Tropospheric Gas Composition and Cloud Structure of the Jovian North Equatorial Belt

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High spatial resolution Voyager infrared interferometer spectrometer spectra of the North Equatorial Belt (NEB) reveal longitudinal variability of 5-µm brightness temperatures of order 100°C. These observations are used to investigate spatial variations in the gas composition and cloud structure of the NEB. We use an anisotropic multiple scattering radiative transfer model to calculate synthetic spectra for comparison with the IRIS observations. The spectral dependence of cloud extinction from 180 to 2300 cm^{-1} is modeled using Mie theory. The entire spectral range of the IRIS observations (180–2300 ¹) is used to constrain the cloud properties and vertical structure of the NEB. Within the model, cm⁻ cloud base locations vary with assumed gas abundances according to thermochemical equilibrium. We find that spatial variations in the abundance profiles of the condensible species, para hydrogen profiles and cloud optical depths can be used as tracers of the local and large-scale dynamics. Based on the spectral dependence of NH₃ cloud extinction that is required to fit the IRIS observations, we conclude that the bulk of the NH₃ cloud extinction is provided by large particles, effective radii $\approx 100 \ \mu m$; however, a small particle mode may also be present. We find that the observed 5-µm brightness temperature structure can be reproduced by spatial variations in cloud opacity and water relative humidity. NEB hot spots, due to their low cloud opacity, provide a unique opportunity to study the deep cloud structure in the Jovian atmosphere. Cloud opacity is required at P > 4 bars (coincident with the location of the thermochemically predicted H₂O cloud) to reproduce the observed continuum level near 2130 cm⁻¹, as well as to model the overall shape of the continuum between 2100 and 2300 cm^{-1} . Water relative humidity is found to vary spatially above the base of the water cloud increasing from $\approx 15\%$ in hot spots to 100% in colder spectral ensembles. The variation of relative humidity is strongly correlated with the variation of cloud opacity, suggesting dynamic depletion of water vapor above the cloud forming level as the most plausible model to explain the spatial variation in the water profile within the NEB.

1. INTRODUCTION

Knowledge of the bulk composition, elemental abundances, and their isotopic ratios provides important constraints for models of the formation and evolution of Jupiter as well as the solar system as a whole. Vertical and horizontal variations in the gas composition are important for understanding the physical and chemical processes active in the Jovian atmosphere. Analysis of the Voyager infrared interferometer spectrometer (IRIS) observations of the Jovian North Equatorial Belt (NEB) over the whole spectral range 180–2300 cm⁻¹ allows us to constrain simultaneously the thermal profile, gas abundances, and the properties of the multilayered clouds in the Jovian atmosphere, as well as their vertical and horizontal variability.

In this paper, we describe the analysis model and present the tropospheric gas composition and cloud structure results from our analysis of the Voyager IRIS observations of the NEB. We report here the results of our analysis for NH₃, H₂S, PH₃, CH₃D, and GeH₄ and the NH₃, NH₄SH, and H₂O clouds. Due to the importance of water, *Carlson et al.* [1992*a*] provided a detailed description of our analysis of its abundance, vertical distribution, and horizontal variability, and we briefly summarize those results here. Our results for the height-dependent parahydrogen fraction profile and its horizontal variations are presented by *Carlson et al.* [1992*b*]. While all of these molecules have been previously detected in groundbased, airborne, and spacecraft observations [e.g., *Kunde et al.*, 1982; *Bjoraker et al.*, 1986*a*, and

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Marten et al. [1981] were the first to consider a realistic cloud model in their analysis of the far-infrared (180–1200 cm⁻¹) IRIS observations of the equatorial region. They used the successive orders of scattering method to include isotropic multiple scattering effects; for the spectral dependence of NH₃ cloud extinction, they used Mie theory and the complex refractive index of NH₃ ice. With this model, Marten et al. concluded that the NH₃ cloud is composed primarily of large particles with radii larger than 30 μ m and that spatial variations in NH₃ cloud opacity alone cannot account for the observed 5- μ m brightness temperature variations. Thus, at least one additional cloud layer is required in the Jovian atmosphere, at pressures greater than 1.5 bars, the maximum depth to which far-infrared observations are sensitive.

Within the IRIS spectral interval $(180-2300 \text{ cm}^{-1})$ only the measurements in the 5- μ m region $(1800-2300 \text{ cm}^{-1})$ are sensitive to the deeper cloud structure. Previous investigators [e.g., *Bjoraker et al.*, 1986*a*, *b*; *Lellouch et al.*, 1989] have assumed that they could ignore the influence of spectrally dependent cloud extinction from multiple cloud layers by utilizing a single absorbing cloud of spectrally uniform

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Fig. 1. Particle single scattering albedos for NH₃ ice (dotted lines), NH₄SH ice (dashed line), and H₂O ice (solid line) as a function of wavelength for the 5 μ m region. These single scattering albedos have been calculated for particle size distributions with effective radii of 3 and 100 μ m for NH₃, 3 μ m for NH₄SH, and 10 μ m for H₂O.

optical depth to reduce the continuum level of a gas-only model to match the IRIS observations. Clouds, however, alter the outgoing radiation not only through extinction but also through scattering and self-emission. Thus, if the single scattering albedo of the cloud particles is $\tilde{\omega}_0 > 0$, direct and diffuse transmission, reflection of downwelling radiation, and reduced emission from nonblack cloud particles must all be specifically accounted for as a function of wavelength.

Figure 1 shows the spectral dependence of the cloud particle single scattering albedos in the 5- μ m region for the three thermochemically predicted cloud constituents, NH₃, NH₄SH, and H₂O. In all cases, the single scattering albedos are >0.5, clearly indicating that multiple scattering effects are important for all three clouds. At these wavelengths the absorbing cloud approximation greatly overestimates the cloud self-emission and does not include the diffusely transmitted and reflected contributions. Also a spectrally uniform cloud optical depth and single scattering albedo does not adequately represent the 5- μ m spectral dependence of NH₃, NH₄SH, or H₂O cloud extinction. Thus the simple expedient of using a grey-absorbing cloud does not adequately model the radiative processes that occur in the Jovian cloud layers.

The inadequacy of an absorbing cloud model is illustrated by Figure 2 which compares synthetic spectra calculated with (solid line) and without (dashed line) the inclusion of multiple scattering. As can be seen from the difference plot in the lower portion of Figure 2, multiple scattering effects are clearly important throughout the entire spectral range of the IRIS observations. Many of the difficulties encountered by previous investigations can be traced to an inadequate treatment of cloud extinction. Kunde et al. [1982] performed the only study utilizing the full spectral range of the IRIS observations, since they were interested in retrieving the tropospheric gas composition and not the cloud structure, Kunde et al. sorted the NEB observations to isolate the hottest spectra. These hot spot spectra, for the purposes of their investigation, were assumed to be cloud-free. Nevertheless, Kunde et al. were compelled to include an absorbing haze in their model for the 1800-2300 cm⁻¹ region in order to reduce the gas-only model continuum to match that of the IRIS observations. Kunde et al. noted that their ad hoc haze model was not unique and that other types of haze or cloud models could be developed to account for the residual continuum not modeled with their parameterization. Indications that an absorbing haze parameterization is not correct come from the difficulties Kunde et al. encountered in deriving self-consistent gas abundances for the well-mixed species in the Jovian atmosphere. For example, the Q-branch region of the ν_2 CH₃D band at 2200 cm⁻¹ suggested a mole fraction in the range of 0.5 to 4.0×10^{-7} with 1.0×10^{-7} providing the best fit, while the P-branch lines between 2120 and 2145 cm⁻¹ required a higher value of 4.0 to 5.0 \times 10⁻⁷. Kunde et al. attributed this difference to unspecified uncertainties in establishing the continuum.

Even larger difficulties were encountered by Kunde et al.



Fig. 2. Comparison of synthetic spectra of the (a) 200–1200 cm⁻¹ and (b) 1800–2300 cm⁻¹ regions calculated with (solid line) and without (dashed line) the inclusion of multiple scattering. In both spectra, cloud extinction is spectrally dependent. The difference between the synthetic spectra is shown in the lower portion of the figure, illustrating the importance of multiple scattering effects. The radiance units are ergs s⁻¹ cm⁻² sr⁻¹/cm⁻¹.

[1982] when retrieving the gas abundances of the condensible species (i.e., NH₃ and H₂O). For example, gaseous NH₃ is the dominant source of molecular opacity from 180 to 300 cm^{-1} where the emission originates between 0.6 and 1.0 bar and from 850 to 1100 cm^{-1} where the emission originates between 0.2 and 0.8 bar. Since these two spectral regions provide redundant information regarding the NH₃ profile in the 0.6- to 0.8-bar region, a single NH_3 profile should simultaneously fit both regions. However, having assumed that the spectra correspond to cloud-free regions and therefore having omitted the spectrally dependent extinction associated with the NH₃ cloud, Kunde et al. were unable to simultaneously fit both spectral regions. Their inferred NH₃ profile fits only the 850–1100 cm^{-1} interval to within the noise of the IRIS measurements, while the difference between the synthetic and observed spectra is of order 1 to 2 K for the 180–300 cm^{-1} region where the instrumental noise is of order 0.03 K.

Kunde et al. [1982] did not identify the expected location of the NH₃ cloud base in their retrieved NH₃ profile, even though NH₃ is close to saturation in the 0.2- to 0.4-bar region, nor did they explain how NH₃ becomes increasingly subsaturated to a maximum depletion by a factor of ≈ 3 at 0.7 bar and then converges to the solar value at 0.8 bar. The near-saturated values retrieved in the 0.2- to 0.4-bar region and the failure of a single gas profile to fit both the 180–300 and 850–1100 cm⁻¹ spectral regions strongly suggest that NH₃ cloud opacity should have been included in the gas abundance retrieval.

Since most gas abundance retrievals rely heavily on fitting line-to-continuum ratios, it is essential that the continuum be properly modeled. This implies that the atmospheric cloud structure must be physically consistent with the vapor profiles of the condensible species and that multiple scattering effects between the different cloud layers be properly modeled. Our model for the far infrared is similar to that used by *Marten et al.* [1981], although we use the doubling and adding method [*Hansen and Travis*, 1974] and include the effects of anisotropic multiple scattering. The primary difference between our radiative transfer model and those used in previous analyses is the inclusion of spectrally dependent cloud extinction for all three cloud layers.

Our approach to atmospheric modeling also differs from previous studies in that we include thermochemical equilibrium calculations along with our radiative transfer model. We set the gas abundances in the deep atmosphere and use thermochemical equilibrium equations to determine the cloud base locations. Extra layer boundaries are then inserted into the model structure coincident with these cloud base locations. Above the condensation level, the vapor profile of the condensible species either follows saturation or a prescribed relative humidity profile. Thus the vapor profiles and cloud locations are interactive and, perhaps more important, self-consistent with the inferred gas abundances. The effects of photochemistry on the profiles of PH₃ and NH₃ are modeled by specifying vapor-to-gas scale height ratios above a specified pressure level. The value of these ratios are constrained by the IRIS observations. Even though the IRIS observations are instantaneous, the effects of the mean circulation can be inferred from variations of hydrogen para fraction, vertical distribution of cloud opacity and water relative humidity.

The signal-to-noise ratio of the IRIS data precludes the

analysis of individual spectra. Thus it is advantageous to group the observations according to emission angle and brightness temperature selection criteria that increase the homogeneity of the samples. Grouping spectra with similar shapes and radiances is a more powerful approach than averaging over regions or latitude zones because it allows for a more detailed analysis of vertical structure and horizontal variability than possible when horizontally heterogeneous observations are mixed together. The details of the IRIS observations and our selection criteria are provided in section 2. Details of our radiative transfer/atmospheric model of the entire IRIS spectral range are given in section 3. The results of our investigation are presented in section 4 followed by our conclusions, and a discussion of some of the unanswered questions is given in section 5.

2. Observations

The IRIS instrument obtained infrared measurements covering the spectral range from 180 to 2500 cm⁻¹ (4–55 μ m) with a spectral resolution of 4.3 cm^{-1} . The details of the instrument design are described by Hanel et al. [1977]. The average noise equivalent radiance for individual Voyager spectra at the time of Jupiter encounter falls off steeply from 3×10^{-8} W cm⁻² sr⁻¹/cm⁻¹ at 180 cm⁻¹ to a minimum of 4×10^{-9} at 400 cm⁻¹ and rises nearly linearly to 1.2×10^{-8} at 2000 cm⁻¹ [Hanel et al., 1979]. Thus one of the primary advantages of the IRIS data set is the high wavelength-towavelength precision of the measurements. Figure 3 shows a representative IRIS spectrum plotted as brightness temperature. The spectrum corresponds to the NEB of Jupiter and consists of an average of 35 individual IRIS spectra. As can be seen, instrumental noise characteristics (dashed line) and atmospheric emission characteristic divide the spectral range into two well-separated intervals. Thermal emission in the spectral interval from 180 to 1500 cm^{-1} originates at pressures less than 1.5 bars, while the 1800-2500 cm^{-1} region contains emission mostly from below the 1.5-bar level.

This natural division has prompted previous investigators to employ different radiative transfer techniques with different simplifying assumptions to analyze the two spectral ranges separately. We too make use of this natural division by using the 180–1200 cm⁻¹ region to constrain the properties of the upper ($P \le 1.5$ bars) troposphere, but then we use the same radiative model and the entire spectrum to analyze the deep atmospheric structure. Thus our final gas profiles and cloud properties provide the best fit to the entire spectrum.

Since the signal-to-noise ratio for the average spectra, corresponding to each of our ensembles, is smaller than the standard deviation of the spectra within that ensemble, we interpret this observed variability as a measure of the spatial inhomogeneity of the Jovian atmosphere combined with the effects of emission angle variations. Accordingly, we assess the quality of our model fit by requiring that the difference between the synthetic and observed spectra be smaller, at all wavelengths, than the standard deviation of the individual spectra comprising the average of an ensemble of IRIS spectra. Furthermore, the low noise level associated with the measurements means that the shape of the average spectrum is meaningful. Thus we can use the shape of the spectrum to provide strict constraints on our retrieved parameters.



Fig. 3. Representative IRIS NEB spectrum (solid line). The noise equivalent radiance is shown (dashed line). This NEB spectrum consists of the average of 35 individual IRIS spectra. The gaseous absorption features are labeled.

To provide homogeneous subsets for our spectral comparisons, we group the NEB data according to the selection criteria shown in Table 1, which also shows the number of spectra in each group. The first three selection criteria are similar to those used by Kunde et al. [1982] and define spectra with near-vertical viewing conditions and with sufficient spatial resolution to analyze the longitudinal structure of the NEB. The data used were obtained between ±1 day of closest approach (Flight Data Subsystem Count (FDSC) values between 16359.24 and 16419.24). Additional radiance criteria are used to further subdivide our spectral ensembles. The average brightness temperatures near 226 cm⁻¹ (45 μ m), and over the interval 1950–2150 cm⁻¹ (5- μ m region) are good indicators of cloud opacity located at $P \leq 1$ bar and P \leq 5 bars, respectively. Since we are also interested in the deep cloud structure, our 226 cm⁻¹ brightness temperature criterion is necessarily coarse to ensure the highest signalto-noise ratio and spatial homogeneity possible for the 1800-2300 cm⁻¹ region. Inspection of the values in Table 1 shows clearly that no spectra are simultaneously hot at 226

TABLE 1. Selection Criteria for the Voyager IRIS Spectra

Category	$T_B \ (226 \ {\rm cm}^{-1})$	T_B (1950–2150 cm ⁻¹)	Number of Spectra
1	140-149	180-200	19
$\overline{2}$	140-149	200-230	76
3	140-149	230-250	74
4	140-149	≥ 250	1
5	≥ 149	180-200	0
6	≥ 149	200-230	8
7	≥ 149	230-250	66
8	≥ 149	≥ 250	52

Criterion 1, restricted to the North Equatorial Belt (7-13°N); criterion 2, restricted to emission angle $\leq 30^{\circ}$ ($\mu \geq 0.866$); and criterion 3, selected by brightness temperature at 226 ± 2 cm⁻¹ and 2050 ± 100 cm⁻¹.

 cm^{-1} ($T_B \ge 149 \text{ K}$) and cold at 1950–2150 cm⁻¹ ($180 \le T_B$) \leq 200 K). Conversely, there is only one spectrum that is cold at 226 cm⁻¹ and hot at 1950–2150 cm⁻¹. This suggests a connection, also noted by Gierasch et al. [1986], between the optical depth of the NH₃ cloud and the total atmospheric opacity at 5 μ m. We will examine this connection in detail in the following sections. B. E. Carlson et al. (Belt zone variations in the Jovian cloud structure, submitted to Journal of Geophysical Research, 1992) we show the frequency distributions of observed brightness temperatures at key wavelengths over $\pm 25^{\circ}$ latitude (planetographic). This survey identifies only a small number of distinct spectral ensembles with most spectra representing linear variations of cloud opacity and relative humidity between several "end-members." By separating spectra into distinct, relatively homogeneous, ensembles, we are able to benefit from averaging to reduce measurement noise without mixing different atmospheric structures.

