaps in Table 2.3.1 (H₂O, CO, GeH₄), no H₂-broadening data was available and air-broadening coefficients are used instead.

For most of the molecules, a Voigt line shape is used with an arbitrary wing cut-off of 35 cm⁻¹, following the approach of Fouchet (2000). Extensive testing showed that varying the location of this cut-off had a negligible impact on the spectrum. The one exception was NH₃; the distribution of different line intensities meant that the low resolution Cassini VIMS spectrum was particularly sensitive to the shape of the NH₃ absorption lines. Bailly et al. (2004) have shown that the NH₃ line wings are significantly sub-Lorentzian, and this empirical model for the shape of the wings was used in place of the Voigt line shape.

In addition to the opacity due to ro-vibrational transitions, there is also some gaseous opacity due to collision induced absorption. Homonuclear diatomic molecules like H₂ do not possess a dipole moment, so do not have ro-vibrational spectra. However collisions between molecules can induce a transitory dipole moment, when subsequently leads to absorption. This absorption is very weak, but H₂ is so abundant that it can still have an observable effect on the spectrum (Irwin, 2008). Collision induced absorption data is included from Orton et al. (2007) and Borysow et al. (1988).

For line-by-line calculations (Chapters 5 and 6) the line data described in this section is used directly. For correlated-k calculations (Chapter 4), the line data is used to pre-calculate the ‘k-tables’ that are subsequently used in the radiative transfer code.

2.4 Scattering

Section 2.3 deals with the absorption and emission of gaseous species. Jupiter’s atmosphere contains several cloud decks, and these also have a significant effect on the observed radiation from the planet. Firstly, cloud particles can absorb and emit light, just like gaseous species. Secondly, cloud particles can scatter light (both reflected sunlight and emission from the planet itself), which significantly complicates the radiative transfer model. The NEMESIS radiative transfer code has the capability to model a multiple-scattering atmosphere, both with line-by-line calculations and with the correlated-k approximation. It has already been shown by Roos-Serote and Irwin (2006) that scattering has a strong influence on Jupiter’s 5- μm spectra, so it is important to include scattering effects in the model.

The scattering properties of the cloud particles are defined by the single-scattering albedo, ω , and the phase function, $P(\theta)$. The single-scattering albedo describes how likely a particle is to scatter a photon of light, and it is defined by (e.g. Irwin, 2008):

$$\omega = \frac{\sigma_{sca}}{\sigma_{sca} + \sigma_{abs}} \quad (2.11)$$

where σ_{sca} is the scattering cross-section and σ_{abs} is the absorption cross-section. If $\omega = 0$, the particle only absorbs light and if $\omega = 1$, the particle only scatters light. The phase function, $P(\theta)$ describes the probability that a photon of light will be scattered by an angle θ relative to its original direction of motion. For perfectly spherical particles, the phase function can be calculated using Mie theory. As Jupiter’s cloud particles are primarily made of ice crystals and solid particles rather than liquid droplets, they are likely to be non-spherical, but Mie theory remains a reasonable first guess for an array of randomly orientated non-spheres (e.g. Irwin, 2008). The phase functions calculated from Mie theory do not have simple analytical expressions, so parametrised forms are typically used instead. NEMESIS uses an approximation to Mie theory known as the

double Henyey-Greenstein phase function (Henyey and Greenstein, 1941). As well as speeding up the calculation, the Henyey-Greenstein phase function smooths over the features of purely spherical particles, such as the ‘rainbow’ and the ‘glory’, which makes it particularly suitable to use in the case of solid cloud particles. The double Henyey-Greenstein phase function is defined as:

$$P(\theta) = \frac{1}{4\pi} \left[f \frac{1 - g_1^2}{(1 + g_1^2 - 2g_1 \cos \theta)^{3/2}} + (1 - f) \frac{1 - g_2^2}{(1 + g_2^2 - 2g_2 \cos \theta)^{3/2}} \right] \quad (2.12)$$

where g_1 and g_2 are known as the asymmetry parameters and f is a weighting factor. A fully isotropic scatterer would have $f = 1$ and $g_1 = 0$ while a fully forward-scattering particle would have $f = 1$ and $g_1 = 1$. The presence of two terms in Equation 2.12 is to allow for the possibility of a scatterer with both a forward- and back-scattering peak. However, Chapter 4 shows that Jupiter’s tropospheric clouds can be modelled using a single peak alone i.e. using $f = 1$. This means that the scattering properties of the cloud particles can be reduced to two parameters: the single-scattering albedo, ω , and the asymmetry parameter, g .

When scattering is included in the calculations, Schwarzschild’s equation (Equation 2.1) contains an extra term that describes scattering into and out of the beam. This equation does not have an analytical solution, and must be instead solved numerically (Irwin, 2008). In NEMESIS, the numerical approach used is the matrix-operator method, which is described in Plass et al. (1973). This method assumes a plane-parallel atmosphere, which is applicable for near-nadir viewing geometries (and applies throughout this thesis).

2.5 Retrieval theory

Sections 2.2–2.4 describe the components of the radiative transfer model. This calculation is also known as a ‘forward model’; for a given atmospheric profile (temperature, composition and cloud structure), the radiative transfer code can calculate the synthetic spectrum that would be observed. The ultimate purpose of NEMESIS is to invert this model, in order to retrieve the atmospheric profile from an observed spectrum.

The atmospheric profile can be described by a set of parameters $\mathbf{x}$, and the spectrum can be described by set of measurements $\mathbf{y}$. In the forward model, $\mathbf{x}$ is used to calculate $\mathbf{y}$, which for a linear problem, can be expressed as (e.g. Irwin, 2008):

$$\mathbf{y} = \mathbf{K}(\mathbf{x} - \mathbf{x}_a) + \epsilon \quad (2.13)$$

where $\mathbf{K}$ is the matrix of functional derivatives (which describes the rate of change of radiance with respect to an atmospheric parameter), $\mathbf{x}_a$ is the a priori estimate of $\mathbf{x}$ (i.e. our initial assumptions about the atmospheric parameters) and ϵ is the measurement error.

In reality, observations of the planet are used to measure $\mathbf{y}$, and $\mathbf{x}$ is the desired end product. There are several different options to solve this problem. The best solution is generally not a simple inversion; this can cause a magnification of measurements errors, leading to an unphysical solution. Instead, NEMESIS uses a optimal estimation approach, which gives a balance between fitting the data and maintaining physically realistic profiles that do not deviate too far from the a priori (Rodgers, 2000). For the linear case, the optimal estimation solution would be

$$\mathbf{x} = \mathbf{x}_a + (\mathbf{K}^T \mathbf{S}_\epsilon^{-1} \mathbf{K} + \mathbf{S}_a^{-1})^{-1} \mathbf{K}^T \mathbf{S}_\epsilon^{-1} (\mathbf{y} - \mathbf{K} \mathbf{x}_a) \quad (2.14)$$

where $\mathbf{S}_a$ is the a priori covariance matrix (the error assigned to the a priori assumption) and $\mathbf{S}_\epsilon$ is the measurement covariance matrix (the combination of measurement and modelling error). In reality, however, problems are rarely linear. For the non-linear problem, the forward model can be linearised about a reference state by using a Taylor expansion. This initial reference state, $\mathbf{x}_a$, is used to calculate a new estimate of the solution which is used as the reference state in the next iteration. This process continues until it converges on a final best-fit solution. This is the process used by NEMESIS to retrieve the atmospheric parameters from the observed spectrum. As described in Irwin et al. (2008), NEMESIS minimises the cost function, ϕ , where

$$\phi = (\mathbf{y}_m - \mathbf{y}_n)^T \mathbf{S}_\epsilon^{-1} (\mathbf{y}_m - \mathbf{y}_n) + (\mathbf{x}_n - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x}_n - \mathbf{x}_a) \quad (2.15)$$

where $\mathbf{y}_m$ is the measured spectrum and $\mathbf{y}_n$ is the synthetic spectrum calculated

from the n th solution, $\mathbf{x}_n$. The first term describes how well the spectrum fits the data, and the second term describes how far the solution deviates from the a priori. The cost function is minimised by using the iterative equation

$$\mathbf{x}_{n+1} = \mathbf{x}_a + \mathbf{S}_a \mathbf{K}_n^T (\mathbf{K}_n \mathbf{S}_a \mathbf{K}_n^T + \mathbf{S}_\epsilon)^{-1} (\mathbf{y}_m - \mathbf{y}_n - \mathbf{K}_n (\mathbf{x}_a - \mathbf{x}_n)) \quad (2.16)$$

where $\mathbf{K}_n$ is the weighting function matrix for the n th iteration. Because the differences between successive $\mathbf{K}_n$ can be large, the simple iteration scheme in Equation 2.16 can become unstable. NEMESIS therefore modifies this scheme according to the Marquardt-Levenberg principle (Press et al., 1992), where the modified state vector used in the next iteration, $\mathbf{x}'_{n+1}$, is actually a weighted average of $\mathbf{x}_{n+1}$ and $\mathbf{x}_n$ that tends towards $\mathbf{x}_{n+1}$ as the convergence approaches. For approximately linear inversions, convergence can be achieved in 2–3 steps and for cases where $\mathbf{K}_n$ varies greatly between steps, 10–20 steps could be required (Irwin et al., 2008). The quality of the final fits can be assessed by considering the goodness-of-fit parameter, χ^2/n . This is the weighted sum of squared errors, χ^2 , divided by the number of degrees of freedom, n . χ^2/n is expected to be of order 1. A much larger value suggests the model is underfitting the data, while a much smaller value suggests the model is overfitting the data (e.g. Bradt, 2003).

The error on the retrieved parameters, $\hat{\mathbf{S}}$, is estimated by (Rodgers, 2000):

$$\hat{\mathbf{S}} = (\mathbf{S}_a^{-1} + \mathbf{K}_n^T \mathbf{S}_\epsilon^{-1} \mathbf{K}_n)^{-1} \quad (2.17)$$

which takes into account the measurement error, modelling error, and error on the a priori assumptions. However, this estimate of the error has several limitations. Firstly, the measurement and modelling errors are not always very well known; in particular, the accuracy of the line data is often poorly understood. Secondly, optimal estimation assumes that the posterior errors on the retrieved parameters are Gaussian, which is not always true. However, Line et al. (2013) has shown that this is a valid assumption when the data is of relatively high spectral resolution (as is the case in this thesis). Finally, it assumes that the model is parametrised correctly, and contains all the relevant variables. For example, a gas may be assumed to be well-mixed and therefore allowed to

vary via a single abundance; this error calculation does not take into account errors due to ‘incorrect’ parametrisation. Similarly, the calculation cannot take into account any missing gaseous species or cloud decks. The formal retrieved errors should therefore be viewed with some caution, as they are likely to provide an underestimate of the true uncertainty in the retrievals. Where possible in this thesis, different assumptions about gaseous abundances and cloud structures are tested, in order to provide a deeper understanding of the uncertainties involved.

2.6 Reference atmosphere

For the analysis of Jupiter’s 5- μm spectra, the jovian atmosphere was divided into 39 levels between 30 bar and 50 mbar, equally spaced in $\log(p)$. 39 levels is the maximum allowed by NEMESIS in a scattering calculation, and the base pressure of 30 bar was chosen to be well below where the weighting functions peak at 5- μm . The temperature and volume mixing ratio of each gas species is defined at each pressure level.

The temperature profile is taken from Cassini CIRS observations (Fletcher et al., 2009). The CIRS temperature profile is sensitive down to a pressure of 800 mbar, and has been extrapolated below this using a dry adiabat, which is consistent with the temperature profile found by the Galileo probe (Seiff et al., 1998) (see Figure 1.2.2). This temperature profile varies with latitude according to the variations in the dry adiabatic lapse rate, and is held fixed during the retrievals. The Galileo entry probe entered a ‘hot-spot’ in Jupiter’s North Equatorial Belt, and is therefore not necessarily representative of the bulk atmosphere. However, the difference between a dry and a moist adiabat is only ~ 2 K in Jupiter’s troposphere (Lewis, 1995) and this difference will have a negligible impact on the observed spectrum compared to the cloud cover (see Section 4.3 for more details).

The a priori NH_3 and PH_3 profiles are taken from Fletcher et al. (2009). NH_3 has a deep volume mixing ratio (VMR) of 1.862×10^{-4} up until 0.9 bar, and then it drops off with a fractional scale height of 0.1 relative to the atmospheric scale height. PH_3 has a deep VMR of 1.86 ppm, which drops off with a fractional scale height of 0.4 above 1 bar. The CH_4 a priori abundance is 1.81×10^{-3} and was obtained from Niemann et al. (1998). The CH_3D profile assumes a constant ratio of $\text{CH}_3\text{D}/\text{CH}_4$ of 8×10^{-5} (Lellouch

et al., 2001). The profile of H_2O is poorly constrained. For this study, a deep volume mixing ratio, fixed at 1×10^{-3} ($\sim$ the solar abundance) is assumed, with a constant relative humidity at higher altitudes. For the reference profile, this relative humidity is set to 10% (order of magnitude based on Irwin et al. (1998)). The remaining minor atmospheric species, CO , GeH_4 and AsH_3 were assumed to be well mixed in the troposphere, with volume mixing ratios of 1.0 ppb, 0.45 ppb and 0.24 ppb respectively (Bézard et al., 2002). Unless otherwise stated, the abundances of each of these gases are allowed to vary via a single scaling parameter.

In a fully variable vertical profile, the solution must be constrained to lie close to the a priori value in order to prevent ill-conditioning (Irwin et al., 2008). However, in the analysis in this thesis, the retrieved quantities are parametrised so the retrieved profiles are always smooth. This means that the a priori errors can be set to a large value, and Equation 2.15 becomes a simple least-squares fit to the data (Irwin et al., 2008)

2.7 Summary

This chapter provided an introduction to the NEMESIS retrieval software. This software is made up of a radiative transfer code that models the top-of-atmosphere radiation and a retrieval algorithm that iteratively converges on a best-fit solution to the observed data. This forms the basis of the analysis of the Cassini VIMS and VLT CRRES observations. Chapter 3 explores these two instruments in depth, and NEMESIS is used to analyse the observations in Chapters 4–6.

Chapter 3

Data

3.1 Introduction

In this thesis, observations from two different instruments are studied: the VIMS instrument on the Cassini spacecraft and the CRIRES instrument at the Very Large Telescope. VIMS provides relatively low spectral resolution observations, which simultaneously cover the entire 5- μm spectral range. These observations provide global coverage of both the nightside and the dayside of the planet. In contrast, CRIRES has very high spectral resolution and provides latitudinally resolved spectra of the planet's dayside. This chapter describes the instruments, data acquisition process, data reduction and calibration for each of the datasets. Cassini VIMS is described in Section 3.2 and VLT CRIRES is described in Section 3.3.

3.2 Cassini VIMS

3.2.1 The Cassini mission

Cassini-Huygens is a joint NASA/ESA/ASI spacecraft with the primary objective of exploring the Saturn system. It is a two-part mission, comprising the Cassini orbiter (equipped with 12 science instruments) and the Huygens entry probe. It was launched in 1997, reached Saturn in 2004, and has been studying the planet and its moons for the last 12 years. To aid the journey to its final target, the spacecraft performed four gravity-assist flybys: Venus (1998 & 1999), Earth (1999) and Jupiter (2000–2001). The Cassini flight trajectory is shown in Figure 3.2.1. Although the primary purpose of the

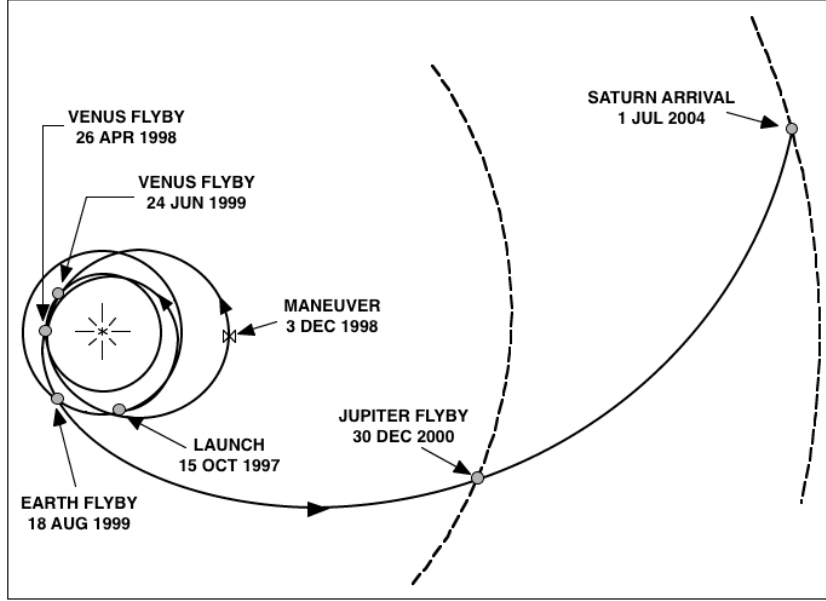


Figure 3.2.1: The Cassini-Huygens flight trajectory, from launch in 1997 to arrival in 2004. The Jupiter flyby took place between October 2000 and March 2001. Credit: NASA/JPL

flybys was to gain a boost from each planet's gravitational field, they also offered the opportunity to make scientific measurements.

Cassini's closest approach to Jupiter was on December 30, 2000 when it reached a distance of 10 million kilometres from the planet. Between October 2000 and March 2001, tens of thousands of images of the planet were taken, which have been used for both scientific and calibration purposes. Discoveries made using instruments on-board Cassini include large numbers of individual storm cells seen in the belts (Porco et al., 2003), the first detection of CH_3 and C_4H_2 in the jovian stratosphere (Kunde et al., 2004), and measurements of Jupiter's magnetosphere showing the response to changes in solar-wind pressure (Kurth et al., 2002). The first science results from the VIMS instrument were reported in (Brown et al., 2003), and included measurements of H_3^+ emission at $3.5 \mu\text{m}$ and NH_3 absorption at $0.93 \mu\text{m}$. The $1.0\text{--}3.2 \mu\text{m}$ segment of the observations was subsequently analysed by Sromovsky and Fry (2010), but the the $5\text{-}\mu\text{m}$ observations of Jupiter have not been previously studied.

3.2.2 The VIMS instrument

The Visual and Infrared Mapping Spectrometer (VIMS) is one of the 12 instruments on-board the Cassini spacecraft and is described in detail in Brown et al. (2004). It is

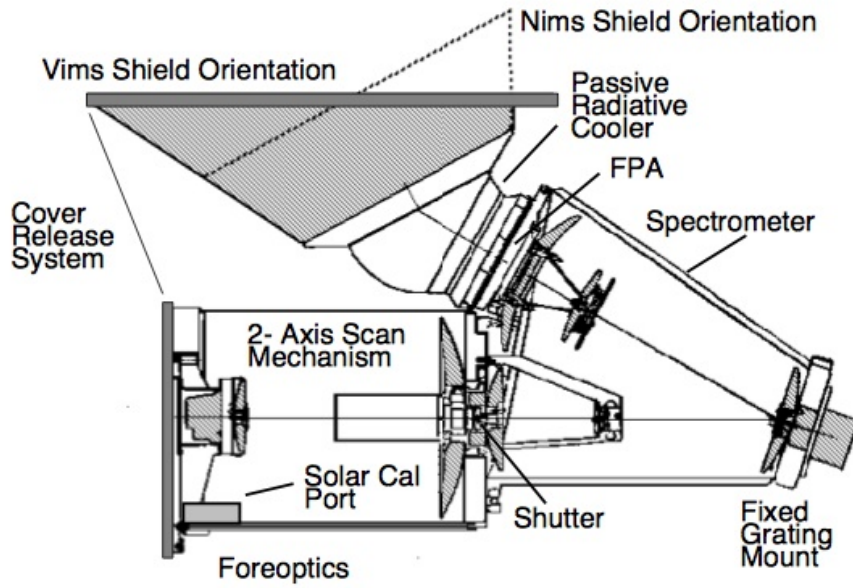


Figure 3.2.2: The VIMS-IR optical design (Miller et al., 1996).

Wavelength range	0.85–5.1 μm
Number of channels	256
Spectral resolution	18.7 nm (at 5- μm)
Spectral sampling	16.6 nm/spectral pixel
Instantaneous field of view	0.25×0.5 mrad
Effective instantaneous field of view	0.5×0.5 mrad
Total field of view	32×32 mrad
Detector science array	1×256 array of InSb detectors

Table 3.2.1: VIMS-IR specifications (Brown et al., 2004).

an imaging spectrometer with one channel in the visible (VIMS-V; 0.35– 1.07 μm) and one channel in the infrared (VIMS-IR; 0.85–5.1 μm). The work presented here uses data from VIMS-IR.

The VIMS-IR optical design is shown in Figure 3.2.2 and its specifications are summarised in Table 3.2.1. The foreoptics consists of a Ritchey-Chrétien telescope with a diameter of 23 cm, where the secondary mirror has a two-axis scan mechanism. The effective instantaneous field of view is 0.50×0.50 mrad, and this is stepped across the scene in orthogonal directions to produce a 64×64 pixel image. The total field of view is 32×32 mrad, which is several times larger than the size of Jupiter’s disk at closest approach (Jupiter had a diameter of 15 mrad at a distance of 9.7 million km).

After reflecting from the secondary mirror, light is collimated using a Dall-Kirkham collimator (Kirkham, 1938) and then hits a triple-blaze grating spectrometer. The

spectrum is then imaged onto a 1×256 array of InSb detectors; these 256 wavelength channels cover the range 0.85–5.1 μm at a spectral sampling of 16.6 nm/spectral pixel. The spectral resolution varies slightly across the wavelength range and has an average value of 18.7 nm in the 5- μm window (K. Baines, personal communication).

An initial spectral calibration was performed during ground-testing, using materials with well-defined spectral features. Post-flight spectral calibration made use of stars and other objects and showed a consistent 21 nm (1.3 channel) shift across the entire wavelength range. This change limits the accuracy of the wavelength calibration to about 5 nm (0.3 channel). Radiometric calibration also consisted of both pre- and post-launch tests. The ground calibration used a glow bar and a tungsten lamp as sources, while the in-flight calibration used observations of Jupiter, the moon, and several stars. In-flight updates to both the spectral and radiometric calibrations are described in McCord et al. (2004).

3.2.3 Observations and data reduction

VIMS data were accessed through the NASA Planetary Image Atlas¹. The data are in the form of ‘data cubes’ with two spatial dimensions and one spectral dimension. VIMS observations were made both on the dayside of the planet as the spacecraft approached in late 2000 and on the planet’s nightside as the spacecraft moved away from Jupiter in early 2001. Figure 3.2.3 shows the phase angle vs. distance trajectory of the spacecraft during December 2000 and January 2001, generated using the JPL HORIZONS on-line ephemeris system². The overplotted crosses show the locations of all of the VIMS observations that were made using the normal sampling mode that is supported by ISIS3³ (Integrated Software for Imagers and Spectrometers), the software package provided by the United States Geological Survey that is used to reduce the data.

When selecting a subset of these data cubes to study, several factors are important. Nightside observations simply show the thermal emission from the planet, whereas the dayside observations also include a component from reflected sunlight, which further complicates the analysis. The initial study therefore focussed on nightside cubes only,

¹pds-imaging.jpl.nasa.gov

²ssd.jpl.nasa.gov/horizons.cgi

³isis.astrogeology.usgs.gov

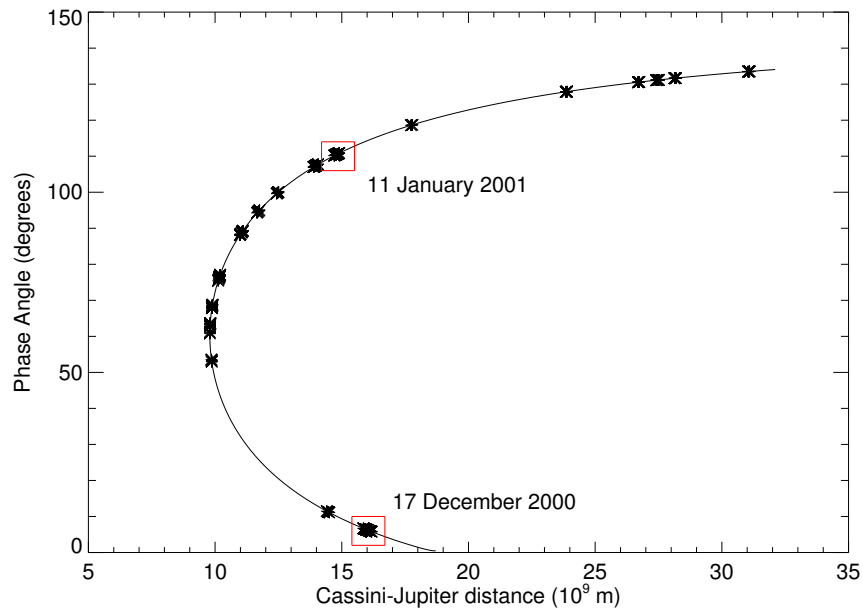


Figure 3.2.3: The trajectory taken by the Cassini spacecraft, with crosses marking the point of all VIMS observations. The selected observations are marked by a red box. Nightside cubes were taken from 11 January 2001 and dayside cubes were taken from 17 December 2000.

with a minimum phase angle of 100° to ensure that the centre of the disk would be far enough from the day-night boundary to avoid contamination from reflected sunlight. This requirement was balanced against the desire to use measurements from as near to the point of closest approach as possible, in order to maximise the spatial resolution. The compromise settled on was a subset of 30 data cubes from a 7 hour period on 11 January 2001. The use of multiple cubes vastly increases the number of available data points, while the fairly limited time period means that atmospheric conditions are unlikely to have changed. For the subsequent dayside analysis, a set of data cubes from 17 December 2000 was used, when the spacecraft was approaching the planet. These observations were made at a similar distance from the planet as the 11 January ones, giving a comparable spatial resolution of ~ 8000 km.

The selected VIMS cubes were processed using the ISIS3 software. This software includes the radiometric and spectral calibration provided by the VIMS Science Team (McCord et al., 2004). The final result is a calibrated data cube of dimensions $64 \times 64 \times 256$ (corresponding to the two spatial and one spectral dimensions).

In addition, ISIS3 makes use of SPICE data kernels in order to provide geometric information about the observations. SPICE is an information system used across a

range of space missions, which provides navigation and other ancillary information in a standardised format (Acton, 1996). ISIS3 takes this information and uses it to produce a backplane data cube, with dimensions $64 \times 64 \times 13$ (corresponding to the two spatial and one geometric dimensions). The 13 pieces of geometric information contained in the backplane cube include the latitude, longitude, phase angle and emission angle.

3.3 VLT CRIRES

3.3.1 The CRIRES instrument

The Very Large Telescope (VLT) is a telescope located in the Atacama Desert in Chile, and run by the European Southern Observatory (ESO). The four 8.2-metre individual telescopes that make up the VLT are primarily used separately, but can also be used together in order to increase the spatial resolution even further.

CRIRES is a cryogenic high-resolution infrared echelle spectrograph (Käufl et al., 2004), located on Antu, one of the four VLT telescopes. The instrument provides long-slit spectroscopy across the wavelength range 0.95–5.38 μm , with a resolving power of 96,000. Since commissioning in 2006, CRIRES has been used to study a wide range of subjects, including stars (Ryde et al., 2009), exoplanets (Bean et al., 2010), molecular gas (Pontoppidan et al., 2011) and comets (Villanueva et al., 2009).

The optical design of CRIRES is shown in Figure 3.3.1 and its specifications are summarised in Table 3.3.1. The light from the telescope initially meets the de-rotator and the adaptive optics system. The de-rotator acts to minimise the effect of telescope field rotation caused by the altazimuth telescope mount, and the adaptive optics system can be used to correct for atmospheric turbulence, but was not used for these observations. The pre-disperser limits the light passing through to a single echelle order, which is then collimated by the three-mirror anastigmat. The grating itself is a 40×20 cm, 21.6 lines/mm, 63.5° blaze echelle. The three-mirror anastigmat also acts as the camera, imaging the spectrum onto the detector mosaic, which consists of four InSb Aladdin arrays. The cryogenic optical bench, shown in grey in Figure 3.3.1, is housed in a vacuum and kept at ~ 65 K, and the detectors are cooled to ~ 25 K. These low temperatures are required to limit the detector noise.

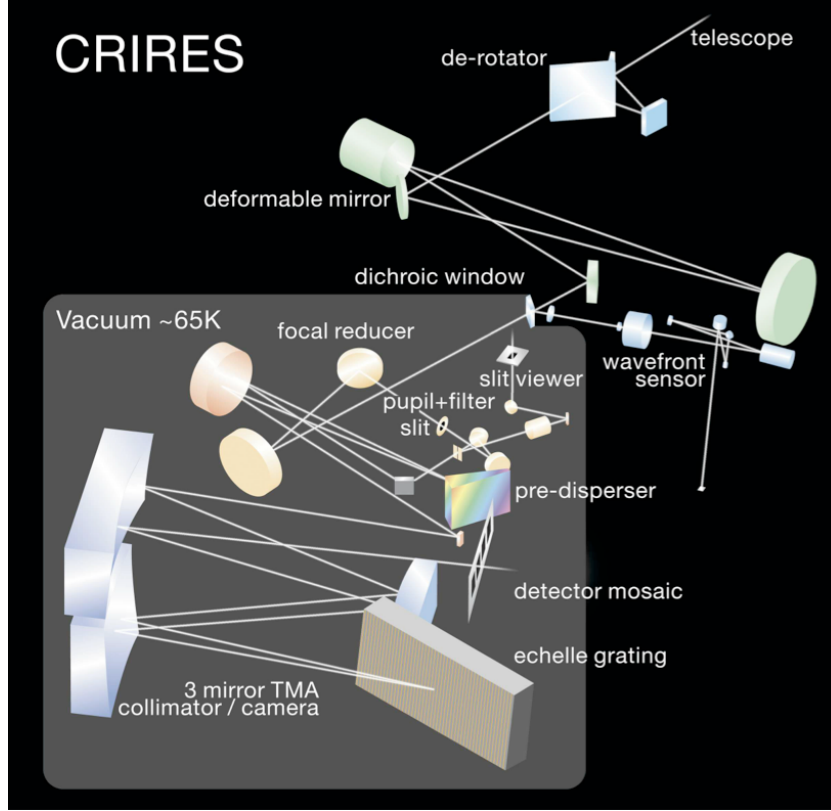


Figure 3.3.1: The CRIRES optical design (Käuffl et al., 2004).

The slit length is 40", and the slit width can be adjusted to 0.2"–1" (a width of 0.2" was used for these observations, which is equivalent to 600 km). The detector has a size of 4096×512 pixels: 512 pixels in the spatial direction, along the length of the slit, and 4096 pixels in the spectral direction. Because the detector is made up of four separate arrays with small gaps between them, the spectral coverage in a single observation is split into four segments (1024 pixels each), with small wavelength gaps ($\sim 0.007 \mu\text{m}$) between each segment.

Further details about the design, calibration and operation of the CRIRES instrument can be found in Käuffl et al. (2004).

3.3.2 Observations

Observations of Jupiter were made using VLT CRIRES on 12 November 2012 (05:00–05:40 UT) and 1 January 2013 (02:50–03:30 UT). The atmospheric conditions were better on 12 November 2012, so the majority of this thesis focuses on those observations. However, the observations from 1 January 2013 are used in Chapters 5 and 6 to show that the same conclusions can be drawn from both datasets.

Wavelength range	0.95–5.38 μm
Resolving power (2 pixels)	96,000
Slit width	0.2''–1''
Slit length	40''
Pixel scale	0.1''
Adaptive optics	MACAO system (60 actuator deformable mirror)
Calibration system	2 blackbodies, 2 spectral lamps, gas cells
Pre-disperser	ZnSe prism
Echelle grating	40 $\times$ 20 cm, 31.6 lines/mm, 63.5° blaze
Detector science array	4096 $\times$ 512 pixels using 4 InSb Aladdin detectors

Table 3.3.1: CRIRES specifications (Käufl et al., 2004).

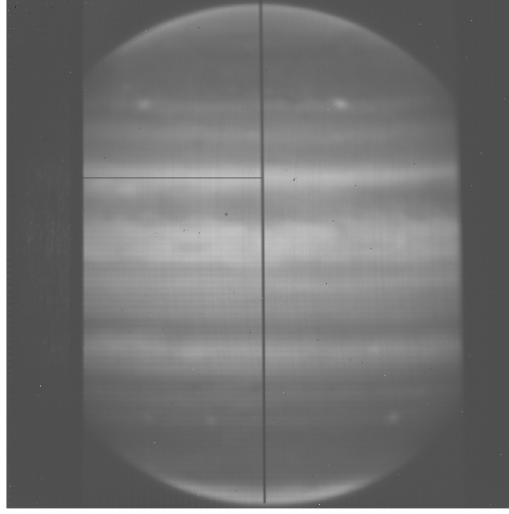


Figure 3.3.2: Acquisition image of Jupiter made at 05:02 UT on 12 November 2012, immediately before the 5- μm observations. This image was made using a Ks-band filter (2.2 μm). The vertical black line shows the alignment of the narrow slit.

On both dates, the slit was aligned north-south on Jupiter, along the planet’s central meridian, allowing us to measure spatial variability with latitude, but not longitude. Figure 3.3.2 shows the acquisition image of Jupiter made on 12 November, using a Ks-band filter (2.2 μm). The vertical black line shows how the narrow slit was aligned. The slit length (40'') was smaller than the angular diameter of Jupiter on the observation dates (47–48''), so the observations cut off the southern polar region.

The Doppler shift on 12 November was -10.25 km s^{-1} , corresponding to a wavelength shift of $1.7 \times 10^{-4} \mu\text{m}$ at 5 μm . The Doppler shift on 1 January was 15.75 km s^{-1} , corresponding to a wavelength shift of $2.6 \times 10^{-4} \mu\text{m}$ at 5 μm .

On each date, observations were made in 14 different wavelength settings. Each wavelength setting has a width of $\sim 0.12 \mu\text{m}$ and is centred on a slightly different wave-

length. Together, these observations cover the entire wavelength range 4.55–5.26 μm . The time lag between the first and last observation was ~ 40 minutes; during this time period, Jupiter rotated $\sim 25^\circ$, so the 14 different observations of different spectral regions do not correspond to precisely the same locations on the planet. The CRRES wavelength settings are listed in the Appendix of this thesis.

In addition to Jupiter, a standard star of known irradiance was observed at each wavelength setting in order to provide radiometric calibration. For the 12 November observations, this star was Pi-2 Orinis (HIP 22509), a spectrophotometric standard from Hamuy et al. (1994). For the 1 January observations, the star used was Zeta Tauri (HIP 26451), a single-lined spectroscopic binary system.

Observations of both Jupiter and the standard star were made in nodding mode. For Jupiter, this consisted of nodding between the planet and the sky. For the star, this consisted of nodding between two different star positions on the slit. In addition, dark images and flatfields were taken.

3.3.3 Data reduction

Reduction using EsoRex

The initial part of the data reduction was performed using EsoRex, the ESO Recipe Execution Tool (Ballester et al., 2006). EsoRex is a command line utility which can be used to process observations made using various instruments at the VLT.

The flatfields, darks, and nodded pairs of observations were used to produce a single combined observation of Jupiter for each of the 14 wavelength segments. The data from each segment has dimensions 4096×512 . The same process was applied to the observations of the standard star. In both cases, spectral calibration was achieved by comparing telluric absorption lines with the HITRAN line database (Rothman et al., 2009). This comparison is performed across a range of spatial positions, and leads to the production of a wavelength calibration map, with the same 4096×512 dimensions as the data.

EsoRex was also used to calculate the background noise on both the stellar and the planetary observations. When these observations were combined (see below), the fractional errors were added in quadrature.

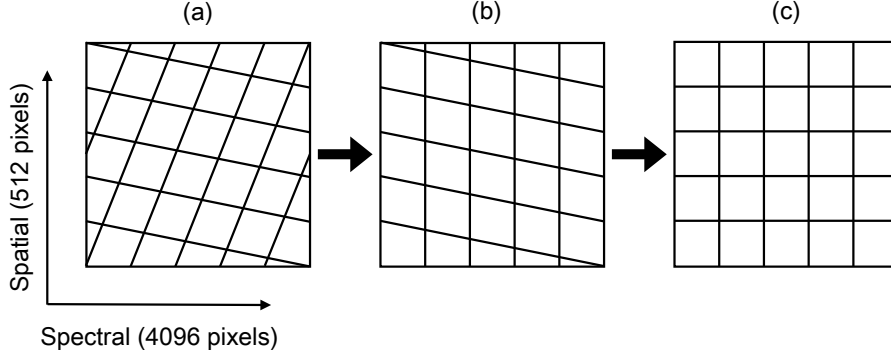


Figure 3.3.3: Correction of the spatial and spectral shears in the VLT CRRES data. (a) The original data. (b) Corrected for the spectral shear. (c) Corrected for the spatial shear.

Subsequent data reduction was performed independently of the EsoRex software.

