

FINAL REPORT
and
SUMMARY OF RESEARCH

ENTITLED

**LABORATORY EVALUATION AND APPLICATION OF MICROWAVE
ABSORPTION PROPERTIES UNDER SIMULATED CONDITIONS FOR
PLANETARY ATMOSPHERES**

To the

Planetary Atmospheres Program of the
National Aeronautics and Space Administration

For Grant NNG05GQ87G

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I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments, entry probe radio signal absorption measurements, and earth-based or spacecraft-based radio astronomical (emission) observations can be used to infer abundances of microwave absorbing constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or the use of laboratory measurements of such properties taken under environmental conditions that are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. For example, laboratory measurements completed recently by Mohammed and Steffes (2003 and 2004) under the predecessor grant (NAG5-12122, 5/1/02-4/30/05), have shown that the millimeter-wavelength opacities from both gaseous phosphine (PH_3) and gaseous ammonia (NH_3) under simulated conditions for the outer planets vary significantly from that predicted by theory over a wide range of temperatures and pressures. These results have directly impacted the study of Saturn's atmosphere conducted with the Cassini Radio Science Experiment, and the application of our results to the data from the first Cassini radio occultation experiments was recently presented by Mohammed et al. (2005, attached). The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both spacecraft entry probe and orbiter (or flyby) radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements *and* to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

II. PROGRESS REPORT

This project represents the latest stage of an ongoing activity begun in February 1984 to conduct laboratory measurements of the microwave and millimeter-wave properties of simulated planetary atmospheres, in support of NASA missions and ground-based microwave and millimeter-wave observations of planetary atmospheres. The project has also included application of the laboratory results to data from missions such as Voyager, Pioneer-Venus, Magellan, Galileo, and Cassini (see Mohammed et al., 2005, attached) and to earth-based observations (such as those from the NRAO/VLA), as well as direct involvement in mission-based microwave measurements and earth-based measurements.

From February 1984 through December 1996, this activity was supported by NASA Headquarters under Grant NAGW-533. From April 1, 1997 through December 31, 2001 this activity was supported by Grant NAG5-4190 from the NASA Goddard Space Flight Center. From May 1, 2002 through April 30, 2005, the project was supported by grant NAG5-12122 from the NASA Goddard Space Flight Center. A one-year, reduced-cost

“extension” was provided by the grant covered by this report (NNG05GQ87G), allowing us to continue working in this area at a reduced level during the period September 15, 2005 through September 14, 2006. The current technical status of this project is described in Section III below, and was also highlighted in a proposal to the NASA Planetary Atmospheres Program for a recently-selected three-year successor project (NNG06GF34G, September 15, 2006-September 14, 2009).

III. RECENTLY COMPLETED LABORATORY MEASUREMENTS AND APPLICATION STUDIES

A. Millimeter-Wavelength Measurements of Phosphine and Ammonia in the Atmospheres of the Outer Planets

In laboratory work conducted during our predecessor grant (NAG5-12122, 5/1/02-4/30/05), we showed that our new model for opacity from phosphine which was based on our previous centimeter-wavelength measurements (Hoffman *et al.*, 2001) and which indicated that the opacity from PH₃ was greater than indicated by theory, also applied at millimeter wavelengths (7.5 – 9.2 mm, Mohammed and Steffes, 2003, and 3.2 mm, Mohammed and Steffes, 2004). This higher-than-expected opacity was attributed to an underestimation of the strength of the collisionally-induced rotational lines of phosphine, whose intensities had never been measured. Additionally, it was found that most current models used for calculating the 9.2-mm opacity of ammonia (NH₃) in a hydrogen/helium atmosphere understate the actual opacity, which would result in an overestimation of ammonia abundance. These results fundamentally changed the expected capabilities and the approaches to interpreting data from the Cassini three-frequency radio occultation studies at Saturn.

When applied to data from the Cassini three-frequency radio occultation studies of Saturn, our results showed that accurate retrievals of the abundance PH₃ in the upper troposphere of Saturn can be obtained from radio occultation experiments using the new 32 GHz (9.3 mm, or Ka-band) transmitter included in the Cassini radioscience system. Results from our laboratory work showed that while 13-cm and 3.6-cm radio occultation experiments from Voyager were only able to detect phosphine at pressures greater than about 0.8 Bars (see Hoffman *et al.*, 2001), the increased opacity and higher spatial resolution obtainable at 9.3 mm have now made detection of phosphine abundance profiles possible up to altitudes with pressures as low as 500 milliBars, and with vertical resolution on the order of 2 km. (These results were presented at the 2005 AAS/DPS meeting, Mohammed *et al.*, 2005, attached.) This clearly demonstrates the key role laboratory measurements play in mission success.

Under the previous grant, we also completed an extensive measurement campaign at 3.2 mm (94 GHz) of the absorption and refraction of both phosphine and ammonia under simulated conditions for the outer planets. Since our current 94 GHz system is an order of magnitude more sensitive than the system we used a decade ago, we repeated the measurements of the opacity from NH₃ in a H₂/He atmosphere conducted by Joiner and

Steffes (1991), under the same temperature and pressure conditions as for phosphine, so as to more tightly constrain the model for ammonia opacity used in interpreting the 3.2 mm radio emission measurements of the outer planets.

The results of both sets of measurements are significant, in that they show that our model for phosphine opacity which was based on our previous centimeter-wavelength laboratory measurements (Hoffman *et al.*, 2001) is accurate even at short (3.2 mm) millimeter wavelengths. Additionally, the measurements show that at pressures greater than 1 Bar, current models for ammonia opacity significantly understate the opacity from ammonia at the 3.2 mm wavelength, presumably due to an underestimation of the contribution from the rotational lines at and above 572 GHz (19 cm^{-1}). Thus, a model was developed for use at 3.2 mm to account for these effects (Mohammed and Steffes, 2004). However, the effectiveness of this model over a more extended wavelength range (of interest to a number of observers) will require experimental verification. (This work will be conducted by a new graduate student, Kiruthika Devaraj, under our successor grant from the Planetary Atmospheres program, NNG06GF34G, September 15, 2006-September 14, 2009).