Figure 4 shows the spatial distribution of the individual IRIS observations in the NEB. In this figure, the characters plotted correspond to one of the eight categories defined in Table 1. The size of the character is proportional to the size of the circular IRIS field of view in degrees along the horizontal axis. (Note the difference in the scale of the axes.) The observations are fairly well distributed in longitude and thus representative of the overall NEB cloud structure. Note, however, that the NEB is spatially quite heterogeneous. Thus spatially averaging the data will not provide an accurate measure of the inhomogeneities present within the NEB.

The range of observed variation within the NEB is illustrated in Figure 5. The NEB hot spot ensemble (category 8) is shown by the solid line and the coldest NEB spectral ensemble (category 1) by the dotted line. The offset between the category 8 and category 1 spectra indicates an overall temperature shift of ≈ 1.5 K in the hydrogen portion of the



Fig. 4. Spatial distribution of the IRIS NEB observations. The characters 1–8 refer to the spectral ensembles defined in Table 1. Character size is proportional to the size of the IRIS field of view in degrees longitude. The apparent lack of latitudinal overlap is an artifact the difference in the scale of the axes.

spectrum (300–800 cm⁻¹). Larger shifts in brightness temperature due to variations in the NH₃ vapor profile and NH₃ cloud opacity are evident in the 200–300 cm⁻¹ and 900–1200 cm⁻¹ regions of the spectrum. The more extreme difference in the 1800–2300 cm⁻¹ region of the spectrum is due to a substantial increase in the deep cloud opacity and water relative humidity in the colder spectral ensemble.

3. MODELING PROCEDURE

Our procedure for modeling the Jovian IRIS spectra consists of generating synthetic spectra and comparing them with the Voyager observations. The first step in the development of our radiative transfer model is the modeling of line-by-line molecular absorption in a vertically inhomoge-



Fig. 5. Comparison of the average IRIS spectra corresponding to the extreme NEB spectral ensembles: category 1 (dotted line) and category 8 (solid line). The nearly constant offset with wavelength throughout the hydrogen region of the spectrum (300-800 cm⁻¹) is indicative of an ≈ 1.5 K temperature difference between these two regions.

neous atmosphere. This involves integration over the vertical extent of the atmosphere using some appropriate spectral resolution $(\Delta \nu)$ for the monochromatic calculations. Exploratory calculations showed that a spectral resolution of 0.5 cm^{-1} for the wavelength intervals 180-900 and 1800-2500 cm^{-1} and 0.1 cm^{-1} for the interval 900-1200 cm^{-1} were sufficient to resolve the tropospheric absorption features; that is, higher spectral resolution produced no discernable differences when the spectra are convolved with the IRIS instrument function to a resolution of 4.3 cm^{-1} . Stratospheric emission features due to acetylene and ethane require much higher spectral resolution to model accurately, but since ethane and acetylene do not directly impact the tropospheric gas composition and cloud structure analysis, we defer study of these species for a separate study and ignore their emission features in the spectral comparisons.

The opacity due to H_2 is calculated using an algorithm supplied by B. J. Conrath (personal communication, 1986) which uses absorption coefficients based on the work of Birnbaum and Cohen [1976] but with a temperature dependence based on the work of Bachet et al. [1983] and Dore et al. [1983]. Contributions from H₂-He collisions are included assuming a helium mole fraction of 0.12 [Gautier et al., 1981]. For the remaining gases, line-by-line calculations are performed using the Goddard Space Flight Center (GSFC) line atlas [cf. Bjoraker et al., 1986a, b; Kunde et al., 1982]. For PH_3 , we use the Gestion et Etude des Informations Spectroscopiques Atmosphériques (GEISA) line atlas [Husson et al., 1982] for the v_1 and v_3 bands and new line data for the $2\nu_2$, $\nu_2 + \nu_4$, and $2\nu_4$ bands [Tarrago et al., 1987]. For NH_3 we use the line data for the 1800-2100 cm⁻¹ region from Lellouch et al. [1987] for our calculations. For CH₃D, we include both the ν_2 and $2\nu_6$ bands using the HITRAN data base [Rothman et al., 1987].

The line shapes are modeled using a Voigt profile [*Pierluissi et al.*, 1977; *Drayson*, 1975]. To account for the sub-Lorentzian behavior of the far wings, we have adopted the common expedient of truncating the Lorentz profile 50 cm⁻¹ from the center of each line. We have verified that this choice of line cutoff does not effect our conclusions regarding gas abundances and cloud properties provided that the actual wing contribution extends beyond 10 cm⁻¹. While our spectral integration over $\Delta \nu$ is standard [*Oinas*, 1983] and similar in many respects to the direct integration techniques developed by *Kunde and Maguire* [1974] and *Scott* [1974], our approach to the integration over the vertical extent of the atmosphere is different.

The vertically inhomogeneous atmosphere is divided into 15 "homogeneous" atmospheric layers per decade of pressure spanning the pressure range 0.001 to 20 bars. Extra layer boundaries, coincident with the thermochemically predicted cloud base locations, are inserted into the baseline vertical structure by subdividing the original layers. (We have tried as many as 300 layers for the vertical resolution to verify that the vertical inhomogeneity of the atmosphere is adequately resolved.)

Mixing ratios of all gases are set at 20 bars; variations at higher levels are calculated for gases that condense, react chemically or photochemically. We use Raoult's law

$$P_i = K x_i$$

TABLE 2.Thermodynamic Data at 298.15 K

Reaction		Δ <i>H</i> , kJ/mol	ΔG , kJ/mol	ΔS, J/mol K
$H_2O(q)$	\rightarrow H ₂ O(l)	-45.00	~8.57	-122.20
$H_2O(q)$	$\rightarrow H_2O(s)$	-51.25	-7.97	-145.17
$NH_3(q) + 1$	$H_2S(q) \rightarrow NH_4SH(s)$	-89.97	-0.38	-282.45
$NH_3(g)$	$\rightarrow \mathrm{NH}_3(\mathrm{s})$	-31.67	9.86	-139.29

Data have been compiled from Weast [1983], Washburn [1928], Kelley and King [1961], and Wagman et al. [1968].

to determine the saturation vapor pressure P_i (atm) of species *i*, where *K* is the equilibrium coefficient and x_i is the mole fraction. The equilibrium coefficient is calculated from

$$\ln K = \Delta G/RT$$

where ΔG is the change in Gibbs free energy for the reactions as given in Table 2, R is the gas constant, and T the temperature in Kelvins [cf. *Carlson et al.*, 1987].

Above cloud base, the gas abundance profiles of NH_3 and H_2O can be set to follow constant relative humidity profiles representing either saturation (100% relative humidity) or subsaturation (reduced relative humidity), or as in the case of water where other data suggest that the relative humidity follows a more complex profile, a more detailed profile can be prescribed by specifying a pressure interval over which the relative humidity. The resultant relative humidity profile is a simplified version of the more complex multilayered profiles found on earth [e.g., Sassen et al., 1989] and is similar to that predicted from studies of moist convection on Jupiter [cf. Del Genio and McGrattan, 1990].

We evaluate absorption line coefficients at the temperatures and pressures corresponding to layer edges and assume that the absorption coefficients vary linearly between the upper and lower layer boundaries. In the case of hydrogen the pressure squared dependence of the absorption coefficient is explicitly included by means of analytic weighting factors that accurately account for the pressure-temperature layering structure used in the model.

Though seemingly innocuous, a very basic problem involves specification of the temperature gradient within individual layers of the model atmosphere to represent the overall atmospheric temperature profile. The direct consequence of choosing a particular form for this intralayer temperature gradient can be costly in computer time and/or numerical accuracy. Often, atmospheric layers are approximated as being isothermal. While this simplifies Planck function computations, the isothermal layer approximation is valid only in the optically thin limit. In cases with substantial opacity, isothermal layering can introduce large "digitization" features in the computed spectra. To avoid these problems, we assume that the temperature gradient within each atmospheric layer can be represented as being linear in Planck function. Differences in the brightness temperature calculated with this assumption and with a linear in temperature intralayer gradient at 2130 cm⁻¹, the most transparent spectral region and therefore the most sensitive to the vertical resolution of the model, are less than 0.01 K for all cases. In addition, this choice offers a number of numerical advantages.

The rationale for using the linear-in-Planck intralayer

temperature gradient is the following. Thermal radiation emitted by a thin element of an isolated atmospheric layer (in a nonscattering atmosphere) is given by

$$dE(\mu) = B_{\nu}(\tau')e^{-\tau/\mu} d\tau'/\mu \qquad (1)$$

where μ is the cosine of the emission angle and $B_{\nu}(\tau')$ is the local Planck function. Within the given layer, the local temperature at point τ' on the relative optical thickness scale τ is prescribed by its assumed linear dependence on the Planck function, i.e.,

$$B_{\nu}(\tau') = B_t + \tau'/\tau(B_b - B_t) \tag{2}$$

where we use B_t to denote the Planck function evaluated at the top edge of the layer at temperature T_t and frequency ν and with B_b similarly defined at the bottom edge of the layer. For this assumed temperature gradient the thermal emission from the entire layer is obtained in closed form: thermal radiation emitted in the direction μ by the isolated atmospheric layer is given by

$$E(\mu) = B_t - B_b + [B_b + \mu/\tau(B_b - B_t)](1 - e^{-\tau/\mu})$$
(3)

for the upward directed beam and

$$E^{*}(\mu) = B_{b} - B_{t} + [B_{t} - \mu/\tau(B_{b} - B_{t})](1 - e^{-\tau/\mu})$$
(4)

for the downward directed intensity.

For small optical thicknesses a few terms of the series expansion form of (3) and (4) are sufficient to remove the apparent singularity in the thermal emission, i.e.,

$$E(\mu) = \sum_{n=1}^{\infty} (-1)^{n+1} \frac{(B_t + nB_b)}{(n+1)!} \left(\frac{\tau}{\mu}\right)^n$$
(5)

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$$E^*(\mu) = \sum_{n=1}^{\infty} (-1)^{n+1} \frac{(B_b + nB_t)}{(n+1)!} \left(\frac{\tau}{\mu}\right)^n \tag{6}$$

respectively.

For nonscattering layers of the atmosphere, equations (3) and (4) describe the upward and downward directed emission from layers of intermediate optical depth. For small optical depths, $\tau < 10^{-3}$, the first two terms of (5) and (6) suffice. For optically thick gas layers, $\tau > 15$, the asymptotic forms of (3) and (4) are used, i.e.,

$$E(\mu) = B_t + [B_b - B_t]\mu/\tau \tag{7}$$

and

$$E^*(\mu) = B_b - [B_b - B_t]\mu/\tau.$$
(8)

The above equations are evaluated at angles corresponding to the Gauss quadrature points used in the doubling and adding calculations, with the extra point corresponding to $\bar{\mu}$, the cosine of the average emission angle of a given IRIS spectral ensemble. For atmospheric layers containing particulate matter, the full doubling equations are used to calculate the effects of multiple scattering on the reflection, transmission, and thermal emission. Adding equations, described below, are then used to obtain the reflection, transmission, and emission for a composite layer formed by combining two layers one on top of the other. By letting the subscripts a and b refer to the top and bottom layer, respectively, the procedure is

$$Q_1 = R_a^* R_b \tag{9}$$

$$Q_n = Q_1 Q_{n-1}$$
 (10)

$$S = \sum_{n=1}^{\infty} Q_n \tag{11}$$

where S is the interlayer multiple scattering function defined at the interface between the two layers. In the above, Q, R, and S represent functions of two angles, i.e., $Q_1 = Q_1(\mu, \mu_0)$, and the product of two functions implies the integration over the adjoining angles of the form

$$Q_{1}(\mu, \mu_{0}) = R_{a}^{*}R_{b} = 2 \int_{0}^{1} R_{a}^{*}(\mu, \mu')R_{b}(\mu', \mu_{0})\mu' d\mu'$$
(12)

which we perform using Gaussian quadrature.

The diffuse reflection and transmission functions R_{ab} and T_{ab} for the combined layers are obtained by

$$D_{\rm ex} = T_a + SX_a + ST_a \tag{13}$$

$$U_{\rm ex} = R_b X_a + R_b D_{\rm ex} \tag{14}$$

$$R_{ab} = R_a + X_a U_{\text{ex}} + T_a^* U_{\text{ex}}$$
(15)

$$T_{ab} = X_b D_{\text{ex}} + X_b T_b + T_b D_{\text{ex}}.$$
 (16)

Here, D_{ex} and U_{ex} are the downward and upward diffuse radiances, respectively, at the layer interface due to external illumination (to layers *a* and *b*) from above. X_a and X_b denote the direct transmission, for example, $e^{-\tau_a/\mu_0}$. In the above, the unstarred terms represent the case of illumination from above, while the starred terms denote illumination from below. A parallel set of relations to those above is used to evaluate bottom illuminated quantities such as R_a^* and T_a^* [cf. Lacis and Hansen, 1974].

The above relations define the numerical procedure for obtaining the reflection and transmission functions for scattered radiation from a two-layer stack. The upward and downward diffuse radiances at the layer interface permit evaluation of the upward and downward fluxes at layer boundaries, and allow computation of heating and cooling rates. To compute thermal emission, the procedure continues with evaluation of the downward and upward emitted radiances (due to illumination sources internal to layers a and b) at the layer interface:

$$D_{\rm in} = E_a^* + SE_a^* + R_a^* E_b + SR_a^* E_b \tag{17}$$

$$U_{\rm in} = E_b + R_b D_{\rm in}. \tag{18}$$

Here, because of azimuthal symmetry, D_{in} and U_{in} are functions of only one angle. The upward and downward directed thermal emission from the combined layers is then given by

$$E_{ab} = E_a + X_a U_{\rm in} + T_a^* U_{\rm in} \tag{19}$$

$$E_{ab}^{*} = E_{b}^{*} + X_{b}D_{in} + T_{b}D_{in}.$$
 (20)

The above set of equations provides the recipe for combining (or adding) the radiative properties of individual atmospheric layers. To obtain the reflection, transmission, and emission of each individual layer, the above equations are used in their "doubling" form by starting with the analytic expressions for first- and second-order scattering [*Hansen and Travis*, 1974], which are "exact" in the optically thin limit ($\tau \approx 10^{-5}$) used to initialize the doubling procedure. In the case of layers with gas only absorption, (3) and (4) fully describe the upward and downward thermal emission.

Within each model layer, gaseous absorbers and cloud particles are taken to be uniformly distributed; that is, each model layer is characterized by its extinction optical depth, its single scattering albedo, and scattering phase function. For each model layer, at each wavelength, the monochromatic radiative parameters are computed as weighted averages of contributing constituents. Thus

$$\tau_n = \tau_{\text{ext},c} + \tau_{\text{ext},a} + k_i u_i + k_j u_j + \cdots$$
(21)

$$\bar{\omega}_n = (\tau_{\text{scat.}c} + \tau_{\text{scat.}a})/\tau_n \tag{22}$$

$$g_n = (g_c \tau_{\text{scat},c} + g_a \tau_{\text{scat},a})/(\tau_{\text{scat},c} + \tau_{\text{scat},a})$$
(23)

where τ_n is the total extinction optical depth (cloud, aerosol, and gaseous absorption contributions) within the given layer. Similarly, $\bar{\omega}_n$ is the ratio of total scattering to extinction optical depth, and g_n is the weighted average of the asymmetry parameter according to contributing scattering optical depths.

In the course of adding the stack of atmospheric layers to obtain the outgoing radiance, we also compute the radiance contribution function to determine the fractional contribution of each model layer to the outgoing radiance at each wavelength. This is accomplished numerically by differencing the outgoing radiance as successive atmospheric layers are added, starting at the top, going down. Thus

$$C_a(\mu) = E_a(\mu) \tag{24}$$

$$C_b(\mu) = E_{ab}(\mu) - E_a(\mu)$$
 (25)

$$C_n(\mu) = E_{mn}(\mu) - E_{lm}(\mu)$$
(26)

By normalizing the C_n , we obtain the normalized contribution function which fully incorporates the effects of atmospheric temperature on Planck emission, including as well the effects of multiple scattering from contributing cloud layers. This depicts the level of peak emission in the atmosphere more accurately than the traditional weighting function approach [e.g., *Conrath et al.*, 1970] limited to gaseous opacity distribution alone. Aside from its diagnostic value of identifying the atmospheric region from which the outgoing radiation originates, the normalized contribution function is an essential element of the temperature retrieval algorithm.