Straightening

If the data were perfectly straight along the spectral dimension (the x-direction on the detector), then the HITRAN-generated wavelength map would also be perfectly straight; the wavelength scale at $y = 0$ would be identical to the wavelength scale at $y = 511$. However, there is an offset observed between the wavelength scale at different spatial points, giving a ‘shear’ in the spectral dimension. In order to correct this, the wavelength calibration map was used to interpolate the data onto a ‘straight’ grid. The effect of making this correction is shown in Figure 3.3.3b.

There is a similar ‘shear’ in the spatial direction. For each wavelength point, a north-south latitudinal radiance profile can be extracted. These latitudinal profiles show maxima in the warm belts and minima in the cool zones, and these maxima and minima should be located at the same position for all wavelengths. However, an offset was once again observed; a given y-position could correspond to a belt at $x = 0$ but correspond to a zone at $x = 4095$, introducing an artificial slope into the spectrum. The extent of this shear was determined using cross-correlation, and was corrected by interpolating onto a straight grid. The effect of making this correction is shown in Figure 3.3.3c. This ensures that the belts and zones occur at the same y-locations for all wavelengths.

These shear corrections were applied to both the planetary observations and the observations of the standard star.

Radiometric calibration

Observations of the standard star were used to divide out the telluric lines from the jovian spectrum and to convert Jupiter’s photon count into spectral radiance. In the case of Pi-2 Orionis, a standard spectrum was provided in the EsoRex software. This was not available for Zeta Tauri, so the irradiance was estimated by scaling a black-body curve to match the observed magnitudes. The shape of the black-body curve was defined by the star’s effective effective temperature, based on its spectral type (B2, 21000 K)⁴. This was then scaled to match the magnitudes obtained from the 2MASS all-sky catalogue of point sources for three different filters: J (1.24 μm), H (1.66 μm) and K (2.16 μm). For Zeta Tauri, these magnitudes are 3.001, 3.047 and 2.808 respectively (Skrutskie et al., 2006).

Using the known stellar irradiance, Equation 3.1 was then used to convert Jupiter’s photon count into spectral radiance:

$$L_J(\lambda) = \frac{N_J(\lambda)}{N_*(\lambda)} \frac{F_*(\lambda)}{\Omega} \quad (3.1)$$

$L_J(\lambda)$ is the jovian spectral radiance, $N_J(\lambda)$ is the jovian photon count for a single pixel, $N_*(\lambda)$ is the total stellar photon count (across several pixels), $F_*(\lambda)$ is the stellar spectral irradiance (provided by EsoRex) and Ω is the solid angle subtended by a single pixel at the detector.

For this calibration to be accurate, $N_*(\lambda)$, the stellar photon count, must include all of the incident photons for the star. In the direction along the slit, this is straightforward; a Gaussian profile is fit to the observed photon count, and the area underneath is summed. However, flux is also being lost due to the narrow width of the slit. To account for this, the profile is assumed to also be Gaussian in the direction perpendicular to the slit, with the same proportions. Assuming the star is centrally placed in the slit, the ‘missing’ flux can then be calculated.

Once this calibration has been applied, the 5- μm brightness temperatures varies between ~ 190 K in the coolest parts of the planet and ~ 260 K in the brightest regions. This is roughly consistent with the Cassini VIMS 5- μm observations, which showed a 180–240 K range. An offset in the location of the star would decrease the calculated

⁴<http://www.uni.edu/morgans/astro/course/Notes/section2/spectraltemps.html>

CRIRES brightness and could account for the difference between the CRIRES and the VIMS measurements. A systematic error in the radiometric calibration could increase or decrease the average radiance, but will not alter the shape of the spectra. Chapter 4 shows that the opacity of the upper cloud has the same effect on the spectrum: scaling it up or down, without altering the shape. Any errors in the radiometric calibration will slightly alter the retrieved opacity of the upper cloud, but will not affect the remainder of the analysis in Chapters 5–7.

For reference, the entirety of the calibrated CRIRES spectrum is shown in the Appendix, alongside a plot of the telluric transmission. Although the division by a standard star partially removed the telluric lines, some residuals remain and the regions of high telluric absorption have large error bars. In the retrievals in Chapters 5 and 6, small spectral regions with large error bars are removed.

Assigning geometries

Geometric data was assigned using information from JPL HORIZONS. Planetocentric latitudes were assigned to each pixel using information about the angular size of the planet, the angular size of each pixel and the location of the planet’s limb; the slit is assumed to be aligned with the central meridian, and this calculation takes into account the planet’s oblateness. Information obtained from JPL HORIZONS was used in order to calculate phase angles, emission angles, solar zenith angles and azimuthal angles for each pixel.

Due to Jupiter’s rotation, each of the 14 wavelength segments observed a slightly different longitude, with a $\sim 25^\circ$ difference between the first and last segment. Because Jupiter is longitudinally inhomogeneous, this means that each wavelength segment is observing a different cloud opacity; this prevents the different segments from being joined together into a single continuous observation.

3.4 Summary

In this chapter, the two data sources used in this thesis were introduced. The first data source is Cassini VIMS. These observations provide global coverage of Jupiter, and cover the entire 5- μm spectral range. This makes this dataset ideal for studying

broadband spectral features, such as clouds. The Cassini VIMS data will be analysed in Chapter 4. VIMS is not sufficiently high spectral resolution to break the degeneracies between molecular species. In order to do this, the second data source, VLT CRILES, is required. CRILES provides very high spectral resolution observations, allowing individual molecular absorption lines to be resolved. The CRILES observations will be analysed in Chapters 5-7.

Chapter 4

Tropospheric clouds: part 1

4.1 Introduction

In this chapter, 5- μm VIMS spectra with a range of phase and emission angles are used to investigate Jupiter's tropospheric cloud structure, and conclusions are drawn about the cloud locations and scattering parameters. In addition, this chapter explores the extent to which these observations can be used to constrain the tropospheric gaseous composition.

The VIMS observations are particularly suited to studies of Jupiter's cloud structure. The instrument provides simultaneous observations across a relatively broad wavelength range (4.5–5.1 μm), allowing the spectral effects of clouds to be disentangled from gaseous species. The VIMS data also provide full global coverage with a spatial resolution of ~ 8000 km, which allows us to compare and contrast the 5- μm spectra from the belts and the zones. Additionally, VIMS made observations on Jupiter's nightside and dayside within the space of a few weeks, giving us the opportunity to study the effect of reflected sunlight.

Previous studies of Jupiter's tropospheric cloud structure consist of both theoretical models and observational work. As described in Chapter 1, a typical theoretical cloud condensation model from Atreya et al. (1999) produces three main cloud levels: NH_3 ice, solid NH_4SH and H_2O ice. If a three-times-solar enrichment is assumed, the bases of these cloud levels are located at 0.8 bar, 2.6 bar and 7.2 bar respectively. Although models such as this provide valuable insights into the approximate condensation levels for each species, the clouds are not necessarily expected to form at these precise loca-

tions. The models make assumptions about the bulk composition of the atmosphere and typically do not include the presence of photochemical products. They also do not account for the complex dynamics of the troposphere, neglecting the vertical mixing which brings up material from below and rains down material from above. All of these factors will act to significantly alter the vertical cloud structure away from the expectations of simple equilibrium models. Observations are therefore fundamental to understanding Jupiter’s tropospheric cloud decks. In the following paragraphs, the conclusions that have been drawn from previous observational studies are briefly summarised.

There are many observational studies supporting the existence of the uppermost NH_3 -ice cloud deck. 5- and 45- μm observations from Voyager IRIS were analysed by Gierasch et al. (1986), who concluded that there was a highly variable cloud deck at ~ 700 mbar, near the expected ammonia condensation level. Imaging data from the Galileo spacecraft showed evidence for thick, variable clouds in the 750 ± 200 mbar region (Banfield et al., 1998), again consistent with NH_3 ice. Further support came in the form of spectroscopic identification of ammonia-ice features, using the 1–3 μm region of the Galileo NIMS spectra (Baines et al., 2002) and 9 μm data from Cassini CIRS (Wong et al., 2004). Baines et al. (2002) found that their ammonia-ice spectroscopic signatures were present in less than 1% of Jupiter’s clouds, and it has been suggested that this may be due to a hydrocarbon haze coating the ammonia-ice particles and masking the absorption features (Kalogerakis et al., 2008).

There are additional studies which support the existence of both an upper NH_3 cloud and a lower cloud formed of NH_4SH . Matcheva et al. (2005) found clouds in the 900–1100 mbar region from Cassini CIRS observations at 7.18 μm , which they concluded were probably composed of an upper layer of NH_3 and a lower layer of NH_4SH . Sromovsky and Fry (2010) analysed 3- μm data from Cassini VIMS, and found that a 500 mbar cloud composed of both NH_3 and NH_4SH provided the best fit. 500 mbar is well above the expected condensation point for NH_4SH , but they suggested that this could be explained by rapid upwelling. Further evidence for a NH_4SH cloud comes from analyses of the 5- μm Galileo NIMS data. Irwin et al. (2001) performed retrievals on the NIMS data and determined that the cloud that provides the majority of the 5- μm opacity is located at around 1.5 bar. This was supported by Irwin and Dyudina (2002), who

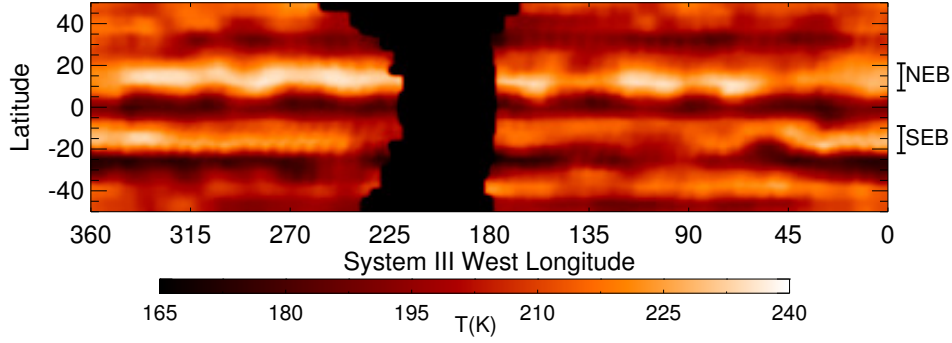
performed a principal component analysis on the same data and once again found that the dominant opacity variations occur in the 1–2 bar region. Based on the location, both studies concluded that the dominant cloud was likely to be composed of ammonium hydrosulphide.

Because of its predicted location deep in the lower troposphere, the water-ice cloud has been difficult to observe. Banfield et al. (1998) used observations from the Galileo Solid State Imaging experiment to identify a deep cloud (located at a pressure greater than 4 bar) in one region close to the Great Red Spot, which they concluded was likely to be composed of water. Similarly, Simon-Miller et al. (2000) found evidence for a water cloud in very small regions of the planet by identifying a water-ice feature near $44\text{ }\mu\text{m}$ in Voyager IRIS spectra. They suggested that this limited detection may be due to the fact that the water cloud is only visible in small regions where the rest of the atmosphere is particularly cloud-free. More recently, Bjoraker et al. (2015) compared the line shapes of CH_3D absorption features between belts and zones and concluded that the variable line width could be explained by the presence of an optically thick water cloud in the zones.

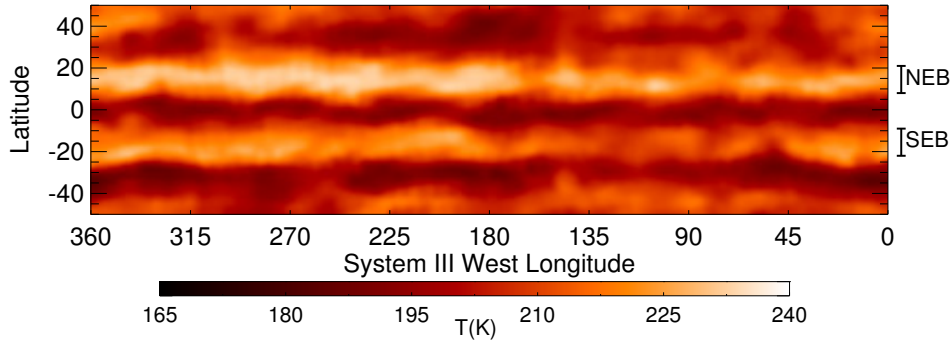
This chapter analyses the $5\text{-}\mu\text{m}$ observations of Jupiter made by the VIMS instrument. Section 4.2 introduces the observations, Section 4.3 explores the sensitivity of these observations to the different atmospheric parameters, and Section 4.4 introduces the basic retrieval model for these observations. Having established that the VIMS data are primarily sensitive to the cloud structure, Section 4.5 explores the constraints that the nightside observations place on the cloud parameters. This information is then used for a global retrieval in Section 4.6, which also analyses the spatial variability of PH_3 and H_2O . Section 4.7 then compares the results from the nightside and the dayside of the planet. Finally, the results from this chapter are summarised in Section 4.8. This chapter is taken from Giles et al. (2015).

4.2 Observations

As described in Section 3.2.3, two sets of data cubes were selected for analysis: nightside observations from 11 January 2001, which only show thermal emission from Jupiter itself, and dayside observations from 17 December 2000, which also have a component of



(a) Nightside data from 11 January 2001.



(b) Dayside data from 17 December 2000.

Figure 4.2.1: Brightness temperature maps from 5- μ m VIMS data on two dates, giving both nightside and dayside observations of Jupiter. Smoothing has been applied to these maps. Black regions correspond to segments where no data was obtained. The GRS is located at $\sim 50^\circ$ W and did not move significantly between the two datasets.

reflected sunlight. Sections 4.4, 4.5 and 4.6 focus on the nightside data, and Section 4.7 compares these results to the dayside observations.

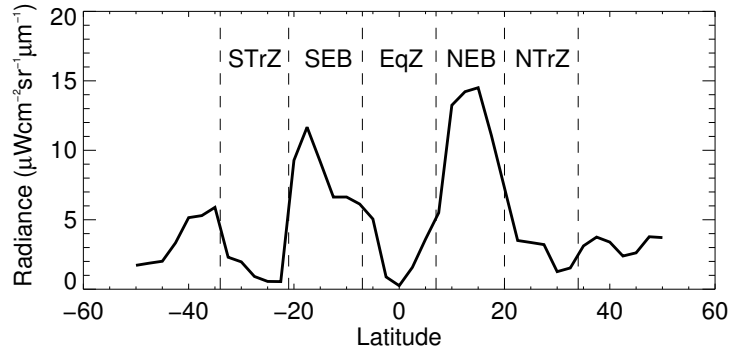
Cylindrical projections of both the nightside and the dayside datasets are shown in Figure 4.2.1. In both panels, the difference between the warm belts and the cooler zones can be seen. In the nightside observations, there is a ~ 70 K brightness temperature difference between the warmest and the coolest regions of the planet. For the dayside observations, this contrast is reduced to ~ 50 K; the cloudy zones reflect more light than the relatively cloud-free belts, meaning that the presence of sunlight has a greater impact on the cool zones than on the warm belts. Zonal mean radiances were then extracted from both the nightside and the dayside datasets. The spectra were divided into forty-one latitudinal bins covering the range -50° to $+50^\circ$ at 2.5° intervals, each with a 5° width. For each latitudinal bin, all spectra with an emission angle within 10° of the minimum value were averaged to produce one final nadir spectrum for each latitude segment.

Following Fletcher et al. (2011), the final errors assigned to these averaged VIMS spectra were 12% of the mean radiance in the 4.5–5.2 μm range. This is a conservative value, including both quadrature-estimated errors due to pre-flight calibration and forward-model uncertainties on the spectral line data. The constant error margin throughout the window prevents retrievals being weighted towards low-radiance regions. The size of the VIMS errors is further discussed in Section 4.6.

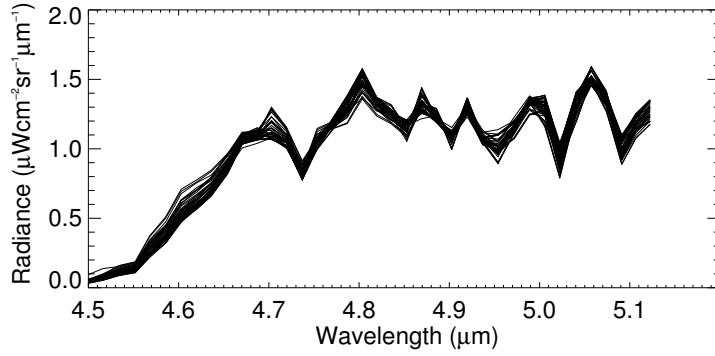
The results of this process for the nightside data can be seen in Figure 4.2.2. Figure 4.2.2a shows the absolute radiances at 5.0 μm as a function of latitude. This figure also labels the spatial regions of the planet: the central zone is the Equatorial Zone (EqZ), the adjacent belts are the North Equatorial Belt (NEB) and the South Equatorial Belt (SEB), and these belts are then flanked by the North Tropical Zone (NTrZ) and the South Tropical Zone (STrZ). Figure 4.2.2a shows that there is a factor of 50 difference between the hottest spectra from the NEB and coldest spectra from the EqZ. Figure 4.2.2b shows the forty-one spectra from the different latitudes, with the average radiance normalised to 1.0 to allow a comparison of spectral shapes. Despite the large difference in absolute radiance between different parts of the planet, Figure 4.2.2b shows that the shape of the spectra remains almost identical throughout. This cannot be explained by variations in the chemical composition or temperature profile of the atmosphere. The only parameter capable of reproducing the huge variability in the radiance while retaining the same spectral shape is the cloud optical thickness, as is shown quantitatively in the following sections.

4.3 Sensitivity analysis

The spectral resolution of the VIMS data is 18.7 nm, which is not high enough to resolve the individual molecular lines of the tropospheric species. This means that there are likely to be degeneracies in the retrievals, where two or more parameters have a similar effect on the spectrum and are therefore indistinguishable. In order to carefully assess this, the radiative transfer model described in Chapter 2 was used to perform a sensitivity analysis, where each parameter was individually altered in order to observe its effect on the spectrum. Initially, a reference synthetic spectrum was produced using the a priori atmospheric profile described in Section 2.6. This a priori



(a) 5.0 μm radiance as a function of latitude.



(b) The normalised spectrum for each latitudinal bin.

Figure 4.2.2: VIMS nightside spectra, divided into 5° wide latitudinal bins and zonally averaged. The upper panel shows how the absolute radiance at 5.0 μm varies with latitude. The lower panel shows the full spectrum for each latitudinal bin, normalised to allow a comparison of their shapes.

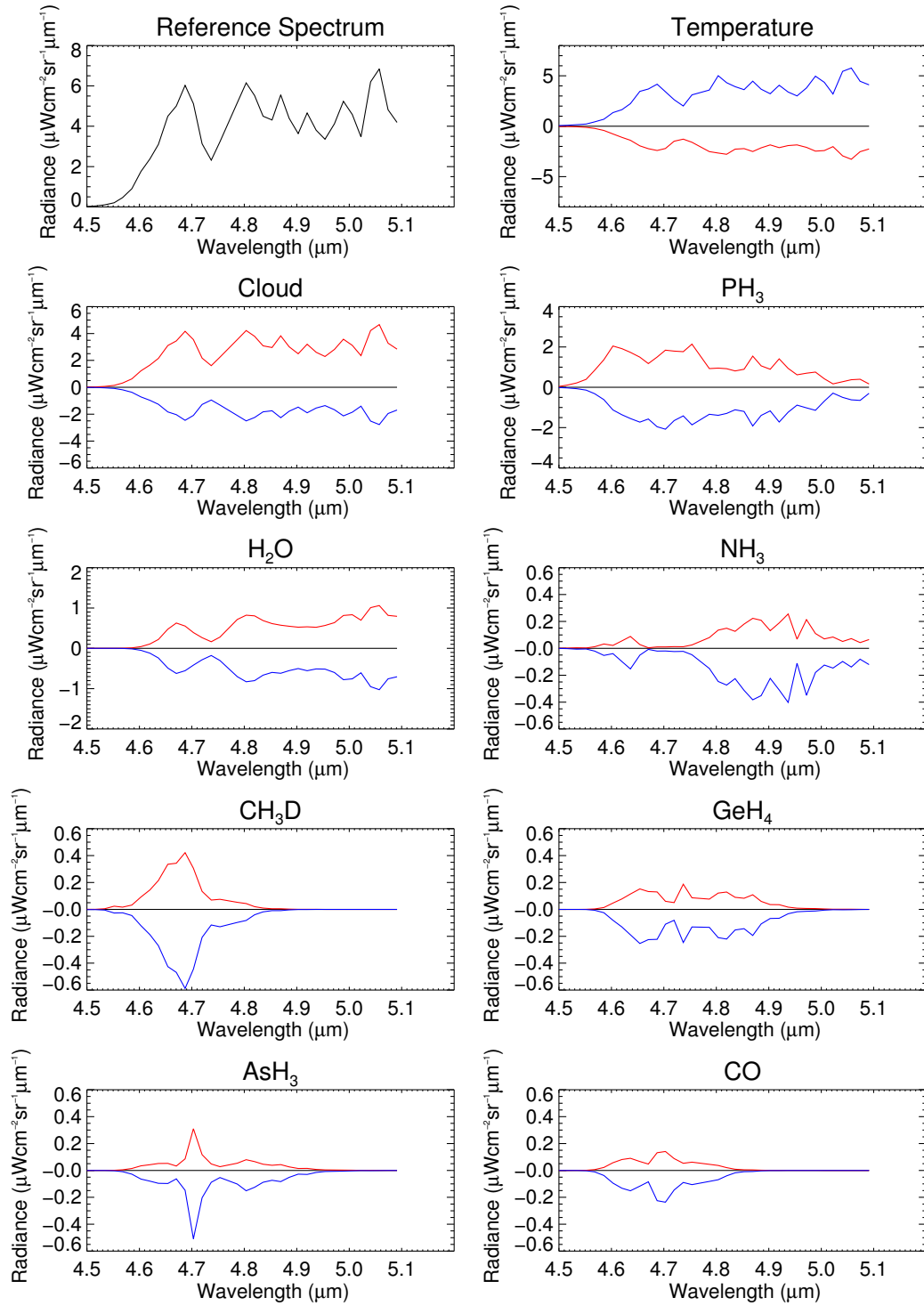


Figure 4.3.1: The sensitivity of VIMS-resolution spectra to changes in different atmospheric parameters. The reference spectrum shows a synthetic spectrum computed for the atmospheric profile described in Section 2.6, and the remaining panels show the (relative) changes to this spectrum that are caused by varying each parameter. The red (blue) lines correspond to a decrease (increase) of 20 K for the temperature, a factor of 20% decrease (increase) for the cloud opacity and a factor of 2 decrease (increase) for the gaseous species.

atmospheric profile also included a single, spectrally-flat cloud layer at 0.8 bar with an optical thickness of 10. The atmospheric parameters were then individually altered, and the new spectra were compared to the reference spectrum. For the molecular species, the entire a priori profile was scaled by factors of 0.5 and 2.0. For water, the deep volume mixing ratio was held constant at the solar abundance and the relative humidity was varied by factors of 0.5 and 2.0. The cloud opacity was varied by factors of 0.8 and 1.2, and the temperature profile was uniformly shifted by ± 20 K. The results of this process can be seen in Figure 4.3.1, where the initial reference spectrum is shown, alongside nine frames showing the difference between the altered profiles and the reference.

The gaseous species with the most significant impact on the shape of the VIMS-resolution 5- μm spectrum are PH_3 and H_2O . Although NH_3 , CH_3D , GeH_4 , AsH_3 and CO all have spectral lines in this region, the relatively low spectral resolution means that they have a minor impact on the VIMS spectra.

The single parameter that has the largest impact on the spectrum is the opacity of the cloud. Because the model cloud is spectrally-flat and located well above the weighting function peak, the entire wavelength range is affected equally; changing the optical thickness scales the entire spectrum up or down, and a relatively minor change in the opacity has a significant impact on the average radiance. Shifting the temperature profile similarly scales the entire spectrum up or down, but to a lesser extent; to reproduce an effect which is comparable to a 20% change in cloud opacity, the temperature shift would have to be as large as 20 K. A more realistic tropospheric temperature difference of 2 K between a moist and a dry adiabat (Lewis, 1995) produces a negligible effect on the spectrum when compared to the variability in cloud opacity. The temperature profile is therefore held fixed at the dry adiabat used in the reference profile, but it should be noted that this may produce a small error ($\sim 2\%$) in the retrieved cloud opacities.

In addition to the degeneracy between cloud and temperature, there are further degeneracies between gaseous species which complicate 5- μm retrievals at this spectral resolution. The regions of sensitivity for each parameter are not unique, but instead overlap, leading to difficulties in disentangling the effects of each individual parameter to produce a reliable retrieval. This is further illustrated by Figure 4.3.2, which shows the results of a retrievability test. 75 radiative transfer models were run with random

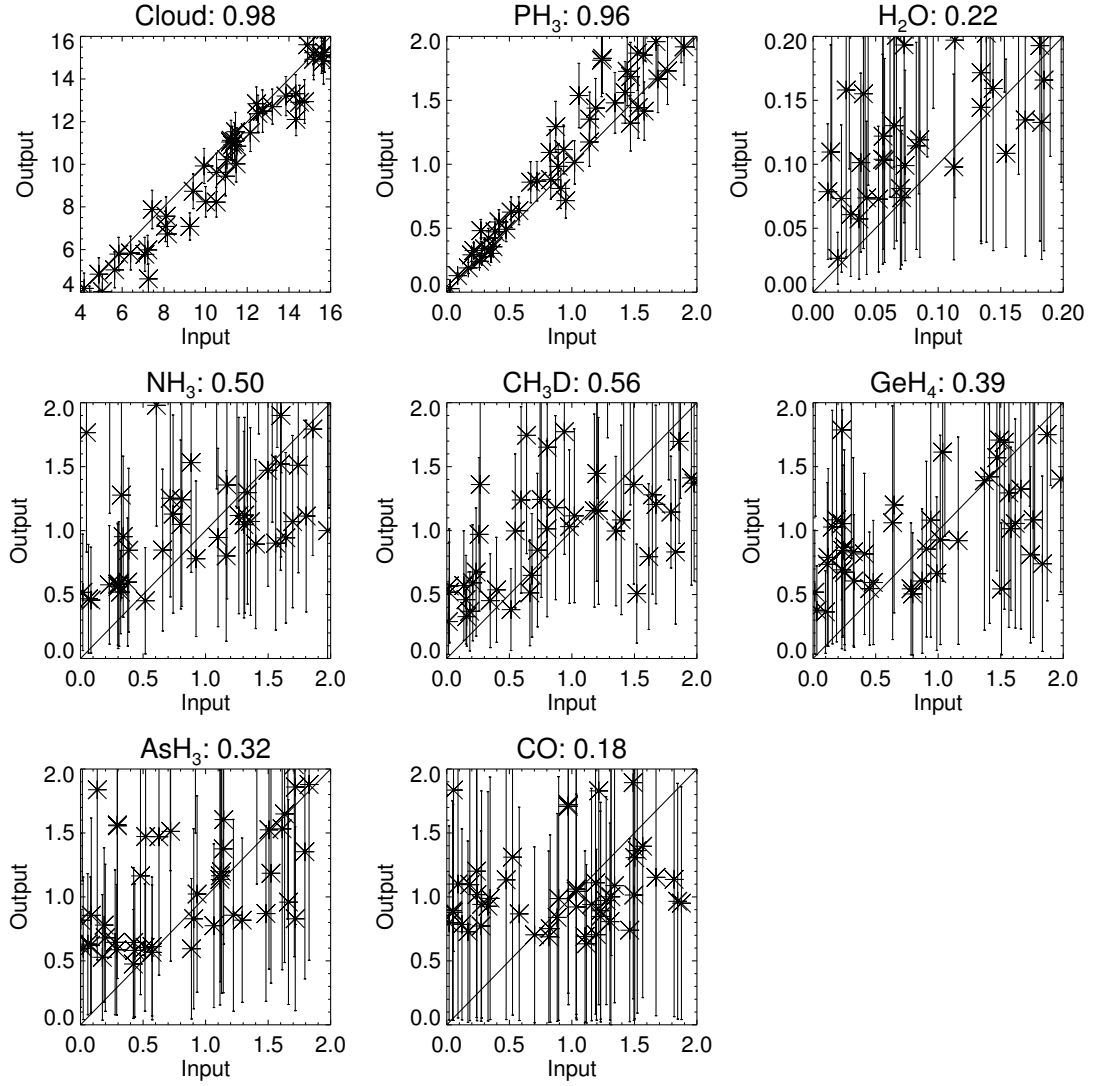


Figure 4.3.2: The retrievability of different atmospheric parameters for VIMS-resolution spectra. In each case, the true ‘input’ value is plotted against the retrieved ‘output’ value. For H₂O, the input and output values correspond to the relative humidity, for the cloud, they correspond to cloud opacity and for the remaining gaseous species, they correspond to the scaling factors of the a priori profile. A perfect retrieval would result in the plotted points lying along the diagonal line. The Pearson product-moment correlation coefficients are given in the titles.

perturbations applied to the reference profile. Random noise was then added to these synthetic spectra, in accordance with the 12% noise estimate for VIMS. The noisy spectra were then used as the input for NEMESIS retrievals. Figure 4.3.2 compares the output from the NEMESIS retrievals with the ‘true’ input values, and provides the Pearson product-moment correlation coefficient (e.g. Snedecor and Cochran, 1989) for each parameter. A correlation coefficient of 1 indicates a perfect positive correlation, while a coefficient of 0 indicates that there is no linear relationship between the two variables.

The results tally with Figure 4.3.1; changes in cloud cover and the PH_3 volume mixing ratio have the most significant impact on the spectrum and can therefore be accurately retrieved, with Pearson product-moment correlation coefficients that are close to 1. The retrievals of H_2O are fairly poor; although H_2O has a significant impact on the spectrum, it can be seen in Figure 4.3.1 that its effect is very similar to that of the cloud opacity, but is smaller in magnitude. This is further shown by the correlation matrix of the retrieved parameters, which gives a high correlation of ~ 0.9 between the H_2O relative humidity and the cloud opacity. This degeneracy between the two parameters limits the reliability of the H_2O retrievals. However, there is no reliable a priori profile for water vapour, so the relative humidity of water is allowed to vary, along with the volume mixing ratio of PH_3 and the cloud optical thickness. The retrievals of the remaining gaseous species are very poor, and the retrieved values are only weakly correlated with the ‘true’ input values. This is unsurprising, as Figure 4.3.1 shows that they have minimal impact on the spectrum. Based on these results, the abundances of NH_3 , CH_3D , GeH_4 , AsH_3 and CO are held fixed at their a priori values during the remainder of this study.

4.4 Basic retrievals

Having explored the sensitivity of the 5- μm region to different parameters and the potential limitations of any retrievals, this section now analyses the VIMS data itself using the NEMESIS retrieval algorithm. As with the sensitivity analysis, PH_3 and H_2O were each allowed to vary via a single parameter. For PH_3 this was a scaling parameter that effectively altered the deep volume mixing ratio, and for H_2O this was the relative

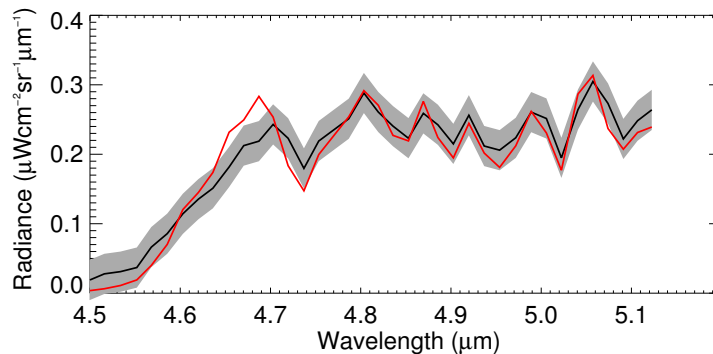


Figure 4.4.1: Fit obtained using nightside data from the EqZ. The VIMS data is shown in black (with the error shown in grey) alongside the best-fit retrieved spectrum in red.

humidity above a fixed deep volume mixing ratio. For each variable, large errors were assigned to a priori values, such that the fit becomes a simple least-squares fit to the data (as described in Section 2.6). Testing was carried out to determine whether allowing additional degrees of freedom in these gases would improve the fit, but this did not make a difference in either case.

As an initial model, a single, compact cloud is used, located at 0.8 bar. This location is well above the weighting function peak (4–8 bar) and is the approximate location of the predicted NH_3 -ice cloud. Figure 4.2.2 shows that the shape of the spectra are almost identical at all latitudes, despite large variations in the absolute radiance. The cloud parameters therefore cannot vary strongly with wavelength, as otherwise any spectral features would be more evident in the cool, optically thick spectra than in the warm cloud-free spectra. In this initial model, a spectrally-flat cloud is therefore used, as has been previously suggested by both ground-based and space-based studies, including Drossart et al. (1982), Bézard et al. (1983), Bjoraker et al. (1986) and Roos-Serote and Irwin (2006). The scattering parameters (see Section 2.4) for this cloud were initially fixed at values that are broadly representative of moderately-sized NH_3 particles (Roos-Serote and Irwin, 2006): single-scattering albedo, $\omega = 0.9$ (highly scattering) and asymmetry parameter, $g = 0.8$ (mostly forward scattering).

An example of the results of these retrievals can be seen in Figure 4.4.1; a good fit to the VIMS 5- μm data can be obtained despite using a very simple cloud model and relatively few free parameters. The one exception to the otherwise good fit is the 4.65–4.75 μm region of the spectrum, where there is an apparent offset between the VIMS data

and the retrieved spectrum. Many attempts were made to solve this issue, including allowing additional gaseous species to be retrieved, altering the parametrisations for the gases (including PH_3), inserting multiple cloud decks and varying the vertical cloud profile. Similar issues can be seen in previous 5- μm studies of Jupiter using Galileo NIMS, including Roos-Serote et al. (1998) and Nixon et al. (2001). Roos-Serote et al. (1998) also had some difficulties in fitting a similar region of the high-resolution ISO/SWS spectra. The consistency of the problem suggests that the problem is with the models rather than the data. The mismatch is similar for both the cool and the warm spectra, suggesting that it is not due to a cloud spectral feature, and is more likely to be due to inaccurate spectral lines from a gaseous species. When the high resolution CRIRES observations were being analysed (Chapters 5-7), this problem was re-examined. There were no obvious problems in fitting any of the absorption features in the 4.65–4.75 μm range that would suggest missing spectral lines. However, the CRIRES observations are made up of a series of small segments, which prevents the entire spectral range from being analysed simultaneously. This means that there could still be inaccuracies in relative strengths of different lines. As with Roos-Serote et al. (1998), this part of the spectrum is excluded to ensure that the retrievals are not driven by an attempt to fit this one region.

4.5 Cloud constraints

Instead of making assumptions about the composition of Jupiter’s clouds, the range of cloud parameters consistent with the VIMS data was explored. Section 4.4 showed that a simple cloud model, consisting of a single, compact, spectrally-flat cloud deck, is able to achieve a good fit to data. This section further explores the cloud parameter space in order to constrain the cloud properties.

4.5.1 Vertical structure

Preliminary conclusions about the vertical location of the tropospheric cloud decks can be drawn by considering the VIMS spectra alone. As described in Section 4.2, Figure 4.2.2 shows that the shape of the spectra are almost identical at all latitudes, despite large variations in the absolute radiance. This immediately tells us that the main

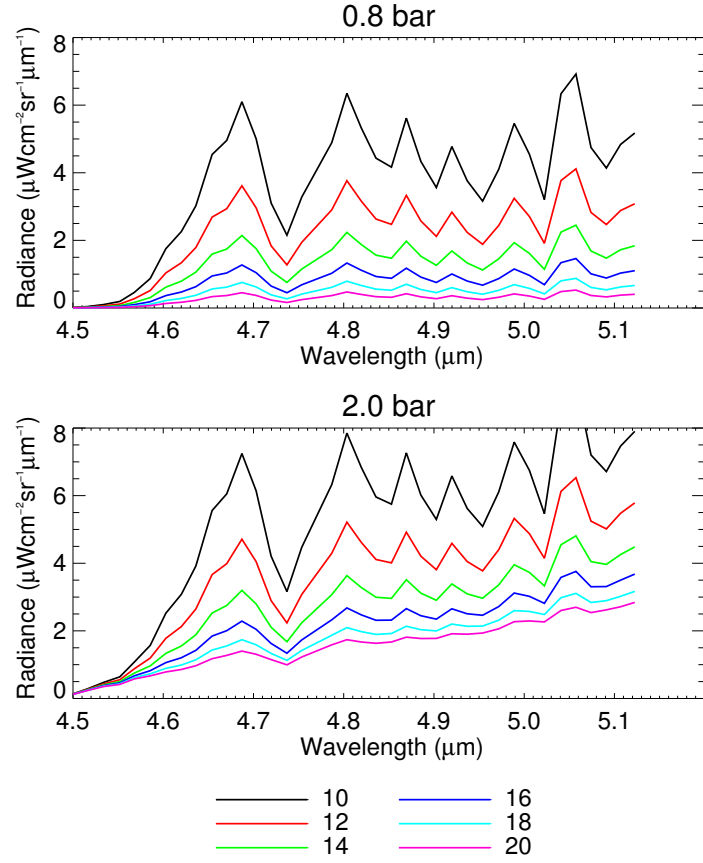


Figure 4.5.1: Sensitivity of spectra to cloud locations. Each panel shows the spectrum for a variety of different cloud optical thicknesses, ranging from 10 to 20. The upper panel corresponds to a compact cloud located at 0.8 bar and the lower panel corresponds to a compact cloud located at 2.0 bar.

cloud decks must be located well above the 4–8 bar region where the weighting functions peak (see Figure 1.2.3); otherwise, thick clouds would block out the radiation from some parts of spectrum more than others, giving a different shape to the 5- μ m spectrum depending on the brightness temperature. Additionally, having an optically thick cloud at a deeper, hotter part of the atmosphere will also start to introduce a blackbody slope into the spectrum, which is not observed in any of the cool spectra. These effects can be seen in Figure 4.5.1, which shows the results of inserting a cloud at two different locations in Jupiter’s troposphere and gradually increasing the optical thickness. In the upper panel, the cloud is located relatively high up in Jupiter’s atmosphere, and increasing the optical thickness decreases the observed radiance, but maintains the same spectral shape, just as is observed in the VIMS spectra. In comparison, the lower panel shows the results for a cloud that has been placed deeper in the atmosphere, at 2.0 bar. Although the spectral shape remains similar for the lower opacities, the spectral shape starts to change for high optical thicknesses. In addition to changing the shape of the spectrum, this second case simply cannot reproduce the low average radiances observed in the cool zones in Figure 4.2.2; at these deeper pressures, the thermal emission from the cloud itself is higher, and even an optically thick cloud therefore cannot reproduce the observed low radiances.