B. Centimeter-Wavelength Opacity from Ammonia in the Atmospheres of the Outer Planets

While our laboratory work conducted under the predecessor grant showed that the model we developed for the microwave and millimeter-wave opacity of *phosphine* under conditions for the outer planets is indeed accurate over a wide range of pressures, temperatures, and wavelengths, our work also shows that a single, unified model for the microwave and millimeter-wavelength opacity of ammonia under conditions characteristic of the outer planets remains illusive. Our previous work has shown that a group of models, each valid over a limited range of wavelengths, temperatures, and pressures must be employed in order to accurately interpret radio opacity data from radio occultation or entry probe link experiments or to accurately interpret microwave emission data measured by spacecraft or earth-based radiometers or radio telescopes. For example, in their recent paper on the microwave retrieval of Jupiter's tropospheric ammonia abundance, the UC-Berkeley Group (Gibson *et al.*, 2005) used 3 different models for ammonia opacity (Spilker, 1990; Joiner and Steffes, 1991; and Mohammed and Steffes, 2004) in their analysis of the complete microwave and millimeter-wave spectrum. While the "three model" approach was certainly the most accurate that could be used given the available collection of laboratory measurements, the authors expressed concern that at the one wavelength (1.1 cm) where the most precise measurement of the microwave flux from Jupiter was conducted ($\pm 1.6\%$), the apparent uncertainty in the model exceeded 20%. In their words, "The errors in the absorption formulas will translate directly into errors in the abundances deduced from model fitting." In order to improve the accuracy of the models for ammonia opacity in a H_2/He atmosphere, we have undertaken a new laboratory measurement program in the 1.1-20 cm wavelength range using an upgraded system. (See Hanley and Steffes, 2006, attached.)

Since support was provided by Georgia Tech to upgrade the 1.1-20 cm measurement system described in the attached paper (Hanley and Steffes, 2006), the achievable

sensitivity is now far better than that achieved in any previous work in this wavelength range. As in our previous work, the opacity of the gas mixtures tested are determined by measuring the change of the “Q”, or quality factor, of the resonances under test induced by the presence of a lossy gas mixture. The instrumentation supporting the new system has been significantly upgraded so that uncertainties in the pressure, temperature, and mixing ratio of gas under test are dramatically reduced, as discussed in the attached paper.

As described in the attached paper (Hanley and Steffes, 2006), the upgraded system is functional at 24 resonant frequencies in the 1.1-20 cm wavelength range. Over the past year, the opacity from a custom-manufactured (Airgas) mixture of 85.517% hydrogen (H_2), 13.5% helium (He), and 0.983 ± 0.02 % ammonia (NH_3) has been measured at these 24 different wavelengths with pressures in the 0.5 to 6 Bar pressure range and temperatures in the range from 180 K to ambient temperature (~ 297 K). It should be noted that while ~ 1 % is the abundance of ammonia in the test mixture being loaded into the test system, the actual abundance being measured is substantially lower due to *adsorption* of gaseous ammonia into the metallic walls of the test system. One key problem with all previous centimeter-wavelength measurements of ammonia is the uncertainty in mixing ratio due to *adsorption* of ammonia gas by the test chambers. At the higher pressures of these measurements, it is necessary to use a metallic pressure vessel to contain the simulated deep atmospheres. Ammonia is easily adsorbed onto the metal surfaces during such measurements, which makes knowledge of the measured ammonia concentration difficult, especially at colder temperatures. Our new laboratory system employs an additional, glass-envelope Ka-Band (7-10 mm) Fabry-Perot resonator, kept at room temperature (minimizing adsorption), to enable differential measurements of the gas mixtures both before and after admission to the pressure vessel. With this method, we have been able to constrain the uncertainty in the ammonia mixing ratio of the gas mixture under test to less than 8% of the stated value under most measurement conditions. Additional monitoring of the precise quality factor (or Q) of the resonators in the system has also been used to characterize both the time variability and the spatial variability of adsorption within the system, further reducing uncertainties.

Results of these measurements will be presented at the October 2006 AAS/DPS meeting in Pasadena, CA. While final processing of the data is not yet complete, it appears as if the model of Berge and Gulkis (1976) gives a more accurate prediction of the centimeter-wavelength opacity from NH_3 in an H_2 /He atmosphere in the 1-6 Bar pressure range than do other more recent models (e.g., Spilker 1990).

As mentioned, these results have direct application to entry probe radio link planning and to retrieval of ammonia abundances from entry probe radio signal extinction measurements (e.g. Galileo). If the measurements to be conducted under the successor grant (NNG06GF34G, September 15, 2006-September 14, 2009) at even higher pressures are consistent with the measurements conducted to date, the deep atmospheric abundance of ammonia inferred from the Galileo entry probe radio link measurements (Folkner et al., 1998) will be reduced by over 30 percent.

These measurements also directly impact interpretive studies of radio occultation measurements of the atmosphere of Saturn conducted with the Cassini spacecraft. The laboratory measurements just completed indicate that the sub-cloud abundance of ammonia initially deduced to be ~104 ppm (see Mohammed et al., 2005, attached) is more likely on the order of ~60 ppm. Similarly, conclusions regarding the subsaturation of ammonia in the upper atmosphere of Jupiter (Gibson *et al.*, 2005) may be subject to significant revision.

Finally, these measurements will be of direct use in the planning and modeling of centimeter-wave measurements of the Jovian atmosphere to be conducted by the Juno mission.

IV. STUDENTS SUPPORTED

Over the course of this one-year grant, one Ph.D. student has been supported as a graduate research assistant. (Mr. Thomas J. Hanley). He has been involved in the publication of papers in both refereed journals and at conferences, such as AAS/DPS and the 4th International Planetary Probe Workshop. (See Section V.) As the new successor grant commences (NNG06GF34G), a new graduate student will join our research group to pursue additional work in this area (Ms. Kiruthika Devaraj, BS, 2006).

V. PUBLICATIONS

The following journal publications, and conference presentations were supported (or partially supported) by Grant NNG05GQ87G:

Journal Publications

T.R. Hanley and P.G. Steffes, "Laboratory Measurements of the Microwave Opacity of Hydrochloric Acid Vapor in a Carbon Dioxide Atmosphere," Icarus, vol. 177, pp. 286-290, September 2005.

T.R. Hanley and P.G. Steffes, "An Advanced System for Measurement of the Microwave Properties Ammonia under conditions for Jovian Atmospheres," to be submitted to Radio Science, September 2006.

Conference Presentations with Published Proceedings or Abstracts

A. Kliore, N. Rappaport, A. Anabtawi, S. Asmar, J. Armstrong, E. Barbinis, G. Goltz, D. Johnston, D. Fleischman, D. Rochblatt, J. Anderson, E. Marouf, K. Wong, F. Thomson, F.M. Flasar, P. Schinder, R. French, C. McGhee, P. Mohammed, P. Steffes, A. Nagy, L. Iess, P. Tortora, R. Ambrosini, and E. Flamini, "An Overview of Cassini Radio Science," Bulletin of the American Astronomical Society, vol. 37, no. 3, 2005, p. 626. Presented at the 37th Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society, Cambridge, UK, September 5, 2005.