For our angle integration, we use three Gauss quadrature points with the phase matrix renormalization described by *Hansen* [1971]. The particular viewing geometry emission angle is specified as an "extra" angle as described by *Lacis* and *Hansen* [1974]. For reflected solar radiation, three quadrature points are far too few, but since the thermal sources are diffusely distributed, good accuracy can be obtained. Comparisons using a larger number of Gauss quadrature points indicate angle integration errors for the three-point quadrature to be $\leq 1\%$ for the intended applications. A more complete description of the doubling and adding method is given by *Lacis and Hansen* [1974].

Our radiative transfer model also allows for the vertical variation of the parahydrogen fraction. The strength of the S(0) hydrogen line near 360 cm⁻¹ is a direct indicator of the parahydrogen fraction, while the wing of the S(0) line is sensitive to the vertical gradient of f_p . Since NH₃ and PH₃ are photochemically reactive, the abundances of these gases in the upper troposphere (in the region of the temperature minimum) are determined by vapor to gas scale height ratios, above the base of the NH₃ cloud ($P \approx 0.5$ bar) for NH₃ and above the 1.0-bar level for PH₃, initially constrained to match the profiles determined by *Kunde et al.* [1982].

Our procedure for determining the temperature profile is similar to the standard inversion procedures described by Conrath et al. [1970]; however, we have replaced the weighting functions with normalized contribution functions. Since the normalized contribution functions include the effects of atmospheric temperature in Planck emission as well as the multiple scattering effects from the contributing cloud layers, it more accurately depicts the peak emission level than the traditional weighting function approach which is based solely in the distribution of atmospheric gaseous opacity. An initial thermal profile, supplied by B. J. Conrath (personal communication, 1986) provides the starting point for our investigations. This profile is iteratively adjusted until the synthetic and observed spectra agree to within the noise equivalent radiance of the average IRIS spectrum at selected wavenumbers within the 200-1200 cm^{-1} interval. The specific frequencies used are 287, 310, 340, 475, 520, 540, 560, 580, 600, 900, 980, 1060, 1098, and 1147 cm^{-1} . As in previous analyses [e.g., Kunde et al., 1982], at pressures greater than 1 bar, we have initially extrapolated the profile with an adiabatic lapse rate calculated for a hydrogen mole fraction of 0.88 and a helium mole fraction of 0.12 and taking into account the variation of specific heat with temperature. We have also examined the temperature profile in the deep atmosphere by inverting the 5- μ m measurements.

Once the gas profiles are specified and the cloud condensation levels determined, the radiative and physical properties of the clouds must be prescribed. The spectral dependence of the cloud particle absorption and scattering characteristics is determined from Mie theory using a twoparameter gamma function [Hansen and Travis, 1974, equation 2.56] to represent the cloud particle size distributions for a variety of values of the effective particle radius a. Since the scattering dependence on the effective variance is found to be small for broad size distributions, we use a nominal value of b = 0.25 for the effective variance. The optical properties for NH₃ ice [Martonchik et al., 1984], NH₄SH ice (G. Sill and J. Pearl, personal communication, 1988), water [Downing and Williams, 1975] and water ice [Warren, 1984] are used to determine the scattering and extinction efficiency factors, Q_{scat} and Q_{ext} , respectively, for the NH₃, NH₄SH, and H₂O cloud layers as a function of wavelength.

The optical depth of clouds is specified as an extinction optical depth τ_{ref} at a reference wavelength ($\lambda = 0.5 \ \mu m$), where the Mie calculated extinction efficiency factor Q_{ext} is normalized to unity. At other wavenumbers of interest, the monochromatic cloud optical depth is given by the product of τ_{ref} and the value of Q_{ext} at that particular wavenumber. Retrieved cloud parameters are thus expressed in terms of an effective particle radius *a* and optical depth referenced at

0.5 μ m (τ_{ref}) for each cloud, but the full wavelength dependence of their optical effects is included in the calculations.

Since it is well known that ice cloud particles are not spherical and tend instead to come in a wide variety of shapes which depend on both temperature and cloud environment, it may seem surprising that Mie calculated scattering parameters using a Henyey-Greenstein phase function are capable of representing the scattering properties in a meaningful way. It should be stressed, however, that to first order, it is the cloud composition that matters most, which is represented through the spectral dependence of the refractive indices and the corresponding Mie scattering parameters $(Q_{\text{ext}}, g, \tilde{\omega}_0;$ the extinction efficiency factor, asymmetry parameter, and single scattering albedo, respectively). Second, it is the ratios of the particle size to the wavelengths of interest that are important (i.e., size parameter, $2\pi r/\lambda$). In this regard, Hansen and Pollack [1970] have shown that terrestrial cirrus cloud particle properties at near-infrared wavelengths could be modeled by using "equivalent spheres" to represent the ice particles. This also means that the broad spectral coverage of the IRIS measurements is well-suited for particle size determination. Furthermore, thermal emission implies a diffusely scattered contribution which would ameliorate differences in scattering angle dependence between spherical and nonspherical particles that are much more apparent in polarization and single scattering cases. Finally, if, as occurs in terrestrial ice clouds, complex and variable ice crystal shapes are found, then this complexity may lead to an additional smoothing of the scattering phase function. Wielicki et al. [1990] have studied the effect that phase function has on retrieved cirrus cloud properties and conclude that a simple Henyey-Greenstein phase function may prove to be appropriate.

The vertical distribution of cloud opacity is modeled using a particle-to-gas scale height ratio H_p/H_g . Thus an individual cloud can physically extend through several layers in our vertical structure, can physically overlap, and can be composed of several different size distributions.

The model output is in the form of synthetic spectra for the $180-2300 \text{ cm}^{-1}$ spectral interval convolved with the IRIS instrument function. A key feature of this analysis is that we analyze the whole IRIS spectral range, using the same radiative transfer model and atmospheric cloud structure for all wavelengths. Use of the entire IRIS spectral range combined with the high precision of the IRIS measurements allows us to use the shape of the spectrum to analyze the composition and cloud structure of the Jovian atmosphere. The model also permits full access to the results of monochromatic calculations at any wavenumber.

To the extent possible, our model retains explicit (unparameterized) dependence on physical variables, such as temperature profile, gas abundance, cloud particle composition, size, and optical depth, the cloud base condensation level and ratio of the particle-to-gas scale height, and, in the case of photochemically reactive species, a reference pressure above which the gas abundance falls off according to specified ratios of the vapor to gas scale heights. This allows for a more careful and self-consistent investigation of the vertical distributions of gas and particle opacity.

4. **RESULTS**

Retrieval of gas abundances requires knowledge of the pressure-temperature profile and the vertical distribution of

TABLE 3. Ratios Relative to H₂

Element	Abundance		
H ₂	1		
He	1.35×10^{-1}		
U C	1.36×10 8 35 × 10 ⁻⁴		
Ň	1.74×10^{-4}		
S	3.76×10^{-5}		
P	4.89×10^{-7}		
Ge	8.80×10^{-9}		

Solar composition based on work by Cameron [1982].

cloud opacity, as well as some indication of the vertical distribution of the gaseous absorbers. Due to their low vapor pressures, low solubilities, and long photochemical lifetimes, the gases H_2 , He, CH_4 , and CH_3D are expected to maintain a constant mixing ratio within the troposphere. This is not the case for the condensible and chemically reactive species for which the mixing ratio can, and does, vary rapidly with altitude.

Table 3 lists the solar elemental abundances relative to H_2 for some of the cosmically most abundant elements [*Cameron*, 1982]. Analysis of the Voyager IRIS data gives an H_2 mole fraction of 0.897 \pm 0.030, which implies a helium mass fraction of 0.19 \pm 0.05 [*Gautier et al.*, 1981]. The fully reduced forms of O, C, N, and S in the Jovian atmosphere are H_2O , CH_4 , NH_3 , and H_2S ; these compounds are expected to be the predominant gas phase form of these elements in the upper troposphere. Also of interest and included in Table 3 are phosphorous and germanium which are disequilibrium species in the Jovian atmosphere.

We define the location of the peak emission level as the level at which the cumulative contribution function is equal to 0.5, such that half of the outgoing radiation originates above this level and half originates below this level. Location of the peak emission level as a function of wavelength is shown in Figure 6 for the 180–1200 cm⁻¹ (Figure 6a) and 1800–2300 cm⁻¹ (Figure 6b) regions of the IRIS spectrum with (solid line) and without (dotted line) the inclusion of cloud opacity. The addition of cloud opacity shifts the location of the peak emission level upward to lower pressures, the degree to which the outgoing radiation is affected depends on the location of the peak emission level relative to the cloud. Figure 7 compares the normalized contribution functions calculated with and without the inclusion of NH₃ cloud opacity at 360 and 600 cm^{-1} , corresponding to the centers of the S(0) and S(1) hydrogen lines, respectively. At these wavelengths, the normalized contribution functions show no change between Figures 7a and 7b, confirming that radiation in the centers of the S(0) and S(1) hydrogen lines is insensitive to the NH₃ cloud. However, at 230 cm⁻¹ and in the wing of the S(0) hydrogen line at 300 cm⁻¹, the emission originates deeper and contains a significant contribution from the NH₃ cloud as evidenced by the double peaked structure of the contribution functions in Figure 7arelative to the single peaked (gas only) contribution functions in Figure 7b.

4.1. Parahydrogen Distribution

Radiatively, the orthohydrogen $(J = 1, 3, 5, \dots)$ and parahydrogen $(J = 2, 4, 6, \dots)$ components behave as a



Fig. 6. Location of the peak emission level, corresponding to a cumulative contribution function value of 0.5 as a function of wavelength for the (a) 180–1200 cm⁻¹ and (b) 1800–2300 cm⁻¹ spectral intervals with (solid line) and without (dotted line) the inclusion of cloud opacity.

mixture of two separate gases with the S(0) line near 360 cm⁻¹ formed by transitions between para levels and the S(1) line near 590 cm⁻¹ originating from transitions between ortho levels. Since transitions for which $\Delta J = 1$ are highly forbidden, a sample of pure H₂ cooled from temperatures in excess if 300 K tends to retain its "normal" 3:1 high-temperature para fraction value of 0.25. Thus, a para fraction of 0.25 observed near 0.2 bar would imply that the time scale for the equilibration process is larger than the time scale for upwelling hydrogen from deeper warmer levels. In contrast, a para fraction of ≈ 0.35 observed near 0.2 bar (characteristic of thermodynamic equilibrium near the NH₃ cloud top at ≈ 115 K) would imply that the time scale. The fact that some

degree of equilibration, though not complete, is observed [Conrath and Gierasch, 1983] implies that the time scale for equilibration is comparable to the time scale for upwelling hydrogen. Furthermore, retrieval of para fractions in excess of 0.25 near 0.2 bar implies that a gradient exists in the vertical parahydrogen profile. The location of this gradient provides insights into the nature of the equilibration process. A review of the known equilibrium mechanisms is given by Massie and Hunten [1982].

Since the NH₃ cloud opacity alters emission in the wing of the S(0) hydrogen line, the value of the parahydrogen fraction must be retrieved simultaneously with the NH₃ cloud properties. This effect is illustrated in Figure 8, where synthetic spectra for uniform para fractions of 0.25, 0.27,



Fig. 7. Comparison of the normalized contribution functions calculated at 230 (solid line), 300 (dashed line), 360 (dotted line) and 600 cm⁻¹ (dash-dotted line) (a) with and (b) without the inclusion of NH₃ cloud opacity. The lack of any change in the shape of the normalized contribution function between Figures 7a and 7b at 360 and 600 cm⁻¹, corresponding to the centers of the S(0) and S(1) hydrogen lines, respectively, illustrates that radiation at these wavelengths is insensitive to the presence of the NH₃ cloud layer. Radiation at 230 cm⁻¹ and in the wing of the S(0) hydrogen line at 300 cm⁻¹ contains a significant contribution from the NH₃ cloud as evidenced by the double-peaked structure of the normalized contribution function.

0.30, and 0.35 with (Figure 8*a*) and without (Figure 8*b*) NH₃ cloud opacity are compared to the average category 8 spectrum. Differences between the observed and synthetic spectra are plotted in the lower portions of Figures 8*a* and 8*b* along with the standard deviation of the category 8 ensemble. As shown by this comparison, none of these synthetic spectra provide an acceptable fit to the category 8 spectrum. In all cases the difference between the synthetic and observed spectra is larger than the standard deviation of the individual spectra composing the average ensemble.

 NH_3 cloud opacity is clearly required in the 200-260 cm⁻¹ region of the spectrum to reduce the gas-only model continuum to match that of the IRIS observations. In the absence of NH_3 cloud opacity (Figure 8b), a para fraction of 0.30 appears to provide the closest fit to the IRIS observations. However, once spectrally dependent cloud opacity is included, as in Figure 8a, a more complicated f_p profile is required. Radiation in the centers of the S(0) and S(1)hydrogen lines is relatively insensitive to the presence of the NH₃ cloud, where an f_p of 0.30 still provides the closest fit. However, radiation in the wing of the S(0) line near 300 cm^{-1} , which originates from deeper layers, is substantially affected by NH₃ cloud absorption. Here, a para fraction of 0.27 provides the best fit to the IRIS observations. Based on this shift in para fraction with the change in depth of the peak emission level from the S(0) line center to the wing (as illustrated by the normalized contribution functions in Figure 7), we conclude that a height-dependent para fraction profile is required to fit the IRIS observations.

As discussed above, the vertical distribution of f_p can be inferred from the differential absorption strength from line center to line edge of the S(0) line. Gierasch et al. [1986] also used the differential absorption strength but used only three wavelengths to evaluate the parahydrogen profile. While, the depth of the S(0) hydrogen line is primarily controlled by f_p , the shape of the wing is controlled by the variation of f_p with altitude. Figure 9 shows a schematic representation of the height-dependent para fraction profile that provides the best fit to the average category 8 spectrum. The height-dependent profile starts with the high-temperature equilibrium value of $f_p = 0.25$ at depth. In this simplified form, specification of the inflection points $P_{\rm L}$ and $P_{\rm U}$, defines the gradient of the parahydrogen fraction between the deep atmosphere "normal" value below $P_{\rm L}$ and the equilibrated cloud top value above the pressure level $P_{\rm U}$.

Figure 10 compares the average category 8 spectrum (dotted line) and our best fit synthetic spectrum (solid line) calculated with the height-dependent para fraction profile in Figure 9. The difference between the synthetic and observed spectra is plotted in the lower portion of the figure (solid line) along with the standard deviation of the IRIS spectra (dotted lines). The poorer fit near 200 cm^{-1} arises in part from uncertainties in the treatment of the line shapes for the far-infrared rotational lines of NH₃, from increased noise in the IRIS observations near this extreme in its spectral range, and from possible inhomogeneities in the vertical distribution of NH_3 . Absorption by the H_2 dimer, evident in the difference spectrum near 350 cm⁻¹, has also been neglected in these calculations. Thus our best fit parahydrogen profile for the NEB hot spots has $P_{\rm L} = 0.5 \pm 0.1$, $P_{\rm U} = 0.2 \pm 0.1$, and $f_p = 0.32 \pm 0.01$ aloft. In determining the uncertainties associated with the retrieved parameters, we varied each parameter until the difference between the synthetic and observed spectra is no longer less than the standard deviation of the individual IRIS spectra comprising the specific ensemble. We also allowed for all compensating adjustments in the other model parameters. For example, in addition to the trade-offs between the effects of simultaneously varying $P_{\rm L}$ and $P_{\rm U}$, it is also possible to partially offset the spectral effect of shifting $P_{\rm U}$ from 0.2 to 0.1 bar by increasing the vertical extent of the NH₃ cloud (i.e., increasing H_p/H_g from 0.15 to 0.25).

Spatial variations in the parahydrogen profile exist within the NEB. Figure 11 shows a comparison of our best fit synthetic spectrum (solid line) to the average category 2 IRIS spectrum (dotted line). The colder 45- μ m brightness temperatures, as evident in the 200-300 cm^{-1} region of the spectrum, are indicative of an optically thicker NH3 cloud in this spectral ensemble, relative to the NEB hot spots. While the retrieved hot spot para fraction profile provides a technically acceptable fit (i.e., the difference between the synthetic and observed spectra falls within the standard deviation of the individual spectra), a more equilibrated para fraction profile improves the quality of the fit. Our best fit para fraction profile for this colder spectral ensemble has $P_{\rm L}$ $= 0.55 \pm 0.05$ bar, $P_{\rm U} = 0.10 \pm 0.05$ bar, $f_p = 0.33 \pm 0.01$ for the cloud top (equilibrated) para fraction and $f_p = 0.25$ at depth. While the results presented above were calculated with $f_n = 0.25$ at depth, values in the range of 0.25-0.26 provide an acceptable fit to the IRIS NEB observations.