The constraints on the vertical location of the clouds were further explored by running retrievals with a wide range of base pressures for the single, compact cloud. This was done for both a cool spectrum from the EqZ and a warm spectrum from the NEB, and the results for three of the altitudes tested are shown in Figure 4.5.2 (EqZ) and Figure 4.5.3 (NEB). When the cloud is located at 0.8 bar, good fits are obtained for both the EqZ and the NEB. As the cloud is moved to progressively deeper pressures, the NEB fits initially remains good, but the EqZ fits worsens significantly. The cut-off for a reasonable EqZ fit is approximately 1.2 bar, while the NEB fits start to deteriorate for cloud base pressures of 3.0 bar or higher. The poor fit at deeper pressure levels is a result of the phenomena shown in Figure 4.5.1; as the cloud is moved deeper into the atmosphere, it starts to introduce a slope into the spectrum that is not seen in the data. In order to achieve a good fit for both warm and cool spectra using a single cloud deck, the clouds must be located at or above the 1.2-bar pressure level. The 1.2-bar pressure

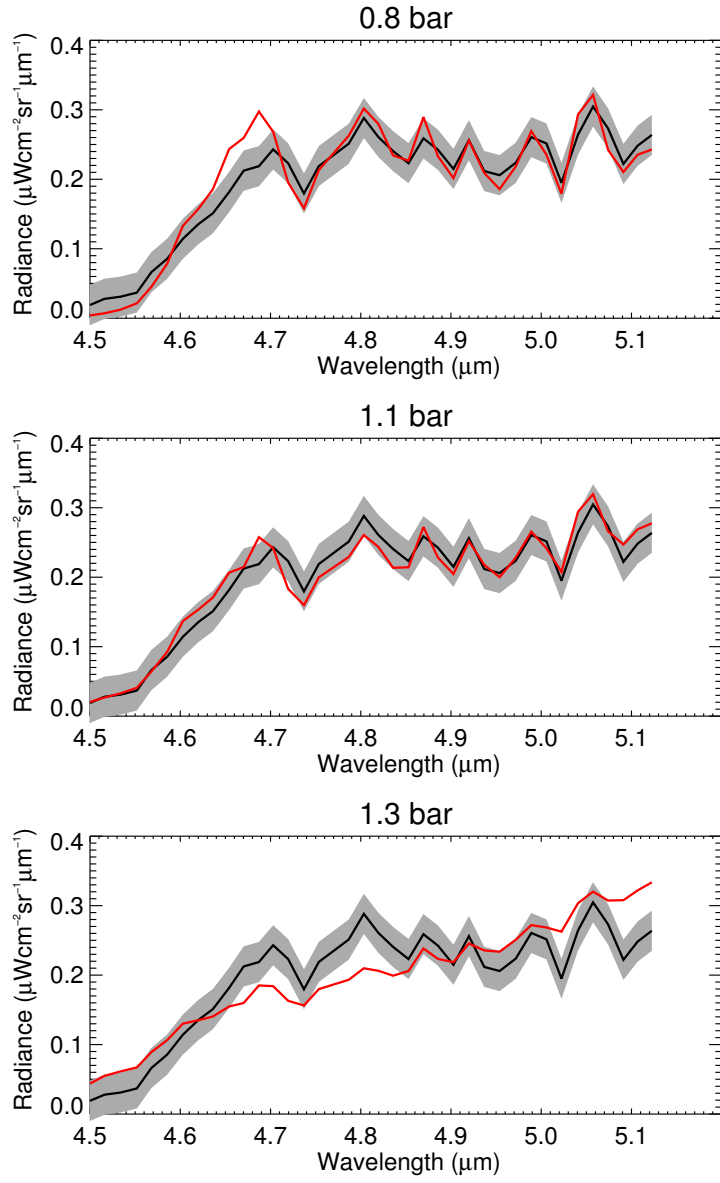


Figure 4.5.2: Retrievals of VIMS spectra from the EqZ. Each panel corresponds to a different cloud base location. The VIMS data is shown in black (with the error shown in grey) alongside the best-fit retrieved spectrum in red.

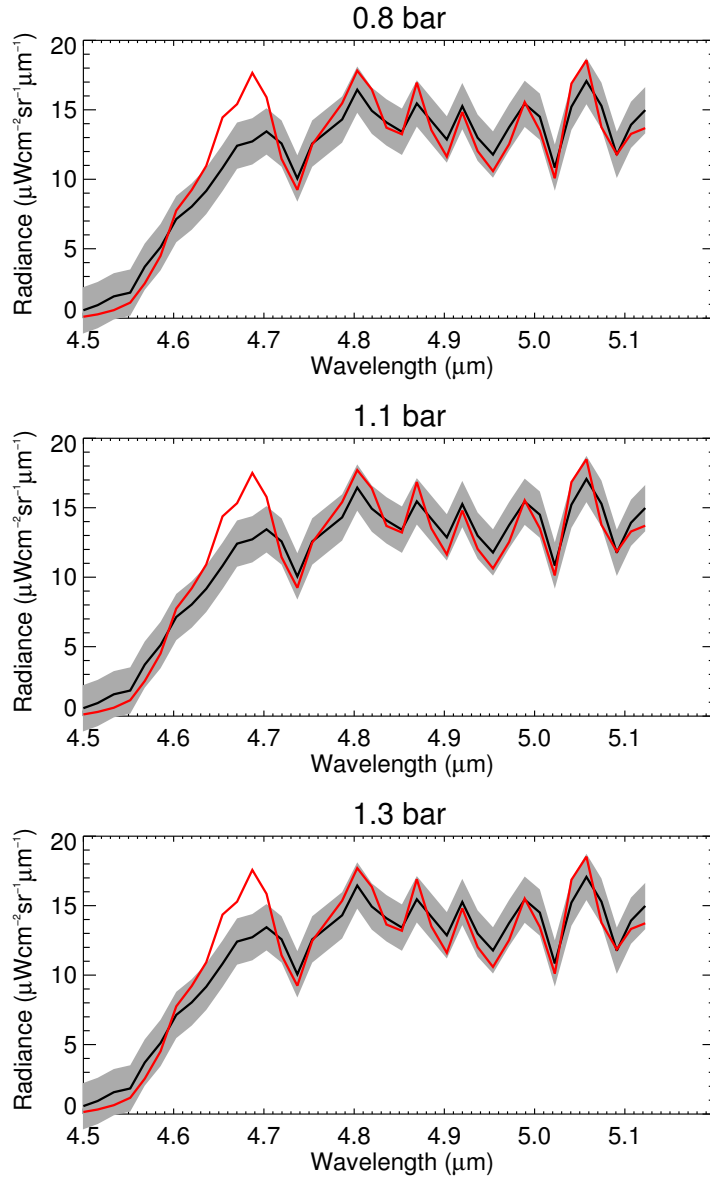


Figure 4.5.3: Retrievals of VIMS spectra from the NEB. Each panel corresponds to a different cloud base location. The VIMS data is shown in black (with the error shown in grey) alongside the best-fit retrieved spectrum in red.

level corresponds to an atmospheric temperature of 180 K, which is approximately equal to the minimum observed brightness temperatures.

In addition to exploring the location of the primary cloud opacity, testing was also carried out to investigate the vertical profile of the cloud. Additional cloud decks, with independently variable opacities, were included in the retrievals, but this made very little difference to the quality of the fits obtained. Similarly, extended cloud decks were used instead of vertically compact clouds, but this did not improve the fit either.

In summary, the VIMS 5- μm nightside data can be fit using a simple cloud model, consisting of a single, compact, spectrally-flat, scattering cloud deck whose opacity varies as a function of latitude, provided that this cloud deck is located at pressures less than 1.2 bar. It is important to note that this does not rule out more complicated cloud structures, including multiple cloud layers at pressures less than 1.2 bar. The existence of deeper cloud decks is also possible, provided that the bulk of the 5- μm opacity still originates from the upper clouds.

Similar analyses have previously been performed using observations from Galileo NIMS which cover the same wavelength range. Roos-Serote and Irwin (2006) used a grey cloud model to fit the NIMS data, and found it had to be located above the 2-bar level, i.e. at pressures less than 2 bar. The analysis in this chapter confirms and tightens this constraint ($p < 1.2$ bar) because the global VIMS data includes spectra from very cold regions, while the NIMS study was restricted to warmer regions. It is the cooler spectra that provide the stronger constraint on the cloud location. Other NIMS studies have this same discrepancy; Irwin et al. (2001), Nixon et al. (2001) and Irwin and Dyudina (2002) all place their principal cloud decks at pressures of 1–2 bar, but since they only use warmer spectra, they are able to place the clouds deeper in the atmosphere and still achieve good fits.

The 5- μm VIMS results are consistent with Cassini observations made at other wavelengths. Sromovsky and Fry (2010) analysed the 3- μm segment of the VIMS data. Using their four-layer multiple-scattering model, they found that the deepest, highest opacity cloud had a spatially variable cloud base that was located between 0.79 and 1.27 bar. Using a narrow spectral window from the CIRS instrument centered on 7.18 μm , Matcheva et al. (2005) found that the cloud absorption coefficient peaks at 0.9–1.1

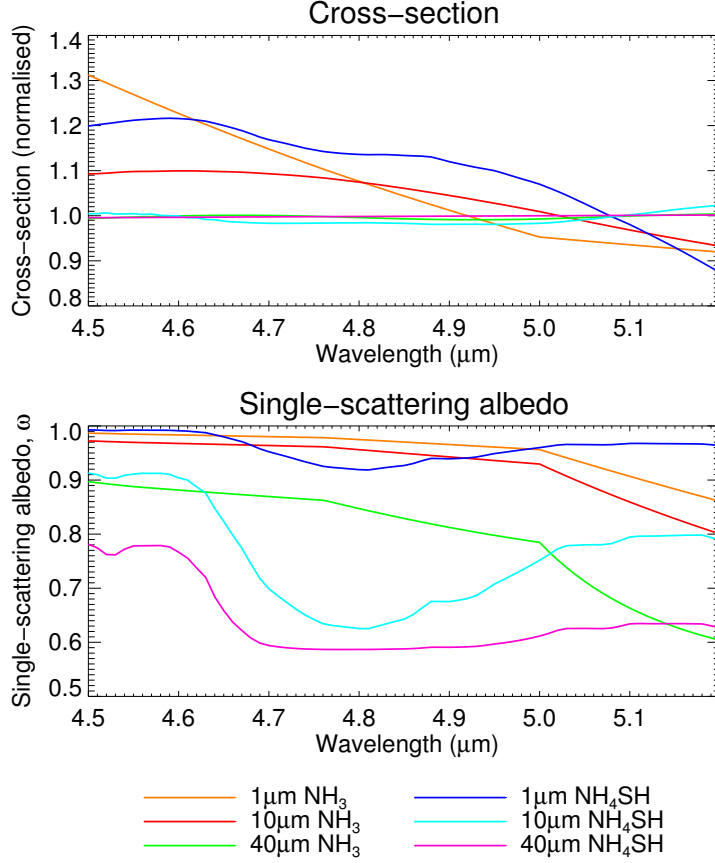


Figure 4.5.4: Spectral parameters for NH_4SH and NH_3 particles of different sizes, calculated using Mie theory and the Henyey-Greenstein approximation.

bar. These locations are broadly consistent with the VIMS data, but this analysis can neither confirm nor rule out the latitudinal variability in the cloud base pressures; a good fit can be achieved with a single cloud base pressure of 1.2 bar or lower, but a range of pressures is also possible, provided the cloud is located above 3.0 bar for the warm spectra from the belts and above 1.2 bar for the cool spectra from the zones.

4.5.2 Spectral features

As described in Section 4.4, the similar spectral shapes in Figure 4.2.2 suggest that the clouds providing the majority of the 5- μm opacity must be relatively spectrally-flat, at least at the VIMS spectral resolution. Having established that a completely grey cloud is able to produce a good fit to the VIMS data, this section now explores the extent to which spectral features are compatible with the observations.

The two expected cloud materials in the middle troposphere are NH_3 ice and solid NH_4SH . Figure 4.5.4 shows the extinction cross-section and single-scattering albedo

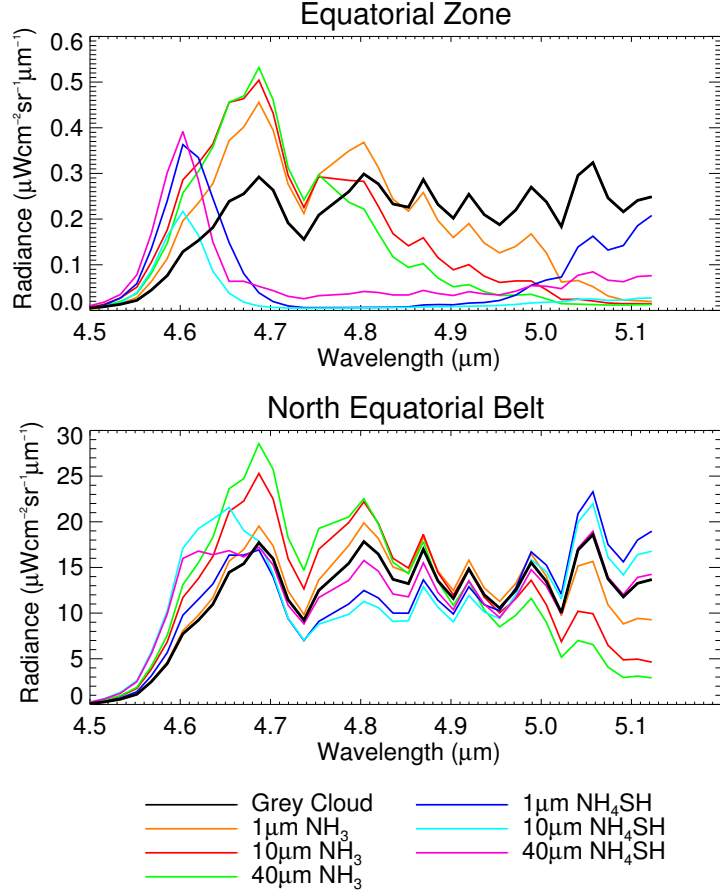


Figure 4.5.5: The effect of different cloud particles on the shape of the spectra. The black line in each case gives the fit obtained using a spectrally-flat cloud. The spectral features are more pronounced in the EqZ, where the clouds are optically thick.

as a function of wavelength for a range of particle sizes. These functions have been calculated using Mie theory and the Henyey-Greenstein approximation, as described in Chapter 2. From this figure, it is clear that the scattering and absorption parameters of particles of pure NH_3 and pure NH_4SH vary significantly with wavelength within the 5- μm window. Larger particles have a relatively flat extinction cross-section, but a strongly wavelength-dependent single scattering albedo, while this is reversed for smaller particles.

The effect of the wavelength-dependent cloud parameters on the spectral shape can be seen in Figure 4.5.5. In each panel, the black line shows the model fit to the VIMS data that is obtained using a spectrally-flat cloud. The remaining lines show the effect of using a ‘real’ cloud material, rather than a grey cloud. To make the impact of the clouds clear, the atmospheric composition (H_2O and PH_3) has been held constant for each cloud material, and the cloud opacity alone has been varied in order to produce an

average radiance as close to the true value as possible. Performing a full retrieval does not however improve the fit sufficiently, and in some cases the algorithm attempts to retrieve an unphysically high abundance of the gaseous species in order to compensate for the poor cloud fit.

Figure 4.5.5 clearly shows that these ‘real’ clouds are unable to reproduce the shape of the VIMS spectra. This is particularly the case for the EqZ, where the clouds are optically thick and any spectral features therefore become more prominent. The varying single-scattering albedo is the primary cause of these poor fits; the sharp gradient at $\sim 4.65\ \mu\text{m}$ seen in Figure 4.5.4 for the NH_4SH particles translates into the sharp gradient seen in Figure 4.5.5, while the gradual slope for the NH_3 produces a slope in the spectra.

Despite being the most plausible materials for the thick tropospheric clouds, pure NH_3 ice and pure solid NH_4SH are not consistent with the VIMS $5\text{-}\mu\text{m}$ observations. However, there are several possible explanations for this. The presence of several cloud decks made up of particles of different materials and/or sizes can act together to ‘blur out’ the individual spectral features, giving a net effect of a roughly grey cloud. This effect, along with the fact that the study focussed on warmer regions where the clouds are thinner, allowed Irwin et al. (1998) to fit the Galileo NIMS $5\text{-}\mu\text{m}$ spectra using a 4-level cloud structure, made up of $0.5\ \mu\text{m}$ tholins, $0.75\ \mu\text{m}$ NH_3 particles, and $0.45\ \mu\text{m}$ and $50\ \mu\text{m}$ NH_4SH particles.

Alternatively, the clouds could be made of a different material altogether, or be masked by deposits from another material which causes the cloud to be spectrally-flat. Coating of ammonia particles by other substances has been suggested by Baines et al. (2002) as an explanation for the absence of spectroscopically identifiable ammonia clouds across the majority of the planet and Kalogerakis et al. (2008) found that thin layers of hydrocarbons are able to alter the spectral features at 3 and $9\ \mu\text{m}$.

The $5\text{-}\mu\text{m}$ data alone cannot distinguish between these various possible scenarios, but the net effect in each case is a roughly grey cloud. The simplest option is therefore chosen, and the analysis continues using a single spectrally-flat cloud.

4.5.3 Scattering properties

The choice of scattering parameters affects the thermal scattering on the planet’s night-side. This section now explores the range of scattering parameters that are consistent with the VIMS nightside observations. As described in Section 2.4, there are two scattering parameters to be studied: the single-scattering albedo, ω , which describes how likely a particle is to scatter a photon of light instead of absorbing it, and the asymmetry parameter, g , which describes the probability that light will be scattered at different angles. Using the same nightside nadir data used in Sections 4.5.1 and 4.5.2, retrievals were performed using many values of ω and g , covering the full range of physically realistic particles. Although the numerical results of the retrievals varied with these different values (a higher single-scattering albedo requires a higher cloud optical thickness for the same spectrum), the fits produced were very uniform. Many different cloud parameters were capable of reproducing the nadir VIMS spectra, so the values of ω and g cannot be constrained from the nadir data alone.

Additional insights can be gained by considering observations made at a range of emission angles, rather than just using nadir observations. In the jovian troposphere, temperature decreases with altitude, leading to limb darkening: the radiance observed decreases towards the edge of the disc. The extent of this limb darkening is heavily dependent on the cloud decks that are present in the troposphere; by comparing the spectra at different emission angles, the scattering parameters of these clouds can be constrained.

The relatively low spatial resolution of the VIMS data means that it is not possible to isolate a single atmospheric feature and compare its appearance at different emission angles. Instead, the scatter plots in Figure 4.5.6 show the limb darkening observed in the equatorial region of the planet (-2.5° to 2.5°). The equatorial region was chosen as it provides the largest range of emission angles, and exhibits less spatial inhomogeneity than other parts of the planet. Each point corresponds to the mean $4.5\text{--}5.2\text{ }\mu\text{m}$ radiance from a single pixel.

The large spread of points is due to the inhomogeneity of the cloud thickness, even within a single latitudinal band (as seen in Figure 4.2.1). Nevertheless, the general limb-darkening trend can be seen - as the emission angles increases towards the limb,

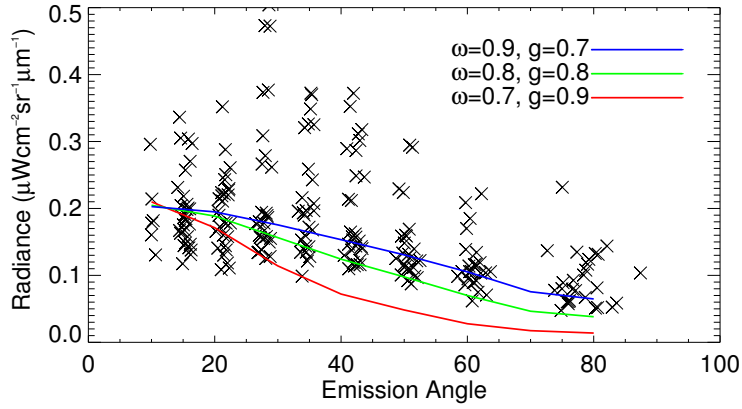


Figure 4.5.6: Limb darkening for different cloud parameters. The scatter plots show the VIMS data from the EqZ as a function of emission angle, averaged over the 5- μm spectral window. The large spread is due to the spatial inhomogeneity of the clouds, even within a single latitude band. Overplotted are the synthetic limb-darkening curves for three example sets of cloud parameters (see Figure 4.5.7 for the full range of parameters tested).

the radiance decreases.

The broad scatter of points in Figure 4.5.6 means that a simultaneous retrieval of data at different emission angles is unlikely to be productive. Two points at different positions along the disc are likely to have very different cloud opacities, in addition to having different emission angles. However, forward modelling does allow us to place some constraints on the cloud parameters that are consistent with the observed data.

For a range of cloud parameter combinations (single-scattering albedo, ω and asymmetry parameter, g), retrievals were performed using nadir (low-emission angle) data. The retrieved parameters from the nadir data were then used to forward-model spectra corresponding to higher emission angles. The results of this process for three example combinations are shown by the solid lines in Figure 4.5.6.

Each limb-darkening curve is anchored to the same point because of the initial retrieval, but they all vary differently with emission angle. Despite the broad scatter of points, it is immediately apparent from Figure 4.5.6 that the case $\omega = 0.9$ and $g = 0.7$ is consistent with the data, but $\omega = 0.7$ and $g = 0.9$ is not.

This retrieval and forward-modelling process was repeated for more parameter combinations. For each case, the radiance at an emission angle of 60° was recorded and is plotted in Figure 4.5.7. The red line corresponds to a radiance of $0.062 \mu\text{Wcm}^{-1}\text{sr}^{-1}\mu\text{m}^{-1}$, the lowest observed radiance at 60° in the VIMS equatorial data. Scattering parameters

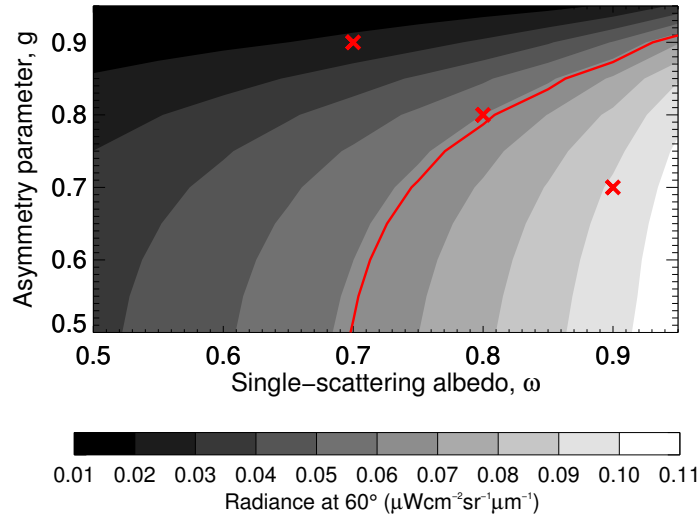


Figure 4.5.7: The synthetic curve radiances at an angle of 60° , for a range of scattering parameters. The red crosses mark the three example parameters used in Figure 4.5.6. The red line marks the minimum observed radiance from the VIMS data; scattering parameters above this line are not consistent with the data. Cases where ω is low and g is high are ruled out.

which produce radiances larger than this value are consistent with the data; scattering parameters which produce smaller radiances are not. Looking at Figure 4.5.6, the blue curve falls within the acceptable fit region, the green curve is on the borderline, and the red curve does not give an acceptable fit. While a fairly large range of ω and g values give an acceptable fit to the VIMS limb-darkening data (the bottom-right segment of the plot), this figure rules out the case where ω is low and g is high.

For this analysis, the location of the cloud deck was held constant at 0.8 bar. The effect of varying the altitude of the cloud deck was investigated, and it was found that no additional constraint was provided on top of the results from Section 4.5.1, i.e. the clouds can be located anywhere above 1.2 bar. The conclusions drawn from Figure 4.5.7 are independent of the cloud location, provided that it is above the 1.2-bar level.

These results support the conclusions of Roos-Serote and Irwin (2006), who performed a limb-darkening analysis using pairs of $5\text{-}\mu\text{m}$ data cubes from Galileo NIMS and found that Jupiter's tropospheric clouds must be highly scattering. With g fixed at 0.8, they found that an optimum fit was obtained with $\omega = 0.9 \pm 0.5$, which is consistent with the results of this section. Roos-Serote and Irwin (2006) noted that the same feature observed at both low and high emission angles had the same overall spectral shape,

and they therefore concluded that they must both be scattered versions of the same generally upwelling spectrum. If the cloud was non-scattering, then at high emission angles, the weighting functions would move to higher altitudes, leading to a change in the spectral shape. The same effect can be seen in Figure 4.2.2b, where the spectra all have the same shape, regardless of latitude and emission angle. This further builds on the conclusions of Roos-Serote and Irwin (2006) by showing that the clouds must be scattering at all latitudes.

4.6 Global retrievals

Using the best-fit cloud model from the previous section (a single, compact grey cloud located above 1.2 bar), retrievals were run across the entire planet in order to investigate variability in the atmospheric parameters. As before, the free parameters in the retrievals were the cloud opacity, the water vapour relative humidity, and the scaling factor of phosphine (effectively the deep volume mixing ratio). Retrievals were run for each of the forty-one 5° latitudinal bins described in Section 4.2 and the results are shown in Figure 4.5.8. The initial error estimate of 12% led to overfitting the data, so the errors were reduced to 6% in order to give a goodness-of-fit value (χ^2/n) of ~ 1 . The initial estimate of 12% was fairly arbitrary, and by reducing this value, a more accurate estimate of the errors on the retrieved quantities can be obtained. The formal errors on the retrieval, determined using the 6% error value, are shown in Figure 4.5.8.

4.6.1 Water vapour

For the initial retrieval, the H_2O abundance was included as a free parameter. The results for this process are shown in black in Figure 4.5.8. If allowed to vary, the zonally-averaged relative humidity ranges from $\sim 0.2\%$ to $\sim 3\%$, with a maximum around the equator and a somewhat asymmetric appearance. However, Figure 4.3.2 previously showed that H_2O cannot be accurately retrieved due to extreme degeneracy and correlation with the other parameters, so these results should be taken with caution.

To further test these results, the H_2O relative humidity was fixed at various different levels and the latitudinal retrievals were re-run. Figure 4.5.8 shows the results for three of these relative humidities: 0.2% (blue), 1% (red) and 3% (green). The fourth panel of

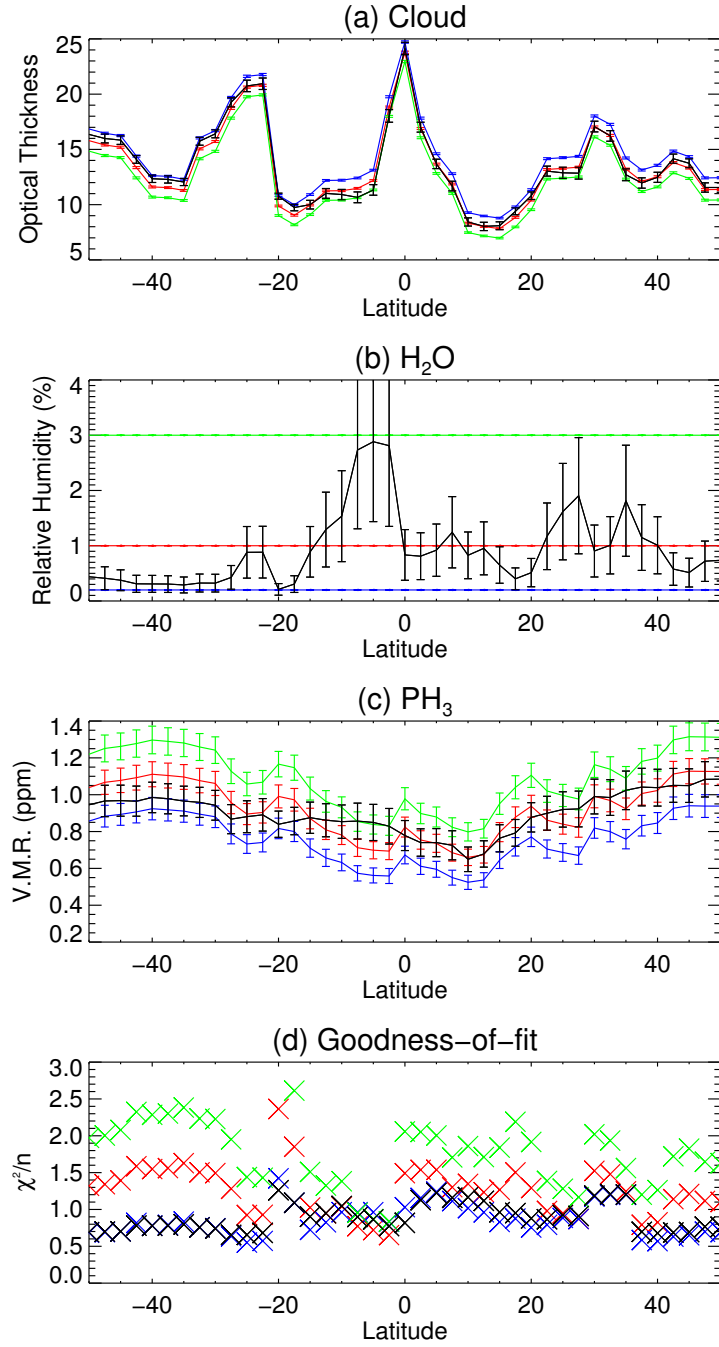
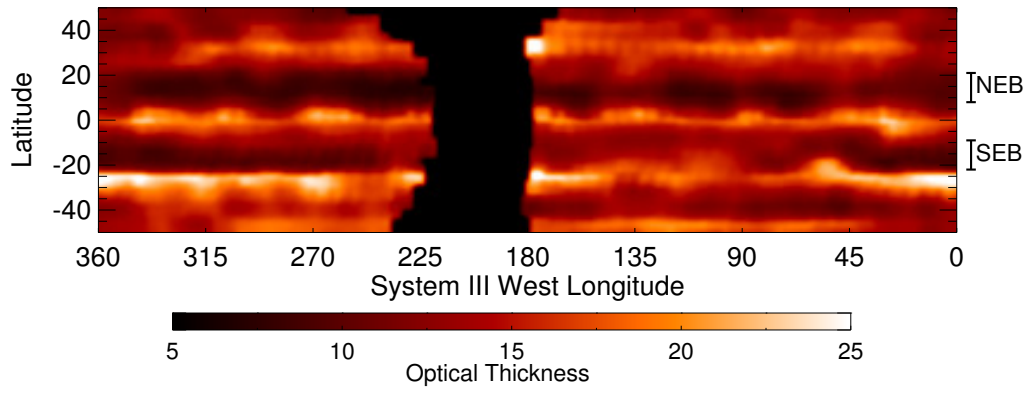
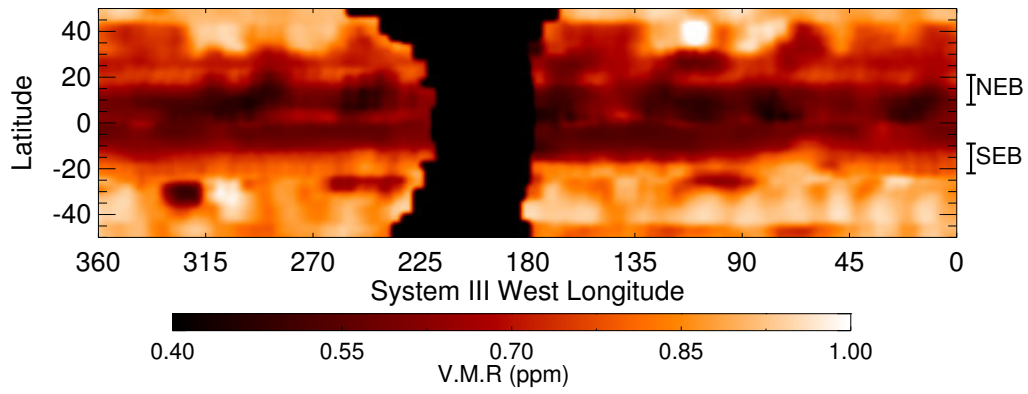


Figure 4.5.8: North-south latitudinal retrievals of the cloud opacity, the H_2O relative humidity and the PH_3 abundance from Jupiter's nightside. The different colours correspond to the four different conditions placed on the H_2O relative humidity: allowed to vary (black), fixed at 0.2% (blue), fixed at 1% (red), fixed at 3% (green). Along with the retrieved quantities is the goodness-of-fit as a function of latitude for each condition.



(a) Cloud opacity.



(b) PH_3 abundance.

Figure 4.5.9: Global maps of the retrieved cloud opacity and PH_3 abundance from the VIMS nightside observations. The H_2O relative humidity has been held fixed at 0.2%. Smoothing has been applied to these maps.

this figure shows the impact that each fixed value has on the goodness-of-fit. Although the higher fixed relative humidities (1% and 3%) lead to significantly worse fits at certain latitudes, the lowest value (0.2%) produces fits that are comparable to the free retrieval. Removing water vapour entirely from the retrievals significantly worsened the fit.

For certain fixed H₂O relative humidities, the cloud opacity and PH₃ abundance are able to adjust and provide an equally good fit to the data. By considering the changes in the goodness-of-fit parameter, the maximum fixed relative humidity allowed is approximately 0.5%. Since a good fit can be achieved using a single H₂O profile at all latitudes (provided that the relative humidity is less than 0.5%) it can be concluded that the VIMS 5- μ m spectra do not provide any evidence for latitudinal variability in water vapour. Previous studies using NIMS data (Roos-Serote et al., 2000) have, however, suggested that there is considerable local variability in the H₂O humidity; since these NIMS analyses made use of high spatial resolution observations of small regions of the planet, this discrepancy may be due to sub-pixel and zonal inhomogeneity in the relatively low resolution VIMS observations. Any small regions of elevated water may be rendered invisible by averaging over the large areas covered by the VIMS pixels, although a thorough exploration of gas and cloud degeneracies in the NIMS dataset is required to confirm this hypothesis.

Assessments of spatial variability of tropospheric H₂O require higher spectral resolution measurements to distinguish between the competing effects of water, cloud opacity and phosphine. Although the CRIRES observations that are analysed in the subsequent chapters provide very high spectral resolution, they are made using a ground-based telescope and the presence of water vapour in the earth's atmosphere complicates retrievals of jovian H₂O. A sufficiently high Doppler shift can allow the telluric and jovian lines to be separated, but the CRIRES observations were made at a time when the Doppler shift is relatively low. High resolution space-based measurements would provide the ideal data to constrain the tropospheric water abundance.

4.6.2 Phosphine

The latitudinal distributions of PH₃ for the different H₂O conditions are shown in Figure 4.5.8. Figure 4.5.9 shows the global distribution for the case where the H₂O

relative humidity is fixed at 0.2%. These global maps were produced by binning the spectra into $10^\circ \times 10^\circ$ size bins and running a retrieval at each latitude and longitude point.