P.N. Mohammed, P.G. Steffes, A. Kliore, A. Anabtawi, S. Asmar, E. Barbinis, G. Goltz, D. Johnston, and E.A. Marouf, "Vertical Profiles of Phosphine and Ammonia on Saturn Derived from the first Cassini RSS Occultation Observation using Forward Modeling," Bulletin of the American Astronomical Society, vol. 37, no. 3, 2005, p. 657. Presented at the 37th Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society, Cambridge, UK, September 6, 2005 (presentation attached).

T.R. Hanley and P.G. Steffes, "New Laboratory Measurements of the Microwave Absorption of Ammonia under Jovian Conditions," Bulletin of the American Astronomical Society, vol. 37, no. 3, 2005, p. 774. Presented at the 37th Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society, Cambridge, UK, September 9, 2005.

T.R. Hanley and P.G. Steffes, "Method of Characterizing Ammonia Opacity in Jovian Atmospheres with Application to Entry Probe Radio Links," Proceedings of the 4th International Planetary Probe Workshop (IPPW-4), in press. Presented at the 4th International Planetary Probes Workshop, Pasadena, CA, June 28, 2006 (reprint attached).

VI. CONCLUSION

Over the 1-year duration of this grant, an effective program integrating microwave and millimeter-wave laboratory measurements with observations conducted from spacecraft experiments and earth-based radio astronomical observations has been continued. Application of laboratory results achieved during the predecessor grant were applied to results from the Cassini radio science activity to detect phosphine and ammonia in the upper troposphere of Saturn. Moreover a new laboratory measurement system has been developed for accurately characterizing the *centimeter-wavelength* opacity from ammonia under simulated conditions for the outer planets. These results have played a significant role in mission planning. It is expected that similar successes will continue as our new grant from the NASA Planetary Atmospheres Program commences.

VII. REFERENCES

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Gibson, J., W.J. Welch, and I. de Pater 2005. Accurate Jovian radio flux density measurements show ammonia to be subsaturated in the upper atmosphere. *Icarus* **173**, 439-446.

Hanley, T.R. and P.G. Steffes 2005. The Microwave Absorption Properties of Hydrochloric Acid Vapor in a Carbon Dioxide Atmosphere," Icarus, **177**, 286-290.

T.R. Hanley and P.G. Steffes 2006. Method of Characterizing Ammonia Opacity in Jovian Atmospheres with Application to Entry Probe Radio Links, Proceedings of the 4th International Planetary Probe Workshop (IPPW-4), in press. Presented at the 4th International Planetary Probes Workshop, Pasadena, CA, June 28, 2006 (reprint attached).

Hoffman, J.P., P.G. Steffes, and D.R. DeBoer 2001. Laboratory measurements of the microwave opacity of phosphine: Opacity formalism and application to the atmospheres of the outer planets. *Icarus* **152**, 172-184.

Joiner, J. and P.G. Steffes 1991. Modeling of Jupiter's millimeter wave emission utilizing laboratory measurements of ammonia (NH₃) opacity. *Journal of Geophysical Research (Planets)* **96**, 17,463-17,470.

Mohammed, P.N. and P.G. Steffes 2003. Laboratory Measurements of the Ka-band (7.5mm to 9.2mm) Opacity of Phosphine (PH₃) and Ammonia (NH₃) under Simulated Conditions for the Cassini-Saturn Encounter." *Icarus*, vol. 166, pp. 425-435.

Mohammed and P.G. Steffes 2004. Laboratory Measurements of the W-band (3.2 mm) Properties of Phosphine (PH₃) and Ammonia (NH₃) under Simulated Conditions for the Outer Planets. *Journal of Geophysical Research*, vol 109, no. E07S13, pp 1-9.

Mohammed P.N., P.G. Steffes, A. Kliore, A. Anabtawi, S. Asmar, E. Barbinis, G. Goltz, D. Johnston, and E.A. Marouf 2005. Vertical Profiles of Phosphine and Ammonia on Saturn Derived from the first Cassini RSS Occultation Observation using Forward Modeling, *Bulletin of the American Astronomical Society*, vol. 37, no. 3, p. 657. Presented at the 37th Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society, Cambridge, UK, September 6, 2005 (presentation attached).

Spilker, T. R. 1990. *Laboratory Measurements of the Microwave Absorptivity and Refractivity Spectra of Gas Mixtures Applicable to Giant Planet Atmospheres*, PhD thesis, Stanford University, CA.

Vertical Profiles of Phosphine and Ammonia on Saturn Derived from the First Cassini RSS Occultation Observation Using Forward Modeling

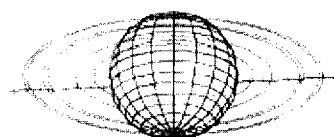
P.N. Mohammed, P.G. Steffes (Georgia Institute of Technology)

A.J. Kliore, A. Anabtawi, S.W. Asmar, E. Barbinis, G. Goltz, D. Johnston (Caltech-JPL)

E.A. Marouf, (San Jose State Univ)

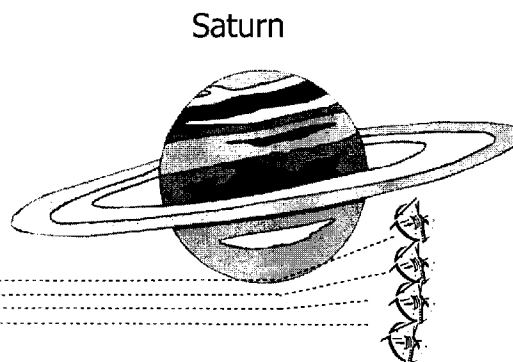
PNM and PGS acknowledge the partial support for this work received from the NASA Planetary Atmospheres Program under grant NAG5-12122