If parahydrogen were in thermal equilibrium then para fractions in excess of 0.27, the equilibrium value at the 1-bar level ($T = 167 \pm 3$ K) would be found at pressures larger than 0.5 bar. The fact that para fractions in excess of 0.26 are inconsistent with the IRIS measurements allows us to conclude that ortho-para equilibration is not a gradual process that begins at depth. Moreover, the fact that the parahydrogen gradient is located within the NH₃ cloud layer strongly suggests that the NH₃ cloud particles play a crucial role in the equilibration process. Finally, we find that larger f_p values are associated with larger NH₃ cloud optical depths. Both of these findings suggest that paramagnetic conversion in NH₃ cloud particle surfaces is the dominant equilibration process consistent with the work by Massie and Hunten [1982]. A more detailed discussion of the parahydrogen fraction and its belt zone variations is given by B. E. Carlson et al. (submitted manuscript, 1992). This result helps to illustrate both the importance of using the spectral coverage of the IRIS measurements and of including spectrally dependent cloud extinction in the analysis.

4.2. NH₃ Distribution

In order to determine the well-mixed abundance of NH₃ it is necessary to probe below the levels at which photochemistry, condensation, and aqueous chemical reactions are expected to occur. Figure 12 shows homogeneous path transmission spectra for NH₃ calculated for an optical path of 10 cm atm (1 cm atm = 2.687×10^{19} molecules/cm² column) at 125 K and a pressure of 0.4 bar for the 200–1200 cm⁻¹ (Figure 12*a*) and for the 1800–2300 cm⁻¹ spectral regions (Figure 12*b*). In the 5- μ m region, we have included an additional transmission spectrum calculated for an optical path of 500 cm atm at 200 K and a pressure of 2.0 bars. Obviously, the larger abundance results in stronger absorption. As can be seen, NH₃ absorption is important in the



Fig. 8. The effect that NH_3 cloud opacity has on the retrieval of the parahydrogen fraction is illustrated through the comparison of synthetic spectra calculated for uniform para fractions of 0.25 (solid line), 0.27 (short dashed line), 0.30 (dash-dotted line), and 0.35 (long dashed line) (a) with and (b) without the inclusion of NH_3 cloud opacity. The average IRIS hot spot spectrum (dotted line) is shown for comparison. Difference spectra are plotted in the lower portion of the figure along with the standard deviation of the IRIS spectral ensemble.



Fig. 9. Schematic representation of our best fit heightdependent para fraction profile to the NEB hot spot spectral ensemble. The inflection points P_L and P_U define the gradient of the para fraction between the deep atmosphere "normal" value below P_L and the equilibrated cloud top value above P_{IL} .

far-infrared from 200 to 260 and 700 to 1200 cm⁻¹ and in the 5- μ m region from 1800 to 2050 cm⁻¹. Since radiation at far-infrared wavelengths originates at pressures less than 1.5 bars, the 5- μ m observations provide the best estimate of the deep NH₃ abundance. Absorption by water vapor dominates from 1930 to 2100 cm⁻¹, thus only the 1800 to 1930 cm⁻¹ region of the 5- μ m spectrum can be used to constrain the deep NH₃ abundance. Note, however, that the location of the peak emission level (Figure 6) in the 1800–1930 cm^{-1} interval indicates that this radiation originates at pressures less than 3 bars, i.e., well above the level at which aqueous chemical reactions would deplete the atmospheric abundance of NH₃. Therefore our "well-mixed" deep abundance of NH₃ is only a lower limit; the actual well-mixed abundance could be as much as 30% larger, depending on the details of the aqueous chemistry [Carlson et al., 1987].

Figure 13 shows a comparison of three synthetic spectra calculated for the simplest NH₃ profile: a constant mixing ratio below the NH₃ condensation level, above which the vapor profile follows saturation. The well-mixed NH₃ abundances are 0.89×10^{-4} (solid line), corresponding to the lower limit determined by *Kunde et al.* [1982], solar (1.78×10^{-4} , dashed line), and 2.5 times solar (4.45×10^{-4} , dash-dotted line), roughly corresponding to the deep mixing ratio determined by *Marten et al.* [1980] from their analysis of radio observations at 6 and 11 cm. The average category



Fig. 10. Comparison of our best fit synthetic spectrum calculated with the height-dependent para fraction profile shown schematically in Figure 9 (solid line) and the average category 8 IRIS spectrum (dotted line). The difference between the observed and synthetic spectra is shown in the lower portion of the figure (solid line) along with the standard deviation of the individual IRIS spectra composing the ensemble (dotted lines). The difference is less than the standard deviation of the IRIS spectra at all wavelengths.



Fig. 11. Comparison of our best-fit synthetic spectrum calculated with the height-dependent para fraction profile in which $P_U = 0.1$, $P_L = 0.55$ with a cloud top para fraction of 0.33 (solid line) and the average category 2 IRIS spectrum (dotted line). The difference between the observed and synthetic spectra is shown in the lower portion of the figure (solid line) along with the standard deviation of the individual IRIS spectra composing the ensemble (dotted lines).

8 IRIS spectrum (dotted line) is also shown for comparison. From an inspection of Figure 13a, it is clear that all three mixing ratios provide too much continuum opacity from 180 to 200 cm^{-1} , too much opacity in the lines from 200 to 250 cm^{-1} and from 1100 to 1200 cm^{-1} . However, in the 5- μm region the situation is quite different. Looking first at the synthetic spectra, we see that none of them reproduce the observed spectrum very well. For example, a solar abundance provides the best fit from 1827 to 1850 cm^{-1} and from 1860 to 1880 cm⁻¹; however, it does not provide an acceptable fit from 1850 to 1860 cm⁻¹ where a 2.5 times solar abundance provides the only acceptable fit. An examination of the difference plot in the lower portion of the figure reveals that an abundance which is 2.5 times solar provides an acceptable fit to these 5- μ m observations (i.e., the difference between the synthetic and observed spectra is less than the standard deviation of the ensemble spectra).

Based on these comparisons, we conclude that a heightdependent NH₃ profile is required. While a 2.5 times solar abundance provides an acceptable fit to the 5- μ m observations, it provides too much opacity in the far infrared. Therefore the abundance of NH₃ must decrease between 3 and 1 bars.

We use the IRIS observations in the far infrared to infer the NH_3 profile in the upper troposphere. Since our model includes thermochemical equilibrium equations, the location of the NH_3 cloud base is automatically controlled by the NH_3 vapor profile. As previously shown in Figure 13*a*, a saturated vapor profile and a well-mixed solar abundance of NH_3 do not provide an acceptable fit to the IRIS NEB (category 8) hot spot observations, consistent with the findings of *Kunde et al.* [1982]. Kunde et al. used the retrieval algorithm of *Smith* [1970] to infer the NH₃ profile from radiance measurements of 967, 970, and 980 cm⁻¹. This direct inversion approach is preferable to our best-fit iterative modeling procedure. Thus, it is gratifying that for pressures less than 0.8 bar, our retrieved profile is in excellent agreement with the results of Kunde et al.

Our preferred NH₃ profile has a constant NH₃ mole fraction of 2.8×10^{-5} in the 0.5 to 1.0 bar region. Above 0.5 bars, NH₃ condenses and is also photochemically destroyed. Consistent with the results of Kunde et al. [1982], we find that the vapor profile above 0.5 bar is subsaturated. We find that the mole fraction of NH₃ decreases from 2.8×10^{-5} at 0.6 bar to 8.8×10^{-6} at 0.45 bar and 2.4×10^{-7} at 0.3 bar. By comparison, Kunde et al. found NH₃ abundances decreasing from 3.0×10^{-5} at 0.6 bar to 8.9×10^{-6} at 0.45 bar and finally to 1.4×10^{-6} at 0.3 bar. The major difference between our preferred NH₃ profile and that inferred by Kunde et al. is that Kunde et al. assumed that the NH₃ abundance increased to the solar value, 1.78×10^{-4} at 0.8 bar which we have already shown does not provide an acceptable fit to the IRIS observations. This assumption would also shift the location of the NH₃ cloud base from 0.5 bar with our temperature profile to 0.7 bar.

An additional distinction is that *Kunde et al.* [1982] could not simultaneously fit both the 180–300 and 900–1200 cm⁻¹ regions of the spectrum with a single NH₃ profile. Based on the location of the peak emission levels, radiation in the 180–300 cm⁻¹ region contains information from the 0.6- to



Fig. 12. Comparison of homogeneous path transmission spectra for NH_3 calculated (a) for an optical path of 10 cm atm at 125 K and 0.4 bar for the 200–1200 cm⁻¹ region and (b) for an optical path of 10 cm atm at 125 K and 0.4 bar and for an optical path of 500 cm atm at 200 K and 2 bars for the 1800–2300 cm⁻¹ region.

1.0-bar region of the atmosphere while the 900–1200 cm⁻¹ interval contains information from the 0.2- to 0.8-bar region of the atmosphere. Since the 0.6- to 0.8-bar region is common to both spectral intervals, a single NH₃ profile should simultaneously fit both spectral regions. While their fit to the 900–1100 cm⁻¹ region is excellent, their fit in the 180–300 cm⁻¹ region is unacceptable. This is a direct consequence of their assumption, based on their need for additional continuum opacity in the 180–300 cm⁻¹ region of the spectrum, that the NH₃ abundance increases to the solar value at 0.8 bar. As shown in Figure 13*a*, a solar abundance of NH₃ results in a poor fit to the depth of the NH₃ lines and to the overall continuum shape since the calculated spectra exhibit a slope different from observed [cf. *Kunde et al.*, 1982, Figure 7*a*].

Reducing the NH₃ abundance to 2.8×10^{-5} in the 0.8- to 1.0-bar region improves the fit of the synthetic spectrum in the NH₃ line centers but does not alter the continuum level. However, the inclusion of NH₃ cloud opacity does. The contribution of the spectrally dependent NH₃ cloud extinction to the outgoing thermal radiation is shown in Figure 14. The dashed line is the synthetic spectrum for a pure gas atmosphere, while the solid line includes the additional opacity provided the NH₃ cloud. The average IRIS hot spot spectrum (category 8, dotted line) is also shown for comparison. The difference between the synthetic spectra calculated with and without the NH₃ cloud opacity is shown in the lower portion of the figure. The spectral regions that are sensitive to NH₃ cloud opacity are longward of 300 cm⁻¹, in



Fig. 13. Comparison of synthetic spectra calculated for well-mixed abundances of NH₃ equal to 0.89×10^{-4} (solid line), 1.78×10^{-4} (dashed line), and 4.45×10^{-4} (dash-dotted line) and the average category 8 IRIS spectrum (dotted line) for the (a) 180–1200 cm⁻¹ and (b) 1800–2000 cm⁻¹ spectral regions. The NH₃ abundance is constant with height below the NH₃ saturation level, above which the profile follows saturation. The radiance units are ergs s⁻¹ cm⁻² sr⁻¹/cm⁻¹.

the "continuum" between the S(0) and S(1) hydrogen lines, and from 700 to 1200 cm⁻¹. Note that the centers of the S(0)and S(1) hydrogen lines are relatively insensitive to the presence of the NH₃ cloud. Details of the retrieved NH₃ cloud structure are presented in the following section.

Figure 15 summarizes our retrieved NH₃ profile. For comparison, the results of thermochemical equilibrium model calculations assuming an NH₃ mixing ratio of 4.5 \times 10^{-4} with solar and 10 times solar abundances of H₂S are shown. Above 0.6 bar, our profile is in excellent agreement with the results of the Kunde et al. [1982]. We find, however, that the abundance of NH₃ increases to a value of 4.45 \times 10^{-4} near 2 bars. This value depends, however, on the shape of the NH₃ profile in the 1- to 2-bar region of the atmosphere. We note that Kunde et al. did not attempt to retrieve the deep NH₃ abundance due to uncertainties in the 5- μ m line parameters, nevertheless, increasing the well-mixed abundance of NH₃ to super solar values would have improved the agreement between their synthetic and observed spectra in the 1800–1900 cm⁻¹ region of the spectrum [cf. Kunde et al., 1982, Figure 15]. Marten et al. [1980] inferred an NH₃ mixing ratio of 4.4×10^{-4} in the deep troposphere, decreasing to 1.3×10^{-4} near the 1-bar level from their analysis of radio observations at 6 and 11 cm. A similar enhancement at depth was inferred by *de Pater and Massie* [1985]. They found that extra absorption is required at pressure levels greater than 2 bars, which can be accounted for by an inaccuracy in the ammonia absorption profile at long (≥ 6 cm) wavelengths, by an overabundance of ammonia by a factor of 1.5-2 compared to the solar value, or by the presence of a water cloud at pressures between 4 and 13 bar (270 and 370 K). More recently, Jenkins and Steffes [1988] concluded, based on the upper limits for methane and water vapor opacity between 10 and 20 cm, that it is unlikely that either methane or water vapor can account for the required excess absorption. Their conclusion supports the presence of an ammonia abundance which exceeds a solar abundance by a factor of 1.5 in the 2- to 6-bar region of the Jovian atmosphere.

de Pater and Massie [1985] also found NH₃ to be depleted

160

150



Fig. 14. Comparison of synthetic spectra calculated for a pure gas atmosphere (dashed line) and with the inclusion of NH₃ cloud opacity (solid line) with the average category 8 IRIS spectrum (dotted line). The differences spectrum in the lower portion of the spectrum shows the spectral influence of NH₃ cold opacity.



Fig. 15. A summary of our retrieved NH₃ profile (heavy solid line). The lighter solid lines correspond to the results of thermochemical equilibrium model calculations assuming a solar and 10 times solar abundance of H₂S. de Pater and Massie's [1985] results should really be thought of as the triangle defined by the three open points. The results of the Kunde et al. [1982] investigation (solid circles) are in excellent agreement with out results for pressures less than 0.6 bars. While our NH₃ abundance at depth is larger than that inferred by de Pater and Massie [1985] and Kunde et al., it is in excellent agreement with the height dependent NH₃ abundances retrieved by Marten et al. [1980] (solid triangles).

by a factor of 4-5 at pressures less than 1-1.5 atm. This depletion was derived from data centered around 3 cm. This corresponds to a mole fraction in the range $3.0-3.75 \times 10^{-5}$. Their results are best summarized by the triangular region, defined by three points in Figure 15. Finally, they found that a definite cutoff or strong depletion exists at pressures less than 0.5 atm, as also proposed by Klein and Gulkis [1978], where the NH₃ mole fraction is of order 1.5×10^{-10} '. These results are in excellent agreement with our results.

Spatial variations in the NH₃ profile exist, as would be expected due to the interaction of cloud, chemistry, and dynamics. The finding that NH₃ is subsaturated in the upper troposphere (P < 0.5) bars is not limited to the NEB of Jupiter. B. E. Carlson et al. (submitted manuscript, 1992) find a similar subsaturated profile throughout the tropical and equatorial regions on Jupiter, consistent with the results of Marten et al. [1981, Figure 4].

The shaded region in Figure 15 represents the range of variability in the amount of NH₃ vapor within the NH₃ cloud. For all of our spectral subsets, we find that NH₃ is subsaturated with longitudinal variations in the degree of subsaturation. In addition, we find longitudinal variations in the abundance of NH₃ present at the 1-bar level in the NEB. These variations are as large as a factor of 3 about the value shown in Figure 15. We also find spatial variations in the deep abundance of NH₃ which are smaller in magnitude than the variations at the 1-bar level. The range of variability in the deep abundance is from 4.4 \times 10⁻⁴ to 4.6 \times 10⁻⁴ corresponding to mixing ratios that are 2.5-2.6 times solar. These longitudinal variations in the deep mixing ratio may be indicative of spatial variations in the degree of aqueous chemical depletion of NH₃ through the interaction of dynamics and aqueous chemistry (suggested by Carlson et al. [1987]). Below the water cloud, the abundance of NH_3 should be well-mixed. We defer discussion of the effect of the assumed H₂S abundances on the NH₃ profile until section 4.4.

4.3. NH₃ Cloud Properties

The effective particle radius of the NH₃ cloud is constrained by the wavelength dependence of the NH₃ cloud extinction. The particle size dependence of Q_{ext} is given in Figure 16a for the 0-1200 cm⁻¹ spectral region and in Figure 16b for the 1800-2300 cm⁻¹ spectral region. The value of Q_{ext} has been normalized to unity at our reference wavelength, $\lambda = 0.5 \ \mu m$. As seen in Figure 16a, the resonant absorption features appear as prominent spikes at \approx 390 and 1060 cm^{-1} . Note the decrease in relative strength of the resonance features with increasing particle size. The absence of these resonance features in the Jovian spectrum led Marten et al. [1981] to conclude that the bulk of the NH_3 cloud particles must be large with effective radii greater than 30 μ m. Since the time of that investigation, it has been argued that irregular particle shapes might suppress the resonance features sufficiently and allow a small particle solution. Calculations for a $1-\mu m$ volume-equivalent radius tetrahedral NH₃ ice crystal by West et al. [1989] demonstrated a reduction in the resonance feature at 390 cm^{-1} from a Q_{ext} value of 3.25 calculated for a sphere to a value of 1.5. They also found that irregular particle shape shifts the resonance feature at 1060 $\rm cm^{-1}$ to a slightly longer wavelength without decreasing Q_{ext} . More recently, Mishchenko [1991] pointed out that irregular particle shapes do indeed alter the strength of Q_{ext} at both 390 and 1060 cm⁻¹. However, these changes in Q_{ext} due to irregular particle shape are not sufficient to suppress both resonance features in the Jovian spectrum.