The different water profiles in Figure 4.5.8 lead to the following global averages for the PH_3 deep volume mixing ratio: 0.90 ± 0.09 ppm (allowing H_2O to vary), 0.76 ± 0.05 ppm (0.2% relative humidity), 0.92 ± 0.6 ppm (1% relative humidity), 1.09 ± 0.06 ppm (3% relative humidity). The best fits therefore suggest a deep volume mixing ratio of 0.76–0.90 ppm. These retrieved global averages are consistent with previous 5- μm studies, including Bjoraker et al. (1986) who gives a value of 0.7 ppm from airborne spectroscopic observations, and Irwin et al. (1998) who gives a value of 0.77 ppm from Galileo NIMS. However, analyses of Cassini CIRS 7.7–16.6 μm observations have reported higher values: Irwin et al. (2004) fit the data using a deep volume mixing ratio that varies between 1.0 and 1.5 ppm, while Fletcher et al. (2009) found that values of 1.8–1.9 ppm produced the best fit. Since CIRS is sensitive to higher altitudes than NIMS and VIMS, and the phosphine abundance should decrease with altitude rather than increase (since it is a disequilibrium species), it is likely that this discrepancy is due to an inconsistency in the database line strengths or a difference in the assumed aerosol opacities between the two spectral regions. The same inconsistency between the 5- μm and 10- μm PH_3 observations also occurs for Saturn (Fletcher et al., 2011). In the future, simultaneous studies of the two spectral regions could solve this apparent discrepancy.

Although the different water vapour profiles shown in Figure 4.5.8 lead to slightly different PH_3 retrievals, they each produce a similar latitudinal pattern: an enhanced abundance at high latitudes compared to low latitudes. This is a phenomenon previously noted in the northern hemisphere by Drossart et al. (1990), who used high-resolution 5- μm spectra to detect a 60% enhancement of PH_3 at high northern latitudes compared to the NEB; the latitudinal retrievals in this chapter give an enhancement of ~ 60 –75%. This high-latitude enhancement in PH_3 is also found in the CRIRES observations; further background and discussion of the physical implications can be found in Chapter 6.

4.6.3 Clouds

Figures 4.2.1 and 4.2.2 both showed that there was significant variation in 5- μm radiance with latitude; as expected, Figures 4.5.8 and 4.5.9 show that this is primarily due to variable cloud thickness. The cloud opacity retrieval shown in Figure 4.5.9 is the inverse of Figure 4.2.1a; where the brightness temperature is high, the opacity is low, and vice versa. The absolute retrieved values of the cloud optical thickness are highly dependent on the chosen cloud parameters (and slightly dependent on the H_2O relative humidity), but the relative changes are always reproduced, with high optical thicknesses in the zones ($0^\circ, \pm 30^\circ$) and low optical thicknesses in the belts ($\pm 15^\circ$).

Figure 4.2.1 also showed that there is considerable variability within latitude bands. One region that has significant longitudinal variation is the SEB, where the brightness temperature varies with distance from the Great Red Spot (GRS). Figure 4.5.9 shows that this is primarily due to cloud opacity. The thickest clouds in the SEB occur to the west of the GRS (50°W , 20°S), in its turbulent wake. It is not until the opposite side of the planet ($\sim 225^\circ\text{W}$), that these clouds thin out and the brightness temperature increases. This phenomenon may be explained by turbulence dredging up material from deeper pressures, causing clouds to form. As the distance from the GRS increases, the atmosphere becomes more quiescent and the ordinary, relatively cloud-free appearance returns. This can also be seen in visible light images taken at roughly the same time (see Figure 4.6.1). Immediately to the west of the GRS, the SEB is visibly turbulent, with lighter coloured regions suggesting the presence of clouds. It is not until the opposite side of the planet, to the east of the GRS, that the SEB returns to its darker, cloud-free colour.

4.7 Reflected sunlight analysis

In addition to the nightside data, VIMS also made measurements on Jupiter's day-side. The majority of this chapter has focussed on the nightside data, as the absence of reflected sunlight considerably simplifies the analysis. This section now seeks to determine whether the results obtained from the nightside are consistent with the dayside observations.



Figure 4.6.1: Visible light image of Jupiter, taken using the Imaging Science Subsystem (ISS) instrument on the Cassini spacecraft in December 2000. Credit: NASA/JPL/Space Science Institute.

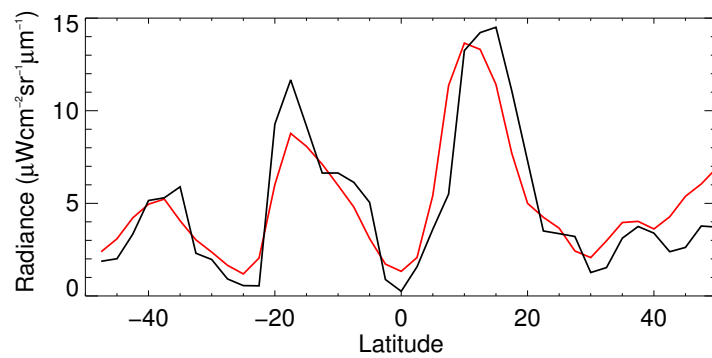


Figure 4.7.1: Zonally averaged 5.0 μm radiance as a function of latitude for both the nightside (black) and the dayside (red).

Figure 4.2.1 compares the global maps from the nightside and the dayside, and it was noted that the contrast between the belts and the zones is smaller on the dayside than on the nightside. This can also be seen in Figure 4.7.1, which gives the zonal averages as a function of latitude. The additional component of reflected sunlight makes little difference in the warm belts and there are even points where the nightside is brighter than the dayside. This is because the belts are relatively cloud-free, so there is less reflection of sunlight.

If the same cloud was observed at different solar zenith angles, there would be a small increase due to reflected sunlight (see Figure 4.7.2 to see the magnitude of this effect); the fact that a small decrease is sometimes observed is due to the fact that the gap of several weeks between the measurements and the low spatial resolution means that the atmospheric conditions are not identical. In the cooler, cloudier regions of the planet, such as the EqZ, the reflected sunlight component becomes more significant. The thicker clouds lead to more reflection, and ensure that the dayside is consistently brighter than the nightside. At the equator, the zonal average radiance at $5.0\ \mu\text{m}$ increases from $0.25\ \mu\text{Wcm}^{-1}\text{sr}^{-1}\mu\text{m}^{-1}$ to $1.33\ \mu\text{Wcm}^{-1}\text{sr}^{-1}\mu\text{m}^{-1}$, an increase of more than 500%. Again, part of this difference may be due to the fact that the observations do not necessarily come from identical regions of the planet, but reflected sunlight clearly accounts for a significant part of the dayside radiance from the zones. This flux difference between the nightside and dayside observations of the equatorial zone is consistent with the analysis of Drossart et al. (1998), who studied the solar reflected component of Jupiter’s $5\text{-}\mu\text{m}$ spectra from Galileo NIMS and found that the minimum flux level was six times greater on the dayside than on the nightside.

Retrievals were run across the range of latitudes on the dayside, using the set of parameters from the previous sections: the volume mixing ratio of PH_3 , the relative humidity of water and the opacity of a single, compact, grey cloud located at 0.8 bar. For these dayside retrievals, the reflected solar component is included in the radiative transfer model. A good fit to the data could be obtained using the same simple cloud model used in the nightside retrievals; an example of the fit obtained in the equatorial region is shown in Figure 4.7.2. This shows that the assumption of purely forward-scattering particles is consistent with the dayside observations.

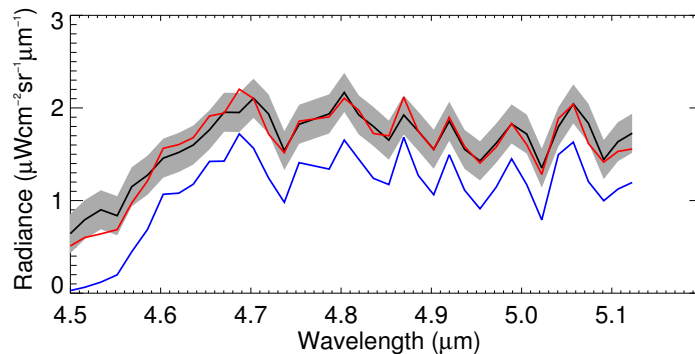


Figure 4.7.2: Example of the fit obtained using dayside data from the EqZ. The VIMS data is shown in black (with the error bars shown in grey) alongside the best-fit retrieved spectrum in red. The blue line shows the spectrum that is obtained from the best-fit atmospheric parameters, with the sunlight turned off, i.e. the equivalent nightside spectrum.

The full results of the zonally-averaged retrievals are shown in Figure 4.7.3. As with Figure 4.5.8, the different colours refer to the different conditions placed on the H_2O profile. The overall shapes of the retrievals are similar to the equivalent nightside plot (Figure 4.5.8); the cloud opacities peak at the same latitudes, and the PH_3 abundance still exhibits an enhancement at high latitudes. There are, however, a few differences. Firstly, the retrieved values of PH_3 exhibit additional minima which correspond to the peaks in cloud opacity. The addition of reflected sunlight accentuates the degeneracy between the cloud parameters and the PH_3 abundance, as it is in the PH_3 absorption wing at the short-wavelength edge of the window where the reflected sunlight component is most significant (see the difference between the red and blue lines in Figure 4.7.2). A small change in the cloud scattering parameters can lead to dramatic changes in the retrieved PH_3 values (a phenomenon not seen on the nightside), so retrievals are unreliable. Higher resolution dayside spectroscopy would reduce this degeneracy, allowing more reliable PH_3 retrievals; PH_3 retrievals of the high-resolution, dayside CRIRES spectra are carried out in Chapter 6.

Secondly, although the cloud opacity has a very similar shape, the absolute values are slightly different. This is likely to be due to a combination of two factors: the opacities may be genuinely different, due to averaging over spatially a inhomogeneous latitudinal band, and the retrievals may be mismatched due to slightly incorrect cloud scattering properties leading to an imbalance between thermal radiation and reflected

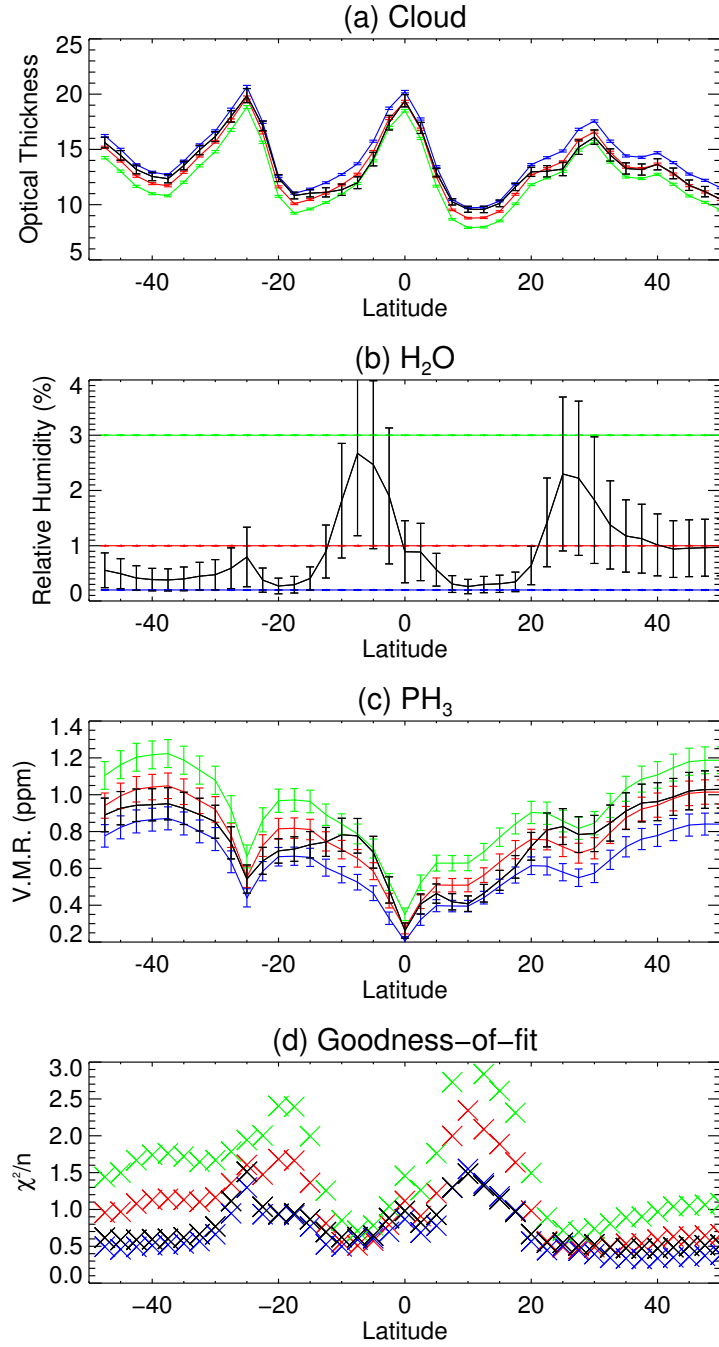


Figure 4.7.3: North-south latitudinal retrievals of the cloud opacity, the H₂O relative humidity and the PH₃ abundance from Jupiter's dayside. The different colours correspond to the four different conditions placed on the H₂O relative humidity: allowed to vary (black), fixed at 0.2% (blue), fixed at 1% (red), fixed at 3% (green). Along with the retrieved quantities is the goodness-of-fit as a function of latitude for each condition.

sunlight. A more comprehensive study, taking into account shorter wavelengths, would be required to jointly constrain the cloud properties.

4.8 Conclusions

This chapter used the 2000/2001 observations of Jupiter made by the Cassini VIMS instrument in the 4.5–5.1 μm range to study the planet’s tropospheric cloud structure and composition. This built on previous work using the Galileo NIMS and Voyager IRIS datasets by making use of (i) the full global coverage afforded by VIMS, covering both warm and cool regions of the planet; (ii) the combination of nightside and dayside observations. The contents of this chapter were published in Giles et al. (2015). The tropospheric cloud structure will be further explored in Chapter 5, and the tropospheric composition will be further explored in Chapter 6.

This chapter showed that VIMS 5- μm data (both nightside and dayside) can be modelled using a very simple cloud model, consisting of a single, compact, spectrally-flat cloud. Section 4.5 showed that bulk of the 5- μm opacity must be located sufficiently high in the troposphere, such that it does not impact the shape of the spectrum. For the coolest regions of the planet, with the thickest clouds, this requirement constrains the clouds to pressures of 1.2 bar or lower. In warmer regions, the clouds can be placed deeper in the atmosphere and still achieve a good fit to the data. The spectra from both cloudy and relatively cloud-free regions have very similar spectral shapes, so the clouds must be relatively spectrally-flat. Pure NH_3 ice and pure NH_4SH do not have sufficiently grey spectra, and are therefore inconsistent with VIMS data. It may be that spectral features in the clouds are masked by coating layers or that multiple cloud decks act together to blur out any features. The relative lack of limb darkening means that the cloud particles must be highly scattering, which supports the previous work of Roos-Serote and Irwin (2006). There is a degeneracy between the single-scattering albedo and the asymmetry parameter, but cases with a low single-scattering albedo and high asymmetry parameter are ruled out.

Global retrievals of cloud opacity, PH_3 and H_2O were performed in Section 4.6. This section showed that the majority of the 5- μm global inhomogeneity can be accounted for by variations in the cloud opacity, with thick clouds in the zones and relatively

cloud-free belts. The retrieved globally-averaged deep volume mixing ratio for PH_3 was 0.76 ± 0.05 ppm (with the H_2O relative humidity fixed at 0.2%), consistent with previous 5- μm studies. The latitudinal retrieval of PH_3 shows an enhancement in the abundance at high latitudes. This will be further studied using CRIREs observations in Chapter 6. The VIMS 5- μm spectra do not provide any evidence for latitudinal variability in the H_2O relative humidity; if the H_2O abundance is held fixed, the cloud opacity and PH_3 abundance are able to adjust in order to produce an equally good fit to the data. The low spectral resolution and high degeneracy between gases mean that H_2O cannot be accurately retrieved, but a fixed relative humidity of less than 0.5% is able to provide a good fit to the data at all latitudes. Unfortunately, the high resolution CRIREs observations cannot be used to produce a reliable retrieval of jovian H_2O due to the presence of telluric H_2O lines.

Section 4.3 showed that the low spectral resolution of VIMS also prevents the accurate retrieval of other gaseous species with weaker signatures in the 5- μm window: NH_3 , CH_3D , GeH_4 , AsH_3 & CO . High resolution spectroscopy is required to retrieve abundances of these species and to search for any spatial variability. CH_3D , GeH_4 and AsH_3 (along with PH_3) are studied using high-resolution CRIREs observations in Chapters 5 and 6.

Chapter 5

Tropospheric clouds: part 2

5.1 Introduction

In this chapter, high spectral resolution observations from the VLT CRIRES instrument are used to further investigate Jupiter's tropospheric clouds. This builds on the conclusions of Chapter 4, which used observations from Cassini VIMS. Chapter 4 made use of the broad spectral coverage from VIMS to constrain the cloud base height and spectral features, and it used the global coverage to study the limb darkening. In contrast, CRIRES provides very high spectral resolution ($R=96,000$) and north-south latitudinal coverage. This means that it can be used to resolve the individual absorption lines of molecular species in Jupiter's troposphere and measure how these line shapes vary with latitude. This chapter focuses on one molecular species: deuterated methane (CH_3D).

Methane (CH_4) was one of the first gases to be discovered in Jupiter's atmosphere when Wildt (1932) identified previously unknown spectral features as methane and ammonia absorption lines. It is the third most abundant gaseous species in Jupiter's upper atmosphere, with a volume mixing ratio (VMR) of 1.81×10^{-3} in the troposphere measured by the Galileo Probe Mass Spectrometer (Niemann et al., 1998). Unlike many other gaseous species, this abundance is not expected to have any spatial variability in the troposphere. CH_4 is chemically stable throughout the troposphere and does not condense at the temperatures found in Jupiter's atmosphere (Taylor et al., 2004), which means that it should be well-mixed.

As a methane isotopologue, CH_3D (deuterated methane) is also constant throughout Jupiter's troposphere. Unlike CH_4 , no in situ measurements have been made of the

jovian CH_3D abundance, but a tropospheric abundance of 0.16 ± 0.05 ppm has been estimated from observations made with the Infrared Space Observatory (Lellouch et al., 2001). Unlike CH_4 , CH_3D has several strong absorption features in the 5- μm window. The observed line shape of the CH_3D features depends on both the CH_3D abundance and the cloud structure. Because CH_3D is expected to be spatially homogeneous, this means that any latitudinal variability in the line shape can be attributed to cloud variability. This makes CH_3D a useful species in the study of Jupiter’s tropospheric cloud structure.

Variability in the CH_3D line shape was recently used by Bjoraker et al. (2015) to infer the presence of a highly variable cloud structure in Jupiter’s deep troposphere. They used observations from the NIRSPEC instrument on the Keck II telescope to show that the CH_3D line shape was much more narrow in the zones than in the belts or hot-spots. They then showed that this could be explained if the belts/hot-spots have no deep cloud opacity, but the zones have an opaque cloud at ~ 5 bar (likely to be composed of water ice). This chapter uses the same principles in order to determine the latitudinal profile of the deep tropospheric cloud structure.

As described in Bjoraker et al. (2015), there are two factors than can affect the shape of the CH_3D absorption lines: the proportion of reflected sunlight and the presence of a deep cloud. In the NEMESIS radiative transfer code, the fraction of reflected sunlight is indirectly controlled by the scattering properties of the cloud. Section 5.2 discusses the impact that these scattering properties have on the retrievals. Section 5.3 subsequently discusses the impact of an additional deep cloud layer. This chapter is based on the first half of Giles et al. (2016a).

5.2 Scattering properties of upper cloud

While the VIMS observations in Chapter 4 covered both the nightside and the dayside of the planet, the ground-based CRISP observations are limited to the planet’s dayside. The extra component of reflected sunlight in the spectrum complicates the analysis. In particular, it introduces a degeneracy between the retrieved abundances of the gaseous species and the scattering parameters of Jupiter’s tropospheric clouds.

This degeneracy can be seen by considering the CH_3D absorption features. Three

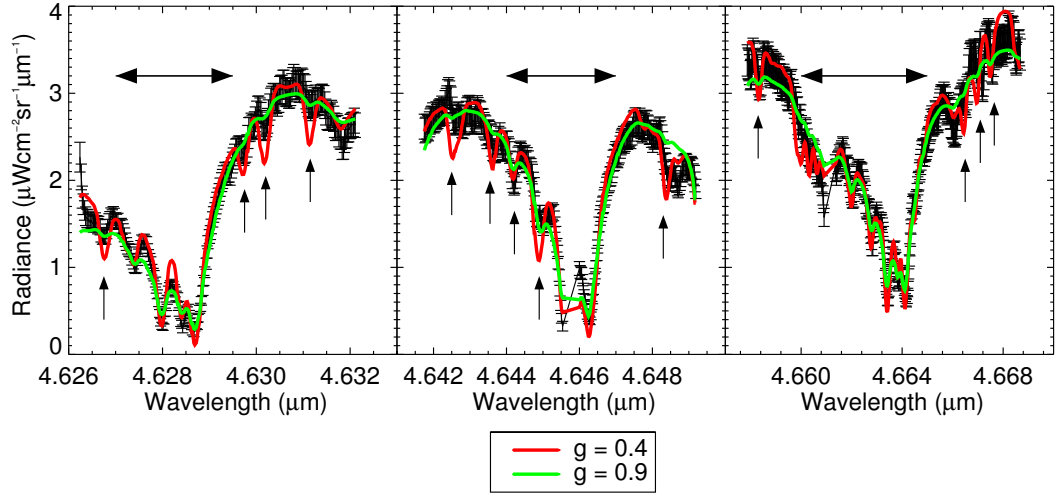


Figure 5.2.1: Fits to three CH_3D absorption features using two different values of the cloud particle asymmetry parameter, g . The observational data (black) is from the EqZ of Jupiter. The location of the CH_3D absorption features are marked by the horizontal double-headed arrows. The location of the most prominent Fraunhofer lines are marked by the vertical single-headed arrows.

of the strongest CH_3D lines observed with CRIRES are shown in Figure 5.2.1. These three features are within the same wavelength segment, which means they are from the same longitude (i.e. were observed simultaneously) and can therefore be analysed simultaneously. The CRIRES data (from 12 November 2012) are shown in black and the arrows show the locations of the CH_3D features. The data are from the Equatorial Zone (EqZ) of the planet, which was chosen as it is one of the cloudiest regions of the planet. Small segments of data with large error bars (due to telluric contamination) have been removed. The three absorption features were fit simultaneously using the NEMESIS retrieval algorithm and the coloured lines show the fits that can be obtained using two different assumptions about the scattering properties of the single cloud deck: firstly, where $g = 0.9$, i.e. the cloud particles are very strongly forward scattering, and secondly, where $g = 0.4$, i.e. the cloud particles are less strongly forward scattering. The single-scattering albedo, ω , was held fixed at 0.9. Both cases can produce a good fit to the broad CH_3D features, but the retrieved CH_3D abundances are significantly different: 0.16 ppm for the case where $g = 0.4$ and 0.08 ppm for the case where $g = 0.9$.

This effect is due to the different amount of reflected sunlight in the two models. A higher asymmetry parameter (more forward scattering) leads to a higher fraction

of radiation originating from the planet, and a lower fraction from reflected sunlight. In contrast, a lower asymmetry parameter (less forward scattering) leads to a higher fraction due to reflected sunlight. A higher g value therefore leads to stronger absorption features for a given gaseous abundance, and hence lower retrieved abundances. There is a similar, but weaker, effect with the single-scattering albedo, ω , with a lower ω value leading to a lower retrieved abundance. It should be noted that this is dependent on the vertical location of the cloud; if the cloud were located below the main line forming region, the effect would be less pronounced. However, Chapter 4 showed that the main tropospheric cloud deck must be located well above the line forming in order to account for the highly variable 5- μm radiation.

This degeneracy between cloud scattering properties and retrieved gaseous abundances is only present in regions of the planet with thick cloud cover (zones like the EqZ). The belts of the planet are relatively cloud-free, so changing the cloud parameters has little impact on the retrievals. This means that different assumptions about the cloud properties can affect the latitudinal profiles of the gases; one set of parameters could suggest that a molecular species has a constant abundance across the planet, while another set of parameters could suggest that the abundance varies between belts and zones. In this section, the degeneracy has been illustrated using CH_3D , but this equally applies to other tropospheric gaseous species. Selecting the correct scattering parameters is therefore of critical importance.

Chapter 4 constrained the scattering parameters of Jupiter’s tropospheric clouds using limb-darkening observations from the VIMS instrument on the Cassini spacecraft. This analysis ruled out strongly forward-scattering particles ($g > 0.9$) and weakly scattering particles ($\omega < 0.7$), but a broad range of values were consistent with the data. CRIRES provides an opportunity to further constrain the scattering properties by using the Fraunhofer lines that can be observed in the spectrum. Fraunhofer lines are the narrow absorption lines that are present in the solar spectrum due to absorption in the sun’s atmosphere. On the planet’s dayside, reflected sunlight makes up a component of Jupiter’s spectrum, and so these lines can be measured. They can be seen in Figure 5.2.1, and are marked by the vertical single-headed arrows. The strength of these features indicates the relative proportions of reflected sunlight and jovian thermal emis-

sion, which in turn depends on the scattering properties. A lower g value (less forward scattering) leads to higher fractions of reflected sunlight, and therefore produces deeper Fraunhofer lines. The asymmetry parameter has a similar, but slightly weaker, effect on the strength of the Fraunhofer lines. By comparing synthetic spectra with different scattering properties to the observed spectrum, the best-fit solution can be found. A similar approach was followed by Bjoraker et al. (2015), who made use of a Fraunhofer line at 4.67 μm to determine the reflectance of the upper tropospheric clouds.

Both ω and g affect the level of the reflected sunlight. In this analysis, ω was held fixed at 0.9, and NEMESIS was used to run retrievals using a range of different g values, in order to determine which value gave the best fit to the CRIRES data. While the resulting best-fit value of g is highly dependent on the assumed value of ω , the combination of the two parameters will produce the true best-fit amount of reflected sunlight.

For this analysis, the entire 5- μm spectral range was used (excluding regions with high error bars due to telluric contamination). A theoretical solar spectrum was obtained from Kurucz (1992). For each value of g , the goodness-of-fit (χ^2/n) was calculated and the results are given in Table 5.2.1. For a single-scattering albedo of 0.9, the best fit was obtained for $g = 0.8$, which is consistent with the results of Chapter 4 and previous studies (Roos-Serote and Irwin, 2006). However, this result should be taken with some caution. Firstly, the minimum at $g = 0.8$ is a fairly broad minimum, and values of 0.65–0.85 produce a reasonably good fit to the data. Secondly, this result is strongly dependent on the solar spectrum used, and any inaccuracies in the theoretical strengths of the Fraunhofer lines will change the inferred value of g . The following section and Chapter 6 will initially use a cloud with an asymmetry parameter $g = 0.8$. However, they will also investigate the extent to which the results are dependent on this assumption.

g	χ^2/n
0.40	2.25
0.45	2.11
0.50	1.95
0.55	1.80
0.60	1.66
0.65	1.54
0.70	1.43
0.75	1.31
0.80	1.19
0.85	1.52
0.90	2.40

Table 5.2.1: The goodness-of-fit values (χ^2/n) for a range of different asymmetry parameters. χ^2/n is minimised when $g = 0.8$ (when ω is fixed at 0.9).

5.3 Deep cloud structure

5.3.1 Spatial variability

Figure 5.2.1 showed three strong CH₃D absorption features in the CRILES data from Jupiter’s cool Equatorial Zone (5°S–4°N). Figure 5.3.1 shows these observations again, and compares them to data from the warm South Equatorial Belt (16°S–6°S). This shows that the CH₃D features are considerably narrower in the EqZ than they are in the SEB.

This difference was further explored by performing retrievals with the NEMESIS retrieval algorithm. Based on the conclusions of Chapter 4 and Section 5.2, the initial cloud model assumes a single compact cloud layer at 0.8 bar, with scattering properties $\omega = 0.9$ and $g = 0.8$. The optical thickness of this cloud was allowed to vary, along with the abundances of all the molecular species described in Section 2.6. For the SEB, the retrieved CH₃D abundance was 0.15 ppm, approximately the same as the value obtained by Lellouch et al. (2001). However, the EqZ produced a much lower value of 0.09 ppm.

As a further test, additional retrievals were run where the EqZ CH₃D abundance was fixed to the higher level (0.15 ppm) and the SEB abundance was fixed to the lower level (0.09 ppm). The results of this are shown by the coloured lines in Figure 5.3.1 and they show that the EqZ and the SEB cannot be fit using the same CH₃D abundance. A VMR of 0.15 ppm fits the SEB data, but is too broad for the EqZ data, while a VMR

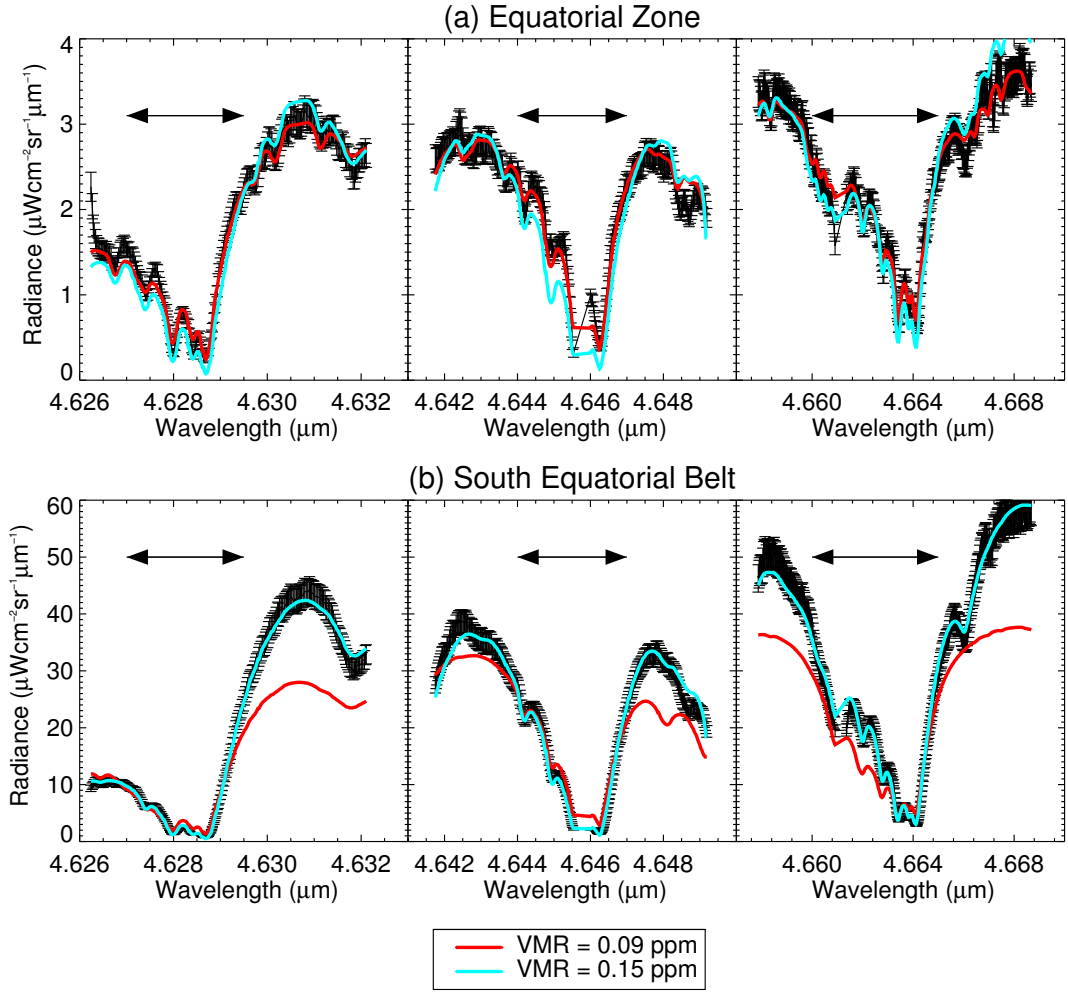


Figure 5.3.1: CH_3D absorption feature fits in two different regions of the planet. CRIRES data is shown in black, and the CH_3D features are shown by the horizontal arrows. Each colour shows the fit obtained using different CH_3D abundances: 0.09 ppm (the best fit value for the EqZ) and 0.15 ppm (the best fit value for the SEB).

of 0.09 ppm fits the EqZ data but is too narrow for the SEB.

However, a factor ~ 2 difference in CH_3D abundance for different regions of the planet is not physically plausible. As described in Section 5.1, methane and its isotopologues are expected to be well-mixed in the troposphere, with no spatial variability. An alternative explanation for this change in CH_3D line shape must therefore be considered; one possibility is spatial variability in the deep cloud structure.

Bjoraker et al. (2015) showed that this belt-zone variation in the CH_3D line shape can be modelled by considering the presence of deep clouds at the pressure levels where water is expected to condense (~ 5 bar). If there is very little cloud opacity at these pressures, the observed top-of-atmosphere radiation originates from deep in the atmosphere, where the pressure is higher. Pressure broadening therefore leads to a line shape with broad wings. Inserting an optically thick cloud layer at 5 bar has two effects on the spectrum: (i) it reduces the continuum radiance, decreasing the apparent depth of the absorption feature and (ii) it moves the weighting function upwards to a region of lower pressures and cooler temperatures, leading to a narrower line shape.

Since the presence of a deep cloud can act to narrow the line shape, this suggests that the narrow EqZ absorption features can be fit, while keeping the CH_3D abundance fixed at the retrieved value for the SEB. To test this, a deep cloud deck was inserted at 5 bar. This deep cloud has the same assumed properties as the upper cloud: compact, spectrally flat, $\omega = 0.9$ and $g = 0.8$. A retrieval was then performed where the opacity of this deep cloud was allowed to vary. The CH_3D abundance was held fixed at the SEB value of 0.15 ppm and, as before, the upper cloud and other gaseous species were also allowed to vary. The results are shown by the yellow line in Figure 5.3.2. For comparison, the case where there is no deep cloud is shown in blue (identical to the blue line in Figure 5.3.1a).

The comparison between the blue and yellow lines shows that this addition considerably improves the fit. The retrieved deep cloud opacity in the EqZ is 230, making the cloud essentially opaque. This agrees with Bjoraker et al. (2015), where the deep cloud in the EqZ was assumed to be opaque. As with Bjoraker et al. (2015), this shows that the difference in CH_3D line shape between the EqZ and the SEB can be accounted for by the variations in the deep cloud structure; the deep cloud is transparent in the SEB

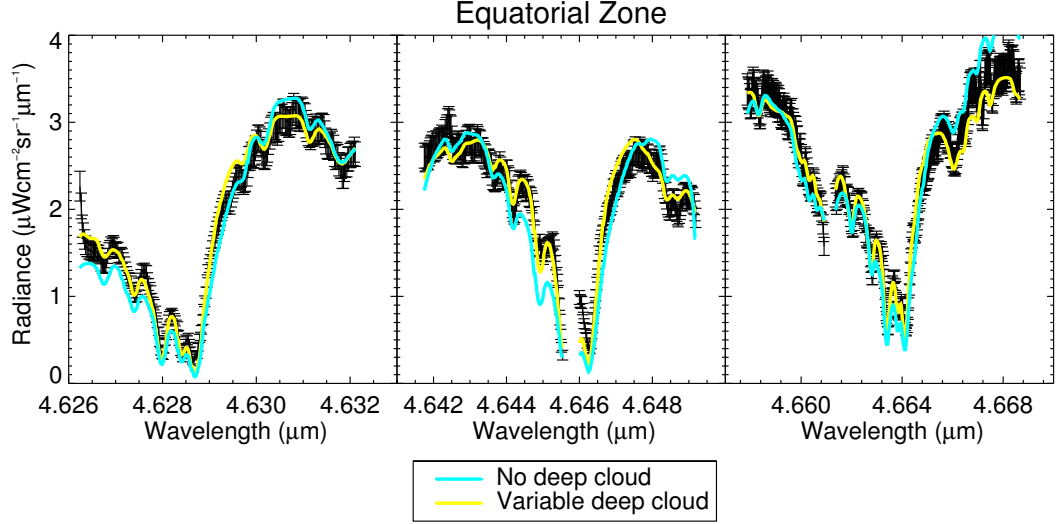


Figure 5.3.2: Retrievals of three CH_3D features in the cool EqZ, with varying the cloud structure. The blue line shows the best fit when there is no deep cloud present, and the yellow line shows the fit where the opacity of a deep cloud at 5 bar is allowed to vary. In both cases, the CH_3D abundance is fixed at 0.15 ppm.

and opaque in the EqZ.

Based on Bjoraker et al. (2015), the initial analysis was performed using an opaque cloud at 5 bar. Further retrievals were carried out in order to test the sensitivity to the cloud location. If the cloud is placed too deep, it falls outside the pressure range probed by 5- μm spectra and can no longer influence the spectral shape. If the cloud is placed too high, it simply plays the same role as the original 0.8-bar cloud and so does not improve the fit. However, between these two extremes, a broad range of cloud locations from 2 to 7 bar can provide a good fit to the data, with deeper pressure levels requiring higher cloud opacities. In the absence of further constraint, the analysis continues using the original 5 bar value.