Radio Occultation Experiment



Rev 7 - May 3, 2005

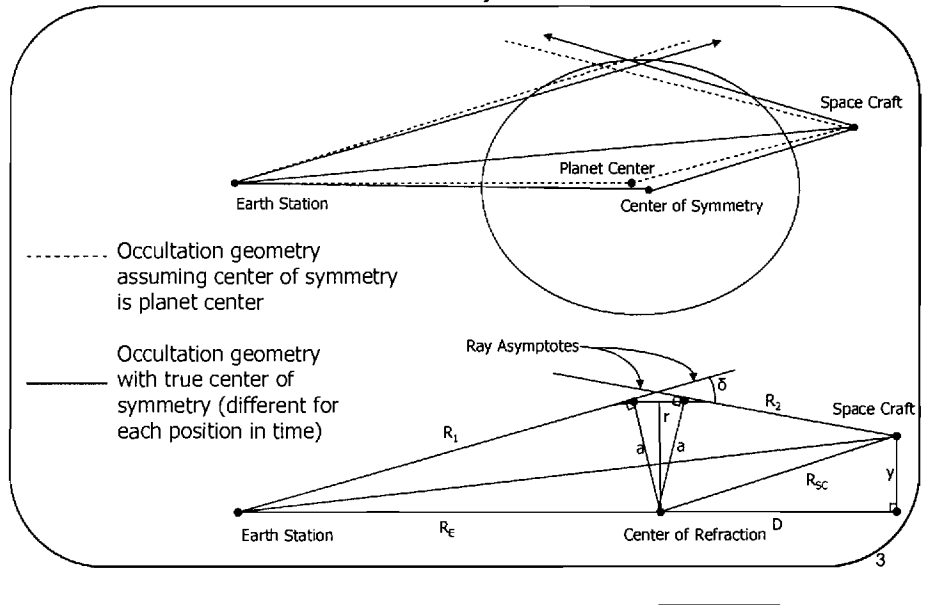
Latitude = 4.9 deg S



Earth

Cassini

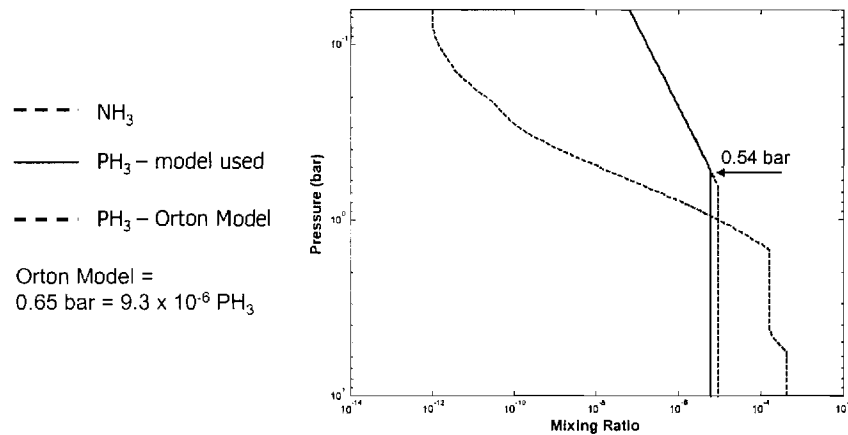
Occultation Geometry for Oblate Planet



Radio Occultation Simulator

- Deep abundances assumed for Thermochemical model
 - $\text{H}_2\text{S} = 308 \times 10^{-6}$
 - $\text{NH}_3 = 412.5 \times 10^{-6}$
 - $\text{H}_2\text{O} = 1 \times 10^{-6}$
 - $\text{CH}_4 = 4.2 \times 10^{-3}$
 - $\text{PH}_3 = 6.2 \times 10^{-6}$
 - $\text{He} = 0.07$
 - Rest = H_2
- PH_3 does not condense in the atmosphere of Saturn
- It is thought to decrease due to photolysis
 - $0.54 \text{ bar} = 6.2 \times 10^{-6} \text{ PH}_3$
 - $0.15 \text{ bar} = 4.1 \times 10^{-7} \text{ PH}_3$
 - Decreases linearly in $\log(P)$ versus $\log(\text{mixing ratio})$ (Orton *et al.* 2000)

Mixing Ratio Profile of PH₃ and NH₃



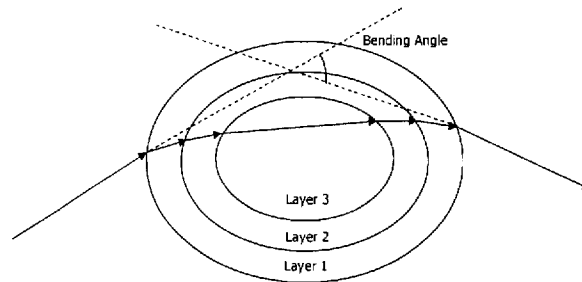
5

Refractivity

- The refractivity of hydrogen, helium and methane were the only constituents considered to cause ray bending in the atmosphere less than 2 bars
 - $N'H_2 = 5.06 \times 10^{-18}$ N-units cm³ molecule⁻¹
 - $N'He = 1.30 \times 10^{-18}$ N-units cm³ molecule⁻¹
 - $N'CH_4 = 1.67 \times 10^{-17}$ N-units cm³ molecule⁻¹
- These were converted to refractive index and used in the ray tracing technique

6

Ray Tracing



The atmosphere is modeled as ellipsoidal shells

1 bar isobaric surface

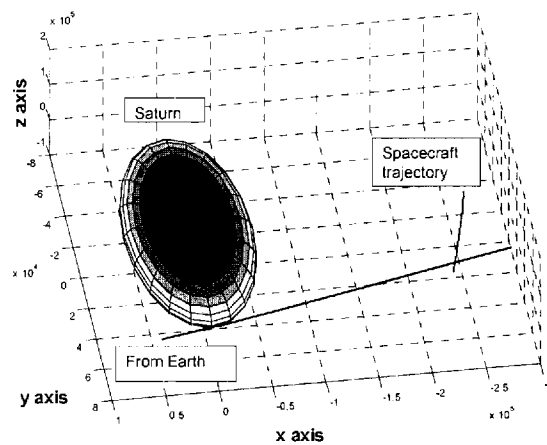
Equatorial Radius = $60\,268 \pm 4$ km

Polar Radius = $54\,364 \text{ km} \pm 10$ km

$$\text{Oblateness} = (R_{\text{eq}} - R_{\text{p}})/R_{\text{eq}} \\ = 0.09796 \pm 0.00018$$

7

Ray Tracing



8

Attenuation Profiles

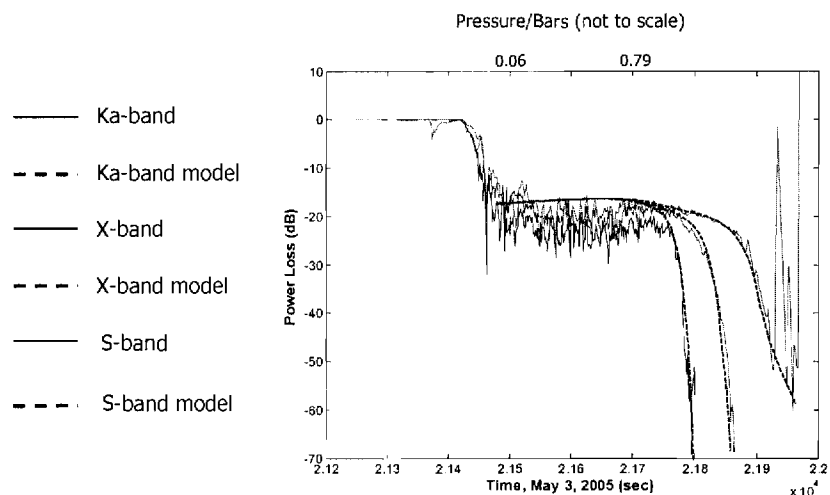
- Attenuation considered
 - Phosphine, using Hoffman *et al.* (2001) model for all bands
 - Ammonia, using Mohammed-Steffes (2003) model for all bands
 - Refractive Defocusing