Alternatively, Shaffer et al. [1986] were able to suppress the small particle resonance features in their synthetic spectra by compressing the vertical cloud extent and thus increasing the gas opacity above the NH₃ cloud top. This is illustrated in Figure 17 which compares synthetic spectra calculated with an NH₃ cloud composed of particles with an effective radius of 3 μm distributed according to particle-togas scale height ratios of 0.15 (solid line) and 0.05 (dashed line) to the average IRIS hot spot spectrum (dotted line). Decreasing the value of the particle-to-gas scale height ratio compresses the cloud layer, i.e., moves the cloud top deeper to a higher pressure. From this comparison we can see that decreasing the vertical extent of the NH₃ cloud decreases the strength of the resonance features in the synthetic spectrum but does not totally eliminate them. Furthermore, decreasing the vertical extent of the NH₃ cloud worsens the fit to the IRIS observations in the "continuum" region between the S(0) and S(1) hydrogen lines, where highaltitude NH₃ particle extinction is required to reduce the continuum level of the synthetic spectrum to match that of the IRIS observations.

While the lack of observed resonance features implies that the NH₃ cloud is composed of large particles, a more compelling argument for large particle sizes can be found in the spectral dependence of Q_{ext} . From Figure 16*a*, it is clear that large particles provide almost an order of magnitude more extinction in the far-infrared than small particles. Thus the best fit in the 180-230 cm⁻¹ region is achieved with $\tau_{ref} = 2.0$ for 3.0 μ m particles versus $\tau_{ref} \approx 0.2$ for 100 μ m particles. However, in the 5- μ m region, the 3- μ m cloud opacity is an order of magnitude larger than that of the 100- μ m particle cloud, far too much to allow a fit to the observed radiances.

Figure 18 compares synthetic spectra calculated with an NH₃ cloud composed of $3-\mu m$ particles (dashed line) and 100- μm particles (solid line) to the average IRIS hot spot spectrum. The values of τ_{ref} have been chosen to provide sufficient particle extinction to provide an acceptable fit to the continuum observations near 200 cm⁻¹. As expected, based on the spectral dependence of Q_{ext} , $3-\mu m$ particles do not provide a good fit to the observations. In particular, in the far-infrared (Figure 18a) $3-\mu m$ particles produce too much absorption in the wing of the S(0) hydrogen line, due to the sharp increase in the value of Q_{ext} from 230 to 330 cm⁻¹, and in the regions of the resonance absorption features at ≈ 390 and 1060 cm⁻¹. The large particle cloud provides a much better fit to the far-infrared observations.

Figure 18b compares the synthetic spectra to the IRIS observations in the 1800-2300 cm⁻¹ region. As expected from the wavelength dependence of Q_{ext} , shown in Figure 16, the 3- μ m particles provide too much opacity in the 5- μ m region. Therefore, based on the spectral dependence of the NH₃ particle extinction, we conclude that the NH₃ cloud on Jupiter is primarily composed of large particles. This is consistent with the conclusions of the studies by *Marten et al.* [1981] and *Orton et al.* [1982]. In fact, Orton et al. noted, when they compared model calculations with measured radiances at 246, 400, 1160, and 2100 cm⁻¹, that very large particles (100- μ m) may be the most suitable, since they run into the fewest difficulties in comparison with the various measurements.

We find that the IRIS observations are best fit with a bimodal particle size distribution with most of the cloud mass concentrated in the large particle mode. While the small particle mode is not a strict requirement (i.e., synthetic spectra calculated with only 100- μ m particles fit the IRIS measurements to within the standard deviation of the individual spectra), the inclusion of a small particle mode minimizes the difference between the synthetic and observed spectra. Therefore our preferred size distribution is a mixture of 3- and 100- μ m effective radius particles. Bimodal size distributions of this type are also characteristic of terrestrial cirrus clouds [e.g., Sassen et al., 1989]. Nevertheless, the bimodal size distribution, per se, may be partly a modeling artifact because the commonly used size distributions are generally unable to describe (in a single distribution) the extremely broad range of particle sizes that appear to be characteristic of ice clouds.

The IRIS observations also provide constraints on the



Fig. 16. NH₃ particle size dependence of the spectral variation of Q_{ext} , the Mie calculated extinction efficiency factor, for the (a) 0-1200 cm⁻¹ and (b) 1800-2300 cm⁻¹ spectral intervals. The sharp peaks near 390 and 1175 cm⁻¹ correspond to the locations of the resonance absorption features.



Fig. 17. Comparison of synthetic spectra calculated with an NH₃ cloud composed of $3-\mu m$ particles with $H_p/H_g = 0.15$ (solid line) and $H_p/H_g = 0.05$ (dashed line) with the average category 8 IRIS spectrum (dotted line). Decreasing the vertical extent of the NH₃ cloud, by using a smaller particle-to-gas scale height ratio, suppresses the NH₃ resonance features.

vertical extent of the NH₃ cloud. First, we reexamine several possibilities initially considered by Orton et al. [1982]. We consider particle-to-gas scale height ratios bracketed by a distribution characterized by a large particle scale height to gas scale height ratio $(H_p/H_g = 0.50)$, suggestive of strong vertical mixing, and a distribution with weak vertical mixing, characterized by a small particle-to-gas scale height ratio $(H_p/H_g = 0.05)$. Orton et al. compared model calculations to disk equivalent brightness temperatures near 246 cm⁻¹. We examine disk resolved measurements and the full spectral coverage of the IRIS observations. Since these ratios prescribe the vertical distribution of cloud opacity in the model atmosphere it is likely that significant horizontal variations occur in the Jovian atmosphere.

The effect of changing the vertical distribution of NH₃ cloud opacity on the synthetic spectrum is proportional to the optical depth of the NH₃ cloud. Increasing the vertical extent of an optically thin NH₃ cloud produces little change in the synthetic spectrum as illustrated in Figure 19a which compares synthetic spectra calculated with particle-to-gas scale height ratios of 0.05, 0.25, and 0.50 to the average IRIS hot spot spectrum (solid line). Our best fit hot spot model has an optically thin NH₃ cloud, $\tau = 0.27$. This comparison shows that particle-to-gas scale height ratios between 0.05 and 0.25, corresponding to cloud tops in the range 0.16 to 0.3 bar, are able to reproduce the IRIS observations. Our best fit model, not shown here, has $H_p/H_g = 0.15$ corresponding to a cloud top at ≈ 0.2 bar. Increasing the vertical extent of the cloud by moving the cloud top to 0.1 bar, using $H_p/H_g = 0.5$, results in too much cloud opacity near the tropopause. With

 $H_p/H_g = 0.5$, the synthetic spectrum no longer fits the IRIS observations in the wing of the S(0) hydrogen line and the small particle mode of the size distribution starts to produce a resonance feature not observed in the IRIS spectrum. While we could suppress the resonance feature by forcing the small particle mode to have a different scale height distribution, i.e., an inhomogeneous vertical distribution of the particles with only the large particles carried aloft, the poor fit to the S(0) line wing would not be improved. Thus particle-to-gas scale height ratios larger than 0.25 are unable to fit the IRIS observations.

These effects are amplified when the optical depth of the NH₃ cloud is larger. In Figure 19b we compare synthetic spectra calculated with particle-to-gas scale height ratios of 0.05, 0.25, and 0.5 to the average category 2 IRIS spectrum. In this case, the total optical depth of the NH₃ cloud is 0.64 with the large particles contributing $\tau = 0.52$. Once again, the cloud top must be at pressures larger than 0.16 bars, $H_{\rm p}/H_{\rm g}$ < 0.25, to provide an acceptable fit to the IRIS observations. Our preferred model fits have cloud tops near 0.2 bar corresponding to particle-to-gas scale heights ratios of 0.15, similar to the value 0.14 retrieved for the NH₃ cloud by Conrath and Gierasch [1984]. These results are also consistent with the finding of Orton et al. [1982] which ruled out small particles and large values of the particle-to-gas scale height ratio as unable to reproduce the infrared radiance at 400 cm⁻¹. Orton et al. concluded that for clouds with 100- μ m particles, the particle-to-gas scale height ratio is probably less than 0.15.

Orton et al. [1982] also performed a cursory comparison of



Fig. 18. Comparison of synthetic spectra calculated with NH₃ clouds composed of $3-\mu m$ particles (dashed line) and our preferred distribution which is bimodal (solid line) to the average category 8 IRIS spectrum for the (a) 180–1200 cm⁻¹ and (b) 1800–2300 cm⁻¹ regions. The particle size dependence of NH₃ cloud extinction with wavelength precludes a solely small particle solution.



Fig. 19. An illustration of the effect of varying the particle-to-gas scale height ratio for the NH_3 cloud. Synthetic spectra calculated with particle to gas scale height ratios of 0.05, 0.25, and 0.50 are compared with (a) the average category 8 IRIS spectrum and (b) the average category 2 IRIS spectrum. The difference between the synthetic and observed spectra is plotted in the lower portion of the figures. The increased optical depth of the NH_3 cloud in the category 2 spectral ensemble increases the effect that the vertical distribution of cloud opacity has on the synthetic spectrum.



Fig. 20. Comparison of a synthetic spectrum (solid line) calculated with the thermochemically predicted NH_3 and H_2S profiles to the average category 8 IRIS spectrum (dotted line). In this case, the NH_4SH cloud base is at 2.2 bars, altering the shape of the NH_3 profile in the 1- to 2-bar region resulting in an unacceptable fit to the IRIS observations as shown in the difference plot in the lower portion of the figure. The radiance units are ergs s⁻¹ cm⁻² sr⁻¹/cm⁻¹.

the angular dependence of the radiance which strongly suggested that $H_p/H_g = 0.05$ is not suitable and that thicker clouds are preferred. This is consistent with our findings, which show that high-altitude NH₃ particle opacity is required to reduce the continuum of the synthetic spectrum to match that of the IRIS observations in the region between the S(0) and S(1) hydrogen lines.

4.4. NH₄SH Cloud Properties

Marten et al. [1981] demonstrated that NH₃ cloud extinction alone cannot account for the observed variation in equatorial and tropical $5-\mu m$ brightness temperatures. They concluded that a second cloud layer must be present at pressures larger than 1.5 bars. Bézard et al. [1983a] analyzed the IRIS observations of the equatorial region by sorting the data into four brightness temperature categories and assuming that the hottest spectra corresponded to "cloud-free" regions. They then used an absorbing cloud model to attenuate the hot spectrum and infer the location and optical depth of the cloud layer required to reproduce their colder spectral ensembles. They concluded that a second cloud layer must be present near the 190 K level, which, based on thermochemical arguments, suggested NH₄SH as the most likely cloud condensate. It is important to note that since Bézard et al. [1983a] assumed NEB hot spots to be cloud-free, they concluded that this cloud is responsible for the observed horizontal contrasts in 5-um brightness temperatures but not for the continuum level of the hot spot spectra.

The location of this cloud layer also corresponds to the

inferred location of the decrease in the NH₃ vapor profile from $\approx 4.4 \times 10^{-4}$ to 2.8×10^{-5} . Based on thermochemical equilibrium models, an NH₄SH cloud is expected to form from the stoichiometric combination of NH₃ and H₂S. In fact, one of the major successes of the early thermochemical equilibrium models is their ability to account for the nondetection of H₂S in the Jovian atmosphere. However, since this reaction involves a one-for-one combination, the abundance of H₂S would have to be 4.1×10^{-4} , a factor of ≈ 10 times solar to decrease the NH₃ abundance from 4.4×10^{-4} at pressures greater than 2 bars to 2.8×10^{-5} at 1 bar. Such a large enhancement of H₂S is difficult to reconcile with the much smaller enhancement of carbon and nitrogen. More importantly, simply increasing the abundance of H₂S to 4.1 \times 10⁻⁴ also shifts the NH₄SH cloud base to 2.2 bars which alters the shape of the NH₃ profile in the 1- to 2-bar region of the atmosphere (cf. Figure 15) and results in an unacceptably poor fit to the 5- μ m spectrum as shown in Figure 20. Thus simply increasing the abundance of H₂S with this chemical scheme is not an acceptable solution.

The formation of cloud particles by chemical reactions in planetary atmospheres is a very poorly understood process, since it depends on the kinetics of the reaction, in this case of NH₃ and H₂S gas to form a solid compound, NH₄SH [Rossow, 1978]. This situation is very different from the change of phase of a relatively volatile substance like NH₃. The saturation vapor pressure of NH₄SH over the solid is extremely low, making this an "involatile" substance [Bézard et al., 1983b]. In such cases, the nucleation rate is controlled more by the number of molecules available for the



Fig. 21. NH₄SH particle size dependence of Q_{ext} with wavelength for the 1800–2300 cm⁻¹ spectral region.

reaction in the gas phase (vapor pressure) than by the thermodynamics of the change of molecular energy with phase and, in fact, the energy of the chemical reaction is more important than that associated with the phase change. The mixing ratios of NH_3 and H_2S imply collision rates about 100 times less than for water vapor near the Earth's surface, suggesting that the times scale for growth of large cloud particles would be hours instead of minutes [Rossow, 1978]. Hence turbulent motions in an atmosphere may produce very large vapor supersaturations because the particle formation process is not effective at consuming vapor: chemical equilibrium may never pertain. Thus the NH₄SH cloud may form well above the level predicted by chemical equilibrium but not at any well-defined location. This effect could explain a much lower cloud base pressure in the presence of the large abundance enhancement of H₂S needed to explain the depletion of NH₃.

Another alternative would be the formation of a different condensate, such as (NH₄)₂S. This could explain both the depletion of NH₃ with lower H₂S abundances, but still enhanced over solar, and a different location for the cloud base. Unfortunately, the thermochemical properties of this compound are not well known, although extensive information exists regarding its formation in aqueous solutions. In solution, $(NH_4)_2S$ requires more energy to form, making NH₄SH the preferred compound, but it is important to remember that the experimental data regarding the formation of NH₄SH have only been collected at or near room temperatures: low-temperature effects may be important. Furthermore, all of these experiments have involved mixing equal molar fractions of NH₃ and H₂S [e.g., Isambert, 1881, 1882] or slight enhancements [Magnusson, 1907]. Thus the reactions may be different when NH₃ is in excess by a factor

of roughly 10 and the temperature is nearly 100°C colder. Both Walker and Lumsden [1897] and Magnusson [1907] commented on the presence of an excess of H₂S upon mixing equal molar volumes of NH₃ and H₂S, suggesting that although a condensate primarily composed of NH₄SH results from the reaction of NH₃ and H₂S, extra NH₃ is consumed in the process. Magnusson [1907] proposed that NH₃ vapor is also lost through adsorption onto the solid NH₄SH particles; his experimental results suggest that approximately 10% of the NH₃ is adsorbed at room temperatures. Colder temperatures may increase the rate of adsorption and the amount of NH₃ adsorbed, just as NH₃ is more soluble in cold water. This effect could enhance the depletion of NH_3 per molecule of H_2S , requiring a smaller abundance enhancement. It would be useful to know how much H₂S was present in excess after combining equal molar amounts to form NH4SH in these experiments; unfortunately, the excess H₂S was released. No satisfactory explanation of the conflict between the cloud base location and the magnitude of the NH₃ depletion has been found.

Figure 21 shows the particle size dependence as a function of wavelength of Q_{ext} for NH₄SH. The lack of significant particle size dependence prevents us from being able to infer the mean cloud particle size in these clouds. We can only retrieve τ_{ext} , the product of τ_{ref} and Q_{ext} , and therefore cannot uniquely determine the particle sizes. For the purposes of this investigation, we have arbitrarily selected a $3-\mu m$ particle size for the NH₄SH cloud. A different particle size assumption for the NH₄SH cloud would result in a different phase function and would only require the value of τ_{ref} to be scaled, but the radiative extinction properties would be the same.

The information content in these IRIS observations is



Fig. 22. Illustration of the effect that NH₄SH cloud opacity has on the 5- μ m spectrum. Synthetic spectra calculated with (solid line) and without (dashed line) the inclusion of NH₄SH cloud opacity are compared with the average category 2 IRIS spectrum. The radiance units are ergs s⁻¹ cm⁻² sr⁻¹/cm⁻¹.

sufficient to provide a constraint on the vertical extent of the NH₄SH cloud layer. We examine a range of particle-to-gas scale height ratios bracketed by distributions characterized by $H_p/H_g = 0.05$ and $H_p/H_g = 0.50$. Since these ratios prescribe the vertical distribution of cloud opacity in the model atmosphere, our results are necessarily dependent on the total optical depth of the cloud in question.