5.3.2 Latitudinal retrieval

The previous section just compared two parts of the planet, the EqZ and the SEB. Similarly, Bjoraker et al. (2015) restricted their radiative transfer analysis to discrete regions of the planet. The analysis is now extended to perform a full latitudinal retrieval of the deep cloud opacity, using the same CH_3D features as in the previous sections. Spectra were smoothed in the spatial direction with a width of 5 pixels in order to reduce the noise. In order to reduce computation time, every third spectrum in the

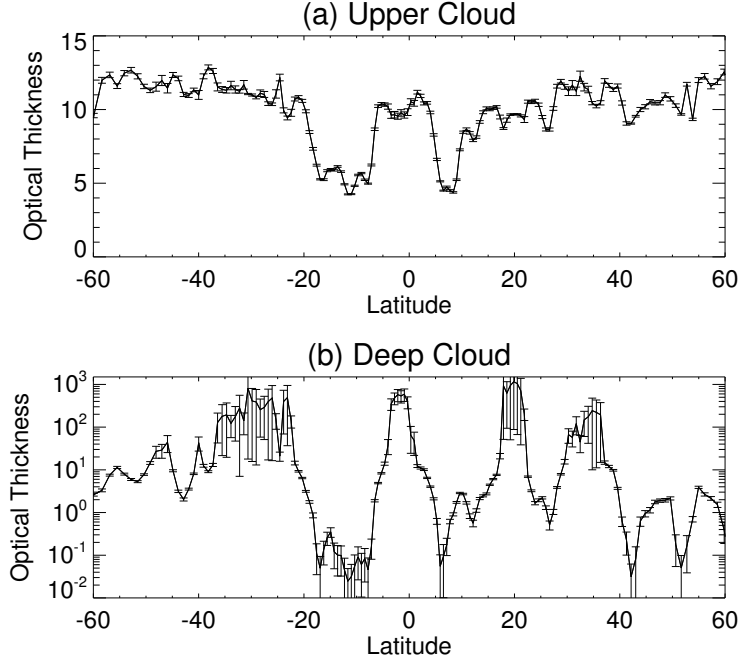


Figure 5.3.3: Retrieved cloud optical thicknesses as a function of latitude for the 12 November 2012 observations. The retrievals show that the cool zones are fit using an opaque deep cloud, while the warm belts are fit using an optically thin deep cloud. The CH_3D abundance is held fixed at 0.15 ppm.

spatial direction was used, reducing the total number of spectra from 512 to 170. For each smoothed spectrum, a retrieval was run following the same procedure as in the previous section.

The retrieved cloud optical thicknesses for both the upper and the deep cloud are shown in Figure 5.3.3. In the belts, the retrieved deep cloud opacity is ~ 0.1 (relatively transparent), and for the zones the retrieved deep cloud opacity is ~ 100 (relatively opaque). The entire analysis so far has used the data from 12 November 2012, but Figure 5.3.4 shows that the same pattern can be seen when the analysis is repeated for the 1 January 2013 data.

The absolute values of both cloud optical thicknesses are very dependent on (i) the accuracy of the radiometric calibration, (ii) the cloud locations and (iii) the choice of cloud scattering parameters. The error bars shown in Figure 5.3.3 do not include these error sources. Changes to either of the first two factors result in a cloud opacity profile that is either scaled up or scaled down, but the relative shape of the latitudinal profile remains constant. The third factor can affect the relative shape of the latitudinal profile, as described in Section 5.3.3.

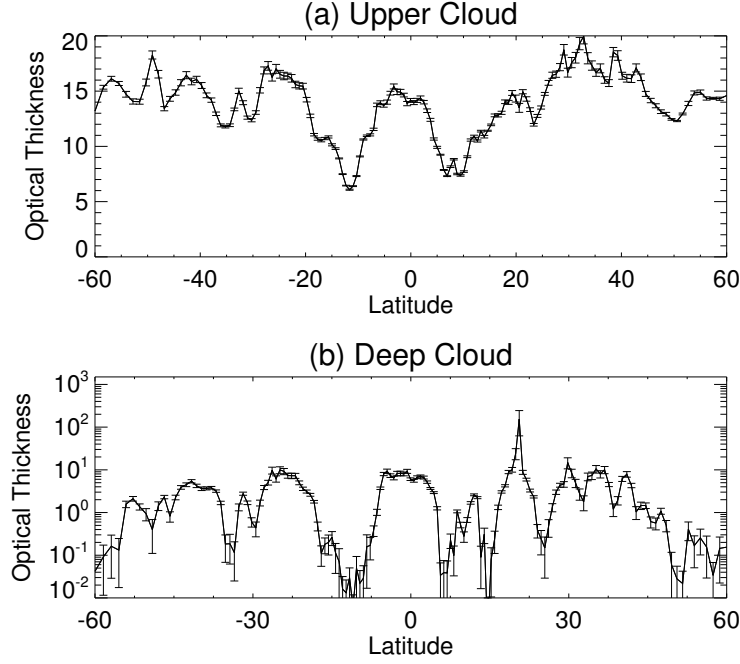


Figure 5.3.4: Retrieved cloud optical thicknesses as a function of latitude for the 1 January 2012 observations. The results agree with the 12 November observations shown in Figure 5.3.3. The CH_3D abundance is held fixed at 0.15 ppm.

5.3.3 Sensitivity to the scattering parameters

Section 5.2 showed that decreasing the asymmetry parameter, g , can act to significantly increase the retrieved gaseous abundances in the cloudy parts of the planet while having a negligible effect on the abundances in the cloud-free regions. This therefore has the potential to solve the problem posed in Section 5.3.1 without having to invoke the presence of a deep cloud deck. With $g = 0.8$, the retrieved CH_3D abundance in the EqZ was 0.09 ppm, compared to 0.15 ppm in the SEB. If instead, $g = 0.4$, the EqZ abundance increases to 0.16, while the SEB abundance remains 0.15 ppm, removing the discrepancy between the two regions. This is shown further by Figure 5.3.5, which shows the same latitudinal cloud profiles as Figure 5.3.3 for the case where $g = 0.4$. Comparing these two figures shows that the change in scattering parameters significantly affects the latitudinal profile. The deep cloud is relatively transparent everywhere, with little difference between the belts and the zones. It is therefore clear that the conclusions about the existence of a cloud deck at 5 bar are very sensitive to the assumptions made about the scattering properties of the cloud deck at 0.8 bar.

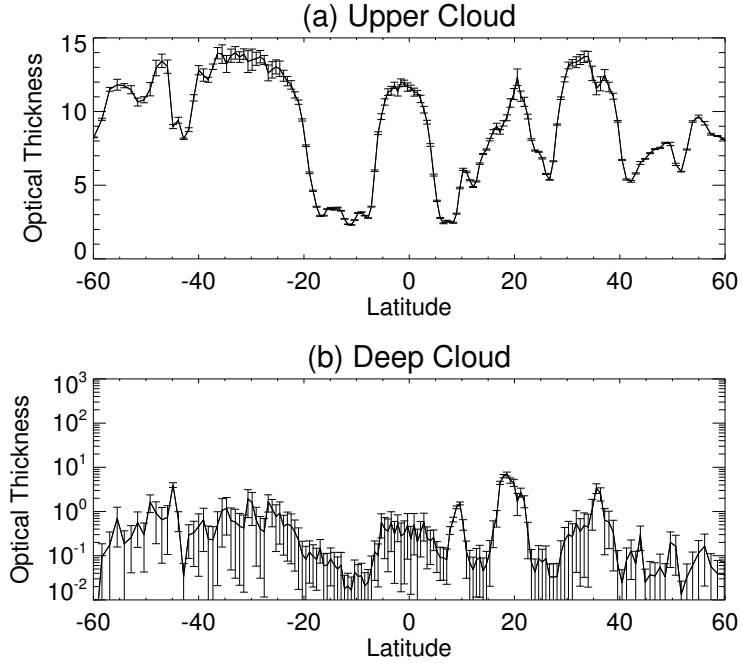


Figure 5.3.5: Retrieved cloud optical thicknesses as a function of latitude, using $g = 0.4$ instead of $g = 0.8$ for the upper cloud. Data is from 12 November 2012. The CH_3D abundance is held fixed at 0.15 ppm.

5.4 Conclusions

This chapter used the high resolution CRIRES observations to analyse Jupiter’s tropospheric clouds. This built upon the work from Chapter 4, and the conclusions from these two chapters form the cloud model used in Chapter 6.

Section 5.2 described a possible degeneracy between the scattering properties of Jupiter’s clouds and the retrieved abundances of CH_3D in the troposphere. Assuming a high asymmetry parameter leads to a low retrieved abundance, and assuming a low asymmetry parameter leads to a high retrieved abundance. The scattering properties of the cloud also control the strength of the Fraunhofer lines in the spectrum; by looking at the strength of the observed lines, an asymmetry parameter of $g = 0.8$ is estimated (assuming a single-scattering albedo of 0.9). However, lower values of g can also provide an adequate fit, meaning that the degeneracy is not entirely broken. Although this section used CH_3D as an example, this degeneracy equally applies to other gases in Jupiter’s troposphere. It will therefore be further explored in Chapter 6, to ensure that conclusions about Jupiter’s disequilibrium species are not heavily reliant on the assumed scattering properties.

Section 5.3 showed that the shape of the CH₃D 5- μ m absorption features varies with latitude. This can be modelled in several different ways: by allowing the CH₃D abundance to vary spatially, by assuming that the cloud particles are not very strongly forward-scattering or by allowing the opacity of a 5-bar tropospheric cloud to vary. The first option was ruled out on the grounds that CH₃D should have a constant VMR throughout the troposphere. The second option was ruled out because an asymmetry parameter of $g = 0.8$ provides the best fit to the Fraunhofer lines. Therefore, the third option is accepted. This suggests that there is an optically thick deep cloud layer present in the EqZ. This agrees with the recent study carried out by Bjoraker et al. (2015). This chapter then expanded on the work of Bjoraker et al. (2015) by performing a full latitudinal retrieval in Section 5.3.2. The opacity of this deep cloud was independently retrieved at each latitude, and the deep cloud was found to be opaque in the zones and transparent in the belts. As previously stated by Bjoraker et al. (2015), the location of this deep cloud suggests that it is likely to be composed of water ice, which is expected to form at the 7.2-bar level in the absence of any vertical motion (see Sections 1.3.2 and 4.1).

The cloud properties discussed in Chapters 4 and 5 will form the basis of the cloud model that is used in Chapter 6 to study gaseous abundances. In addition, Chapter 6 will further explore the degeneracies discussed in this chapter in order to determine their effect on retrievals of gaseous species.

Chapter 6

Disequilibrium species

6.1 Introduction

In this chapter, high resolution observations from VLT CRIRES are used to measure the abundances of three molecular species in Jupiter’s troposphere: germane (GeH_4), arsine (AsH_3) and phosphine (PH_3). GeH_4 , AsH_3 and PH_3 are all disequilibrium species in Jupiter’s troposphere, and their abundances are determined by temperature-dependent equilibrium reactions. Deep in the atmosphere, the temperature is high enough that they are chemically stable. The temperature in the observable troposphere is much lower, so the abundances are expected to rapidly drop off with height, as these gases are converted into other species (e.g. Fegley and Lodders, 1994). However, GeH_4 , AsH_3 and PH_3 have all been detected in Jupiter’s 5- μm window with higher abundances than would be expected from equilibrium chemistry (Fink et al., 1978; Noll et al., 1989; Ridgway et al., 1976). Further background about GeH_4 , AsH_3 and PH_3 are given in Sections 6.2.1, 6.3.1 and 6.4.1 respectively.

The apparently enhanced abundances are indicative of rapid vertical mixing, which brings the disequilibrium species upwards from deeper pressures where they are more abundant. Instead of dropping off rapidly with altitude, the abundance is ‘frozen in’ at a roughly constant value. This observed abundance corresponds to the equilibrium abundance at a much deeper level, known as the ‘quench level’, which depends on both the strength of vertical mixing and the chemical kinetics (rates of reaction) (Lewis and Fegley, 1984). The quench level for AsH_3 is at a temperature of at least 400 K (Fegley and Lodders, 1994), while the quench levels for GeH_4 and PH_3 are near 1000 K (Lewis

and Fegley, 1984). This means that disequilibrium species such as GeH_4 , AsH_3 and PH_3 act as tracers of vertical atmospheric dynamics in Jupiter’s troposphere (Taylor et al., 2004).

Because vertical mixing strengths vary as a function of latitude (Flasar and Gierasch, 1978; Wang et al., 2015), the tropospheric abundances of these species are also expected to vary. Wang et al. (2016) recently carried out a theoretical study where they combined diffusion-kinetics calculations with estimates for how Jupiter’s eddy diffusion coefficient varies with latitude, in order to predict the large-scale latitudinal distribution of disequilibrium species, including GeH_4 , AsH_3 and PH_3 . Because of the different chemical kinetics for the different gases, they predict that the GeH_4 abundance should be a maximum in the equatorial latitudes and should decrease towards the poles, whereas they expect AsH_3 and PH_3 to be latitudinally uniform (see Section 6.5 for more details).

This chapter uses the CRIRES observations to search for evidence of latitudinal variability in Jupiter’s tropospheric composition. Unless otherwise stated, the data used is from the November 2012 observations, as these provided a better signal-to-noise ratio. The cloud model developed in Chapters 4 and 5 is used, and the degeneracies with cloud structure that were discussed in Chapter 5 are further developed. Section 6.2 focusses on GeH_4 , Section 6.3 focusses on AsH_3 and Section 6.4 focusses on PH_3 . The results of these sections are discussed in Section 6.5, and are compared to the theoretical predictions from Wang et al. (2016). This chapter is based on the second half of Giles et al. (2016a).

6.2 GeH_4

6.2.1 Background

Germane (GeH_4) is a molecular species that only exists in thermochemical equilibrium deep in Jupiter’s atmosphere (Taylor et al., 2004). In the 298–2000 K range, GeH_4 is modelled to be the second most abundant Ge species (after GeS), but as the altitude increases and the temperature decreases, it precipitates out to form either GeS or GeSe (Fegley and Lodders, 1994). Because of this, GeH_4 was not expected to be present in observable quantities in the upper troposphere at the pressure levels that are

probed by the majority of the infrared spectrum. However, the 5- μm atmospheric window probes much deeper into the atmosphere, leading Corice and Fox (1972) to predict that GeH_4 absorption features might be visible in this part of the spectrum.

GeH_4 has two infrared active fundamental bands, ν_3 (4.737 μm) and ν_4 (12.210 μm) (Fink et al., 1978). Fortunately, ν_3 is located in the middle of the 5- μm window, providing the perfect opportunity to detect the disequilibrium species. It was the Q-branch of the ν_3 fundamental that was first identified as a jovian GeH_4 feature by Fink et al. (1978), using observations from the Kuiper Airborne Observatory (KAO). They derived a tropospheric GeH_4 abundance of 0.6 ppb. The same spectral feature was subsequently observed in data from the Infrared Interferometer Spectrometer (IRIS) on the Voyager 1 spacecraft (Kunde et al., 1982; Drossart et al., 1982). Using observations from the KAO, Bjoraker et al. (1986) later identified additional GeH_4 features, corresponding to the R3 and R6 transitions. In addition, they identified the Q-branch of the ν_1 fundamental at 4.738 μm ; this band is ‘forbidden’ under symmetry considerations, so the absorption lines are weaker than those belonging to the ν_3 fundamental (Lepage et al., 1981).

For these earlier analyses, the available spectroscopic data was restricted to a single isotopic species, $^{74}\text{GeH}_4$. In fact, there are five naturally occurring isotopes of germanium: ^{70}Ge , ^{72}Ge , ^{73}Ge , ^{74}Ge and ^{76}Ge . The terrestrial relative abundances are 20.6%, 27.5%, 7.8%, 36.5% and 7.7% respectively (Berglund and Wieser, 2011).

Bézard et al. (2002) was the first study to include line data for all five isotopic species of GeH_4 . They used theoretical line data which they scaled to match observations for a single isotope, assuming telluric relative abundances. Their derived VMR of 0.45 ppb was 35–50% lower than previous estimates, which they attributed to the use of the additional isotopes. This thesis takes a similar approach to the GeH_4 line data as Bézard et al. (2002). Theoretical line lists were generated for each isotope using the Spherical Top Data System (Wenger and Champion, 1998) and were then scaled to match the observed $^{74}\text{GeH}_4$ data from the GEISA database (Jacquinet-Husson et al., 1999). As with Bézard et al. (2002), telluric relative abundances were assumed. In the absence of information about the vertical profile, a constant tropospheric VMR was assumed (as in previous studies).

Previous studies of GeH_4 have generally been restricted to a single region of the planet: either an average over the centre of the disc (Fink et al., 1978; Bjoraker et al., 1986) or a focus on the bright NEB (Kunde et al., 1982; Bézard et al., 2002). The exception to this is Drossart et al. (1982), who found that within a factor of 2, the same GeH_4 abundance was able to fit the range of spectra from 30°S to 30°N . This section uses the improved spectral resolution afforded by CRIRES to repeat this latitudinal study and to extend it to higher latitudes.

6.2.2 Identification of GeH_4 features

The first task is to identify the GeH_4 absorption features in the $5\text{-}\mu\text{m}$ spectra. Previous studies show that there is a strong Q-branch feature at $4.737\text{ }\mu\text{m}$. In a preliminary investigation, it was clear that this absorption feature was particularly strong in the bright SEB; this spatial region was therefore used in order to search for additional GeH_4 absorption features.

The identifiable GeH_4 absorption features in the CRIRES spectra are shown in Figure 6.2.1. The NEMESIS retrieval algorithm was used to model the data. The best fit from the case where all gaseous species are allowed to vary is shown in red. In order to highlight the shape and location of the GeH_4 absorption features, the blue line shows what the spectrum would look like in the absence of GeH_4 . Each feature is labelled and consists of a blend of the five isotopologues.

The Q, R3 and R6 bands have been identified in previous studies (Fink et al., 1978; Bjoraker et al., 1986). In addition, the R7 line is identified, located at $4.639\text{ }\mu\text{m}$. From each of these four spectral features, the following GeH_4 abundances are retrieved: 0.51 ppb (R7), 0.62 ppb (R6), 0.45 ppb (R3), 0.58 ppb (Q). These small differences could reflect genuine differences, due to each line probing a slightly different pressure level, or they could be due to inaccuracies in the line data. As each segment was observed at a different time, and therefore a different longitude, a simultaneous retrieval of all four features is not possible.

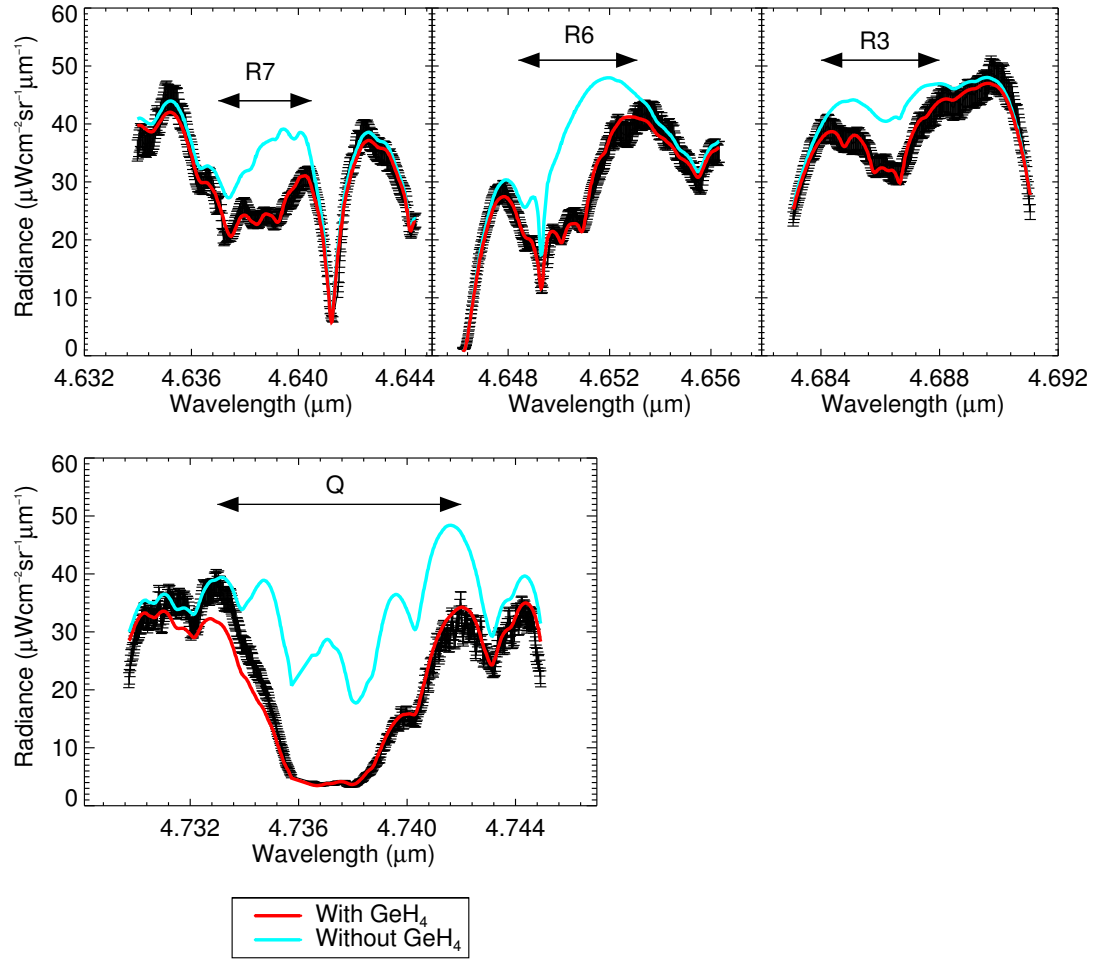


Figure 6.2.1: Identification of GeH₄ absorption features in the 5- μm window. CRIRES data are shown in black and are taken from the SEB. The best fits obtained from two retrievals are shown: with GeH₄ present (red) and with no GeH₄ present (blue).

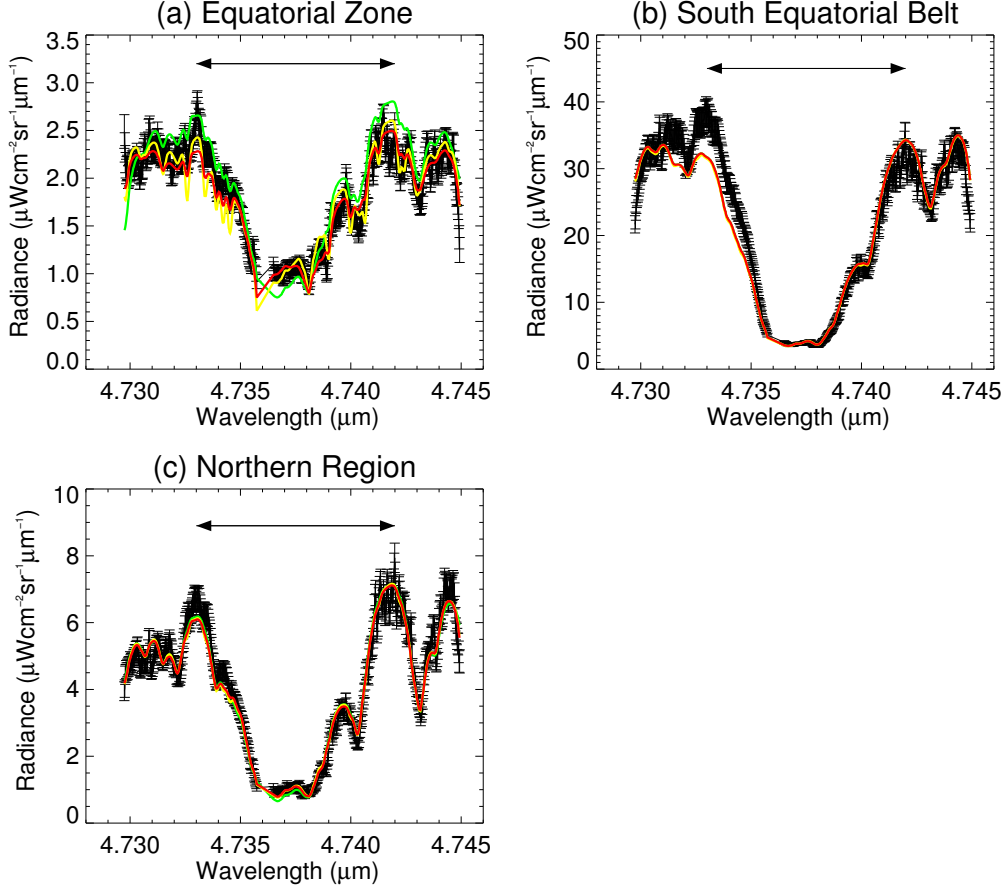


Figure 6.2.2: GeH_4 absorption feature fits in the Equatorial Zone, South Equatorial Belt and Northern Region ($50\text{--}55^\circ\text{N}$). The CRIRES observations are shown in black. The coloured lines show the best fit that can be achieved using different assumptions. Red: single cloud deck with $g = 0.8$, GeH_4 allowed to vary. Yellow: single cloud deck with $g = 0.4$, GeH_4 allowed to vary. Green: upper cloud deck with $g = 0.8$, additional deep cloud deck, GeH_4 held fixed at 0.58 ppb (the SEB best-fit value).

6.2.3 Spatial variability

Having identified GeH_4 absorption features in one discrete region of the planet (the SEB), this section now considers how this varies with latitude. This analysis is restricted to the most prominent GeH_4 feature, the Q-branch at $4.734\text{--}4.742\text{ }\mu\text{m}$.

Figure 6.2.2 shows the same absorption feature for three different regions of the planet: the EqZ, the SEB and a ‘Northern Region’, corresponding to $50\text{--}55^\circ\text{N}$. Comparing the data, shown in black, from the three regions highlights some differences: the absorption feature appears to be deepest in the SEB and shallowest in the EqZ. This would appear to suggest that there is less GeH_4 present in the EqZ. However, Chapter 5 has already demonstrated a degeneracy between cloud properties and retrieved gaseous

abundances. Chapter 5 showed that the depth of an absorption feature can depend on both the scattering properties of the main cloud deck and the presence/absence of a deep cloud deck. Since the primary difference in line shape is between the EqZ and SEB, which have very different cloud structures, it is likely that clouds play an important role.

In order to explore whether or not the CRIRES observations provide evidence for spatial variability in GeH_4 , a series of retrievals were performed on these three spectra, making different assumptions about the cloud structure. The abundances of all the gaseous species were allowed to vary, as described in Section 2.6. The red line shows the case where there is a single upper cloud deck, with an asymmetry parameter $g = 0.8$. The opacity of this cloud is allowed to vary in the retrievals. The retrieved GeH_4 abundances are 0.19 ppb (EqZ), 0.58 ppb (SEB) and 0.44 ppb (Northern Region); in this case, the shallow feature in the EqZ does indeed lead to a lower retrieved abundance.

The yellow line in Figure 6.2.2 shows the case where the asymmetry parameter of the cloud is changed to 0.4. Because the SEB and Northern Region have low optical thicknesses, this has a negligible effect on those retrievals (now 0.59 ppb and 0.47 ppb respectively). In contrast, there is a significant effect for the EqZ, increasing the retrieved abundance from 0.19 ppb to 0.47 ppb. Changing the asymmetry parameter (i.e. making the cloud particles less forward scattering) can therefore account for much of the apparent difference between the EqZ and the SEB. While Chapter 5 did show that $g = 0.8$ provides a better fit to the data than $g = 0.4$, it should be noted that this does provide a possible explanation for variability in line shape.

A more plausible explanation for the different line shapes is the presence/absence of a deep cloud. In Section 5.3.1, the CH_3D abundance was fixed to the SEB value, and the EqZ was fit by adding a deeper cloud deck. The same process is repeated here; the upper cloud is returned to an asymmetry parameter of 0.8, the GeH_4 abundance is fixed to the SEB value (0.58 ppb) and the opacity of a deep cloud at 5 bar is allowed to vary in the retrievals (along with the opacity of the upper cloud). The green line in Figure 6.2.2 shows the fits obtained for this case. The retrieved deep cloud is opaque in the EqZ and transparent in the SEB, just as in Section 5.3.1. This shows that the deep cloud structure studied Chapter 5 can account for the difference in line shape between the EqZ and SEB.

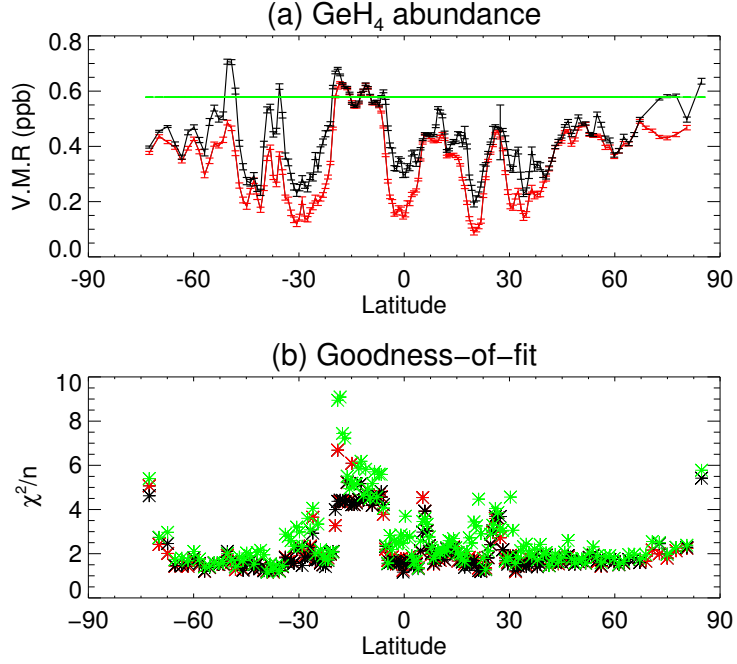


Figure 6.2.3: GeH₄ abundance and goodness-of-fit values as a function of latitude, for three different assumptions. Red: single cloud deck with $g = 0.8$, GeH₄ allowed to vary. Black: additional deep cloud deck with opacities derived from Chapter 5, GeH₄ allowed to vary. Green: deep cloud deck allowed to vary, GeH₄ held fixed at 0.58 ppb.

6.2.4 Latitudinal retrieval

Section 6.2.3 showed that there is no evidence for spatial variability in GeH₄ between three discrete regions of the planet, as any differences in line shape can be accounted for by changes in the cloud structure alone. In this section, this analysis is extended to cover the full range of latitudes available. Figure 6.2.3 shows the GeH₄ abundance as a function of latitude, retrieved with different assumptions. Alongside this is the goodness-of-fit (χ^2/n), which describes the quality of the fit at each latitude point. As with Figure 6.2.2, the three colours correspond to different assumptions made in the retrievals.

The red line corresponds to the simplest case, where there is a single upper cloud deck (with $g = 0.8$) and the GeH₄ abundance is allowed to vary, i.e. the same as the red line in Figure 6.2.2. Section 6.2.3 showed that this simple case led to a variability in the retrieved GeH₄ abundance, with a depletion in the EqZ and an enhancement in the SEB. Figure 6.2.3 shows that this applies at all latitudes, and so this simple assumption leads to an apparent belt-zone variability.

The black line corresponds to the case where an additional deep cloud is included in the model and its latitudinal profile is fixed to the values retrieved in Chapter 5 (shown in Figure 5.3.3). It should be noted that this previously determined deep cloud latitudinal profile relates to a different longitude, so it is not necessarily expected to be identical to the cloud profile at this longitude. Nevertheless, the overall latitudinal trend is likely to be very similar and it provides an insight into the role of the deep cloud on the retrieved abundances. Once again, the GeH_4 abundance is allowed to vary in the retrievals. It can be seen in Figure 6.2.3a that the presence of this deep cloud increases the retrieved abundances in the zones, decreasing the apparent belt-zone variability shown by the red line. Figure 6.2.3b shows that this change does not alter the quality of the fits.

For the green line, the GeH_4 abundance is held fixed as a function of latitude and the opacity of the deep cloud is allowed to vary, instead of being held fixed at the values determined in Chapter 5. This is the same as the green lines in Figure 6.2.2. As in Section 6.2.3, the GeH_4 abundance chosen is the best-fit value from the SEB (0.58 ppb). Since the SEB is relatively cloud-free, there are no complications from the cloud structure and this should represent the ‘true’ abundance. The goodness-of-fit in Figure 6.2.3 shows that this constant abundance is able to provide a similar fit to the data at all latitudes. Section 6.2.3 previously showed that variability in the deep cloud opacity could account for the difference in line shape between three discrete regions of the planet, and Figure 6.2.3 shows that this remains true across the full range of latitudes.

This is further confirmed by Figure 6.2.4, which shows (in green) the retrieved deep cloud opacity in this last case, where the GeH_4 abundance is held constant. This is compared to the earlier results (in black) from Chapter 5, obtained using CH_3D . Both lines show a very similar trend of high opacity in the EqZ and low opacity in the adjacent belts. There are some small differences between the two lines, but this is unsurprising as the observations relate to two different longitudes. The latitudinal cloud profile required to ‘balance out’ a flat GeH_4 distribution appears consistent with the earlier results.

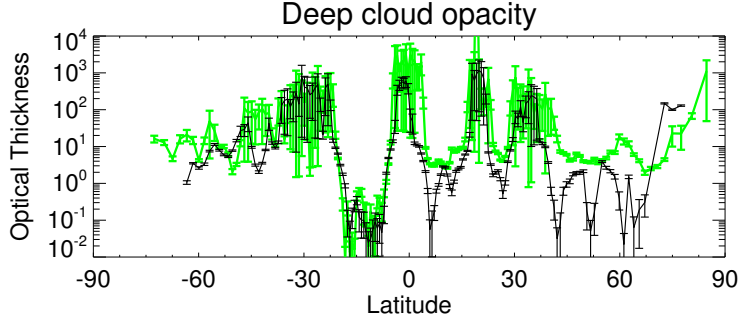


Figure 6.2.4: Deep cloud opacity as a function of latitude. Green: the retrieved deep cloud abundance required to fit the GeH_4 absorption features, when the GeH_4 abundance is held fixed. Black: the retrieved deep cloud abundance required to fit the CH_3D absorption features, when the CH_3D abundance is held fixed (identical to Figure 5.3.3).

6.2.5 Summary

This section identified four GeH_4 absorption features in the CRIRES observations, one of which has not been previously identified. The strongest absorption feature was used to search for latitudinal variability and it was found that there are some differences in the line depth between the EqZ, the SEB and the Northern Region of the planet. However, this difference in line shape does not necessarily indicate a difference in the abundance of GeH_4 , as the variation in line shape could instead be accounted for by Jupiter's tropospheric cloud structure. This analysis was then extended to all latitudes, and it was found that a single fixed GeH_4 abundance could be used to fit all spectra without compromising the quality of the fits. The CRIRES observations therefore do not provide any evidence for spatial variability in GeH_4 . This highlights the high level of degeneracy present in the problem. The deep cloud structure, which has not been included in any previous studies of GeH_4 , significantly complicates the analysis. Future observations that simultaneously cover strong CH_3D and GeH_4 absorption lines could provide an opportunity to study GeH_4 variability; if the CH_3D line shape remains constant between different spatial regions of the planet, but the GeH_4 line shape varies, this could indicate genuine GeH_4 variability.

6.3 AsH₃

6.3.1 Background

Arsine (AsH₃) is the most abundant arsenic gas on Jupiter. Deep in the atmosphere it exists in thermochemical equilibrium but, in the absence of vertical motion, it precipitates into As₄(s) or As₂S₂(s) at ~ 400 K (Fegley and Lodders, 1994). Strong upwelling can bring the gas further upwards into the middle troposphere. As with GeH₄, the 5- μ m atmospheric window provides an opportunity to probe deep into the atmosphere where the species is present in observable quantities.

AsH₃ has two fundamental bands within the 5- μ m window: ν_1 at 4.728 μ m and ν_3 at 4.704 μ m. Of these, the ν_3 band is stronger and its Q-branch led to the first detection of AsH₃ on Jupiter by Noll et al. (1989), using spectra obtained from the United Kingdom Infrared Telescope. They initially estimated the deep abundance to be $0.7^{+0.7}_{-0.4}$ ppb, which was then revised down to 0.22 ± 0.11 ppb the following year, after acquisition of new airborne and ground-based spectra and additional laboratory line data (Noll et al., 1990). More recently, Bézard et al. (2002) used data from the Canada-France-Hawaii Telescope to obtain an abundance of 0.24 ppb. The Q-branch of the ν_3 band remains the only AsH₃ feature to have been identified on Jupiter; this section searches for evidence of additional AsH₃ absorption lines. As with GeH₄, it is assumed that AsH₃ is well-mixed in the troposphere.