Refractive defocusing
computed using

$$p_n = 10 \log_{10} \left[\frac{\gamma}{a} \left(1 - R_2 \frac{d\delta}{da} \right) \right]$$

9

Tri-band Power Attenuation



10

Conclusion

- Depths penetrated by
 - Ka-band – 0.85 to 0.88 bar
 - X-band – 1.17 bar
 - S-band – 1.89 bar
- Results are consistent with a deep PH_3 abundance of 10x solar and an NH_3 abundance of 104 ppm below the base of the ammonia cloud, corresponding to abundances of 2.2x solar NH_3 and 10x solar H_2S in the deep atmosphere

11

METHOD OF CHARACTERIZING AMMONIA OPACITY IN JOVIAN ATMOSPHERES WITH APPLICATION TO ENTRY PROBE RADIO LINKS

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ABSTRACT

The link budgets used when designing the communication system on a probe rely heavily on attenuation of the medium through which the radio signals travel. This is especially important in the denser atmospheres of the Jovian planets. Knowledge of the temperature, pressure and concentrations of the various atmospheric constituents all factor into determining the attenuation of a radio signal through a specified path length assuming a thorough understanding of each constituent's microwave absorbing properties. Ammonia is one of the most common and strongest microwave absorbing constituents found in the atmospheres of the Jovian planets, yet discrepancies in its current opacity models require that new experimental data be taken to better constrain its true properties.

1. INTRODUCTION

One of the major surprises of the Galileo Probe Mission to Jupiter was the determination of an ammonia abundance in the deep atmosphere which was 4 times the solar abundance in the 7-16 Bar pressure range. This abundance was retrieved from the signal attenuation measurements on the 1.387 GHz radio link from the entry probe to the Galileo Orbiter during the probe descent [1]. This result was problematic for atmospheric modelers, since earth-based observations of radio emission from the Jovian atmosphere indicated a significantly lower, disk-averaged, deep atmospheric abundance of ammonia, and the ammonia abundance retrieved from the Galileo Entry Probe Net Flux Radiometer (NFR) never exceeded 1.5x solar down to the 4 Bar pressure level with a maximum of 3x solar deeper in the atmosphere [2]. While this disparity could possibly be explained by a localized instability creating the elevated ammonia abundance detected by the entry probe radio link, [2] suggested that it may also be due to

inaccuracies in the models for constituent radio opacities used by [1].

The models used by [1] for the opacity of ammonia in the Jovian atmosphere were from [3] and from [4]. While the opacity from ammonia predicted by the two models is consistent (within 5%) when calculated under the deep atmospheric conditions encountered by the Galileo Probe, both models are based on laboratory measurements which were made at significantly higher frequencies and at significantly lower pressures than those for the Galileo Probe radio link. An improved microwave measurement system has been developed at Georgia Tech (similar to that used by [5]) to measure ammonia opacity in a hydrogen and helium atmosphere at wavelengths from 1.1 to 20 cm, pressures from 0.5 to 16 bars and temperatures from 185 to 550 K.

One key problem with all previous measurements of ammonia is the uncertainty in mixing ratio due to adsorption of ammonia gas by the test chambers. At the higher pressures of these measurements, it is necessary to use a metallic pressure vessel to simulate the deeper atmosphere. Ammonia is easily adsorbed onto the metal surface during measurements, which makes knowledge of the measured ammonia concentration difficult, especially at colder temperatures. Our new laboratory system uses a glass Ka-Band Fabry-Perot resonator kept at room temperature (minimizing adsorption) enabling differential measurements of the gas mixtures both before and after admission to the pressure vessel. With this method we have been able to constrain the uncertainty in the ammonia concentration to a maximum of 10% of the stated value and generally less than 5% of the stated value.

This new measurement system will enable development of a more accurate model of ammonia opacity at microwave frequencies that can be used in the design of communications links between planetary probes and orbiters as well as improving the scientific return from Galileo Entry Probe data and future missions.

2. MEASUREMENT THEORY

The method used to measure the microwave absorptivity of a gas is based on the lessening of the quality factor (Q) of a resonance in the presence of a lossy gas. The technique of monitoring the changes of Q of different resonances of a cavity resonator in order to determine the refractive index and the absorption coefficient of an introduced gas or gas mixture (at those resonant frequencies) has been used for over one half of a century. (See, e.g., [6]-[9])

The Q of a resonance is a unitless quantity defined in Eq. 1 by [10] as

$$Q = \frac{2\pi f_0 \times \text{Energy Stored}}{\text{Average Power Loss}} \quad (1)$$

where f_0 is the frequency of the resonance and can be measured directly as the frequency divided by its half-power bandwidth (f_0/BW). The opacity or absorptivity (α) of a gas is related to the Q of that gas by

$$\alpha = \frac{\varepsilon''\pi}{\varepsilon'\lambda} = \frac{1}{Q_{\text{gas}}} \frac{\pi}{\lambda} \text{ (Nepers/km)} \quad (2)$$

where ε' and ε'' represent the real and imaginary permittivity of the gas and λ is the wavelength in km [10]. The quality factor of a resonator loaded with a test gas can be represented by

$$\frac{1}{Q_{\text{loaded}}^m} = \frac{1}{Q_{\text{gas}}} + \frac{1}{Q_{\text{vac}}} + \frac{1}{Q_{\text{ext1}}} + \frac{1}{Q_{\text{ext2}}} \quad (3)$$

where Q_{loaded}^m is the measured quality factor of the gas filled resonator, Q_{gas} is the quality factor of the gas itself, Q_{vac} is the quality factor of the evacuated cavity resonator, and Q_{ext1} and Q_{ext2} represent the external coupling losses from the antenna probes in the resonator [10]. Since only symmetric resonators are used, we can assume $Q_{\text{ext1}} = Q_{\text{ext2}}$. The Q_{ext} value can be calculated by measuring the transmissivity of the system, $t = 10^{-S/10}$ where S is the insertion loss of the resonator in decibels, and using the relations [10]