The small optical depth inferred for the NH₄SH cloud in NEB hot spots is insensitive to the vertical extent of that cloud, but it is not expected to be larger than that for optically thicker clouds associated with stronger vertical motions. Our colder ensembles limit the vertical extent of the NH₄SH cloud layer. If the particle-to-gas scale height ratio is 0.5, then the NH₄SH cloud particles would be mixed into the upper tropospheric NH₃ cloud layer. Since the 180–1200 cm⁻¹ observations of the NEB do not contain the spectral signature of NH_4SH ice in the continuum regions, notably 450–530 cm⁻¹ and 900–1200 cm⁻¹, we have an upper limit on the vertical extent of that cloud layer. NH4SH clouds with $H_p/H_g = 0.05$ to 0.15 are best able to fit the observations. The situation is quite different in zones, where NH₄SH cloud particles are advected upward into the NH₃ cloud layer (cf. B. E. Carlson et al., submitted manuscript, 1992).

To illustrate the effect that the NH₄SH cloud has on the 5- μ m spectrum, we calculated synthetic spectra with and without the inclusion of NH₄SH cloud opacity. These synthetic spectra are compared with the average category 2 IRIS spectrum in Figure 22. The retrieved optical depths of the NH₃, NH₄SH, and H₂O clouds are 0.64, 2.1, and 6.0, respectively. Large NH₃ cloud particles provide $\tau = 0.52$

distributed following $H_p/H_g = 0.1$, while the small particles are distributed following $H_p/H_g = 0.15$. NH₄SH cloud opacity can clearly account for the bulk of the observed 5- μ m variability as concluded by *Bézard et al.* [1983*a*].

Since atmospheric dynamics influences the formation of the NH₄SH cloud, we might expect that the chemical balance might vary with location and that H₂S might be observable in some locations. *Bézard et al.* [1983*b*] showed that the inclusion of H₂S at 1100–1200 cm⁻¹ improved the agreement between the synthetic and observed spectra. Based on our analysis of different spectral ensembles, we find that we are able to fit all of the observations using a condensation-predicted H₂S profile. However, due to the low abundance of H₂S in the upper troposphere these spectra are virtually indistinguishable from those calculated assuming no H₂S at all.

4.5. H₂O Cloud Properties

Evidence for a third, deeper cloud layer comes from the NEB hot spot observations, in particular, the continuum radiance in the 2100–2200 cm⁻¹ region. Figure 23 shows a comparison between synthetic spectra calculated with an abundance of water which is 2 times solar, with (solid line) and without (dashed line) the inclusion of water cloud opacity to the average category 8 IRIS spectrum (dotted line). NH₃ cloud properties retrieved from the 180–1200 cm⁻¹ region have been included in the calculation of both synthetic spectra. In this calculation, the abundance of water follows saturation (100% relative humidity) above the condensation level at 4.9 bars. While both synthetic spectra



Fig. 23. Comparison of the average category 8 IRIS spectrum (dotted line) to synthetic spectra for 2 times solar water calculated with (solid line) and without (dashed line) water cloud opacity. Condensation occurs at 4.9 bars. A saturated (100% relative humidity) profile is assumed above the condensation level. The optical depth of the water cloud is 4. The difference spectrum in the lower portion of the figure shows the effect of water cloud opacity on the synthetic spectrum. The difference is taken between the synthetic spectrum calculated with and without water cloud opacity. Though water cloud opacity is clearly required to fit the IRIS observations in the 2100-2200 cm⁻¹ spectral interval, a saturated water profile produces too much opacity in the 1900-2000 cm⁻¹ region. The radiance units are ergs s⁻¹ cm⁻² sr⁻¹/cm⁻¹.

have too much water vapor opacity in the 1900-2000 cm⁻¹ interval to provide a good fit to the observations, the additional opacity provided by the water cloud is still required to reduce the synthetic continuum to match that of the observations from 2100 to 2200 cm⁻¹. The poor fit in the 1900-2000 cm^{-1} interval is what prompted Kunde et al. [1982] and Bjoraker et al. [1986b] to reduce the vapor abundance in the line-forming region, 2-4 bars. However, they elected to fit the 1900-2000 cm^{-1} spectral interval by decreasing the well-mixed abundance of water in the Jovian atmosphere to 0.02 times solar. This amount of water still implies the formation of an ice cloud near 2 bars that they did not include in their calculations. As shown by Carlson et al. [1992a], this solution does not provide an acceptable fit to the IRIS observations once a physically more realistic and thermochemically consistent cloud model is used in the analysis.

Since emission in the 1900–2000 cm⁻¹ spectral interval originates at higher altitudes than that from 2100 to 2200 cm⁻¹, an alternative way to decrease the water vapor opacity in the 1900–2000 cm⁻¹ interval is to reduce the abundance of water in only the 2- to 4-bar region of the atmosphere (i.e., above cloud), rather than at all pressure levels. This suggests that decreasing the relative humidity of water above the water cloud would provide an acceptable fit to the IRIS hot spot observations.

Figure 24 illustrates such a fit. In this case, the abundance

of water is 2 times solar with saturation occurring at 4.9 bars. The optical depth of the water cloud is 4.0, the same as that shown by the solid line in Figure 23. The main difference is that in the calculation of this synthetic spectrum, the relative humidity of water above the base of the water cloud decreases to a minimum of 15% at 3 bars above which it increases to 100% at 1 bar. The optical depths of the NH₃ and NH₄SH clouds are 0.27 and 0.02, respectively. Thus water vapor and cloud opacity dominate the 5- μ m spectrum in hot spots. Based on this comparison with the IRIS hot spot observations, Carlson et al. [1992a] found that the well-mixed abundance of water deep in the atmosphere must be at least 1.5 times solar. We have adopted their preferred value of 2.76 \times 10⁻³ corresponding to an abundance of water which is 2 times solar for all of our subsequent calculations with variable relative humidity of water above the cloud.

If drying by atmospheric motions is responsible for the reduced relative humidity in hot spots, we would expect to find significant spatial variations in water relative humidity within the NEB. Figure 25 shows a comparison of our best fit synthetic spectrum calculated with an abundance of water which is 2 times solar and 100% relative humidity above the water cloud which forms at 4.9 bars and the average category 2 IRIS spectrum. The retrieved optical depths for the NH₃, NH₄SH, and H₂O clouds are 0.64, 2.1, and 6.0, respectively. Water cloud opacity is distributed following



Fig. 24. Comparison of the average category 8 IRIS spectrum (dotted line) to a synthetic spectrum calculated with a 2 times solar abundance of water and a reduced relative humidity profile above the cloud (solid line). Optimum fit is obtained with the relative humidity decreasing from 100% at cloud base (4.9 bars) to 15% at 3 bars, then increasing to 100% at 1 bar above which the relative humidity remains constant. The optical depths of the NH₃, NH₄SH, and H₂O clouds are 0.27, 0.02, and 4, respectively. This combination of parameters is our preferred fit to the entire IRIS spectrum for hot spots.

 $H_p/H_g = 0.25$. The larger NH₄SH cloud opacity found in this region, relative to the NEB hot spots, reduces the sensitivity of these observations to the relative humidity profile. Nevertheless, this comparison shows that a saturated water profile is consistent with the IRIS observations.

Intermediate spectral ensembles provide a better glimpse at the deeper levels with sufficient sensitivity to determine the relative humidity in the 2- to 4-bar region; for these cases, we find values intermediate between that of the hot spots and 100%. For the average category 7 spectrum, shown in Figure 26, the relative humidity gradually decreases from 100% at cloud base, 4.9 bars, to 40% at 2 bars, above which it increases to 100% at 1 bar. The optical depths of the NH₃, NH₄SH, and H₂O clouds are 0.39, 0.1, and 5.0, respectively. Cloud opacity is vertically distributed following $H_p/H_g = 0.15$, except for the water cloud for which $H_p/H_g = 0.25$.

The spectral correlation between relative humidity and average $5-\mu m$ brightness temperature make plausible our inference that relative humidities are even nearer to 100% in regions with thicker clouds, presumably associated with stronger vertical mixing, and that drying due to localized intense downwelling motions are responsible for the low relative humidities and cloud opacities found in the NEB hot spots (cf. B. E. Carlson et al., submitted manuscript, 1992).

Measurements in NEB hot spots are particularly sensitive to the vertical extent of the water cloud structure. If H_p/H_g = 0.5, H₂O cloud particles are pushed up into the NH₄SH

cloud layer resulting in too much cloud opacity in the 1- to 3-bar region. This alters the shape of the synthetic spectrum (cf. Figure 1) and we are no longer able to obtain a satisfactory fit to the category 8 IRIS observations. The best fit is found for $H_p/H_g = 0.15$. However, it is important to note that NEB hot spots are anomalous, nearly clear, regions and therefore not representative of even the NEB as a whole. Thus H_p/H_g for the water cloud restricted to values of 0.15 or less in hot spots is consistent with the interpretations of them as regions of localized subsidence. However, a value of $H_p/H_g = 0.25$ provides a better fit to the colder spectral ensembles in the NEB. Thus there appears to be a positive correlation between the vertical extent of the H₂O cloud layer, water relative humidity, and the optical depth of the overlying clouds (see further discussion by B. E. Carlson et al., submitted manuscript, 1992).

Figure 27 shows the spectral dependence of Q_{ext} as a function of particle size for H₂O. The lack of any particle size dependence prevents us from inferring the mean cloud particle size for this cloud. We can only retrieve the value of τ_{ext} , the product of τ_{ref} and Q_{ext} , and therefore cannot uniquely determine the particle size. For the purposes of this investigation we have selected a 10- μ m particle radius for the H₂O cloud particles [Rossow, 1978; Carlson et al., 1988]. A different particle-size assumption would result in a different phase function and would only require that the value of τ_{ref} be scaled to maintain the same radiative extinction properties.



Fig. 25. Comparison of the average category 2 IRIS spectrum (dotted line) to a synthetic spectrum calculated with an abundance of water which is 2 times solar and 100% relative humidity above the water (solid line). The optical depths of the NH_3 , NH_4SH , and H_2O clouds are 0.64, 2.1, and 6, respectively. This combination of parameters is our preferred fit to the entire category 2 IRIS spectrum.

We find that an additional cloud layer is located near 5 bars in the Jovian atmosphere. The location of this cloud layer is consistent with thermochemical predictions of a water cloud near this level. Finally, this result is consistent with the conclusions of *Borucki and Williams* [1986] that the lightning observed in the Jovian atmosphere originates near 5 bars.

4.6. H_2O Distribution

Figure 28 summarizes our retrieved H₂O profile. The results of our investigation are quite different from those reported by Kunde et al. [1982] and Bjoraker et al. [1986b]. Kunde et al. [1982] obtain a profile for the mixing ratio of water varying from 3×10^{-5} at 4 bars to 1×10^{-6} at 2.5 bars, while Bjoraker et al. [1986b] find a similar variation but attribute it to levels deeper than 4 bars. Bjoraker et al. point out, however, that the location of the levels to which the results apply is dependent on the radiative model used in the analysis. Kunde et al. assume the presence of a grey, absorbing haze with a total optical depth of 0.54 at 5 bars, uniformly mixed throughout the line-forming region, and an optically thick lower boundary at 279 K. Bjoraker et al., on the other hand, assume the presence of a grey, absorbing cloud above the line-forming region at 210 K (≈2 bars) with an optical depth of 2.93 and an optically thick lower boundary at 353 K (≈12 bars). These differences in the model boundary conditions and cloud opacities and locations lead to the differences shown in Figure 28 in the H₂O distributions inferred from the same observations. Furthermore, neither of these studies included the spectrally dependent cloud extinction associated with a H_2O ice cloud that is predicted to form near the 2-bar level based on their retrieved H_2O profiles. When the thermochemically consistent clouds are included, the calculated spectra do not fit the IRIS observations [*Carlson et al.*, 1992a].

The primary limitation of these results is that no mechanism has been found to account for an order of magnitude depletion of water abundance below the cloud forming level. Conversely, we argue that this decrease in the H_2O abundance in the line-forming region (2-4 bars) results from a reduction of water relative humidity above the base of the water cloud by dynamic motions. Spatial variations in water relative humidity thus contribute to the spatial variations observed in the 5- μ m brightness temperatures. The region between the two solid curves in Figure 28 corresponds to the range of variability that we have seen in the above cloud relative humidity profile. Analogous variations of relative humidity occur on Earth where at low latitudes the relative humidity, even near the ocean surface, varies by 20% between upwelling and downwelling components of the Hadley circulation and decreases by more than 50% with altitude, even in the convective zone [Newell et al., 1972; Oort, 1983]. The larger differences between the Kunde et al. [1982] and Bjoraker et al. [1986b] profiles result from different lower boundary conditions in their radiative transfer models as well as differences in the vertical distribution of their cloud opacity. Our profiles are significantly different from the results of Kunde et al. and Bjoraker et al. at pressures greater than 2.5 bars, since we allow the relative humidity of water to vary above the cloud and therefore do



Fig. 26. Comparison of the average category 7 IRIS spectrum (dotted line) to a synthetic spectrum calculated with an abundance of water which is 2 times solar and a relative humidity profile that gradually decreases from 100% at cloud base (4.9 bars) to a minimum relative humidity of 40% at 2 bars. The optical depths of the NH_3 , NH_4SH , and H_2O clouds are 0.39, 0.2, and 5, respectively. This combination of parameters is our preferred fit to the entire category 7 IRIS spectrum.

not need to reduce the well-mixed abundance of water in order to decrease the amount of water vapor in the 2- to 4-bar region in NEB hot spots. We note that for pressures less than ≈ 2.5 bars, the Bjoraker et al. profile begins to converge with ours. Above the 1-bar level, water appears to be saturated, consistent with the determination by *Larson et al.* [1984], which may provide the best measure of the water abundance in the upper troposphere, since most of the $5-\mu m$ radiation measured by IRIS originates at deeper levels.

The IRIS measurements do not allow us to uniquely constrain the well-mixed abundance of water. Since water condenses in the Jovian atmosphere, it is only possible to determine a lower limit on the well-mixed water abundance from measurements of the vapor abundance above the condensation level. Increasing the abundance of water merely shifts the cloud base to higher pressures resulting in an increased continuum level, which can be compensated for by increasing the cloud opacity to match the 2130 cm⁻¹ window radiance. Likewise, the relative humidity above the water cloud can be adjusted to match the observed line absorption in the 1900–2000 cm^{-1} region. Nevertheless, we can conclude that the well-mixed abundance of water is at least 1.5 times solar, with 2 times solar our preferred solution based on the shape of the spectrum. Moreover, while the relative humidity profile is not unique, we can confidently conclude that the relative humidity of water is reduced in the 2- to 4-bar region of the atmosphere in the hotter (average 5- μ m brightness temperatures in excess of 230 K) spectral ensembles and that the relative humidity increases with decreasing 5- μ m brightness temperature.

4.7. PH3

Phosphine is important in the Jovian atmosphere because it is a tracer of both dynamics and chemistry. *Prinn and Barshay* [1977] proposed that vertical transport in Jupiter's troposphere is rapid enough to quench chemical reactions that would otherwise destroy PH₃. (This mechanism also accounts for the observed presence of CO and GeH₄ in the upper troposphere.) Prinn and Barshay calculated that an eddy diffusion coefficient $K = 2 \times 10^8$ cm² s⁻¹ would be sufficient to transport CO on a time scale shorter than its conversion to the low-temperature form of carbon, CH₄. *Fegley and Prinn* [1985] studied the kinetics of PH₃ oxidation and concluded that for the same eddy diffusion coefficient, it is quenched at 1240 K, comparable to the 1100 K quench level for CO determined by Prinn and Barshay.

PH₃ was first detected in the 1000 cm⁻¹ region by *Ridg-way et al.* [1976] and in the 2100 cm⁻¹ region by *Larson et al.* [1977] with mixing ratios $\approx 6 \times 10^{-7}$. *Drossart et al.* [1982] determined a mixing ratio of $(4.5 \pm 1.5) \times 10^{-7}$ from their analysis of the 2200–2240 cm⁻¹ region of the IRIS data. This value is also within the range $(6.0 \pm 2.0) \times 10^{-7}$ determined by *Kunde et al.* [1982] but slightly smaller than the $(7.0 \pm 1.0) \times 10^{-7}$ determined by *Bjoraker et al.* [1986a]. The effect that varying the well-mixed abundance of PH₃ has on the synthetic spectra is shown in Figure 29 which compares synthetic spectra calculated with PH₃ model fractions of 2.0 $\times 10^{-7}$ (dashed line), 6.0×10^{-7} (solid line), and 8.0×10^{-7} (dash-dotted line) to the average category 8 IRIS spectrum. From the difference plot in the lower portion of the figure,



Fig. 27. H₂O particle size dependence of Q_{ext} as a function of wavelength for the 1800–2300 cm⁻¹ interval.