Out of the previous studies of AsH₃, only Noll et al. (1990) studied more than one region of the planet. They made observations of both the EqZ and the NEB and found that these spectra matched to within the noise levels. As the CRIRES observations provide both high spectral resolution and high spatial resolution, this section extends the analysis to cover all latitudes.

6.3.2 Identification of AsH₃ features

As with GeH₄, the initial task is to identify the AsH₃ absorption features in the 5- μ m spectrum. The only previously identified feature is the Q-branch at 4.704 μ m; in a preliminary investigation, it was determined that this feature was particularly strong at high latitudes (50–55°N), and so this ‘Northern Region’ is used in order to search for

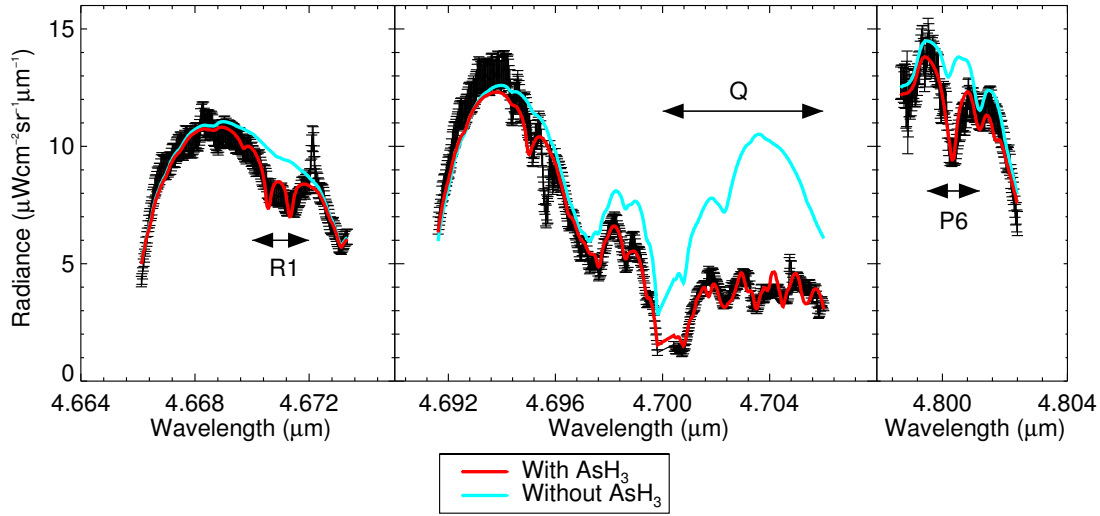


Figure 6.3.1: Identification of AsH_3 absorption features in the 5- μm window. CRIREs data are shown in black, and are taken from the Northern Region ($50\text{--}55^\circ\text{N}$) of the planet. The best fits obtained from two retrievals are shown: with AsH_3 present (red) and with no AsH_3 present (blue).

additional absorption lines.

Two additional absorption features were identified, corresponding to the R1 and P6 lines of the ν_3 band (Dana et al., 1993). These lines are shown in Figure 6.3.1, alongside the Q-branch. The CRIREs data are shown in black. The blue lines show the best fit that can be achieved when no AsH_3 is present in the atmosphere and the red lines show the best fit that can be achieved when the AsH_3 abundance is allowed to vary. As in Section 6.2.2, the cloud opacity of the single cloud deck and the gaseous abundances were also allowed to vary. In each case, the addition of AsH_3 significantly improves the fit. The following abundances were obtained from each retrieval: 0.60 ppb (R1), 0.49 ppb (Q) and 0.46 ppb (P6).

6.3.3 Spatial variability

This section now considers how the AsH_3 line shape varies with latitude, and how this affects the retrieved abundances. As in previous studies, the most prominent AsH_3 absorption feature in the 5- μm window is used: the Q-branch of the ν_3 fundamental, located at 4.704 μm . Figure 6.3.2 shows this spectral region for the three spatial regions of the planet considered in Section 6.2. Unlike the previous GeH_4 section, the EqZ and the SEB have similar spectral shapes. In this case, however, there is significant

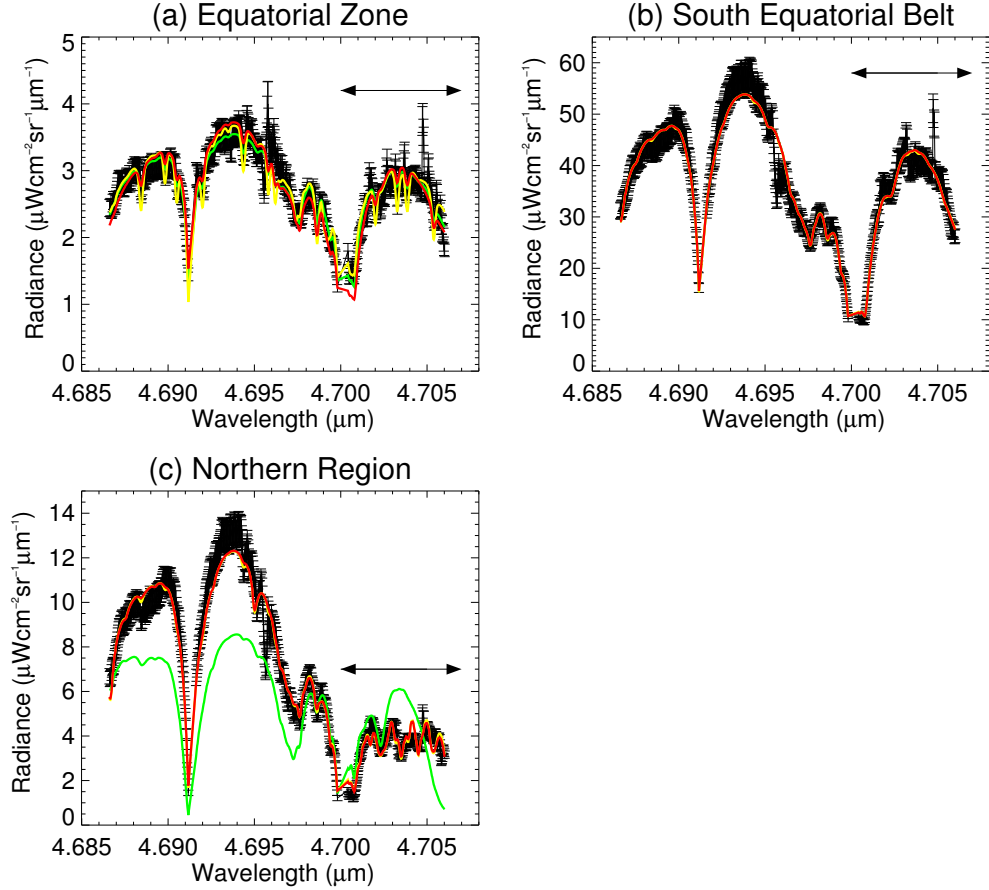


Figure 6.3.2: AsH_3 absorption feature fits at different latitudes. The CRIRCS observations are shown in black. The coloured lines show the best fit that can be achieved using different assumptions. Red: single cloud deck with $g = 0.8$, AsH_3 allowed to vary. Yellow: single cloud deck with $g = 0.4$, AsH_3 allowed to vary. Green: upper cloud deck with $g = 0.8$, additional deep cloud deck, AsH_3 held fixed at 0.02 ppb (the SEB best-fit value).

variation between these spectra and the spectrum from the Northern Region. The absorption feature at $4.704 \mu\text{m}$ is considerably flatter in Northern Region than it is in the SEB and the EqZ, suggesting a higher abundance of AsH_3 at high latitudes.

This is confirmed by a simple NEMESIS retrieval, shown in red in Figure 6.3.2. This fit is analogous to the red line in Figure 6.2.2, where the cloud structure consists of a single upper cloud with $g = 0.8$. With these assumptions, the following retrieved abundances for AsH_3 are obtained: 0.01 ppb in the EqZ, 0.02 ppb in the SEB and 0.49 ppb in the Northern Region.

Section 6.2 showed that the apparent change in GeH_4 line shape could be accounted for by variations in the cloud structure, rather than changes in the GeH_4 abundance itself. However, clouds are unlikely to be responsible here, since the EqZ and the SEB

have very different cloud structures, and yet the AsH_3 features have similar spectral shapes. The observed effect is also unlikely to be due to limb darkening at high altitudes. Firstly, previous studies have shown that Jupiter’s tropospheric clouds are highly scattering, which minimises the effect of limb darkening (Chapter 4; Roos-Serote and Irwin, 2006). Secondly, even if there was a significant limb-darkening effect, the opposite phenomenon would be seen. At higher latitudes, the emission angle increases, and so higher altitudes in the atmosphere are probed, where (as a disequilibrium species) AsH_3 is *less* abundant, not more.

Nevertheless, in order to fully explore any degeneracy, the different retrievals performed in Section 6.2.3 for GeH_4 were repeated. The yellow line in Figure 6.3.2 shows the fits that are obtained when the asymmetry parameter of the upper cloud is changed from 0.8 to 0.4. The retrieved abundance in the EqZ is increased from 0.01 ppb to 0.05 ppb, and the SEB and Northern abundances remain essentially unchanged at 0.02 ppb and 0.50 ppb respectively. The gap between the equatorial regions and the high latitudes cannot be closed by changing the scattering properties of the upper cloud.

The effect of a deep cloud is now considered. The AsH_3 abundance was held constant at the SEB level (0.02 ppb) and the opacity of a deep cloud was allowed to vary. The results are shown by the green line in Figure 6.3.2. As in Section 6.2, the EqZ and SEB can be fit using the same AsH_3 abundance, provided that the deep cloud is optically thick in the EqZ. However, it is clear from Figure 6.3.2c that this constant abundance cannot be applied to the Northern Region of the planet. It can therefore be concluded that the spatial variation in the AsH_3 line shape is indicative of genuine latitudinal variability in the abundance of AsH_3 .

6.3.4 Latitudinal retrieval

Having established that there is evidence for spatial variability in AsH_3 , the analysis is now extended from three discrete sections to a full latitudinal retrieval of AsH_3 . Figure 6.3.3 shows the latitudinal distribution of AsH_3 alongside the goodness-of-fit values. The three different colours correspond to the same assumptions described in Section 6.2.4.

For the latitudinal distribution shown by the red line, there is a single upper cloud

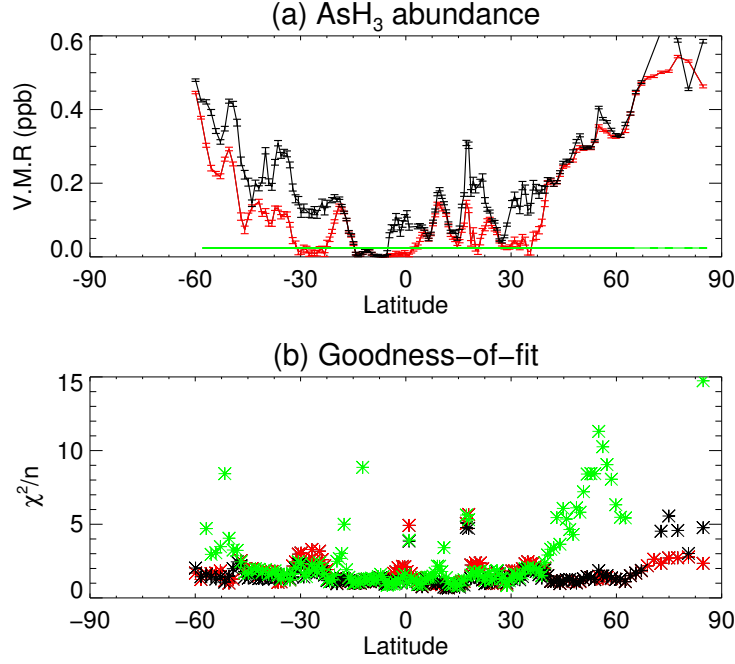


Figure 6.3.3: AsH₃ abundance and goodness-of-fit values as a function of latitude, for three different assumptions. Red: single cloud deck with $g = 0.8$, AsH₃ allowed to vary. Black: additional deep cloud deck with opacities derived from Chapter 5, AsH₃ allowed to vary. Green: deep cloud deck allowed to vary, AsH₃ held fixed at 0.02 ppb.

deck with $g = 0.8$ and the AsH₃ abundance is allowed to vary, i.e. the same as the red line in Figure 6.3.2. Section 6.3.3 showed that this simple case produced similar retrieved abundances in the EqZ and SEB and an enhanced abundance in the Northern Region. This pattern is confirmed by the full latitudinal retrieval; there is a roughly symmetrical distribution, with an increased abundance at high latitudes.

The black line shows the case where an additional deep cloud is included in the model, with the latitudinal profile obtained from Chapter 5. While the precise values of the retrieved abundances do change slightly with the inclusion of this additional cloud deck, the overall trend of an enhancement at high latitudes remains true. The errors shown in Figure 6.3.3 represent the formal errors on the retrieval; the difference between the red and black lines shows that these formal errors are significantly smaller than the errors due to varying assumptions about cloud structure. As with Section 6.2.4 for GeH₄, the inclusion of an additional cloud does not alter the quality of the fits.

This is in contrast to the final case, shown by the green line. In this case, the AsH₃ abundance is held fixed at the SEB value of 0.02 ppb and the deep cloud is allowed to vary. Section 6.3.3 showed that this can provide a good fit in the EqZ and SEB, but fails

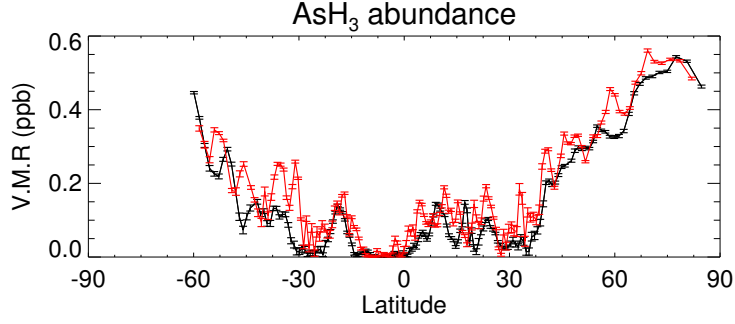


Figure 6.3.4: AsH_3 abundance as a function of latitude from the 12 November 2012 dataset (black) and the 1 January 2013 dataset (red). In both cases, a single cloud deck with $g = 0.8$ is assumed and the AsH_3 abundance is allowed to vary. The two datasets produce consistent results.

to provide a good fit for the Northern Region. Figure 6.3.3 extends this to all latitudes and shows that this constant abundance can provide a good fit between 40°S and 40°N , i.e. there is no evidence for belt-zone variability. However, the fit worsens considerably at high latitudes; an abundance of 0.02 ppb simply cannot fit these regions, regardless of the opacity of the deep cloud. This leads to the conclusion that there is a genuine enhancement at high latitudes.

Figure 6.3.3 is restricted to the data from 12 November 2012. However, Figure 6.3.4 shows that the 1 January 2013 dataset produces consistent results, and that the overall conclusion of an AsH_3 enhancement at high latitudes is reproducible.

6.3.5 Summary

This section identifies three AsH_3 absorption features in the CRIRES observations, two of which have not been previously identified. The strongest absorption feature was used to search for latitudinal variability and it was found that there are significant differences in the line shape between spectra from the equatorial regions (EqZ and SEB) and spectra from the far northern latitudes. These differences cannot be accounted for by Jupiter's cloud structure, and it can therefore be concluded that there is evidence for latitudinal variability in the abundance of AsH_3 . This was further shown by a latitudinal retrieval, which demonstrated that a constant AsH_3 abundance is inconsistent with the observations. Instead, the retrieved AsH_3 abundances have a roughly symmetrical latitudinal profile, with a minimum in the equatorial regions and enhanced abundances at high latitudes. Finally, it was shown that this effect can be seen in both the 12

November 2012 and the 1 January 2013 datasets.

6.4 PH₃

6.4.1 Background

Like GeH₄ and AsH₃, phosphine (PH₃) is a disequilibrium species in Jupiter's troposphere. Deep in the atmosphere it is the primary phosphorus gas but at higher altitudes, thermal decomposition produces PH, which then reacts with OH radicals to form P₄O₆ (Fegley and Lodders, 1994). However, the solar abundance of P is much greater than the solar abundances of Ge or As (Grevesse et al., 2007). This means that even though the VMR of PH₃ decreases rapidly with altitude, it is still present in observable quantities in the upper troposphere and is therefore detectable outside the 5- μ m window.

The strongest PH₃ absorption features in the infrared can be seen at ~ 5 μ m and at ~ 10 μ m. PH₃ was simultaneously detected in both of these spectral regions. Ridgway et al. (1976) noted that including a solar abundance of PH₃ in their model improved their fit to 10- μ m observations of Jupiter from the McMath Solar Observatory. This was then confirmed by Larson et al. (1977), who also found that a roughly solar abundance of PH₃ was able to fit their 5- μ m airborne observations.

The 5- μ m and 10- μ m spectral regions probe different pressure levels in Jupiter's troposphere. At 5- μ m, the spectrum is primarily sensitive to the lower troposphere ($p > 1$ bar) whereas the 10- μ m observations are mostly sensitive to the upper troposphere, at pressures $p < 1$ bar. This pressure difference allows us to start to constrain the vertical profile of PH₃. Fletcher et al. (2009) modelled Jupiter's PH₃ abundance using a constant abundance of 1.86 ppm up to 1 bar, above which it drops off with a constant fractional scale height of 0.3. In the absence of further information, this section assumes the profile from Fletcher et al. (2009) and varies it via a single scaling parameter; because the CRIRES data are from the 5- μ m region, it is probing the deep abundance, i.e. the region with a constant VMR.

Several previous studies have also considered the possibility of spatial variability in the deep PH₃ abundance, as observed in the 5- μ m window. Drossart et al. (1982)

and Lellouch et al. (1989) searched for a correlation between cloud opacity and PH_3 abundance, but did not find any evidence. More recently, Drossart et al. (1990) found evidence for a PH_3 enhancement at high latitudes compared to equatorial- and mid-latitudes, and this was also seen in the results of Chapter 4. The spatially resolved 10- μm studies show a different pattern; the retrieved fractional scale height shows a global maximum over the equator and global minima at the NEB and SEB (Irwin et al., 2004; Fletcher et al., 2009).

6.4.2 Spatial variability

For GeH_4 and AsH_3 , the choice of spectral region was clear, as there were very few absorption features in the 5- μm window. In contrast, PH_3 is one of the primary absorbers at 5- μm , and there are many absorption features. In particular, the short-wavelength part of the 5- μm spectrum (4.50–5.00 μm) is dominated by PH_3 absorption. This might sound ideal, but in fact this prevents the accurate retrieval of PH_3 , as it becomes difficult to distinguish between PH_3 and other broad features such as the cloud opacity and the water vapour abundance. Instead, the long-wavelength edge proved more useful, where PH_3 absorption contributes less to the continuum. There is a clear, well-separated absorption feature at 5.071 μm and this will be used as the basis of the study of the deep PH_3 abundance in the troposphere.

Figure 6.4.1 shows this absorption feature for the same three spatial regions used in Sections 6.2.3 and 6.3.3. The CRIRES spectra in Figure 6.4.1 show variation in the depth of the PH_3 absorption feature; the feature is considerably deeper in the Northern Region than in the EqZ, while the SEB spectrum is between these two extremes.

The coloured lines show the best fit obtained for each spectrum, with the same assumptions as in Sections 6.2.3 and 6.3.3: the red line shows the case where there is a single upper cloud with $g = 0.8$ and the yellow line shows the case where there is a single upper cloud with $g = 0.4$. In the first case (red), the retrieved abundances in the EqZ, SEB and Northern Region are 0.49 ppm, 0.76 ppm and 1.22 ppm respectively. In the second case (yellow), these become 0.81 ppm, 0.75 ppm and 1.21 ppm. As in Section 6.3.3 for AsH_3 , altering the scattering properties of the cloud can account for the discrepancy between the EqZ and the SEB but not between the equatorial regions

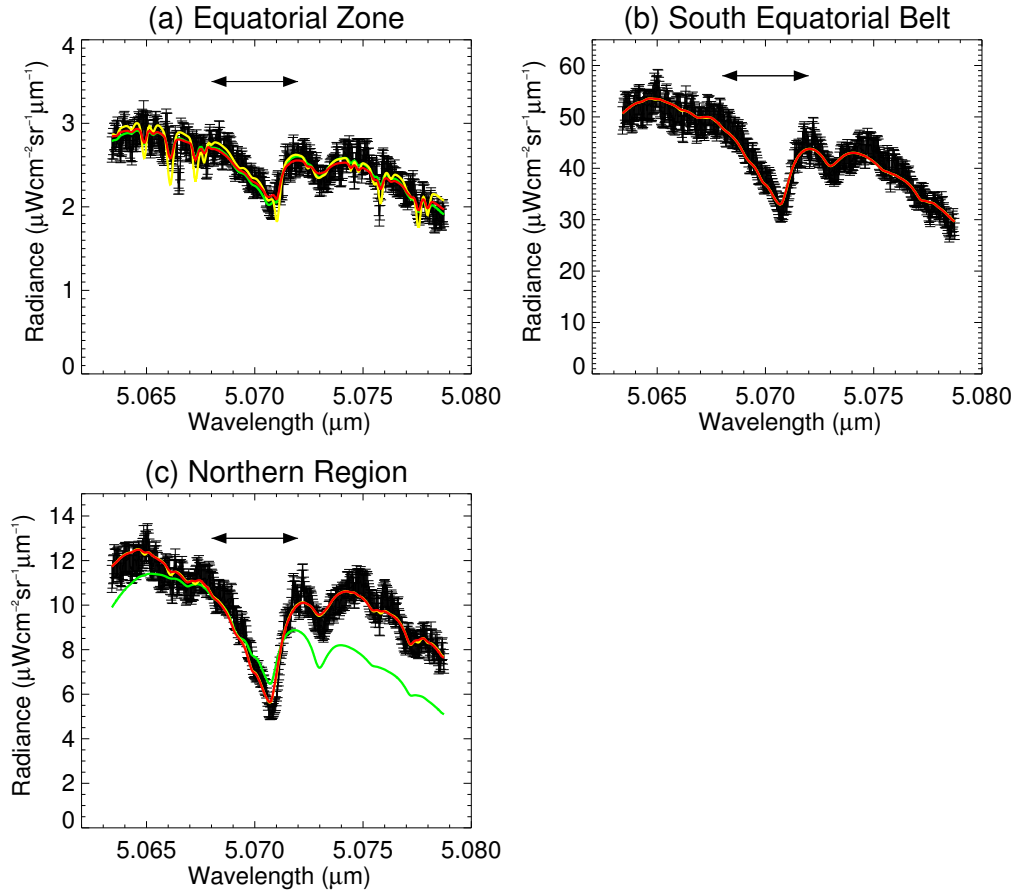


Figure 6.4.1: PH₃ feature fits at different latitudes. The CRIRES observations are shown in black. The coloured lines show the best fit that can be achieved using different assumptions. Red: single cloud deck with $g = 0.8$, PH₃ allowed to vary. Yellow: single cloud deck with $g = 0.4$, PH₃ allowed to vary. Green: upper cloud deck with $g = 0.8$, additional deep cloud deck, PH₃ held fixed at 0.76 ppm (the SEB best-fit value).

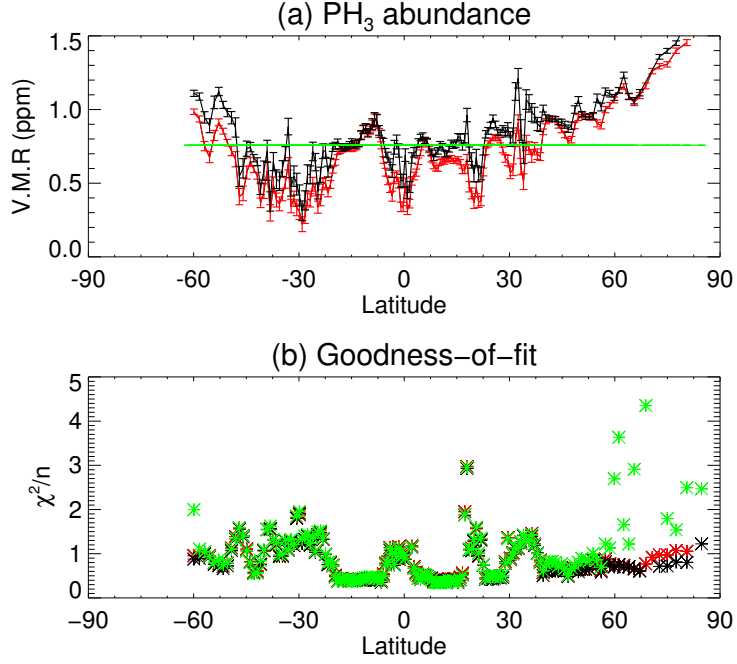


Figure 6.4.2: PH₃ abundance and goodness-of-fit values as a function of latitude, for three different assumptions. Red: single cloud deck with $g = 0.8$, PH₃ allowed to vary. Black: additional deep cloud deck with opacities derived from Chapter 5, PH₃ allowed to vary. Green: deep cloud deck allowed to vary, PH₃ held fixed at 0.76 ppm.

and the polar regions.

The green line in Figure 6.4.1 shows the case where the upper cloud has $g = 0.8$, the PH₃ abundance is fixed to the SEB value (0.76 ppm) and the opacity of a second deep cloud is allowed to vary. Again, the results are the same as for AsH₃ in Section 6.3.3. The presence of an optically thick deep cloud in the EqZ can account for the change in spectral shape between the EqZ and the SEB, but the same PH₃ abundance cannot be used to fit the regions at high latitude. It is therefore concluded that there is evidence for latitudinal variability in PH₃.

6.4.3 Latitudinal retrieval

Having previously focussed on three discrete sections of the planet, full latitudinal retrievals of PH₃ can now be performed. The results of these retrievals are shown in Figure 6.4.2; these results take the same form as Figure 6.2.3 (for GeH₄) and Figure 6.3.3 (for AsH₃) and the same conclusions can be drawn for PH₃ as for AsH₃.

As with AsH₃, the black and red lines in Figure 6.4.2 both show a polar enhancement in PH₃, whether or not the additional deep cloud from Chapter 5 is included in the

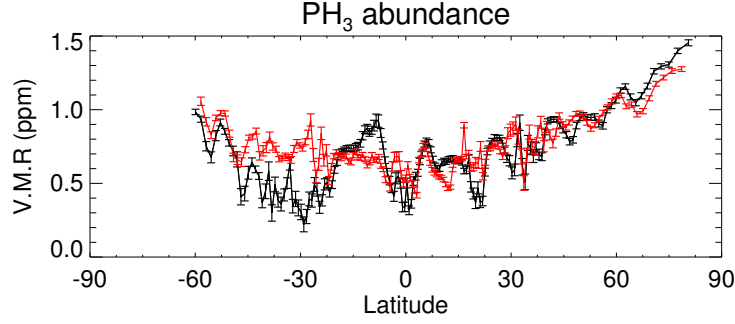


Figure 6.4.3: PH_3 abundance as a function of latitude from the 12 November 2012 dataset (black) and the 1 January 2013 dataset (red). In both cases, a single cloud deck with $g = 0.8$ is assumed and the AsH_3 abundance is allowed to vary. The two datasets produce consistent results.

model. The case where the PH_3 abundance is held fixed at 0.76 ppm is shown in green, and this also follows the same pattern as AsH_3 ; at low latitudes, this model provides a good fit (i.e. there is no evidence for belt-zone variability), but the fit starts to worsen at high latitudes (in this case, above $\sim 60^\circ\text{N}$). This agrees with the conclusion made in Section 6.4.2 that there is evidence for an enhancement in PH_3 at high latitudes, with the retrieved abundance varying from ~ 0.5 ppm near the equator to 1.5 ppm at 80°N .

Figure 6.4.2 is restricted to the data from 12 November 2012. However, as with AsH_3 , the 1 January 2013 dataset produces consistent results (see Figure 6.4.3), showing that the overall conclusion of an PH_3 enhancement at high latitudes is reproducible.

6.4.4 Summary

This section considered how the strength of a well-isolated PH_3 absorption line varies with latitude. The depth of the absorption feature was found to be greater for the high northern latitudes than for the equatorial latitudes. As with AsH_3 , this section showed that this was due to higher PH_3 abundances at high latitudes. A latitudinal retrieval was then performed, and this showed a minimum near the equator and an enhancement at high latitudes.

6.5 Discussion

6.5.1 Abundances

In Sections 6.2–6.4, the abundances of three disequilibrium species in Jupiter’s troposphere were measured: GeH_4 , AsH_3 and PH_3 . For GeH_4 , the initially retrieved abundances varied between 0.19 ppb (EqZ) and 0.58 ppb (SEB). By including a variable deep cloud, all spatial regions could be fit using a tropospheric abundance of 0.58 ppb. This is similar to previous results, which range from 0.45 ppb (Bézard et al., 2002) to 1.0 ppb (Drossart et al., 1982). This is significantly lower than the solar abundance of 6.7 ppb (Grevesse et al., 2007), but this is unsurprising as solid GeS , rather than gaseous GeH_4 , is predicted to be the most abundant Ge-bearing species at the temperatures found in Jupiter’s observable troposphere (Fegley and Lodders, 1994).

Unlike GeH_4 , measurements of the AsH_3 absorption feature showed evidence for an enhancement at high latitudes. The retrieved abundance varies from as low as 0.01 ppb in parts of the equatorial region to 0.55 ppb at 80°N . Previous measurements of the AsH_3 abundance have relied exclusively on data from the low latitudes of the planet, and have led to estimates of 0.2–0.3 ppb (Bézard et al., 1989; Noll et al., 1990; Bézard et al., 2002). Wang et al. (2016) notes that AsH_3 is expected to be the primary As-bearing gas in the troposphere, and that these previous studies therefore suggest that the jovian abundance of As is lower than the solar value (a solar abundance would lead to a volume mixing ratio of 0.34 ppb, Grevesse et al., 2007). This is surprising, as other heavy elements show an enrichment relative to solar abundances (e.g. Taylor et al., 2004). The results from this chapter show that in some parts of the planet, the observed AsH_3 abundance is subsolar, while in other regions, AsH_3 shows an enrichment of up to 1.6 times the solar value. It is possible that this higher abundance is more representative of the deep abundance of As.

As with AsH_3 , the retrieved PH_3 abundance varies with latitude, ranging from 0.5 ppm in the equatorial regions to 1.4 ppm at 80°N . These values are consistent with previous $5\text{-}\mu\text{m}$ observations of PH_3 : 0.7 ppm (Bjoraker et al., 1986), 0.77 ppm (Irwin et al., 1998) and 0.76–0.90 ppm (Chapter 4). As noted in Chapter 4 and in previous studies (Fletcher et al., 2011), measurements at $5\text{ }\mu\text{m}$ appear to consistently lead to lower

retrieved abundances than measurements at 10 μm ; these differences are potentially due to errors in line data or cloud modelling uncertainties. Assuming a solar abundance of P would lead to a PH_3 volume mixing ratio of 0.43 ppm (Grevesse et al., 2007), so the results in this chapter show a variation from roughly solar levels to an enrichment of up to 3.3 times more the solar value, which is consistent with the observed enhancement of other heavy elements in Jupiter’s atmosphere (Lunine et al., 2004).

6.5.2 Latitudinal profiles

In Sections 6.2–6.4, the latitudinal distributions of GeH_4 , AsH_3 and PH_3 were studied. No evidence for spatial variability in the abundance of GeH_4 was found. This result for GeH_4 agrees with the previous study of Drossart et al. (1982) who found no evidence for variability in the 30°S to 30°N region, and this chapter shows that this remains true at higher latitudes. This is in contrast to the results for AsH_3 and PH_3 . While the 5- μm observations provide no evidence for any belt-zone variability in the middle troposphere, both of these species exhibit an enhancement towards the poles. An enhancement at high latitudes has previously been reported for PH_3 (Chapter 4; Drossart et al., 1990), but this is the first time that it has been seen in AsH_3 , as the only previous studies to compare different regions of the planet focussed exclusively on the equatorial latitudes (Noll et al., 1990).

The results in this chapter contrast with a recent study from Wang et al. (2016), who made theoretical predictions for the latitudinal distribution of disequilibrium gases. In their work, they use a diffusion-kinetics code to investigate how the abundances of disequilibrium species in Jupiter’s troposphere depend on the vertical eddy diffusion coefficient, K_{eddy} . They find that GeH_4 has a different behaviour to AsH_3 and PH_3 , which agrees with the CRIRES observations. The GeH_4 abundance is found to be very sensitive to the eddy diffusion coefficient, with a higher value of K_{eddy} leading to a higher tropospheric abundance. In contrast, PH_3 and AsH_3 do not appear to have any dependence on K_{eddy} (for a physically plausible range of K_{eddy}). These differences are due to the interplay between the quench level for the gas (the pressure at which the abundance is ‘frozen in’) and the equilibrium vertical profile. If the quench level is deep in the regime where the gas is well-mixed, then a small change in K_{eddy} will not

alter the observed abundance. If the quench level is in a regime where the equilibrium abundance is rapidly changing with altitude, then a small change in K_{eddy} can have a significant effect on the observed abundance.

Wang et al. (2016) consider how abundances vary as a function of K_{eddy} , while a previous study by the same authors (Wang et al., 2015) looks at how K_{eddy} varies with latitude at a pressure of a few bars. In Wang et al. (2015), they suggest that rotation plays an increasingly important role in suppressing turbulent convection at higher latitudes and that the tropospheric K_{eddy} should decrease towards the poles. This also agrees with earlier theoretical work by Flasar and Gierasch (1978). Incorporating this finding, Wang et al. (2016) therefore predict that GeH_4 should decrease in abundance towards the poles, while AsH_3 and PH_3 should remain constant.

This contrasts strongly with the results of this chapter, which show that GeH_4 is constant with latitude, while AsH_3 and PH_3 increase towards the poles. Although there are several degeneracies that complicate the retrievals of gaseous abundances in Jupiter’s troposphere, these overall latitudinal trends are robust. The differences between the equatorial latitudes and the high latitudes are extremely apparent in the raw observations themselves, even before the spectra are modelled using NEMESIS (Figures 6.3.2 and 6.4.1). There is therefore a clear discrepancy between the theoretical predictions and the observations. There are several factors which could contribute to this discrepancy.

One possibility is that it is due to photolytic destruction, which was not considered in Wang et al. (2016). Many studies have concluded that ultraviolet photons are a significant contribution to the destruction of PH_3 in the upper troposphere (e.g. Strobel, 1977; Ferris et al., 1984). It has also been suggested as a destruction mechanism for GeH_4 (Guillemin et al., 1995) and could plausibly apply to AsH_3 too. Geometry effects mean that higher latitudes receive fewer UV photons from the sun, and the higher opacity of stratospheric aerosols in the polar regions also acts as a shield to UV light (Zhang et al., 2013). Both of these effects could lead to a lower rate of photolytic destruction at high latitudes, and therefore a higher abundance could be maintained over the poles. By increasing the abundances of all three species at high latitudes, the latitudinal profiles from Wang et al. (2016) can be modified to match the results in this chapter. However,

the reason that photolysis was neglected by Wang et al. (2016) is that it is expected to primarily affect the upper troposphere ($p < 0.5$ bar), not the deeper regions probed in the 5- μm region (Strobel, 1977). The PH_3 column density down to the 0.5-bar level is only $\sim 3\%$ of the total column density down to the 4-bar level; even if the entirety of the upper tropospheric PH_3 was removed by photolysis, it would have a negligible impact on the 5- μm spectra.

Alternatively, it is possible that the vertical mixing strength does not in fact decrease towards the poles. Wang et al. (2015) and Flasar and Gierasch (1978) focus on small-scale turbulent convection and how this is affected by the increasing importance of rotation at high latitudes. While turbulent convection is inhibited towards the poles, it is possible that planetary-scale vertical motion increases. Solar heating is at a minimum at the poles, which reduces the static stability of the atmosphere and convection becomes uninhibited (Irwin, 2008). In addition, this would be qualitatively consistent with recent images of Jupiter’s poles, taken by the Juno mission, which show multiple vortices and chaotic behaviour in the polar regions. However, Wang et al. (2016) showed that PH_3 and AsH_3 are not sensitive to the strength of vertical mixing, while GeH_4 is. Even if the trend reported in Wang et al. (2015) and Flasar and Gierasch (1978) were to be reversed entirely, there should be an increase of GeH_4 at the poles and a flat latitudinal distribution of AsH_3 and PH_3 , which is not what is observed.

It is also possible that the sensitivities of the disequilibrium species to K_{eddy} , as described in Wang et al. (2015), are not robust. In several cases, the authors note a lack of available kinetics data which limit the confidence in the results. However, these diffusion-kinetics calculations do provide a possible explanation for the difference in behaviour between GeH_4 and AsH_3/PH_3 . In addition, changes to the chemistry model cannot single-handedly account for the discrepancy between the theory and the observations. If AsH_3 and PH_3 were more sensitive to K_{eddy} , they would be predicted to decrease, not increase, towards the poles, since K_{eddy} decreases at high latitudes (Wang et al., 2015).