$$t = \left[2 \frac{Q^m}{Q_{\text{ext}}} \right]^2 \quad (4)$$

$$Q_{\text{ext}} = \frac{2Q^m}{\sqrt{t}} \quad (5)$$

where Q^m represents a measured quality factor. Substitution of Eq. 5 into Eq. 3 yields

$$\frac{1}{Q_{\text{gas}}} = \frac{1 - \sqrt{t_{\text{loaded}}}}{Q_{\text{loaded}}^m} - \frac{1 - \sqrt{t_{\text{vac}}}}{Q_{\text{vac}}^m} \quad (6)$$

with t_{loaded} and t_{vac} representing the transmissivities of the loaded and vacuum measurements respectively. Calculating Q_{gas} in this manner is slightly flawed, however, as this formula does not account for changes

in the center frequency of a resonance when a gas is present. If measurements are conducted under relatively benign conditions, (e.g., 270 K - 400 K), it is possible to construct a tunable resonator which can be retuned to the original resonant frequency when the test gas is present. (See, e.g., [7] or [8]) However, under conditions of extreme temperature and pressure it is very hard to construct reliable tunable resonators. If "fixed-tuned" resonators are used, the frequency shift, which accompanies the introduction of the gas under test, changes the coupling of the resonator and consequently the quality factor, even in the absence of opacity. Known as dielectric loading (described in more detail in [3] and [11]), this effect requires an additional measurement to be made with an essentially lossless gas that causes the same frequency shift as that from the test gas. Using this measurement in place of that made under vacuum conditions in Eq. 6, substituting into Eq. 2, and converting from Nepers/km to dB/km ($1 \text{ Np/km} = 2 \text{ optical depths/km} = 2 \times 10 \log_{10} e \approx 8.686$) dB/km gives the final formula for calculating absorptivity

$$\alpha = 8.686 \frac{\pi}{\lambda} \left(\frac{1 - \sqrt{t_{\text{loaded}}}}{Q_{\text{loaded}}^m} - \frac{1 - \sqrt{t_{\text{matched}}}}{Q_{\text{matched}}^m} \right) \frac{\text{dB}}{\text{km}} \quad (7)$$

[12].

Measuring the refractivity of a gas is more direct than determining its opacity. Refractivity (N) is calculated as:

$$N = 10^6 \frac{(f_{\text{vac}} - f_{\text{gas}})}{f_{\text{gas}}} \quad (8)$$

where f_{vac} and f_{gas} represent the center frequencies of a resonance measured with the system under vacuum and filled with the test gas mixture respectively [13]. This represents the refractivity of the entire gas mixture, which is the sum of the constituents' refractivities weighted by their percent concentrations. Refractivity is dependent on pressure and temperature and is often presented in a normalized form to exclude these dependencies. Normalized refractivity is calculated as:

$$N' = \frac{NRT}{P} \left(\frac{N - \text{units} \times \text{cm}^3}{\text{molecule}} \right) \quad (9)$$

where T is the temperature in Kelvin, P is the pressure in atmospheres and $R = 1.362344 \times 10^{-22} \text{ (atm} \cdot \text{cm}^3 \text{)/(molecule} \cdot \text{K)}$.

3. SYSTEM DESCRIPTION

The current system is composed of three major parts described herein:

3.1 Planetary Atmospheric Simulator

The planetary atmospheric simulator controls and monitors temperature and pressure along with handling the gases used. The main component of the atmospheric simulator is a pressure vessel capable of handling pressures from vacuum to 8 atm with a volume of approximately 31 liters. The vessel is cylindrical and made of stainless steel with a detachable top plate sealed by a Viton O-ring and vacuum grease. Gases are fed into the vessel through a series of regulators and valves. The pressure vessel itself is contained in a temperature chamber. For temperatures between 173 K and 218 K, the temperature chamber is a Revco ultra-low temperature freezer. At warmer temperatures between room temperature and 550 K, the vessel is placed in a digitally-controlled electric oven.

The temperature inside the pressure vessel is monitored by an Omega Model HH21 Microprocessor Thermometer connected to a T-type thermocouple probe inserted into a sealed and capped inverted 3/8" outer diameter pipe protruding into the vessel, but not in direct contact with the gases inside. The thermometer has a resolution of a tenth of a degree Celsius and an accuracy of $0.3\% \pm 0.6^\circ \text{C}$ at temperatures below 0°C and $0.1\% \pm 0.6^\circ \text{C}$ at temperatures above 0°C . The thermocouple itself has an accuracy of either 0.75% or 1°C , whichever quantity is greater. Positive pressures in the system are measured by an Omega DPG7000 Digital Test Gauge with a resolution of 1 mbar and accuracy of 10 mbar, capable of measuring pressures up to 300 psig, whereas pressures below one atmosphere are measured by a Hastings Model 760 vacuum gauge with a resolution of 1 torr and an accuracy of 1 mbar. A Precision Scientific vacuum pump Model 150 is used to evacuate the gases from the system from 1 bar down to a level of 0.1 mbar. Gases at higher pressures are ventilated through an exhaust valve. The evacuated gas can be sent through a chemical scrubber to remove toxicity as in previous uses with ammonia and phosphine. A combustible gas detector (GasTech model GP-204) can also be used to detect leaks from the pressure vessel when the system contains hydrogen. A glass tube tee capable of withstanding 3 atm of pressure is also connected to the gas handling system for mixture sampling and testing, but is maintained at room temperature.

A PortaSens II gas leak detector is used in the system to measure trace amounts of contaminants that may be left in the system due to lingering adsorption effects. It is attached to a long glass tube chamber that is used for mixing the test gases with air, because the electrochemical sensor module in the PortaSens II requires a minimum of 5% oxygen to operate. The PortaSens II is connected in a feedback configuration with the input sampling from a plastic tube extending to the bottom of the glass chamber and the output into the top of the chamber to maximize the accuracy of its measurement on a mixture of a limited volume.

3.2 Microwave Measurement Subsystem

At the heart of the microwave measurement subsystem are two stainless-steel cylindrical cavity resonators positioned inside the pressure vessel. These resonators have been plated with gold, so as to prevent reactions with corrosive acid vapors that have been measured previously. The larger of the two resonators measures 13.1 cm in radius and stands 25.3 cm tall, thus making it ideal for measurements from 1.5 to 10 GHz. It rests at the bottom of the pressure vessel, whereas the smaller resonator, measuring 2.5 cm in radius and 4.9 cm high, rests on a shelf suspended from the top of the pressure vessel. The small resonator is best used in measurements from 10 to 28 GHz. Each resonator contains two closed-loop antenna probes mounted on their top plates and oriented to maximize the Q or quality factor of TE_{0mn} modes. Both resonators are connected to hermetically-sealed bulkhead feed-throughs on the top plate of the pressure vessel. Each resonator has two horizontal slits on their circular sides near their top plates that act to suppress unwanted TM resonant modes, as well as allowing gases to enter them. The two ports for each resonator are symmetric.