Fig. 28. Summary of our retrieved H_2O profile. The solid curves represent the range of variability observed in the Jovian atmosphere. The upper curve corresponds to saturation above the base of water cloud at 4.9 bars, while the lower curve represents the maximum depletion, the category 8 profile.

we see that a mole fraction of 2.0×10^{-7} does not provide enough absorption, while a mole fraction of 8.0×10^{-7} provides too much absorption. Thus our best fit to the IRIS observations is obtained with a well-mixed (deep) abundance of 6.0×10^{-7} .

Above the 1-bar level, UV radiation photolyzes PH₃ as first discussed by *Prinn and Lewis* [1975]. *Strobel* [1977] pointed out that NH₃ and PH₃ photochemistry are coupled. His calculations suggest that the presence of NH₃ increases the photochemical removal of PH₃; however, the reaction

$$NH_2 + PH_3 \rightarrow NH_3 + PH_2$$

was found to be very slow at Jovian temperatures [Bosco et al., 1982]. Thus, in the absence of dynamics, the PH₃ distribution in the upper troposphere, P < 1 bar, should be intermediate between the photochemical model predictions of *Prinn and Lewis* [1975] and *Strobel* [1977]. Since we are not prepared to include photochemical reactions in our model, we have adopted a parameterized height dependence based on a vapor-to-gas scale height ratio which approximates the photochemical profiles. Thus, at pressures larger than 1 bar, PH₃ is well mixed with a mole fraction of 6×10^{-7} , while at pressure less than 1 bar the abundance falls off following the vapor-to-gas scale height ratio.

The effect of varying the vapor-to-gas scale height ratio is illustrated in Figure 30 which compares synthetic spectra calculated with a well-mixed PH₃ abundance of 6.0×10^{-7} and three different vapor to gas scale height ratios: 0.1 (dashed line), 0.3 (solid line), and 0.55 (dash dotted line) to the average category 8 IRIS spectrum. A comparison of the



Fig. 29. Comparison of synthetic spectra calculated with PH₃ mole fractions of 2.0×10^{-6} (dashed line), 6.0×10^{-6} (solid line), and 8.0×10^{-6} (dash-dotted line) with the average category 8 IRIS spectrum for the (a) 900-1200 cm⁻¹ and (b) 1800-2300 cm⁻¹ spectra intervals. The difference between the synthetic and observed spectra is plotted in the lower portion of the figure. The radiance units are ergs s⁻¹ cm⁻² sr⁻¹/cm⁻¹.

900–1200 cm⁻¹ region shown in Figures 29 and 30 reveals that this region of the spectrum cannot be used to uniquely constrain both the well-mixed abundance of PH₃ and its vertical variation. Therefore we rely on the 5- μ m portion of the spectrum to constrain the well-mixed abundance of PH₃. Varying the vapor-to-gas scale height ratio has little effect on the 5- μ m spectrum since the radiation originates below the 1-bar level. However, as shown in Figure 30, it has a pronounced effect on the 900–1200 cm⁻¹ spectral interval. In this case, vapor-to-gas scale height ratios of 0.1 and 0.55 provide unacceptable fits to the IRIS observations as shown in the difference plot in the lower portion of the figure. Our best fit is a vapor to gas scale height ratio of 0.3.

We find, however, that a single vapor-to-gas scale height ratio does not provide an acceptable fit to all of our IRIS NEB spectral ensembles. The range of observed variability



Fig. 30. Comparison of synthetic spectra calculated for a wellmixed PH₃ abundance of 6.0×10^{-6} at three different vapor-to-gas scale height ratios: 0.1 (dashed line), 0.3 (solid line) and 0.55 (dash-dotted line) above the 1-bar level with the average category 8 IRIS spectrum for the 900–1200 cm⁻¹ region.

is compared with the photochemical model predictions of *Prinn and Lewis* [1975] and *Strobel* [1977] in Figure 31. While most of the observations fall within the theoretical limits, the NEB hot spots have less PH_3 aloft than that predicted based on the coupled PH_3 -NH₃ photochemistry [*Strobel*, 1977] and may be indicative of stronger downwelling motions within the hotter spectral ensembles.

4.8. CH_4 and CH_3D

With the vapor profiles of the condensible species and cloud properties determined, we can focus our attention on



Fig. 31. Comparison of the range of our height-dependent PH₃ profiles (curves A, B, and C) with the photochemical model results of *Strobel* [1977] (curve labeled PH₃-NH₃) and *Prinn and Lewis* [1975] (curve labeled PH₃ only). The PH₃ profile retrieved by *Kunde et al.* [1982] (curve with error bars) is also shown for comparison. The vertical bars for the 900-1200 and 2050-2225 cm⁻¹ intervals show the pressure levels probed by the measurements at these wavelengths. Only the 900-1200 cm⁻¹ measurements are sensitive to the PH₃ distribution above 1 bar.



Fig. 32. Homogeneous path transmission spectrum for CH₄ calculated for an optical path of 1700 cm atm at 125 K and 0.4 bar.

the well-mixed species. For well-mixed gases such as methane, abundance determinations based on analyses of spacecraft and ground-based data at a variety of wavelengths are in good agreement. Gautier and Owen [1983] revised the earlier determination of Gautier et al. [1982] to (2.18 ± 0.18) \times 10⁻³. This CH₄ abundance is consistent with earlier determinations from ground-based observations in the visible (1.8 \pm 0.4) \times 10⁻³ [Sato and Hansen, 1979] and near-infrared at 1.1 μm (2.6 \pm 1.2) \times 10 $^{-3}$ [Buriez and de Burgh, 1980]. Recently, Bjoraker et al. [1986a] reported the first detection of the weak ν_3 - ν_4 hot band of CH₄ in Jupiter's 5- μ m spectrum. From this they were able to infer a mixing ratio for the deep troposphere of $(3.0 \pm 1.0) \times 10^{-3}$ corresponding to the C/H ratio which is 3.6 ± 1.2 times solar. This range is marginally consistent with the other values and points to difficulties in interpreting 5- μ m spectra (see discussion of CH₃D below), even for well-mixed species.

CH₄ absorption contributes significant continuum opacity in the 1150–1200 cm⁻¹ region as shown by the homogeneous path transmission spectrum in Figure 32. Based on this continuum contribution, we prefer a CH₄ mole fraction of 2.2×10^{-3} in excellent agreement with the value (2.2 ± 0.4) $\times 10^{-3}$ determined by *Knacke et al.* [1982]. All of these values are larger than the (1.75 ± 0.2) $\times 10^{-3}$ used by *Kunde et al.* [1982].

In addition to CH₄, CH₃D is also present in the Jovian atmosphere. CH₃D absorbs in both the 1050–1200 cm⁻¹ and 2050–2350 cm⁻¹ regions of the IRIS spectrum as illustrated by the homogeneous path transmission spectra in Figure 33. A comparison between synthetic spectra calculated with mole fractions of 2.0×10^{-7} (dashed line), 6.0×10^{-7} (solid line), and 8.0×10^{-7} (dash-dotted line) and the category 8 IRIS observations is shown in Figure 34. In both the 900–1200 and 2000–2300 cm⁻¹ intervals, a mole fraction of 2.0×10^{-7} provides too little absorption to fit the IRIS observations. The difference between the synthetic and observed spectra, shown in the lower portion of Figure 34, is clearly larger than the standard deviation of the individual spectra comprising the ensemble. In the absence of H₂S, a mole fraction of 6.0×10^{-7} is required to fit the continuum region near 1175 cm⁻¹, while an abundance as large as 8.0×10^{-7} provides too much opacity near 1160 and 2140 cm⁻¹. Thus our best fit to the IRIS observations in both of these spectral intervals corresponds to a mole fraction of $(4.5^{+2.2}_{-1.0}) \times 10^{-7}$. This value is consistent with the mole fraction (3.6 \pm 0.5) $\times 10^{-7}$ derived from an analysis of ground-based observations of Jupiter at 8.5 μ m [Knacke et al., 1982] and with the mole fraction (3.5^{+1.0}_{-1.3}) $\times 10^{-7}$ previously determined by Kunde et al. [1982].

Kunde et al.'s [1982] lower limit was inferred from the 1120-1220 cm⁻¹ region. However, this lower limit was established by extending the far wings of all absorbers to 2000 half widths. This, as noted by Kunde et al., is an extreme lower limit since it implies an unlikely value for the far wing CH₄ contribution [Gautier et al., 1982]. Nevertheless, both of these values are larger than the (2.0 \pm 0.4) \times 10^{-7} inferred by *Bjoraker et al.* [1986a] from their analysis of the Kuiper airborne 5- μ m observations. Differences in the cloud parameterization are responsible for these variations in the retrieved CH₃D abundance. In particular, the absorbing cloud parameterization used in the Bioraker et al. [1986a] analysis provides more opacity in the 2100-2200 cm^{-1} region of the spectrum than our spectrally dependent clouds. While Kunde et al. [1982] also used an absorbing cloud parameterization, their cloud was vertically distributed throughout the atmosphere (P < 5 bars) with a total optical depth of 0.5. In contrast, the Bjoraker et al. cloud is located at 2.1 bars (210 K) with a normal optical thickness of 2.93. This cloud is therefore located within the line-forming region for CH₃D. While Bjoraker et al. used line-tocontinuum ratios to infer the gas abundance, the location of their absorbing cloud causes it to contribute differential opacity to the line and continuum regions of the spectrum;



Fig. 33. Homogeneous path transmission spectra for CH_3D calculated for the (a) 1000–1200 cm⁻¹ spectral region for optical paths of 2 and 5 cm atm at 125 K and 0.4 bar and (b) 1800–2300 cm⁻¹ spectral region for optical paths of 2 and 5 cm atm at 200 K and 2 bars.

thus their absorbing cloud does not act as a neutral density filter and a more rigorous treatment of cloud opacity should have been included. A more complete discussion of the manner in which an absorbing cloud interacts with the radiation field is given by *Carlson et al.* [1992a].

4.9. Other Trace Gas Species

Other carbon compounds observed in the Jovian atmosphere include CO [*Beer*, 1975] and nonmethane hydrocarbons such as C_2H_2 and C_2H_6 [*Ridgway*, 1974]. Since we are concerned with the tropospheric gas composition, we leave C_2H_2 and C_2H_6 as the subject for future work. CO does absorb in the 5- μ m region of the Jovian spectrum. However, its contribution is negligible at the 4.3 cm⁻¹ resolution of the IRIS observations as illustrated in Figure 35 which compares synthetic spectra calculated with and without the inclusion of CO. The difference plot in the lower portion of the figure is the difference between the synthetic spectra calculated with and without the inclusion of CO. Based on the results of previous investigations [e.g., Noll et al., 1988; Bjoraker et al., 1986a], we have included CO using a mole fraction of 1.0×10^{-9} .

Germane is another disequilibrium species in the Jovian atmosphere. The abundance of GeH₄ inferred from this investigation is $(7.0 \pm 3.0) \times 10^{-10}$, in excellent agreement with the $(7 \pm 2) \times 10^{-10}$ inferred by *Kunde et al.* [1982] and similar to the value derived from ground-based observations 6×10^{-10} [Fink et al., 1978] and Kuiper airborne observa-



Fig. 34. Comparison of synthetic spectra calculated with CH₃D mole fractions of 2.0×10^{-6} (dashed line), 6.0×10^{-6} (solid line), and 8.0×10^{-6} (dash-dotted line) with the average category 8 IRIS spectrum for the (a) 1050–1200 cm⁻¹ and (b) 2000–2300 cm⁻¹ spectral intervals. The difference between the synthetic and observed spectra is plotted in the lower portion of the figure. The radiance units are ergs s⁻¹ cm⁻² sr⁻¹/cm⁻¹.

tions $(7.0^{+4.0}_{-2.0}) \times 10^{-10}$ [Bjoraker et al., 1986a]. The observed abundance of GeH₄ is consistent with chemical models provided that vertical mixing rapidly advects GeH₄ from the ≈ 1000 K level.

5. DISCUSSION

We have found that the well-mixed abundances of NH_3 and H_2O are both enhanced relative to solar values and consistent with the previously determined enhancement for carbon in the form of CH_4 [Gautier and Owen, 1983]. Both NH_3 and H_2O have complex height-dependent profiles controlled by the competing influences of cloud physics and dynamics. Therefore we have retrieved the heightdependent profiles of the condensible species and the cloud structure of the NEB in its simplest form. Based on terrestrial analogs and the results of moist convection models [Del



Fig. 35. Comparison of synthetic spectra calculated with (solid line) and without (dashed line) the inclusion of CO with the average category 8 IRIS spectrum (dashed line). The difference between the two synthetic spectra is shown in the lower portion of the figure.

Genio and McGrattan, 1990], it is likely that multiple saturated and subsaturated, with respect to H₂O, layers are present in the Jovian atmosphere. This implies that the water cloud may actually be a multilayered structure. Nevertheless, our conclusion that a water cloud is present near the 5-bar level based on the need for extra opacity in the 2100-2200 cm⁻¹ region is unaffected by the assumption that the water cloud is single-layered. Similarly, the NH₃ cloud may be multilayered. Regardless of the potential complexity of the vertical structure of the individual cloud layers, we find that three discrete cloud layers are required to fit the IRIS observations. Moreover, based on the height dependence of the vapor profiles of the condensible species, our results are consistent with the formation of NH₃, NH₄SH, and H₂O cloud layers as predicted by thermochemical equilibrium models [e.g., Weidenschilling and Lewis, 1973].

The inferred decrease in the abundance of NH_3 in the 2-bar region is consistent with the formation of an NH_3 containing (i.e., NH_4SH) condensate, provided that sulfur is enhanced by a factor of nearly 10 times solar and that significant supersaturations are required to form the condensate. While such a large enhancement of H_2S is difficult to understand, it is still within the range of enhancements suggested by interior and atmospheric evolution models [*Stevenson*, 1982; *Hubbard and Marley*, 1989]. Additional laboratory work is required to understand the reaction between NH_3 and H_2S ; in particular, it is possible that a significant amount of the NH_3 is lost through adsorption. Thus, at present, the abundance of sulfur in the Jovian atmosphere is poorly constrained.

Nevertheless, there is good circumstantial evidence to suspect that significant amounts of sulfur are present. Aqueous chemical reactions involving sulfur chemistry may explain some of the Jovian coloration, and as noted by *West et al.* [1985], sulfur ions in solution may exhibit the correct spectral dependence for the "dark brown" cloud features in the Jovian atmosphere. UV photolysis of NH_4SH and even H_2S itself may provide additional chromophores and add to the observed yellow coloration of the planet. Finally, based on our retrieved gas abundances, we are able to re-determine the Jovian D/H ratio using our retrieved abundances. The D/H ratio may be derived from the CH_3D abundance

$D/H = [CH_3D]/(4f[CH_4])$

where f is the deuterium fractionation factor describing the partitioning of deuterium between HD and CH₃D, and the factor of 4 accounts for the replacement of any one of the four H atoms in CH₄ by one D atom. Beer and Taylor [1978] report an f value of 1.37 ± 0.07 . More recently, Fegley and Prinn [1988] suggested a value of $f = 1.25 \pm 0.05$. For simplicity, we have adopted a value of f = 1.37 so that we can directly compare our D/H ratio with previously reported values. Our best fit CH₃D and CH₄ mole fractions suggest a D/H ratio of $(3.6 \pm 0.5) \times 10^{-5}$. This value is comparable to the value of D/H = $(3.6^{+1.0}_{-1.4}) \times 10^{-5}$ determined by Kunde et al. [1982]; note however, that their CH₄ abundance is significantly smaller than ours (when adjusted for the differences in the CH₄ abundance, the Kunde et al., D/H ratio becomes $(2.8^{+0.8}_{-1.0}) \times 10^{-5}$) while much larger than the value determined by Bjoraker et al. [1986a] (which adjusted for the differences in CH₄ abundance, becomes $(1.6 \pm 0.5) \times$ 10^{-5}). This difference with respect to the Bjoraker et al. results is not surprising, given the factor of 2 difference between the CH₃D mole fractions. The agreement between our CH₃D and CH₄ mole fractions and the D/H ratio is in much better agreement with *Knacke et al.* [1982], who obtained a D/H ratio of $(3.0^{+1.1}_{-0.8}) \times 10^{-5}$.

As discussed extensively in the literature [e.g., Kunde et al., 1982; Bjoraker et al., 1986a; Owen et al., 1986], Jupiter is believed to have retained the deuterium abundance representative of the solar nebula [Hubbard and MacFarlane, 1980]. Independent values for the protosolar D/H value can be estimated from He³ in gas-rich meteorites and in the solar wind: Black [1972] estimated $0.8 \times 10^{-5} < D/H < 3.0 \times 10^{-5}$ for the protosolar nebula from He³ in gas-rich meteorites and Bochsler [1981] obtained D/H ratios of 2–2.5 × 10⁻⁵ from the He³/H ratio in the solar wind. More recently, Anders and Grevesse [1989] suggested that a value (3.4 ± 1.0) × 10⁻⁵ is more appropriate for the protosolar value based on their analysis of the He³/H ratio in the solar wind and gas-rich meteorites.