6.6 Conclusions

In this chapter, the VLT CRIRES observations were used to spectrally resolve the line shapes of three disequilibrium species in Jupiter’s troposphere: GeH_4 , AsH_3 and PH_3 . Because the observations were latitudinally resolved, they were then used to search for latitudinal variability in these species.

Chapter 5 described degeneracies between the cloud structure and scattering properties that complicate the retrievals of tropospheric gaseous abundances. In this chapter, it was shown that these degeneracies can sometimes explain apparent belt-zone variations in composition. This is true in the case of GeH_4 where the change in line shape between belts and zones can be accounted for by the cloud structure alone. Any future studies into belt-zone variability must therefore carefully assess the degeneracies with the cloud structure before any conclusions about spatial variations in gaseous abundances can be drawn.

All spatial regions can be fit with a GeH_4 abundance of 0.58 ppb, which is consistent with previous studies. The AsH_3 abundance varies between 0.01 ppb and 0.55 ppb from equator to pole. This is the first time that supersolar abundances of AsH_3 have been measured in Jupiter’s atmosphere. The PH_3 abundance varies between 0.5 and 1.5 ppm, which is consistent with previous measurements.

GeH_4 appears to have an abundance that is constant with latitude, while PH_3 and AsH_3 show evidence for an enhancement at high latitudes. This partially agrees with a theoretical study from Wang et al. (2016) which suggests that GeH_4 should have a different latitudinal profile to PH_3 and AsH_3 . However, the results of this chapter disagree with the shapes of those profiles, as they predict that GeH_4 should decrease towards the poles while PH_3 and AsH_3 are flat. The explanation for this difference is not clear, but it could be due to the effect of photolytic destruction, increased global-scale convection at the poles, missing chemical kinetics data, or a combination thereof.

Chapter 7

Ionospheric H_3^+ emission

7.1 Introduction

During the analysis of the VLT CRIRES observations in Chapters 5 and 6, several narrow emission lines were detected in Jupiter’s 5- μm spectrum. Molecular species in Jupiter’s troposphere produce absorption lines, rather than emission lines, because temperature decreases with altitude in the troposphere. These emission lines must therefore be due to a species much higher up in Jupiter’s atmosphere, where the temperatures are higher than in the troposphere. These lines were subsequently identified as belonging to H_3^+ , an auroral species in Jupiter’s ionosphere.

Observations of H_3^+ lines provide a valuable way to study Jupiter’s upper atmosphere. The species can be used to measure ionospheric temperatures and to trace energy inputs from high-energy particles and solar radiation. The species itself also directly affects the ionospheric conditions, both acting as a stabilising ‘thermostat’ and providing the main contribution to ionospheric conductivity. Further details on the role of H_3^+ in planetary atmospheres can be found in Miller et al. (2006).

The first spectroscopic detection of H_3^+ emission from Jupiter’s ionosphere was made by Drossart et al. (1989) in the $2\nu_2(l = 2)$ overtone band at 2 μm in the K-band (l is the quantum number for vibrational angular momentum). This was almost immediately followed by detections of the ν_2 fundamental at 4 μm in the L-band (e.g. Oka and Geballe, 1990), and more recently by detection of the $2\nu_2(0)-\nu_2$ (4 μm) and $3\nu_2(3)-\nu_2$ (2 μm) overtones (Stallard et al., 2002; Raynaud et al., 2004). The strongest H_3^+ signature is localised around the northern and southern auroral ovals (Drossart

et al., 1992); in these regions of the planet, high-energy electrons are accelerated along the magnetic field lines into the upper atmosphere where they ionise Jupiter’s neutral gases to produce H_2^+ . H_2^+ and H_2 can then combine to produce H_3^+ and H , a reaction that is so efficient that very little H_2^+ remains in the upper atmosphere. In addition to this auroral effect, there is a planet-wide signal due to extreme ultra-violet radiation from the sun which also causes ionisation (Miller et al., 1997).

Studying the H_3^+ emission lines can provide us with several different types of temperature measurements for Jupiter’s ionosphere. The kinetic temperature, T_{kin} , can be derived from the Doppler broadening of individual lines. The rotational temperature, T_{rot} , can be derived by comparing the relative intensities of different rotational lines within the same vibrational manifold. The vibrational temperature, T_{vib} , can be derived by simultaneously measuring lines from multiple vibrational levels. If the gas is in local thermodynamic equilibrium (LTE), then these three temperatures should be the same. The kinetic temperature of H_3^+ in Jupiter’s ionosphere has been measured once before (Drossart et al., 1993), the vibrational temperature has been measured twice (Stallard et al., 2002; Raynaud et al., 2004), and several studies have measured the rotational temperature (Drossart et al., 1989; Oka and Geballe, 1990; Maillard et al., 1990).

This chapter uses the high-resolution CRIRES observations to measure sixteen H_3^+ emission lines in the 4.5–5.2 μm range. This is the first time that H_3^+ emission lines have been measured in this spectral region. The lines are sufficiently strong that they can be seen even against the bright background radiation from the troposphere. The emission lines are identified in Section 7.2 and consist of fifteen lines from the ν_2 fundamental and one line from the $2\nu_2(2)-\nu_2$ overtone. The broadening of these lines is then measured to evaluate T_{kin} (Section 7.3), and the relative intensities of the ν_2 lines are measured to evaluate T_{rot} (Section 7.4). Finally, the fundamental and overtone lines are compared to evaluate T_{vib} (Section 7.5). This is first time that all three temperatures have been measured simultaneously. The implications for thermodynamic equilibrium in Jupiter’s ionosphere are discussed in Section 7.6. This chapter is based on Giles et al. (2016b).

Line centre (μm)	Line centre (cm^{-1})	Line identification	Doppler broadening ($10^{-5} \mu\text{m}$)
4.59605	2175.78	P(5,0)	8.2 ± 0.7
4.60232	2172.82	P(5,1)	7.5 ± 1.9
4.61180	2168.35	P(5,6+) (overtone)	8.7 ± 1.4
4.62048	2164.28	P(5,2)	8.2 ± 0.6
4.64493	2152.89	P(5,3)	6.5 ± 0.2
4.67213	2140.35	P(5,4)	7.1 ± 0.2
4.68401	2134.92	P(5,5)	7.6 ± 0.2
4.71140	2122.51	P(6,1)	6.9 ± 1.4
4.73206	2113.24	P(6,2)	6.7 ± 1.3
4.76956	2096.63	P(6,3)	7.4 ± 0.6
4.80901	2079.43	P(6,4)	6.9 ± 0.5
4.87446	2051.51	P(6,6)	9.3 ± 0.2
4.91807	2033.32	P(7,4)	9.2 ± 1.6
4.98184	2007.29	P(7,5)	8.2 ± 1.2
5.04318	1982.87	P(7,6)	10.2 ± 0.5
5.16608	1935.71	P(8,6)	9.7 ± 1.8

Table 7.2.1: Identified H_3^+ emission features in Jupiter’s 5- μm window. The identification and centre of each line are obtained from Kao et al. (1991). The Doppler broadening was measured from the CRIRES data.

7.2 Line identification

Following Giles et al. (2016b), this analysis is restricted to the higher-quality CRIRES observations from 12 November 2012. Sixteen H_3^+ emission lines were detected in this dataset. These lines were identified using the spectroscopic line list of Kao et al. (1991) and are listed in Table 7.2.1. Fifteen of the lines belong to the ν_2 fundamental vibrational band, and the remaining line (located at 4.6118 μm) belongs to the $2\nu_2(2)$ - ν_2 overtone. This is the first time that an emission line from this overtone has been observed in Jupiter’s atmosphere. All of the lines are in the P-branch of the vibrational manifold, and the line identification describes the J and K quantum numbers of the lower state, as described by Miller and Tennyson (1988).

The first emission line to be identified was the P(5,5) line at 4.68401 μm . Using this emission line, the strength of the H_3^+ emission was explored as a function of latitude. In order to improve the signal-to-noise, the data was smoothed with a spatial bin width of three pixels. At each pixel, the IDL MPFIT routine (Markwardt, 2009) was used to fit the P(5,5) emission line. As CRIRES is an echelle spectrometer, the instrumental line shape is triangular, and the resolving power of 96,000 translates into a resolution of

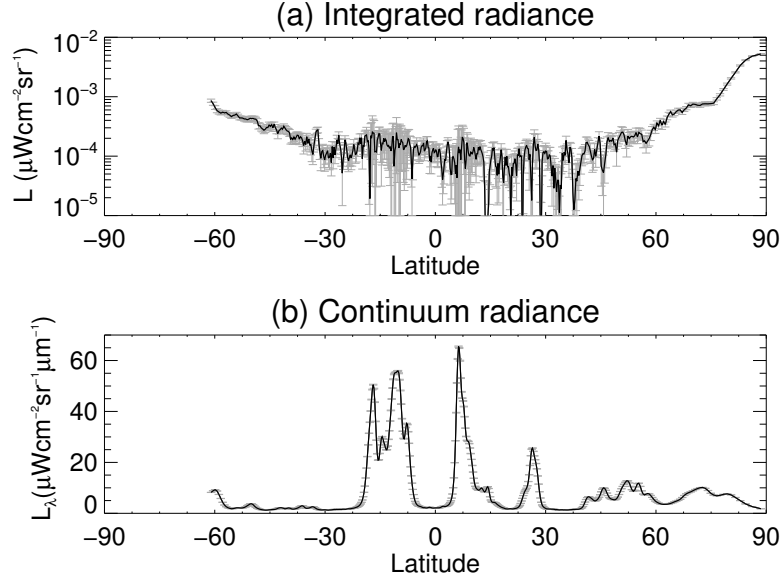


Figure 7.2.1: (a) The integrated radiance of the H_3^+ P(5,5) emission line as a function of latitude. (b) Jupiter's continuum radiance (in the immediate vicinity of the P(5,5) line) as a function of latitude.

$\sim 5 \times 10^{-5} \mu\text{m}$. Because the slit is very narrow, the broadening that occurs from Jupiter's rotation and the horizontal flow of gas is expected to be negligible. The function used to fit the data was therefore a convolution of a known triangular line shape (from the instrument) and an unknown Gaussian curve (from the Doppler broadening). There were six variables used: three for the Gaussian component (line centre, line width, line strength) and three for the background (a quadratic fit).

Figure 7.2.1a shows how the integrated radiance of the emission line varies with latitude. The CRIRES slit length was slightly smaller than the diameter of the planet, so the south pole is not covered. For comparison, Figure 7.2.1b shows the continuum radiance. The strength of the H_3^+ emission is a factor of ~ 100 higher in the polar regions than in the equatorial regions, which is consistent with the fact that H_3^+ is predominantly an auroral species. Since the northern polar region has the strongest H_3^+ signature, data from this region ($84\text{--}86^\circ\text{N}$) was used to search for additional emission lines. The results of that search are the sixteen lines described in Table 7.2.1, and they are each shown in Figure 7.2.2.

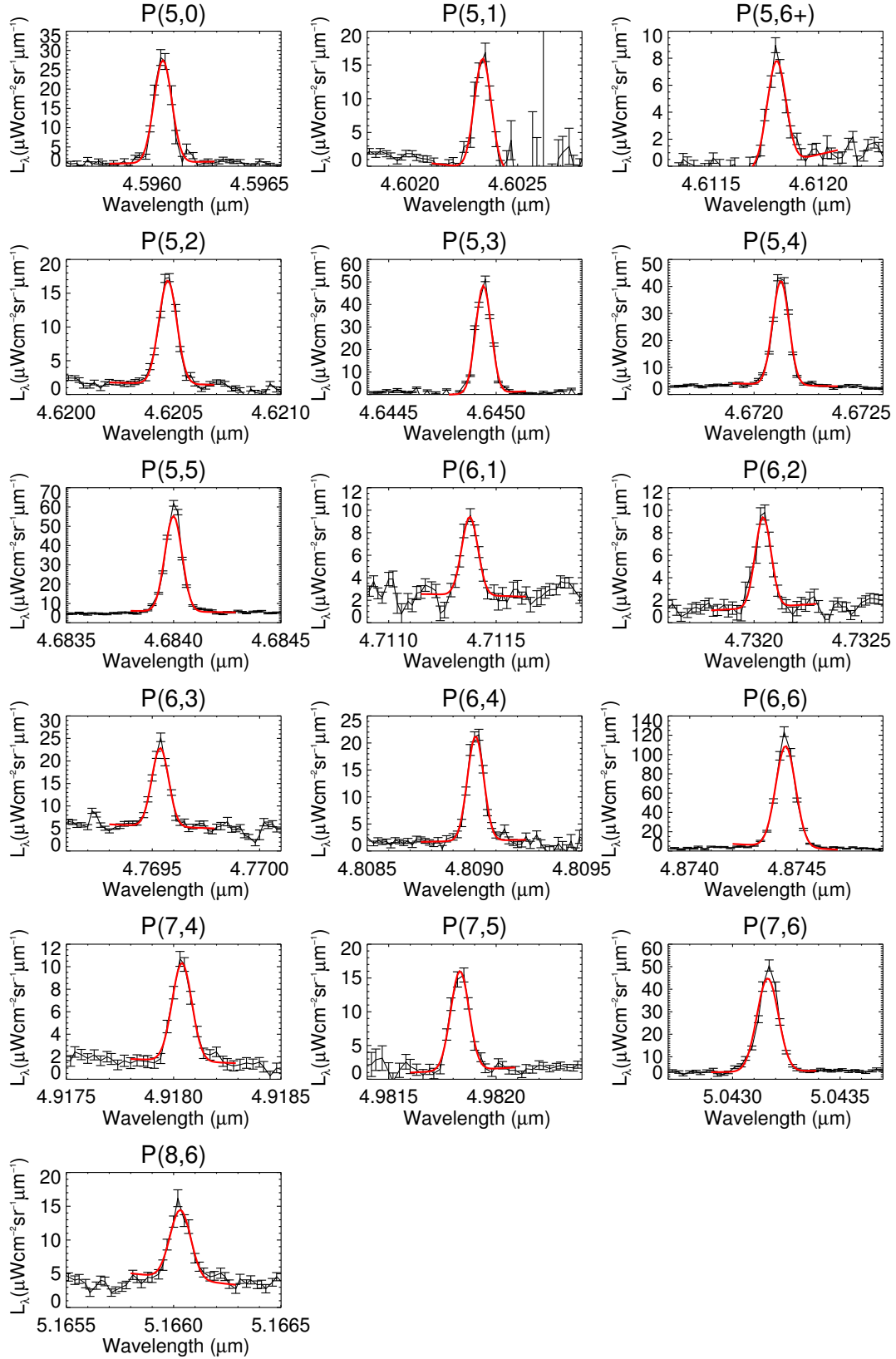


Figure 7.2.2: Identification of H_3^+ emission features in Jupiter's 5- μm window. The data is from the northern polar region ($84\text{--}86^\circ\text{N}$) where the H_3^+ signature is particularly strong. Longitudes range from 87°W to 108°W . The fitted lines are shown in red and the estimated Doppler line widths are listed in Table 7.2.1.

7.3 Kinetic temperature

The kinetic temperature of the H_3^+ ions can be determined by measuring the width of the emission lines. The analysis was restricted to the same northern polar region (84–86°N) shown in Figure 7.2.2, where the emission lines are strongest; at other latitudes, the low signal-to-noise prevents reliable results. As in the previous section, the MPFIT routine was used to fit a convolution of a triangle and a Gaussian to the sixteen observed lines. These fits are shown by the red lines in Figure 7.2.2 and the Gaussian FWHM of each line is given in Table 7.2.1. These Doppler line widths can then be converted into a kinetic temperature, T_{kin} , via Equation 7.1 (e.g Emerson, 1996):

$$T_{\text{kin}} = \frac{Mc^2}{8R \ln 2} \left(\frac{\Delta\lambda}{\lambda_0} \right)^2 \quad (7.1)$$

where M is the molar mass of the molecule, c is the speed of light, R is the molar gas constant, $\Delta\lambda$ is the Doppler line width and λ_0 is the wavelength at the centre of the line.

The individual Doppler broadening widths listed in Table 7.2.1 can be combined to produce a weighted average for the kinetic temperature, along with an error that takes into account the uncertainty in fitting the emission lines. However, this error is relatively small when compared with the error due to uncertainty in the resolving power of the CRIRES instrument. The Doppler broadening widths in Table 7.2.1 assume a resolving power of 96,000; if a 10% error on this value is assumed, and the fitting process is repeated for the upper and lower bound, then the average kinetic temperature is 1390 ± 160 K. This is reasonably consistent with the value of 1150 ± 60 K that was obtained by Drossart et al. (1993) using an emission line at $3.5 \mu\text{m}$, observed with the Fourier Transform Spectrometer at the Canada-France-Hawaii Telescope.

7.4 Rotational temperature

The rotational temperature of the H_3^+ ions can be determined by measuring the relative intensities of different rotational lines within the same vibrational manifold. In this dataset, there are observations of fifteen lines from the same ν_2 fundamental band. Since multiple wavelength settings were used to make these observations, there is not a

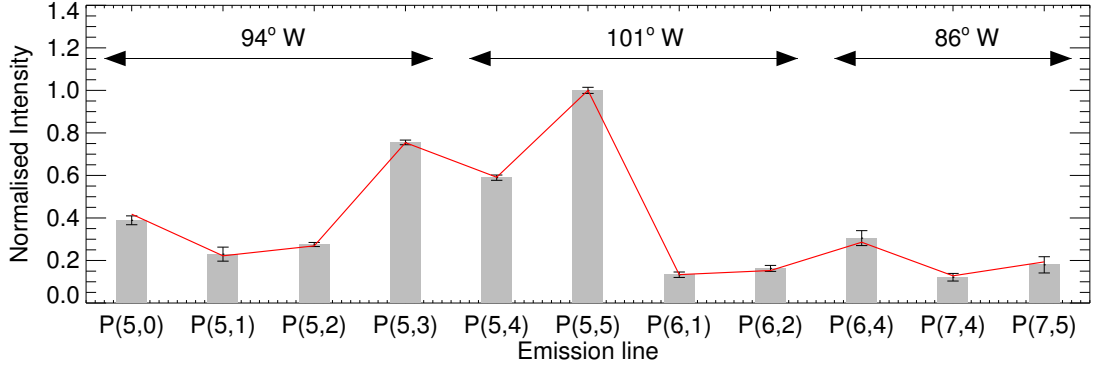


Figure 7.4.1: Relative intensity of observed H_3^+ lines, normalised to the P(5,5) line (grey) and the fit obtained with the retrieved T_{rot} of 960 K (red).

full set of H_3^+ lines at any given spatial location; different lines correspond to different longitudes with potentially different H_3^+ abundances. However, there is some overlap between the segments, such that several emission lines were observed twice. If the H_3^+ temperature is assumed to be constant across this region, and the only variations are in the H_3^+ abundance, then the different lines can be scaled so they can be compared. Once scaled, there are three sets of lines that can be directly compared: (i) P(5,0), P(5,1), P(5,2) and P(5,3) at 94°W (ii) P(5,4), P(5,5), P(6,1) and P(6,2) at 101°W , (iii) P(6,4), P(7,4) and P(7,5) at 86°W . Within each set, the H_3^+ abundance should be consistent, but the abundance may vary between the sets. The normalised intensities of these lines are shown by the grey bars in Figure 7.4.1.

In the optically thin limit, the integrated radiance of the emission line (relative to the continuum) is proportional to the optical depth, and therefore to the line intensity. As described in Stallard et al. (2002), if LTE is assumed, then the line intensity, I , of a particular transition is given by

$$I \propto \frac{\tilde{\nu} g (2J' + 1) A}{Q(T_{\text{rot}})} \exp\left(-\frac{E'}{kT_{\text{rot}}}\right) \quad (7.2)$$

where $\tilde{\nu}$ is the wavenumber of the transition, g is the nuclear spin degeneracy factor, J' is the rotational quantum number of the upper state, A is the Einstein A coefficient, $Q(T)$ is the partition function (calculated for H_3^+ in Miller et al., 2013), E' is the energy of the upper state and T_{rot} is the rotational temperature.

For each emission line, the parameters $\tilde{\nu}$, g , J' , A and E' are known (Neale et al., 1996). A temperature T_{rot} can be found that reproduces the relative intensities shown in

Figure 7.4.1. MPFIT was used to apply a least-squares fit, with four variables: T_{rot} and three scaling factors for each of the three sets of emission lines, which are proportional to the H_3^+ abundance in each case.

This fitting process produced a best-fit T_{rot} of 960 ± 40 K. This fit is shown by the red line in Figure 7.4.1. The retrieved relative abundances are 1.47 (94°W), 1.00 (101°W) and 1.26 (86°W), and the grey bars in Figure 7.4.1 have been scaled according to these values. In order to confirm that this fitting process was reliable, the three sets of lines were also fit independently, using just two parameters in each case (T_{rot} and a scaling factor). This produced results of 880 ± 110 , 980 ± 40 , 830 ± 170 K, which is consistent with the overall value of 960 ± 40 K. This result is also consistent with previously published results for the rotational temperature of H_3^+ in Jupiter’s atmosphere, which range from 670 K (Oka and Geballe, 1990) to 1250 K (Drossart et al., 1993).

7.5 Vibrational temperature

The vibrational temperature of the H_3^+ ions can be determined by measuring the relative intensities of emission lines from different vibrational manifolds. In this dataset, there are simultaneous observations of one emission line from an overtone band, P(5,6+), alongside several emission lines from the fundamental band: P(5,0), P(5,1) and P(5,3). Equation 7.2 compared the intensities of different lines within a vibrational manifold in order to calculate the rotational temperature. Similarly, the intensities of lines from different vibrational manifolds can be compared in order to calculate the vibrational temperature (Stallard et al., 2002). Equation 7.2 can be re-expressed as

$$T_{\text{vib}} = \frac{E'_2 - E'_1}{k} \left[\ln \left(\frac{I_1 (2J'_2 + 1) \tilde{\nu}_2 A_2}{I_2 (2J'_1 + 1) \tilde{\nu}_1 A_1} \right) \right]^{-1} \quad (7.3)$$

where the subscripts 1 and 2 refer to two different emission lines. If the intensity of the overtone line is compared with each of the three fundamental band lines, vibrational temperatures of 940 ± 50 K, 910 ± 80 and 920 ± 30 K are obtained. The mean value of T_{vib} is therefore 925 ± 25 K. Previous studies found T_{vib} values of 900–1250 K (Stallard et al., 2002) and 960 ± 50 (Raynaud et al., 2004).

7.6 Discussion and conclusions

In this chapter, high-resolution ground-based observations from the CRIRES instrument at the VLT were used to identify previously-undetected H_3^+ emission lines in Jupiter’s 5- μm spectrum: fifteen lines from the ν_2 fundamental, and one line from the $2\nu_2(2)$ - ν_2 overtone. By considering the broadening and relative intensities of these lines, the following temperatures were determined: $T_{\text{kin}} = 1390 \pm 160$ K, $T_{\text{rot}} = 960 \pm 40$ K and $T_{\text{vib}} = 925 \pm 25$ K. All of these values are consistent with previous measurements of H_3^+ in Jupiter’s ionosphere.

These temperatures can be compared to gain insight into whether the assumption of LTE is valid. The Einstein-A coefficients of the rotational transitions are very small, so at the temperatures and densities present in Jupiter’s ionosphere, the rotational states are expected to be in LTE (Melin et al., 2005). This in turn means that the derived rotational temperature should match the kinetic temperature; this chapter finds that T_{rot} is slightly lower than T_{kin} , which could suggest that this assumption is not valid. The only previous study to compare T_{kin} and T_{rot} found consistent values (1150K and 1250K, Drossart et al., 1993). However the T_{rot} in this case was larger than the temperature derived by most other studies. It is less surprising that T_{vib} is lower than T_{kin} , as this has already been shown in observational work by Raynaud et al. (2004) also found this to be the case. This agrees with theoretical work by Melin et al. (2005), which shows that vibrational levels depart from LTE in Jupiter’s upper atmosphere; the vibrationally excited levels of H_3^+ are not thermally populated, and the rate of collisional population is low, due to the low H_2 density in the ionosphere.

Departures from LTE will primarily affect the high altitudes (>2000 km above 1 bar), where the atmosphere is both hot and tenuous (Melin et al., 2005). This significantly reduces the ability of H_3^+ to act as a thermostat in Jupiter’s upper atmosphere, as the energy inputs from charged particles and solar radiation are not offset by the cooling provided by H_3^+ . In addition, non-LTE effects alter the apparent H_3^+ column density, which in turn affects the inferred conductivity of the ionosphere. The degree to which LTE holds is therefore important in understanding the energy budget of the upper atmosphere.

It should be noted that this suggested violation of rotational LTE must await direct

confirmation by future measurements. The key challenge of this study was the use of H_3^+ lines in multiple different wavelength settings, observed at different times, meaning different temperatures and ionospheric wind speeds are potentially being convolved, in addition to different H_3^+ abundances. Only an instrument capable of simultaneous observations of multiple H_3^+ lines over a broad wavelength range could allow confirmation that T_{rot} and T_{kin} are genuinely out of equilibrium. This study may be possible with future observations, including IRTF/ISHELL and the refurbished CRIRES.

Previous studies of H_3^+ have focussed on the L-band and the K-band. The detection of additional H_3^+ lines opens up a new atmospheric window for future studies: the M-band. This increases the range of instruments that can be used to observe H_3^+ auroral emission, and will also allow for simultaneous studies of both the upper and deep atmosphere. It will also be important when assessing future 5- μm images of the polar regions, such as those made by the JIRAM instrument on the Juno spacecraft, where the auroral signature may be large enough to be seen against the background.

Chapter 8

Conclusions and future work

8.1 Introduction

This thesis uses 5- μm spectroscopic observations of Jupiter to study the composition and cloud structure of the jovian troposphere. This work is especially timely due to the recent arrival of NASA’s Juno spacecraft at Jupiter in July 2016. Juno will make high spatial resolution observations in a series of narrow swathes of the planet, and the instrument suite includes a mapping spectrometer that covers the 5- μm window (JIRAM, Adriani et al., 2008). This mission is being accompanied by an extensive campaign of ground-based observations, which can provide additional spatial, spectral and temporal coverage (Orton, 2009). As shown in this thesis, these different types of observations provide complementary information. This thesis sets the scene for Jupiter’s deep atmosphere in the run up to the Juno mission and shows the possibilities and limitations for future analyses of the 5- μm spectral region. The conclusions reached in this thesis can be further explored in the data that will be obtained in the next few years.

In the introduction to this thesis, a list of aims was laid out in the form of six questions. Section 8.2 returns to that list and describes the results that were found in each case. Finally, in Section 8.3, suggestions are made for future work.

8.2 Conclusions

1. When studying Jupiter's troposphere using 5- μm spectroscopy, what are the advantages and disadvantages of low-resolution spacecraft data, compared with high-resolution ground-based data?

Chapter 4 focused on observations made by the VIMS instrument on the Cassini spacecraft. VIMS provides simultaneous observations over the entire 4.5–5.1 μm spectral range, making it the ideal dataset for studying broadband features such as clouds. These observations provide global coverage of the entire planet, and the wide range of emission angles allows for a study of limb-darkening effects. VIMS also provides access to the planet's nightside, which simplifies the analysis as it removes the complication of reflected sunlight. Additionally, the fact that the observations are made from a spacecraft means that there are no telluric lines that need to be removed. The primary drawback of the VIMS observations is that the spectral resolution is relatively low, which leads to degeneracies between atmospheric parameters and means that the abundances of minor molecular species cannot be accurately retrieved.

Chapters 5 and 6 used observations from the CRIRES instrument at the Very Large Telescope. These observations are very high spectral resolution, allowing individual absorption lines to be resolved. In addition, these observations are latitudinally resolved (spatial resolution of 0.1") so can be used to search for latitudinal variability. However, these observations do not provide full global coverage, so interesting features like the Great Red Spot cannot be studied. Unlike VIMS, the CRIRES observations do not cover the entire 5- μm window simultaneously. Because different wavelength segments observe different parts of the planet, a simultaneous retrieval of the entire spectral region is not possible. Nevertheless, the high spectral resolution breaks many of the degeneracies between the clouds and gases, allowing minor molecular species such as GeH_4 and AsH_3 be studied.

2. What degeneracies exist between different atmospheric parameters, and what limitations do these place on atmospheric retrievals?

Chapter 4 showed that the relatively low spectral resolution of the VIMS observations introduces several degeneracies between retrieved parameters, particularly for minor molecular species. Because of this, only cloud opacity and PH_3 abundance can be reliably retrieved from VIMS data. Even with these, it should be noted that the absolute values of the retrieved parameters are highly dependent on assumptions made about the parametrisation of the profile, e.g. the location of the cloud deck. However, because these same assumptions are applied to the entire planet, conclusions can still be drawn about global variability (these are described in later in this section). The JIRAM instrument on the Juno spacecraft (Adriani et al., 2008) has a similar spectral resolution to VIMS (10 nm, compared to 17.8 nm), so some these degeneracies should be explored in future analyses of JIRAM observations.

While using very high spectral observations, such as those provided by CRILES, breaks some of these degeneracies between gaseous species, others remain. As described in Chapters 5 and 6, the presence of reflected sunlight in the dayside spectrum introduces a degeneracy between the cloud scattering properties and the retrieved gaseous abundances, which has the potential to give the false impression of belt-zone variability in composition. In addition, there is some degeneracy between the deep cloud structure and retrieved gaseous abundances, such that the presence of a highly variable deep cloud deck can account for apparent differences in absorption line strengths. Both of these degeneracies can be broken to a certain extent; the fraction of reflected sunlight can be estimated using the Fraunhofer lines, and the deep cloud structure can be constrained using a species such as CH_3D that is not expected to vary with latitude. Nevertheless, these degeneracies should be carefully explored when drawing conclusions about spatial variability of gaseous species.

3. What is the vertical structure of Jupiter’s tropospheric cloud decks, and how do they vary across the planet?

Jupiter’s cloud structure was studied in Chapters 4 and 5. Chapter 4 showed that a single cloud deck was sufficient to model the global Cassini VIMS data, provided that

this cloud is located no deeper than 1.2 bar. Clouds around this pressure level are likely to be composed of NH_3 or NH_4SH . Theory suggests that there should be multiple discrete cloud decks; it should be noted that the VIMS data does not rule this scenario out, but also does not provide evidence for multiple clouds. This single cloud layer is highly variable across the planet, with thick clouds in the zones and thin clouds in the belts. In addition to the belt-zone variability, there is also some longitudinal inhomogeneity, particularly in the wake of the Great Red Spot.

Chapter 5 focussed on the VLT CRISP data and showed that this provides some evidence for the presence of an additional deep cloud deck at ~ 5 bar, although this result is fairly sensitive to degeneracies with the cloud scattering properties. Based on its location, this cloud is likely to be composed of water ice. When a full latitudinal retrieval was performed, this deep cloud was found to be essentially opaque in the zones and transparent in the belts.

4. What are the scattering properties of Jupiter’s cloud particles, and are there any spectral features?

Chapter 4 showed that the tropospheric clouds must be relatively spectrally flat. A single cloud deck made of either pure NH_3 ice or pure NH_4SH is inconsistent with the data, as these ices are not sufficiently ‘grey’. This could be due to the presence of multiple cloud decks made of particles of different materials and/or sizes. These could combine to ‘blur out’ the individual spectral features, leading to an overall grey appearance. Alternatively, the clouds could be coated in another material, such as a thin layer of hydrocarbons, which would mask the spectral features.

Both Chapter 4 and Chapter 5 studied the scattering properties of Jupiter’s cloud particles: the single-scattering albedo, ω and the asymmetry parameter, g . A limb-darkening analysis of the VIMS data showed that the particles must be highly scattering (ω greater than ~ 0.7) and not too strongly forward scattering (g less than ~ 0.9). In Chapter 5, the strength of the Fraunhofer lines was used to assess the relative proportions of reflected sunlight and thermal emission. If a single-scattering albedo of 0.9 is assumed, then the best fit to the Fraunhofer lines is when $g = 0.8$, which is also consistent with both the VIMS result and the previous work of Roos-Serote and Irwin

(2006).

5. How do the abundances of disequilibrium species vary across the planet, and what can be inferred about the tropospheric dynamics?

Three disequilibrium species were studied in Chapter 6: GeH_4 , AsH_3 and PH_3 . These gases were retrieved as a function of latitude. There is no evidence for any belt-zone variability in these gases, as changes in the deep cloud opacity can account for the variability in the line shape. The GeH_4 latitudinal distribution can be modelled using a constant abundance, but both AsH_3 and PH_3 require an enhancement at high latitudes. A recent study by Wang et al. (2016) provided theoretical predictions for the latitudinal distribution of disequilibrium gases. This theoretical work partially agrees with the results of the CRIRES observations, in that GeH_4 exhibits a different behaviour to AsH_3 and PH_3 . However, Wang et al. (2016) predict that GeH_4 should decrease towards the poles while PH_3 and AsH_3 are flat, which is not seen in the results of Chapter 6. This difference between the predicted and observed distributions could be due to the effect of photolytic destruction, increased global-scale convection at the poles, missing chemical kinetics data, or a combination these factors.

6. What information can measurements of H_3^+ emission provide about Jupiter's upper atmosphere?

In Chapter 7, the CRIRES observations were used to measure emission lines from H_3^+ , an auroral species in Jupiter's ionosphere. By measuring the widths and relative strengths of the emission lines, three temperature measurements were made: kinetic, rotational and vibrational. This is the first time that all three temperatures have been measured simultaneously, and differences between these values suggest that an assumption of local thermodynamic equilibrium in Jupiter's upper atmosphere may not be valid.

8.3 Future work

8.3.1 Global mapping of disequilibrium species

In this thesis, disequilibrium species were measured using observations from the CRIRES instrument. These observations covered a single, very narrow north-south slice of the planet, allowing latitudinal, but not longitudinal, variability to be studied. In the future, this analysis could be further extended to provide full global coverage, using new observations from the TEXES instrument at NASA’s Infrared Telescope Facility in Hawaii (Lacy et al., 2002). TEXES is a high resolution mid-infrared spectrometer, covering 5–25 μm with a resolving power of 15,000 in its medium mode. While this is a lower resolving power than CRIRES, it is still sufficiently high to measure the abundances of minor species such as GeH_4 and AsH_3 . TEXES can be used in a mapping mode in order to build up global coverage of the planet, instead of the narrow latitudinal swath provided by the CRIRES observations.

The CRIRES instrument is currently unavailable, making TEXES one of the only ground-based instruments capable of making 5- μm spectroscopic observations of Jupiter during the Juno mission time frame. As such, it has already been used to make many observations as part of the ground-based support campaign (including observations in October 2014 for which I was the Primary Investigator), and this observing program is likely to continue. An example of a full, global 5- μm map from TEXES can be seen in Figure 8.3.1. These TEXES observations could be used to measure the distributions of AsH_3 , GeH_4 and PH_3 across the entire planet. This would allow us to measure the spatial variability in gaseous composition that is associated with features such as storms and vortices. By making observations over a period of several years, temporal changes could also be studied. This would help to further understand Jupiter’s dynamics, and would provide vital global context for Juno observations, which also seek to map tropospheric composition in narrow swathes of the planet.

8.3.2 Abundances of condensable species

In this thesis, the primary focus was on clouds and disequilibrium gaseous species. In the future, this work could be extended to cover an additional class of gases: condensable

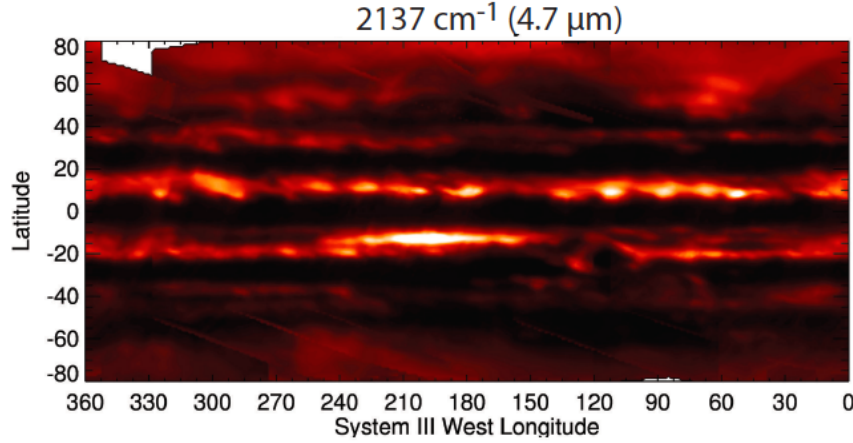


Figure 8.3.1: Example of a raw intensity map from the 5-micron window, obtained using the TEXES instrument in December 2014 (Fletcher et al., 2016).

species. Both NH_3 and H_2O fall into this category, as the temperature of the jovian troposphere means that both gases are expected to condense to form clouds. This causes them to have complicated vertical distributions, and makes their abundance more difficult to measure. As cloud-condensation models are heavily reliant on the abundances of H_2O and NH_3 , an accurate measurement of these gases is needed to understand the formation of tropospheric clouds (Atreya et al., 1999). In addition, both species are among the more abundant minor constituents of the jovian atmosphere, so they are important in understanding Jupiter’s bulk composition. The abundance of H_2O is particularly useful in understanding the origin of Jupiter, as water ice may have played an important role in the transport of volatiles in the early Solar System (Lunine et al., 2004).