The feed-through ports on the pressure vessel connected internally to the large resonator are also connected via low-loss flexible coaxial cables to a 2-port Agilent E5071B Vector Network Analyzer that operates from 300 kHz to 8.5 GHz. The feed-throughs corresponding to the small resonator have one port connected to an input from an HP 83650B Swept Signal Generator and the other to a high-resolution HP 8564E Spectrum Analyzer both via semi-rigid RG-405 coaxial cable. A ferrite isolator is placed between the signal generator and the small resonator to provide a minimum of 10 dB of isolation for measurements up to 18.5 GHz. At higher frequencies, the cables themselves provide enough isolation from reflected signals due to their attenuation. The spectrum analyzer is set in detector positive peak mode, which displays the maximum power level received during the integration time of each

point. The maximum resolution on the spectrum analyzer is 601 points in both the frequency and amplitude axes whereas the network analyzer offers higher precision up to 1601 points.

The signal generator and spectrum analyzer can also be connected to a Fabry-Perot resonator contained in the glass tube tee that operates at Ka-band from 28 to 40 GHz. The Fabry-Perot resonator is the same used by [14] and consists of two gold-plated mirrors. One mirror is flat and contains two symmetric WR-28 waveguide ports and the other mirror is concave, allowing reflection of signals from one port to the other through path lengths up to 10 meters. The waveguides are connected to waveguide-to-coax SMA adapters, which are connected to high-frequency flexible coaxial cables.

Ideally, a network analyzer that could measure up to 40 GHz would replace the signal generator and spectrum analyzer, but at this time it is too cost prohibitive. This would allow greater accuracy along with shorter measurement times.

3.3 Data Handling Subsystem

The data acquisition subsystem consists of a laptop computer connected to the spectrum analyzer, network analyzer, and swept signal generator via a general purpose interface bus (GPIB) connected to a National Instruments NI-488.2 interface card. The suite of instruments is controlled via Matlab and the Standard Commands for Programmable Instruments (SCPI) language. The primary function of the software is to control the instruments and retrieve resonance data from either the spectrum analyzer or network analyzer in the form of received power as a function of frequency. Each resonance is viewed with the amplitude axis extending 10 dBm and with the frequency axis being approximately twice the half-power bandwidth. This "zooming-in" on each resonance allows the best resolution without spreading the resonance over multiple screen widths. The resolution bandwidth (RB) of the spectrum analyzer is set to the value closest to $1/100^{\text{th}}$ of frequency span of a particular resonance under the resonance's broadest condition during the experiment. The value of RB is limited to 1 or 3 times any integer power of ten within the specifications of the device and is kept constant for all measurements of a specific resonance. The software used is similar to the PCSA program created by [15], but with the added flexibility of Matlab and the ability to process the incoming data to directly calculate absorptivity and refractivity.

One problem that arises with the measurements from the spectrum analyzer comes from sweep-on-scan

nulls. Due to difficulty in synchronizing the swept signal generator to the spectrum analyzer, the signal generator is set to sweep at a fast pace while the spectrum analyzer is set at a slower rate. The sweep rate for the signal generator is set at 75 ms at which the device can put out a stable power level at all frequencies used. It can be made to sweep as fast as 10ms, but the signal is noisier. At 75 ms per sweep, in order for the spectrum analyzer to always detect the signal, there must be at least 75 ms during each of the 601 frequency measurement bins. The actual time between sweeps is greater than this, however, as the signal generator is not capable of operating at 100% duty cycle, because it takes a finite amount of processing time to begin each sweep. To detect this signal in each frequency bin it would take close to two minutes per sweep! For each resonance measurement it is beneficial to take multiple sweeps to decrease variance, so it is optimal to have shorter, more frequent sweeps and to use the computer to interpolate the value of the null points that are generated when the swept signal is not present during the measurement interval. If we limit the amount of time for each measurement to 200 seconds, the standard deviation of the set of measurements weighted by the statistical confidence coefficient is minimized for 5 sweeps of 40 seconds each. The data from each set of sweeps is saved in a unique file.

To compensate for this apparent loss of resolution, an algorithm is run to "stuff" the data. The algorithm sets the value of each point equal to the average of the ten nearest points (five higher and five lower) if it is less than that average. For the points within five of either end, the average is done on fewer than ten points to avoid the curve being leveled at the edges. This is done until an iteration is reached where the change at every point is less than 0.02%. This accounts for raising all the original null points but does not account for noisy spikes that stand above the data. To compensate for these, the same algorithm is used, except that it only changes points that are greater than the average of the ten nearest points until the change is less than 10%. This method allows for a smoother curve than doing a simple linear interpolation between the high points.

Once the sweeps have been stuffed, the software calculates the center frequency, half-power bandwidth, power level at the peak, asymmetry figure of merit, and Q of each sweep and averages those values. The standard deviation of each measurement becomes its 1σ uncertainty. The half power or 3 dB bandwidth is calculated by interpolating between the data points to give a frequency value that corresponds to exactly 3 dB below the peak power both higher and

lower than the peak frequency. The asymmetry figure of merit is calculated as in [15].

$$af = 100 \times \frac{(f_r - f_c) - (f_c - f_l)}{f_r - f_l} \% \quad (10)$$

The asymmetry figure of merit is used to determine if any resonances are yielding questionable results. Resonances with asymmetry values greater than 15% generally suggest overlapping of nearby resonances and would likely yield inaccurate results. It is also important to make sure the asymmetry values of both the loaded and matched measurements for a particular resonance data point are consistent within $\pm 2\%$, or this may suggest that there is preferential broadening of one side of the resonance over the other. Resonances that do not meet these criteria are not used.

The measurements taken with the network analyzer are much quicker and require less processing. The network analyzer is capable of generating 30 null-free data sweeps each of both transmissive scattering parameters (S12 and S21) along with one sweep each of the reflective scattering parameters (S11 and S22) in one minute at 1601-point resolution with an IF bandwidth of 10 kHz. The primary data used is from S21, but the others are taken to ensure consistency and enable better examination of each resonance. These 62 sweeps are saved to the laptop and then undergo the same processing to calculate the average and standard deviation of the center frequency, half-power bandwidth, power level, asymmetry and Q.