Assuming that deuterium was originally homogeneously distributed in the solar nebula, the outer planets are expected to show D/H variations due to either fractionation during planetary formation as predicted by *Hubbard and MacFarlane* [1980] or ion-molecule reactions in the interstellar medium prior to solar system formation [*Owen et al.*, 1986]. In either case, the D/H ratio on Jupiter should be similar to that of the protosolar nebula, independent of the formation process. The excellent agreement between our Jovian D/H value of $(3.6 \pm 0.5) \times 10^{-5}$ and the protosolar value $(3.4 \pm 1.0) \times 10^{-5}$ determined from He³ in meteorites and in the solar wind [*Anders and Grevesse*, 1989] tends to confirm these predictions.

There are some conclusions that are common to all of the NEB spectral ensembles. Consistent with the NH₃ cloud analysis performed by *Marten et al.* [1981], we find that the NH₃ cloud is primarily composed of large particles with effective radii $\approx 100 \ \mu\text{m}$. Small particles do not provide sufficient opacity in the spectral region from 200 to 330 cm⁻¹ to fit the observations without increasing the optical depth of

the NH₃ cloud beyond the point that it can simultaneously fit both the far-infrared and 5- μ m observations. The results of *West et al.* [1989] strongly suggest that irregular particle shapes, tetrahedral NH₃ ice in particular, will not alter this conclusion. While particle shape may alter the strength and location of the NH₃ ice resonance features, it does not significantly alter the spectral dependence of Q_{ext} which is the basis of our conclusion that the NH₃ cloud particles are large.

Our preferred NH₃ cloud particle size distribution is bimodal with most of the cloud opacity provided by the large particle mode. The addition of a small particle component improves the overall fit but is not a strict requirement (i.e., spectra calculated with only the large particle mode fit the data). Including the small particle mode minimizes the difference between the synthetic and observed spectra by improving the shape of the synthetic spectrum. However, it is possible that the bimodal nature of the size distribution is a modeling artifact of attempting to fit a nonspherical size distribution with spherical particle parameters. Nevertheless, bimodal size distributions of this type are characteristic of terrestrial cirrus clouds [e.g., Sassen et al., 1989]. Several possibilities could explain the existence of a small particle component. First, the small particle mode may reflect growing particles or may, instead, reflect some aspect of the particle's irregular shape. Finally, it is also possible that if NH₃ can supercool, as does water, the small NH₃ particles might be composed of supercooled liquid NH₃. One of the surprises of terrestrial cirrus investigations, has been detection of supercooled liquid droplets at temperatures near 40°C [Sassen et al., 1989]. Additional microphysical and laboratory work is needed to understand more fully the nature NH₃ cloud in the Jovian atmosphere. The Galileo probe is expected to return in situ measurements of the NH₃ cloud structure.

Local variations in cloud top altitude, vertical distribution of NH₃ cloud opacity, NH₃ vapor profile, abundance of PH₃ above the 1-bar level, and parahydrogen fraction all appear to be correlated, suggesting that these properties can be used as tracers of the local atmospheric dynamics. We find the parahydrogen fraction to be correlated with the optical depth of the NH₃ cloud where regions with larger cloud opacity and more extensive clouds are associated with higher para fractions. This suggests that NH₃ cloud particles play a dominant role in the equilibration of orthohydrogen and parahydrogen, consistent with the work of *Massie and Hunten* [1982]. A more detailed discussion of the retrieved parahydrogen profiles and their implications for dynamics is given by *Carlson et al.* [1992b].

While previous investigations [e.g., Kunde et al., 1982; Marten et al., 1981; Bézard et al., 1983a] have assumed that NEB hot spots correspond to cloud-free regions, we find the NEB hot spots correspond to the clearest, though not cloud-free, regions. Some NH₃ cloud opacity is required to fit the 180–1200 cm⁻¹ observations even in hot spot regions. This result is consistent with the findings of the Pioneer 10 and 11 photometry and polarimetry analysis by Smith and Tomasko [1984], which found no evidence for spatially resolved cloud-free regions. The photometry and polarimetry models all require the presence of a "haze" in the upper troposphere.

Our results are consistent with the presence of the three thermochemically predicted cloud layers in the Jovian atmo-

Parameter	Category 1	Category 2	Category 3	Category 7	Category 8
Number of spectra	19	76	74	66	52
ų	0.976	0.977	0.957	0.977	0.958
NH ₄ cloud					
τ	0.78	0.64	0.50	0.39	0.27 ± 0.03
There portioles	0.70	0.52	0.38	0.29	0.19
Phat	0.56	0.57	0.53	0.52	0.52 ± 0.05
$H_{\rm p}/H_{\rm c}$	0.15	0.15	0.15	0.15	0.15 ± 0.10
NH ₂ SH cloud					
π	4.0	2.1	0.3	0.10	0.02 + 0.08
P	1.95	1.87	1.92	1.90	1.90 ± 0.10
- Dot $H_{\rm p}/H_{\rm c}$	0.15	0.15	0.15	0.15	0.15 ± 0.10
H ₂ O cloud	0.15	0112	0110		
7	10.0	6.0	8.0	5.0	4.0 ± 1.0
, Р.	5.0	4.9	4.9	4.8	4.9
- bot $H_{\rm p}/H$	0.25	0.25	0.20	0.15	0.15 ± 0.10
NH- profile	0.25	0.20	0.20	0110	
NH ₂ H /H	0.14	0 14	0.13	0.11	0.10 ± 0.02
NH. at 1 bar	3.3×10^{-5}	4.2×10^{-5}	3.3×10^{-5}	3.3×10^{-5}	$(2.8 \pm 0.2) \times 10^{-5}$
NH, at 3 bars	4.6×10^{-4}	4.4×10^{-4}	4.5×10^{-4}	4.5×10^{-4}	$(4.5 \pm 0.1) \times 10^{-4}$
PH, profile	4.0 × 10	1. 7 10	4.5 / 10	1.5 / 10	$(1.5 = 0.1) \times 10^{-1}$
	0.45	0.30	0.35	0.40	0.30 ± 0.15
Para profile	0.45	0.50	0.55	0.10	0.50 = 0.15
D	0.10	0.10	0.20	0.20	0.20 ± 0.10
	0.10	0.10	0.20	0.20	0.20 ± 0.10
	0.40	0.33	0.30	0.30	0.30 ± 0.10
JP H.O. profile	0.55	0.55	0.55	0.52	0.52 ± 0.01
Minimum U. O. humidity %	100	100	40	40	15
<i>P</i> minimum humidity $\%$			2.0	2.0	3.0

TABLE 4. Summary of Our Best Fit Model Results

sphere. The properties of the upper tropospheric NH_3 cirrus layer are well constrained by the IRIS observations. The spectral dependence of NH_3 cloud extinction required to fit the IRIS observations requires that the bulk of the cloud opacity be provided by large particles, consistent with the results of *Marten et al.* [1981], but the addition of a small particle component improves the spectral fit and may also be present. This may be partly a modeling artifact in that the commonly used size distributions are generally unable to describe (in a single distribution) the extremely broad range of particle sizes that appear to be characteristic of ice clouds.

The results of our investigation into the spatial variability within the Jovian NEB are summarized, for each spectral ensemble, in Table 4. Categories 4, 5, and 6 have been omitted as there is only one spectrum in category 4, no spectra in category 5, and only eight spectra in category 6. Let us first explain some of the values in Table 4. We have determined the uncertainties associated with our retrieved parameters by varying the parameter of interest until the difference between the synthetic and ensemble-average spectra is no longer less than the standard deviation of the individual spectra comprising the ensemble. In addition, we have allowed for compensating adjustments in other parameters. The range of compensating adjustments that can be made is limited because each parameter has a unique spectral signature. For gas abundances, this approach is reasonable since gases absorb at discrete frequencies. For continuum absorbers, such as clouds, this approach is less satisfactory. For example, the optical depth of the NH₄SH cloud is $0.02^{+0.06}_{-0.02}$ for our category 8 ensemble. However, the shape of the synthetic spectrum calculated with an NH₄SH cloud optical depth of 0.08 is quite different from that of the average spectrum, although based on our criterion that the difference between the synthetic and observed spectra be less than the standard deviation, the synthetic spectrum is an acceptable fit. Thus, in some sense the uncertainties associated with the retrieved cloud optical depths are very conservative.

It is also important to remember that the uncertainties in the cloud optical depths are the uncertainties associated with our best fit to the average spectrum and not representative of the variability within the ensemble. Moreover, given the relatively large field of view (FOV) of the IRIS measurements (>0.8° latitude) and the fact that with increasing spatial resolution (decreasing FOV) that range of the observed brightness temperatures continues to increase, it is likely that cloud opacity is more variable than that suggested by the uncertainties in our retrieved optical depths. Finally, while a single cloud-type extends through multiple model layers, clouds in our analysis are single-layered. Given the subsaturated relative humidity profiles for NH_3 and H_2O , it is possible that the clouds are multilayered (i.e., more than one cloud layer per condensate) or that the clouds are horizontally inhomogeneous within the IRIS field of view. Nevertheless, the values in Table 4 show that we can distinguish between the clouds in our ensembles. The uncertainties associated with the optical depths of the NH₃ and NH₄SH clouds, clearly preclude category 7 results from providing an acceptable fit to the category 8 spectrum.

In order to simplify the presentation, uncertainties associated with our retrieved values are only presented for our category 8 results. The uncertainties associated with the cloud optical depths are representative of the uncertainties associated with the cloud opacities in the other ensembles. Thus the uncertainty associated with the optical depth of the NH₃ cloud is of order 10% in all ensembles. For the NH₄SH cloud, the low 5- μ m radiances combined with the relatively large standard deviation of the category 1 spectra results in a ±2.0 uncertainty in the optical depth of the category 1 NH₄SH cloud. In addition, the increased optical depth of the NH₄SH cloud reduces our sensitivity to water cloud levels in category 1 regions. Thus, for the warmer ensembles, the uncertainty associated with the water cloud optical depth is ±25%, though the uncertainty associated with the optical depth of the category 1 water cloud is larger. The uncertainties associated with the NH₃, PH₃, and parahydrogen profile parameters apply to all of the ensembles. Finally, we emphasize that it is the range of variability found within the NEB that is the issue.

Based on the results presented in Table 4, we see that significant spatial variability exists within the NEB. We find that most of the observed brightness temperature variability can be explained in terms of local variations in cloud opacity. Small temperature variations, of order 2°C, are also present within the NEB.

Going from category 1 to category 3, we see from the values in Table 4 that the NH₄SH and H₂O cloud optical depths decrease with increasing 5- μ m brightness temperatures. In addition, we see that there is a tendency for the NH₃ cloud optical depth to change following the NH₄SH and H₂O optical depths even though the 45- μ m brightness temperature criterion is unchanged. Moreover, we see that in the hotter 45- μ m brightness temperature category the optical depths of the NH₃ cloud are smaller. All optical depths are referenced at 0.5 μ m. Thus the results presented in Table 4 strongly suggest that a vertical correlation of cloud opacity exists within the Jovian atmosphere.

We find that the correlation between NH₃ and NH₄SH cloud opacity, previously noted by *Bézard et al.* [1983*a*] and *Gierasch et al.* [1986], extends also to the water cloud. While this correlation is most apparent in our hotter spectral ensembles (categories 3, 7, and 8), it most likely extends to our colder spectral ensembles (categories 1, 2, and 6) as well. However, the increased opacity of the NH₃ and NH₄SH clouds results in a loss of sensitivity to the water cloud level. Additional evidence for a vertical correlation of cloud opacity is the absence of any spectra that are simultaneously "hot" at 45 μ m (226 cm⁻¹) and "cold" at 5 μ m (average over 1950–2150 cm⁻¹) corresponding to category 5 and only one spectrum for which the converse is true (category 4).

Also correlated with the spatial variations in cloud opacity is the retrieved relative humidity profile for water. This correlation strengthens our conclusion that dynamic depletion of water vapor above the condensation level is the most plausible explanation for the low relative humidities found in the hotter spectral ensembles (categories 3, 7, and 8). Further bolstering this conclusion is the correlation between relative humidity and the inferred vertical extent of the water cloud. A saturated water profile provides the best fit to our colder spectral ensembles (categories 1, 2, and 4). In these regions, water cloud opacity is distributed with particle-togas scale height ratios in excess of 0.15. The increased vertical extent is required to fit the continuum region near 2130 and 2170 cm^{-1} . Conversely, hot spots require the lowest relative humidities and correspond to the driest regions on Jupiter; moreover, cloud vertical extent is less.

Intermediate spectral ensembles (categories 3 and 6) are the most interesting in that water vapor and NH_4SH cloud opacity provide the bulk of the 5- μ m continuum opacity. Even though these spectral ensembles have the same $5-\mu m$ brightness temperature selection criterion, the same atmospheric model cannot fit both regions. The category 3 spectral ensemble requires more NH₃, NH₄SH, and H₂O cloud opacity, while the relative humidity profile is similar to that in category 6 regions.

Dynamics appears to play an important role in determining the cloud and humidity structure of the Jovian atmosphere, as would be expected of condensate clouds [Rossow, 1978]. The difference between the "hot" and "cold" $45-\mu m$ spectral ensembles in most likely a response to the local variations associated with wave motions. If we identify the "updraft" or enhanced mixing phase of the wave (relative to the mean for the NEB) with increased relative humidities of both NH_3 and H_2O , larger cloud optical depths, larger PH_3 abundances above the 1-bar level, increased para fractions, and lower temperatures and if we identify the "downdraft" or suppressed mixing phase of the wave with reduced relative humidities, decreased cloud optical depths, decreased PH₃ abundances above the 1-bar level, decreased para fractions, and higher temperatures, it then follows that the NEB hot spots (category 8) can be interpreted as regions in which the wave downdraft adds to the mean downdraft. Consistent with that notion, the NEB hot spots correspond to the clearest and driest regions in the Jovian atmosphere. Some cloud opacity is present, but the retrieved optical depths of all three clouds are smaller than in other regions. While we find that NH₃ vapor is subsaturated within the NH₃ cloud in all regions, it is most subsaturated in the hot spots. Similarly, we find the PH₃ profile above 1 bar to be depleted. In addition, hot spots are also the driest regions in terms of the relative humidity of water where the minimum water relative humidity of $\approx 15\%$ occurs near 3 bars.

By increasing the optical depth of all three clouds, increasing the relative humidity of water, and allowing more PH₃ above the 1-bar level, we can reproduce the other spectral ensembles that are "hot" at 45 μ m from the NEB hot spot structure. This suggests that while the overall sense of the NEB dynamics is "downwelling", the strength of the downwelling motions is diminished in these "cooler" regions. Also suggestive of this is the relative humidity profile retrieved from the category 7 spectra. While the relative humidity is still subsaturated, the depletion is shifted to lower pressures, where a minimum relative humidity of 40% is found at 2 bars; thus the deeper portions of the atmosphere (2–4 bars) are wetter.

This progression continues for the "cold" $45-\mu m$ spectral ensembles, where it is also necessary to increase the vertical extents of the clouds. We interpret these regions as regions in which a wave updraft subtracts from the strength of the mean downwelling, and in extreme cases (categories 1 and 2), creates a weak updraft. The updraft is, however, less intense than the upwelling motions found in zones (B. E. Carlson et al., submitted manuscript, 1992). The category 3 spectral ensemble clearly corresponds to weak downwelling motions with the relative humidity profile of water similar to that found in the category 7 regions. Nevertheless, the cloud opacity is increased along with the vertical extent of the water cloud. In addition, structure of the NH₃ cloud is changed. In the "cold" 45- μ m spectral ensembles, we find that the large particle mode is more concentrated near the NH3 cloud base (i.e., has a smaller-scale height distribution) similar to that found in zones (B. E. Carlson et al., submitted

manuscript, 1992). Coupled with this change in the NH_3 cloud is an increase in the cloud top parahydrogen fraction. Thus we find that the "cold" $45-\mu m$ spectral ensembles are more zonelike than their hot counterparts as evidenced by the increased cloud opacity, increased vertical extent of the clouds, increased cloud top para fraction, and increased amount of PH_3 aloft, all of which are suggestive of upwelling motions.

There are indications that the actual cloud structure is more complex than that presented here. For example, the retrieved relative humidity profile for water in hot spots is found to start decreasing immediately above the cloud base, suggesting either the existence of multilayered water cloud structure (multiple saturated and subsaturated layers) or of horizontally inhomogeneous (broken) water clouds on spatial scales less than ≈ 900 km. Further suggestions of cloud brokenness are found in the field-of-view dependence of the IRIS observations. With decreasing field of view, the observed brightness temperature extremes are found to increase, indicating that further spatial inhomogeneity exists at spatial scales not resolved by the IRIS observations.

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