Quantifying the distributions of H_2O and NH_3 is one of the key scientific goals of the Juno mission, and in particular, the Microwave Radiometer (MWR, Pingree et al., 2008). By making observations in the microwave region of the spectrum (0.6–22 GHz), MWR can probe down to pressures greater than 100 bar. Instead of measuring a full spectrum, it distinguishes between different gaseous abundances by measuring how brightness varies with emission angle at six discrete frequencies (Janssen et al., 2005). These observations can be complemented by 5- μm measurements which probe the 4–8 bar region, in order to build up a more comprehensive understanding of the vertical structure.

Retrievals of H_2O were shown in Chapter 4, but the low spectral resolution of Cassini

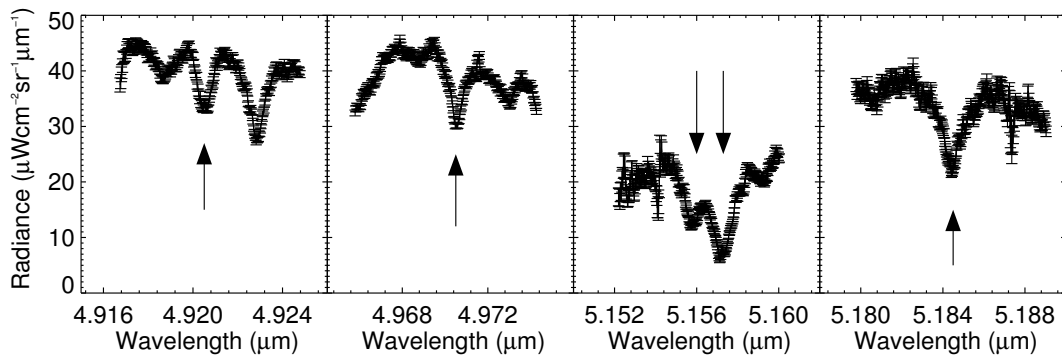


Figure 8.3.2: NH_3 absorption lines in 5- μm observations from VLT CRILES. The arrows mark the line centres.

VIMS meant that these retrievals were not very reliable. In the CRILES dataset, H_2O absorption lines were visible, but due to the low Doppler shift between the earth and Jupiter at the time of the observations, the jovian lines are difficult to distinguish from the telluric lines. In contrast, NH_3 could be initially studied using the same CRILES observations used in this thesis. There are several prominent NH_3 lines (see Figure 8.3.2) that could be used to measure the latitudinal distribution of this species in the middle troposphere. Both species would also benefit from additional observations made using the TEXES instrument.

Unlike the existing CRILES dataset, future TEXES observations can be planned so that there is a sufficient Doppler shift, allowing jovian H_2O lines to be distinguished from telluric H_2O lines. This means that H_2O can be retrieved, in addition to NH_3 . The TEXES observations also have the advantage of providing global coverage, and coinciding with the Juno observations. Water and ammonia play important roles in Jupiter's atmosphere, and combining TEXES observations with MWR will lead to a greater understanding of their global and vertical distribution.

8.4 Final comments

This thesis has shown that both Cassini VIMS and VLT CRILES provide valuable datasets with which to study Jupiter's atmosphere. In the near future, there will be many more exciting opportunities for Jupiter science, and this work can inform future studies and investigations. At the time of writing, the Juno mission has been in orbit around Jupiter for almost three months; in the coming months and years, the data

obtained by this mission will improve our understanding of the planet's formation, evolution and structure. Juno will be followed by JUICE, an ESA mission that is expected to reach Jupiter in 2030. The James Webb Space Telescope is due to launch in 2018 and will provide high spatial resolution observations at wavelengths accessible only from space. Finally, there are several next-generation, extremely large telescopes currently under construction in Chile and Hawaii, that will provide unprecedented spatial resolution, allowing small-scale atmospheric features to be studied. Using these telescopes, we will be able to build on current work to significantly improve our understanding of the processes shaping the jovian weather layer and the implications for giant planet atmospheres throughout our Solar System.

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Appendix

Wavelength setting (μm)	Segment	Wavelength range (μm)	Primary absorption lines	Telluric contamination
4.607	a	4.548–4.573	PH_3 (continuum)	Partially obscured by tellurics
	b	4.581–4.606	CO , PH_3 (continuum)	Fairly clean
	c	4.613–4.637	CO , PH_3 (continuum)	Fairly clean
	d	4.643–4.666	CH_3D , CO , GeH_4 , PH_3 (continuum)	Fairly clean
4.623	a	4.562–4.587	PH_3 (continuum)	Partially obscured by tellurics
	b	4.595–4.619	CO , PH_3 (continuum)	Fairly clean
	c	4.626–4.649	CH_3D , CO , GeH_4 , PH_3 (continuum)	Fairly clean
	d	4.656–4.678	CH_3D , CO , AsH_3 , PH_3 (continuum)	Fairly clean
4.709	a	4.652–4.676	CH_3D , CO , AsH_3 , PH_3 (continuum)	Fairly clean
	b	4.683–4.706	CH_3D , GeH_4 , AsH_3 , PH_3 (continuum)	Fairly clean
	c	4.712–4.734	CO , PH_3 (continuum)	Fairly clean
	d	4.740–4.761	PH_3 (continuum)	Fairly clean
4.723	a	4.666–4.690	CH_3D , GeH_4 , PH_3 (continuum)	Fairly clean
	b	4.696–4.719	CH_3D , CO , AsH_3 , PH_3 (continuum)	Fairly clean
	c	4.725–4.747	GeH_4 , PH_3 (continuum)	Fairly clean
	d	4.753–4.774	PH_3 (continuum)	Fairly clean

4.809	a	4.757–4.779	CO, PH ₃ (continuum)	Partially obscured by tellurics
	b	4.785–4.806	AsH ₃ , PH ₃ (continuum)	Partially obscured by tellurics
	c	4.812–4.832	PH ₃ (continuum)	Partially obscured by tellurics
	d	4.837–4.856	PH ₃ (continuum)	Partially obscured by tellurics
4.822	a	4.770–4.792	PH ₃ (continuum)	Fairly clean
	b	4.798–4.819	AsH ₃ , PH ₃ (continuum)	Partially obscured by tellurics
	c	4.825–4.845	PH ₃ (continuum)	Partially obscured by tellurics
	d	4.850–4.869	PH ₃ (continuum)	Partially obscured by tellurics
4.872	a	4.799–4.829	PH ₃ (continuum)	Mostly obscured by tellurics
	b	4.838–4.867	PH ₃ (continuum)	Partially obscured by tellurics
	c	4.875–4.904	PH ₃ (continuum)	Partially obscured by tellurics
	d	4.911–4.939	PH ₃ (continuum)	Fairly clean
4.886	a	4.813–4.843	PH ₃ (continuum)	Partially obscured by tellurics
	b	4.852–4.881	PH ₃ (continuum)	Partially obscured by tellurics
	c	4.889–4.917	PH ₃ (continuum)	Partially obscured by tellurics
	d	4.925–4.952	PH ₃ (continuum)	Partially obscured by tellurics

4.978	a	4.909–4.938	PH ₃ (continuum)	Fairly clean
	b	4.946–4.974	PH ₃ (continuum)	Partially obscured by tellurics
	c	4.981–5.008	PH ₃ (continuum), H ₂ O (continuum)	Fairly clean
	d	5.015–5.041	H ₂ O (continuum)	Partially obscured by tellurics
4.992	a	4.923–4.952	PH ₃ (continuum)	Partially obscured by tellurics
	b	4.960–4.988	PH ₃ (continuum)	Partially obscured by tellurics
	c	4.995–5.022	PH ₃ (continuum), H ₂ O (continuum)	Partially obscured by tellurics
	d	5.029–5.055	H ₂ O (continuum)	Fairly clean
5.084	a	5.020–5.047	H ₂ O (continuum)	Partially obscured by tellurics
	b	5.054–5.081	PH ₃ , H ₂ O (continuum)	Fairly clean
	c	5.087–5.113	NH ₃ , H ₂ O (continuum)	Partially obscured by tellurics
	d	5.119–5.143	H ₂ O (continuum)	Partially obscured by tellurics
5.098	a	5.034–5.061	H ₂ O (continuum)	Fairly clean
	b	5.068–5.095	PH ₃ , H ₂ O (continuum)	Partially obscured by tellurics
	c	5.102–5.126	NH ₃ , H ₂ O (continuum)	Partially obscured by tellurics
	d	5.133–5.157	H ₂ O (continuum)	Mostly obscured by tellurics

5.191	a	5.131–5.156	H ₂ O (continuum)	Mostly obscured by tellurics
	b	5.163–5.187	NH ₃ , H ₂ O (continuum)	Partially obscured by tellurics
	c	5.193–5.217	NH ₃ , H ₂ O (continuum)	Mostly obscured by tellurics
	d	5.223–5.245	NH ₃ , H ₂ O (continuum)	Mostly obscured by tellurics
5.205	a	5.145–5.170	NH ₃ , H ₂ O (continuum)	Partially obscured by tellurics
	b	5.177–5.201	NH ₃ , H ₂ O (continuum)	Partially obscured by tellurics
	c	5.208–5.230	H ₂ O (continuum)	Mostly obscured by tellurics
	d	5.237–5.258	H ₂ O (continuum)	Partially obscured by tellurics

Table 8.4.1: A description of the CRIRES wavelength settings. There are 14 wavelength settings, and each one is made up of 4 segments. The principal absorption lines in each wavelength setting / segment are listed, along with a qualitative assessment of the level of telluric contamination in each case. For more details about the telluric lines, see Figures 8.4.1–8.4.2.

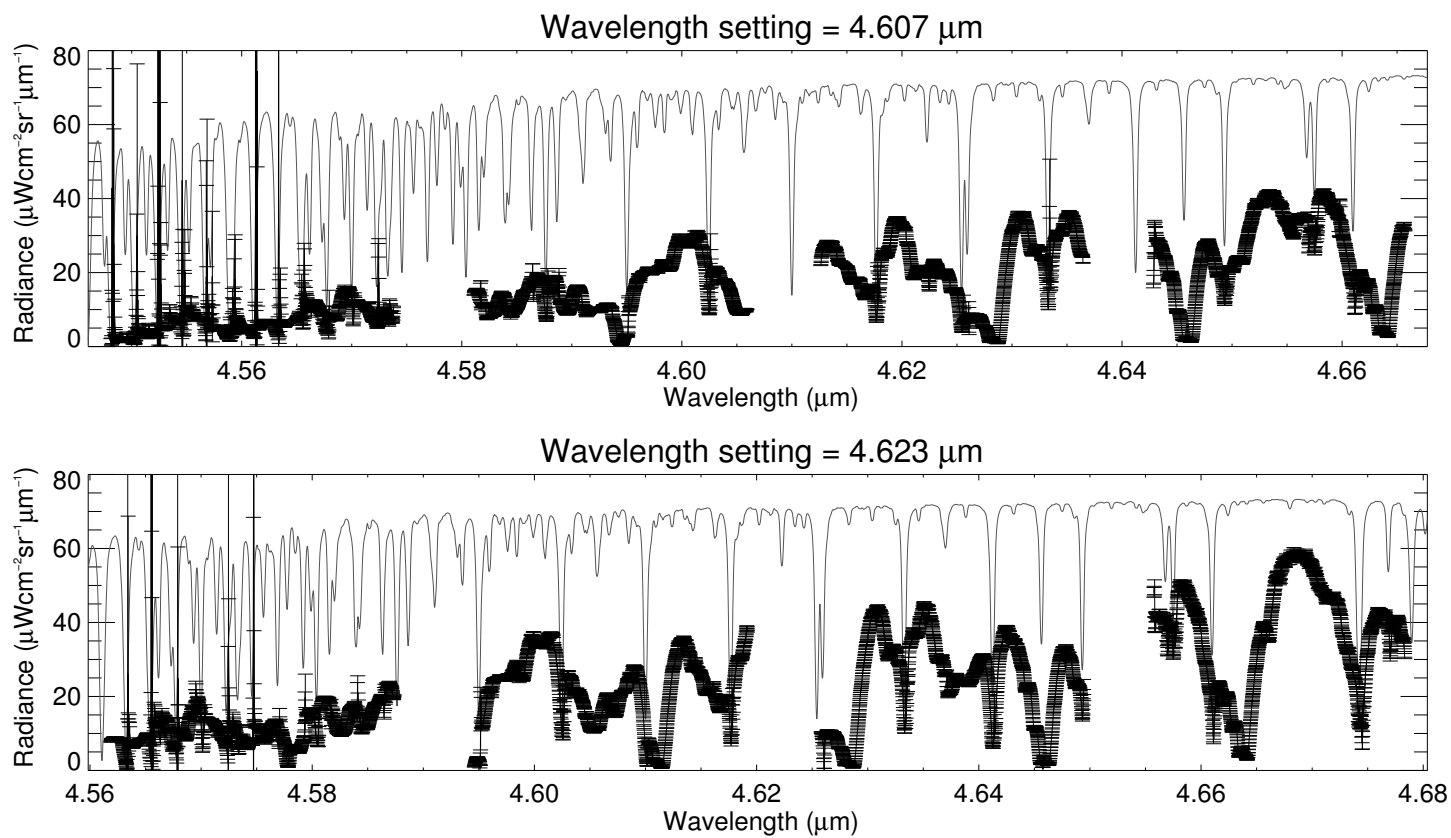


Figure 8.4.1: CRIREs observations from 12 November 2012. CRIREs data from the SEB is shown in black, the telluric transmission is shown in grey.

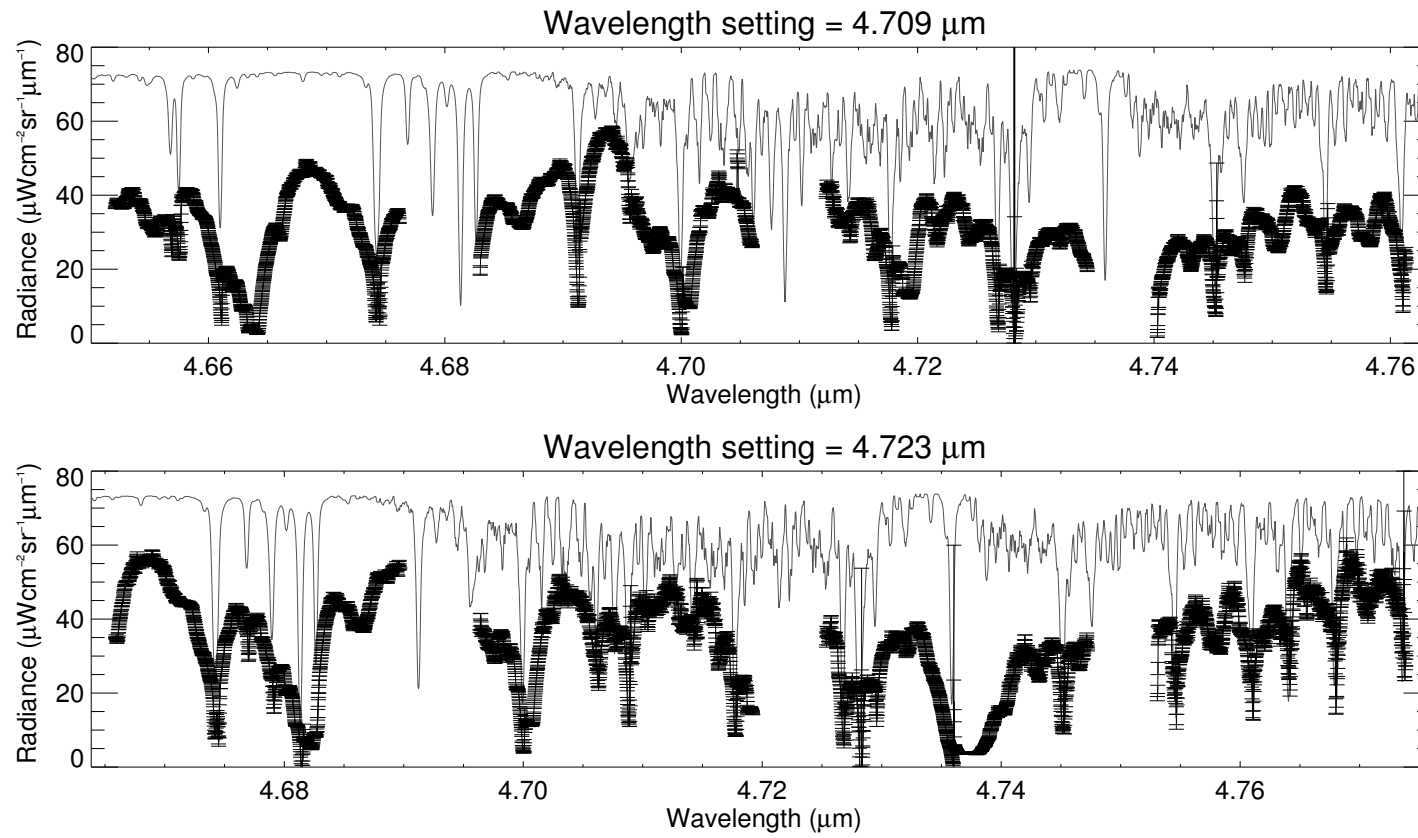


Figure 8.4.2: CRIREs observations from 12 November 2012. CRIREs data from the SEB is shown in black, the telluric transmission is shown in grey.

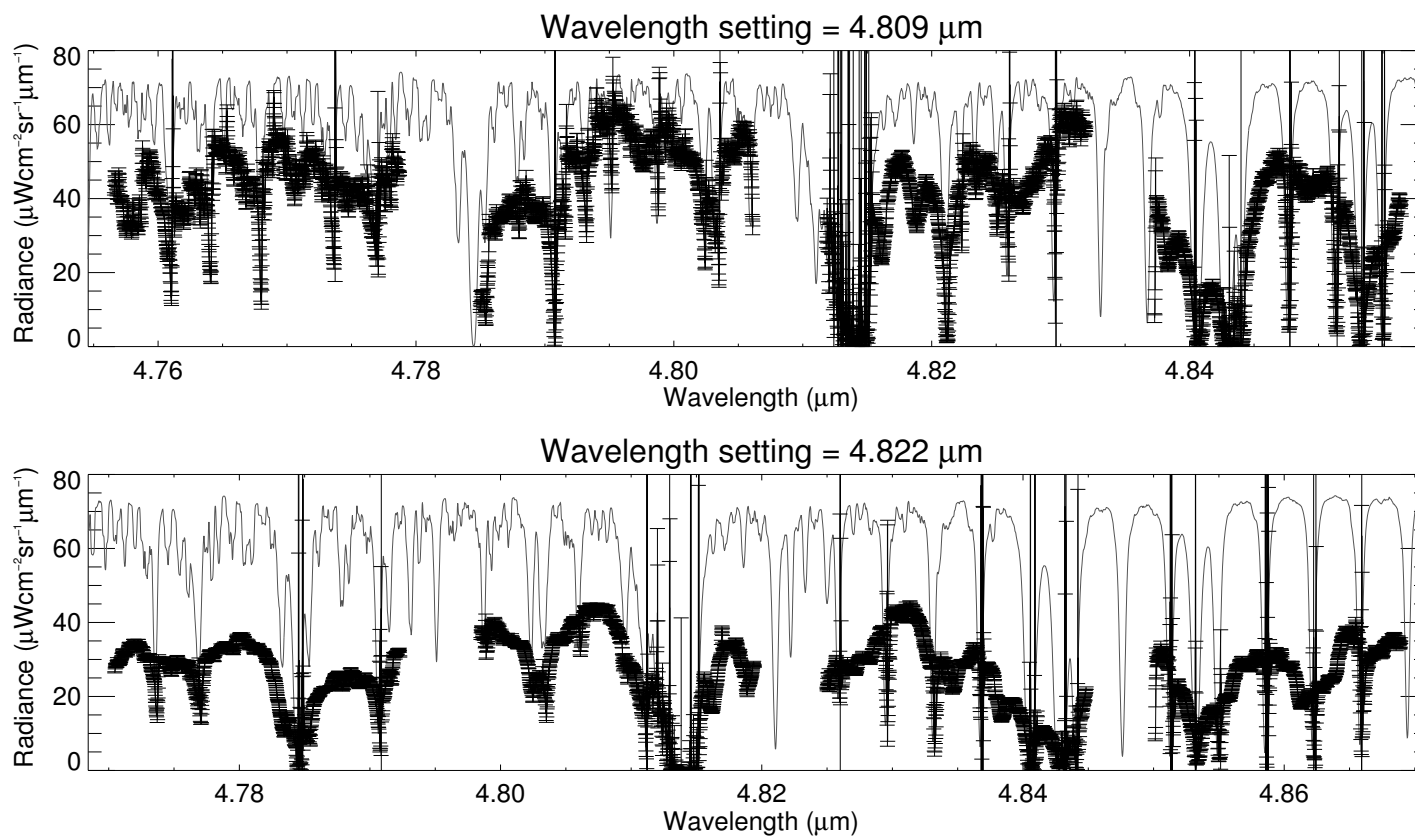


Figure 8.4.3: CRIREs observations from 12 November 2012. CRIREs data from the SEB is shown in black, the telluric transmission is shown in grey.

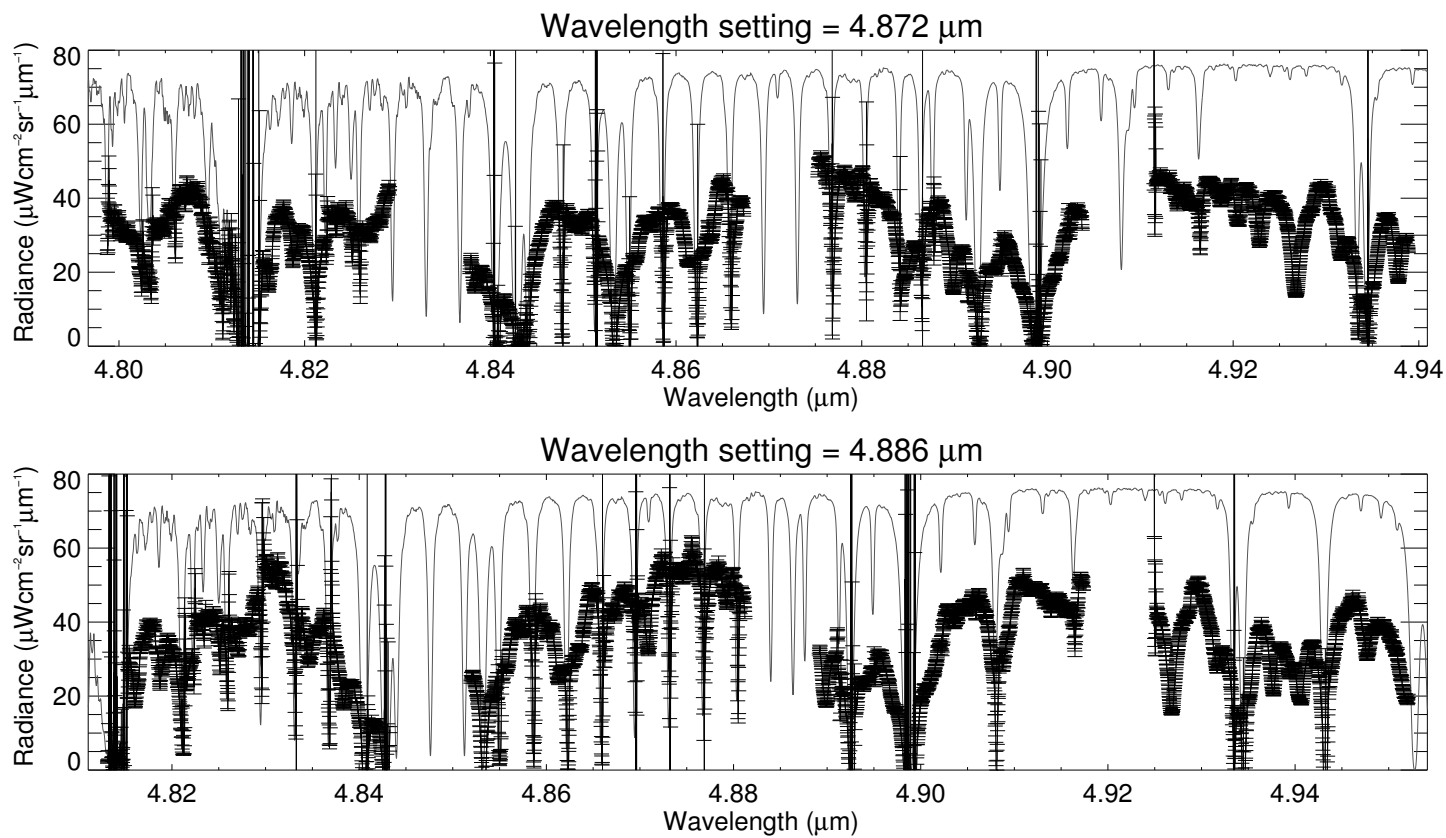


Figure 8.4.4: CRIRCS observations from 12 November 2012. CRIRCS data from the SEB is shown in black, the telluric transmission is shown in grey.

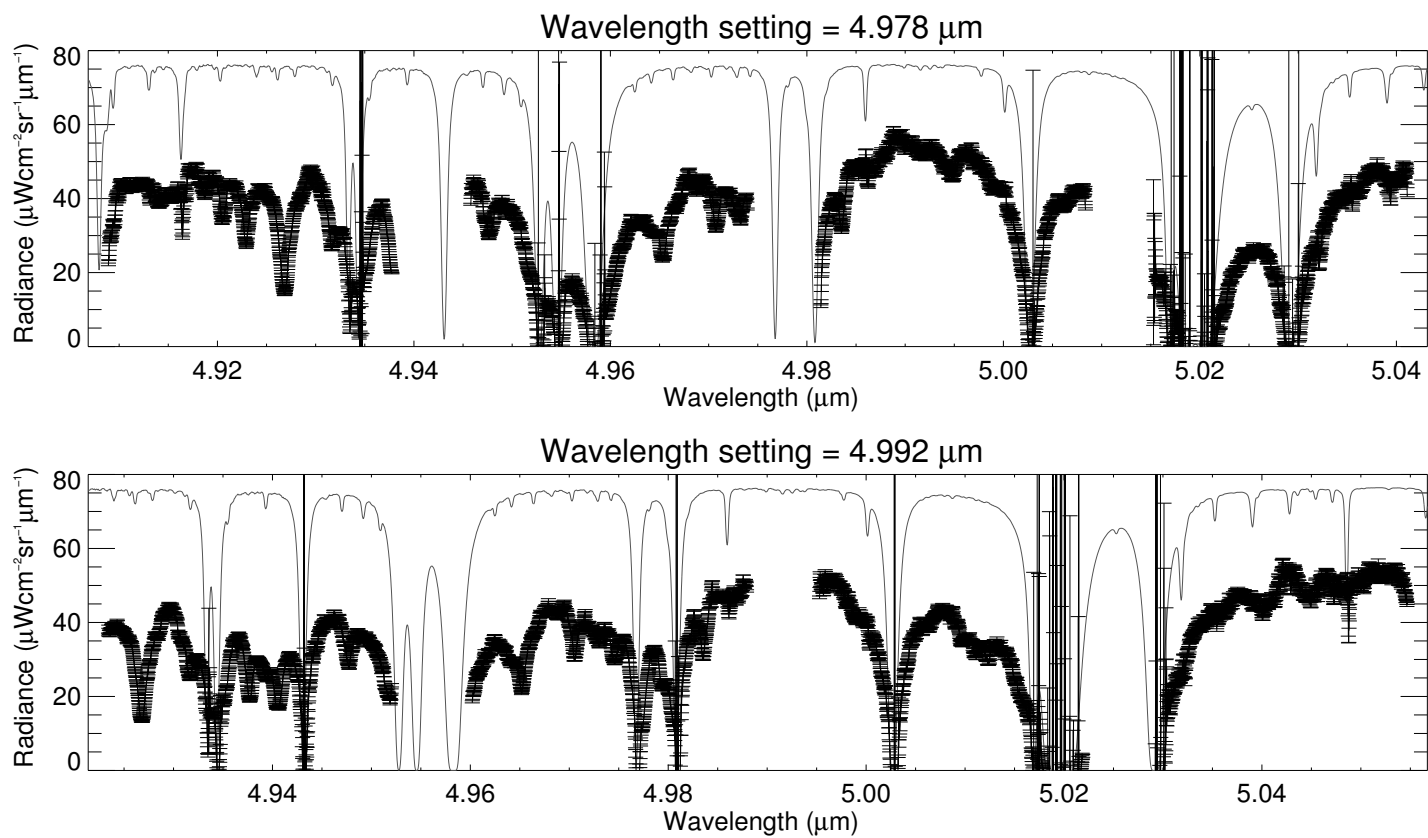


Figure 8.4.5: CRIRRES observations from 12 November 2012. CRIRRES data from the SEB is shown in black, the telluric transmission is shown in grey.

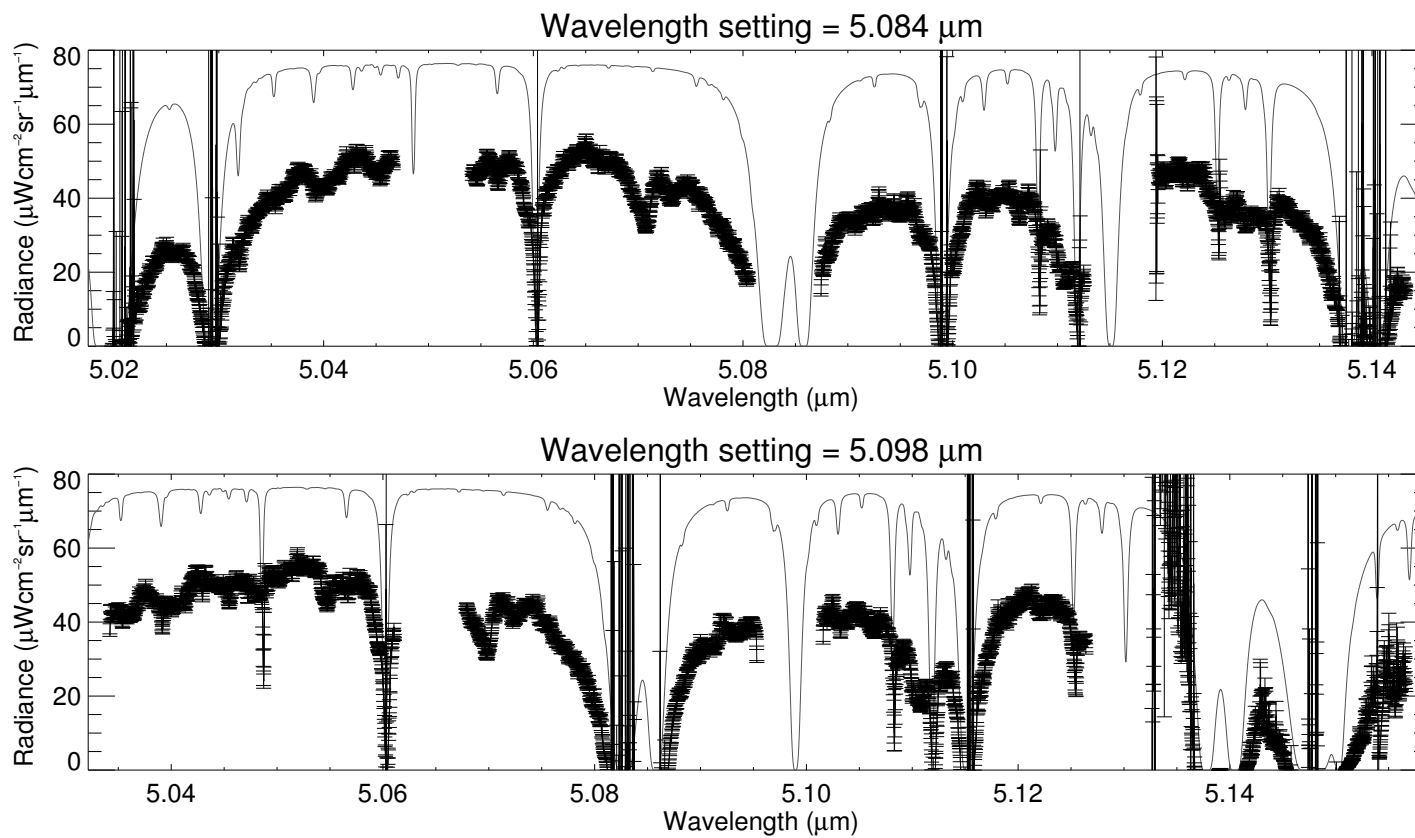


Figure 8.4.6: CRILES observations from 12 November 2012. CRILES data from the SEB is shown in black, the telluric transmission is shown in grey.

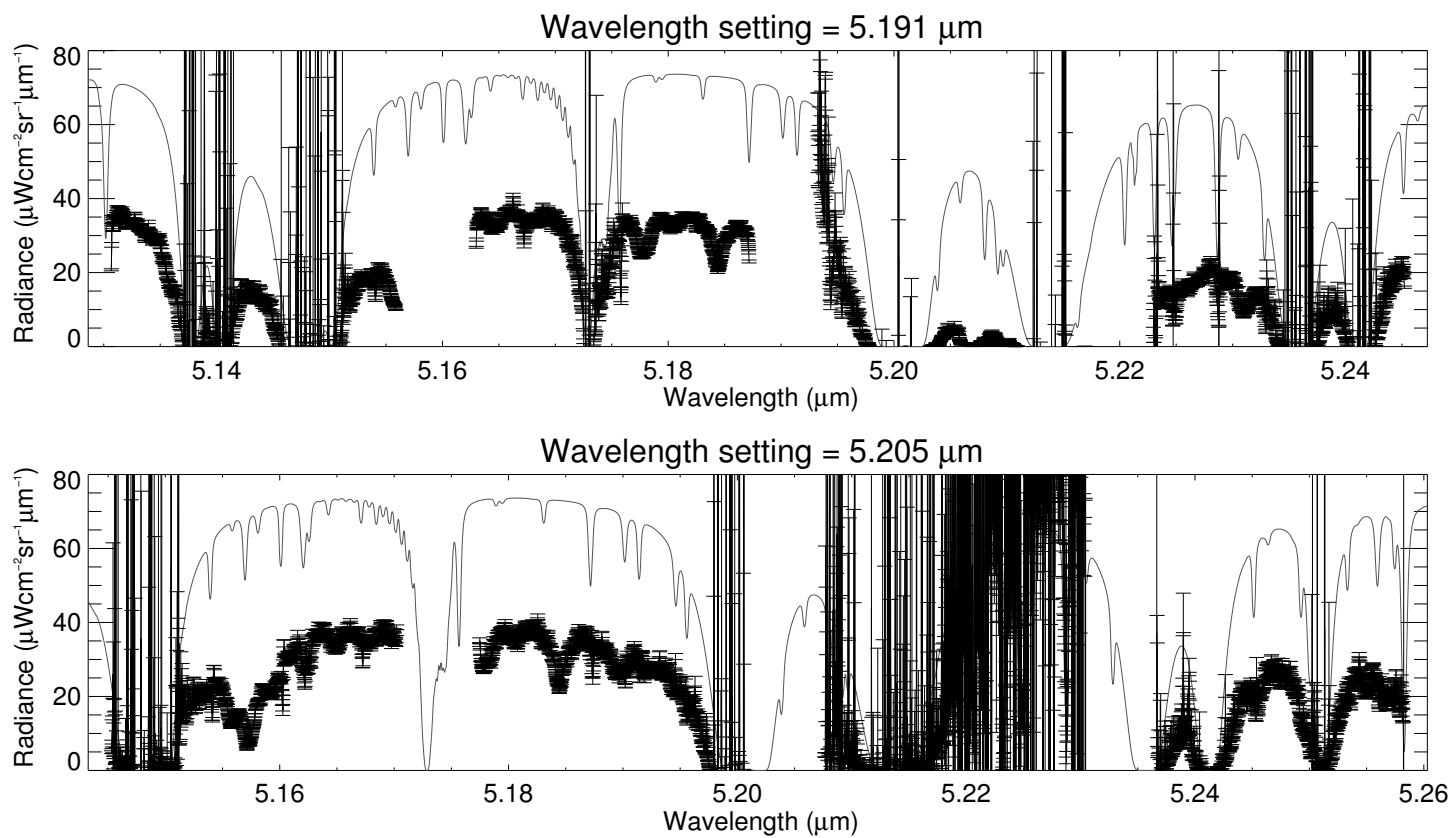


Figure 8.4.7: CRIREs observations from 12 November 2012. CRIREs data from the SEB is shown in black, the telluric transmission is shown in grey.