4. COMPLEXITIES WHEN MEASURING AMMONIA

One complication that arises when measuring ammonia or other highly polarized molecules is its strong tendency to adsorb or stick to the surfaces of the metallic test chamber. This makes knowledge of the ammonia concentration in vapor phase during the measurements very difficult. As the ammonia cools, the molecules have less kinetic energy to escape the attraction to the metallic sides of the pressure vessel and resonators and thus bind themselves to the surfaces due to their large dipole moment. These adsorption effects are seen most strongly at colder temperatures that relate to the upper Jovian atmospheres.

In our measurements, we use a certified premixed cylinder of gas containing hydrogen, helium and approximately 1% ammonia. At 188 K after 2 bars of this mixture have been added to the system, the gaseous ammonia concentration can drop to around 0.25%. To add to the complication, as the ammonia adsorbs in the

vessel and resonators, the concentration of ammonia begins to vary inside the system. This is due to great differences between the surface area to volume ratios of the large resonator, the small resonator, and the rest of the pressure vessel as can be seen in Table 1.

Table1. Surface areas and volumes for the various regions inside the pressure vessel

	Volume (cm ³)	Surface Area (cm ²)	SA / V Ratio
Small resonator	~ 96	~ 116	~ 1.21
Large resonator	~ 13,600	~ 3160	~ 0.23
Remainder of vessel	~ 18,000	~ 9000	~ 0.50

The greater the surface area to volume ratio, the greater the amount of adsorption that will occur, since there will be more sites for the ammonia molecules to bind and less ammonia molecules in a smaller volume. Since the resonators only have two small slits through which to exchange gases, this means that the gases take a long time to thoroughly mix, especially at colder temperatures. This can be charted by monitoring the values of the Q's of resonances in both resonators until those values stabilize. For the coldest ammonia measurements planned (188 K) this stabilization can take upwards of 15 hours.

The Fabry-Perot resonator attached to the measurement system can be used as a means of comparison and verification of the ammonia mixing ratio. Since this resonator is comprised mostly of glass and is maintained at room temperature, the total amount of ammonia adsorbed is less than 1% of the amount added. By measuring the microwave properties of the mixture at Ka-band before the gas is added to the vessel and comparing them to that of the gas mixture that has been in the vessel, the degree of adsorption can be characterized.

5. MEASUREMENT PROCEDURE

Each experiment is designed to measure a gas mixture of a specified concentration at a constant temperature over a range of pressures and at resonances covering a wide range of frequencies. The first step is to make sure the gas handling system is sufficiently leak-proof. Any major leaks will add to the uncertainty of the experiment and in the case of toxic gases present a health hazard. With the system sealed, each resonance is measured under a vacuum of less than 1 torr. For every measurement, the temperatures and pressures inside the pressure vessel before and after each set of sweeps are recorded, averaged, and saved

along with the sweeps. Next, the test gas is slowly added to the pressure vessel until the desired highest pressure to be measured is reached, also providing for enough gas to fill the Fabry-Perot resonator and to allow for some small leaks, which are inevitable at the coldest measurements. The temperature inside the pressure vessel is monitored and the system is given time to thermally equilibrate and to thoroughly mix. The gas mixture is then allowed to enter the Ka-Band Fabry-Perot resonator. Once the mixture has stabilized throughout the system, measurements of each resonance, including six Ka-Band frequencies, are then executed with the test gas in the resonators. Next, the Ka-Band system is evacuated and purged. The gas in the pressure vessel, if at positive pressure, is then vented off or sent through a scrubber until the next desired pressure is reached. Measurements of all test resonances are again similarly carried out at each desired pressure.

A vacuum is once again drawn in the system and argon is added and vacuumed out numerous times to purge the system of any adsorbed ammonia. The argon that is added is periodically drawn off into the gas sensing chamber and mixed with ambient air. This allows for the connected ammonia gas sensor to detect the amount of ammonia that may still be present in the system. This purging process is continued until the ammonia concentration is less than 30 ppm. This minimizes the amount of contamination in the system for the dielectrically matched measurements. A long vacuum is drawn and the resonances measured once more, along with straight-through cable transmissivities under the same conditions on the spectrum and network analyzers as the original measurements. This eliminates the variability in measured power that can occur under slightly different frequency and span conditions. Then, the process of dielectric matching is begun. The frequency that each resonance shifted to while the system was loaded with the test gas is calculated and a loss-less gas (argon) is added until each resonance has shifted to that same frequency. This usually requires a slightly different pressure of gas for each resonance. This is done for every resonance at each measured pressure. Lastly, a final vacuum is drawn and the resonances and transmissivities measured one last time.

6. DATA PROCESSING

The data processing begins after an experiment has been completed. Software was created that loads the experimental data, runs the data stuffing algorithm, and calculates the absorptivity and refractivity of the test gas at the measured frequencies, pressures,

temperature and mixing ratio. Any major variation in cable transmissivity over the frequency range of a resonance is deconvolved from the measured sweeps. The transmissivity of the cables at the frequency of each resonance under each pressure condition is calculated by averaging the values of the average of each set of sweeps from the transmissivity measurements. This value is subtracted from the peak power measured from the test gas and matching gas and the result is the insertion loss (S). The first and last vacuum measurements are compared to the middle one for consistency, but the values of the middle vacuum measurement are the ones used in Eq. 7 because they are temporally closest to the loaded and matched measurements. Then, by using Eqs. 7 and 9, the absorptivity and normalized refractivity are calculated. These values are compared to current models to determine which models work best at each temperature, pressure, concentration, and frequency.

7. CONCLUSION

Planetary probes can provide a wealth of scientific information with in situ measurements that remote sensing options do not offer. In order for these measurements to be of use, the data taken by a probe must be transmitted either to an orbiting spacecraft or directly back to Earth, most often via UHF or microwave radio frequencies. By better constraining the microwave properties of ammonia in Jovian atmospheres, more efficient probe transceivers can be utilized and greater scientific information can be obtained from past as well as future missions.

The High-Sensitivity Microwave Measurement System described has evolved greatly from its first configurations where the measurement sweeps were read on the spectrum analyzer by eye. The sensitivities have vastly improved in response to a demand for more precise radiative transfer models and with more accurate equipment. The measurement procedure has also become more complicated to allow this greater precision, through an improved understanding of the adsorbing and mixing properties of gaseous ammonia.

There are plans to upgrade the pressure vessel in the near future to be able to withstand 16 atm of pressure. In a few more years, a pressure vessel similar to that described in [7] will be built that is capable of maintaining pressures up to 100 atm. After thorough characterization of ammonia is completed, water vapor will be measured to better constrain its microwave absorptive properties.

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