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HIGH-PRECISION LABORATORY MEASUREMENTS SUPPORTING RETRIEVAL OF WATER VAPOR, GASEOUS AMMONIA, AND AQUEOUS AMMONIA CLOUDS WITH THE JUNO MICROWAVE RADIOMETER (MWR)

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Abstract:	The NASA Juno mission includes a six-channel microwave radiometer system (MWR) operating in the 1.3-50 cm wavelength range in order to retrieve abundances of ammonia and water vapor from the microwave signature of Jupiter (see Janssen et al. [2016], this issue). In order to plan observations and accurately interpret data from such observations, over 6000 laboratory measurements of the microwave absorption properties of gaseous ammonia, water vapor, and aqueous ammonia solution have been conducted under simulated Jovian conditions using new laboratory systems capable of high-precision measurement under the extreme conditions of the deep atmosphere of Jupiter (up to 100 Bars pressure and 505 K temperature). This is one of the most extensive laboratory measurement campaigns ever conducted in support of a microwave remote sensing instrument. New, more precise models for the microwave absorption from these constituents have and are being developed from these measurements. Application of these absorption properties to radiative transfer models for the six wavelengths involved will provide a valuable planning tool for observations,

	and will also make possible accurate retrievals of the abundance of these constituents during and after observations are conducted.

RESPONSES TO REVIEWER COMMENTS, SUBMISSION # SPAC-D-16-00001 BY STEFFES ET AL.

Our responses to Reviewer Comments are shown in boldface below each reviewer comment (shown in italics):

RESPONSES TO REVIEWER #1 COMMENTS

Reviewer #1: This work is critical to successfully interpreting the results of measurements of our planets and this publication is important to the community -- particularly right now in the context of Juno. This paper provides invaluable information to the community of the vast array of measurements made in support of this science and is an impressive body of work. I strongly support this work to be published.

My main question with regard to this paper (which may show my ignorance of the goal of a Space Science Reviews paper as well as a somewhat time-fragmented reading of it) is what, if any, new measurements (or formalism updates) are included, or is it a summary paper of where things stand? It would be very helpful if the introduction could provide an overview of the presented material and a table with a 'scorecard' of what is presented (i.e. the different measurement types/systems/configurations with references and comments as to differences from that reference). The head of every section could then provide the overview/references for that constituent and the differences if any. I found myself questioning whether this section was the same system/configuration and measurements, a summary of these, or new measurements or interpretations. I think defining a clear set of systems and configurations will help as well. I think that without this clarification, it is hard to track where things are at. I think with a small amount of additional material (possibility primarily in an Intro table) this can easily provide that guidance so that the full context of these measurements can be understood.

We thank Reviewer 1 for his/her kind comments regarding the value of our work and appreciate the suggestions for improving the paper. The reviewer is correct that this is largely an overview paper with the objective of summarizing the extensive program of microwave laboratory measurements conducted so as to support remote sensing of water vapor, ammonia, and liquid condensates in the jovian atmosphere from the Juno spacecraft. The idea of a "scorecard" at the beginning of the paper is a great idea and has been integrated into the paper as a final paragraph in Section 1. Additionally, we have made more clear the system configurations and have identified how each was used.

Regarding the other minor comments:

Sec. 2 (near end of the paragraph), the '(at these resonant frequencies)' doesn't need the '()'
Corrected as per reviewer's request.

Sec. 2 (just after that), 'refractivity based on frequency' is confusing—maybe 'refractivity based on the shift of the resonant frequency'?
Corrected as per reviewer's request.

Sec. 2.1 (Fig 1), 1a is mostly contained within 1b — I think these could be one figure, maybe by abstracting the gas handling system with 1b?
In the past, we actually attempted to combine Figures 1a and 1b. However, we found that it was too difficult for a reader to keep track of the complex manifold gas system used for the experiments. We do agree that the figure showing the gas handling system (1a) can be improved so as to make more clear the various valve positions. As a result, we have redrawn Figure 1a to highlight the richness of the information contained within that figure. We have also highlighted (in the text) the role that measurements made with the gas handling system (alone) played in establishing a more accurate equation of state for hydrogen, helium, and water vapor under deep Jovian conditions.

What is the SSR policy on English vs metric units? Sec 2.1 in parts shows both, but mostly just metric.
As with most journals, we assumed that SSR employs SI units (metric). The only reason that English units were parenthetically added was that most standard sizes of raw metallic products sold in the US are identified by their ANSI (American National Standards Institute) sizes, which are English units. We felt that providing the ANSI sizes would aid any experimenters wishing to duplicate our system.

Sec. 2.1 (4th para), can drop the 'hydro-tested' or explain it.
Dropped as per reviewer's request.

Sec 3 (1st para), sentences 3 and 4 are repetitive
Corrected as per reviewer's request.

One big question on which it would be good to have some more discussion is the extrapolation of these models, as will inevitably be tried. Are there known issues, to they behave smoothly, are there pressure/temperature bounds etc?
The reviewer makes a very good point. As we state in Section 6 (Future Work), the models for the centimeter-wavelength opacity of ammonia (Devaraj et al., 2014 and Hanley et. al. 2009) are consistent (within 6%) at temperatures up to 500 K, however they diverge by nearly 15 % when extrapolated to 600 K and larger with even higher temperatures. As a result, future work is described which involves measurements of pure ammonia up to 600 K, which will aid in determining accurate extrapolations to higher temperatures. Similarly, as stated in Section 6, the behavior of the Karpowicz et al. 2011 water vapor model becomes non-physical at temperatures above 600 K, since a best-fit "correction term" in the water-vapor self-continuum does not exhibit appropriate physical behavior at high temperatures. As described in Section 6, additional measurements of the opacity of pure water vapor at 600 K will be used to appropriately account for the water vapor self-continuum at high temperature. (Note that the wording in Section 6 has been updated to make this more clear.)

Fig 12, I'm not sure that the picture is needed.
This objective of this paper is to highlight the breadth and magnitude of a large laboratory program supported directly by a NASA mission. Since the Mission Juno papers being

submitted as a group to Space Science Reviews contain a large amount of information regarding mission instruments and hardware, it was felt appropriate to similarly document the supporting hardware for this laboratory program.

I have to confess that at this point my confusion/questions raised above stopped me from reading too deeply for the minutiae as I tried to figure out where things stood. Also, in the figures a number of lines are labelled 'This Work' and I wasn't sure if it pertained to new material for this paper, or was summarizing earlier work from this group (but still post e.g. Joiner).
All references to "this work" have been removed from the figures, and the individual references for each (even though they were conducted as part of this program) are now included.

RESPONSES TO REVIEWER #2 COMMENTS

General Comments:
This paper seems to be more a review paper than a paper reporting new scientific results. The only previously unreported results are the measurements of opacity of H2S in otherwise pure H2, and the text states these were done primarily to verify the utility of extrapolation of previous models. The 12 new measured opacity data are reported only in graphical format; a table of the appropriate results (measured opacity, upper and lower error bar limits, etc.) would be useful. If the editors are comfortable with the review paper emphasis then this paper is acceptable as is, but if they are primarily seeking new research reports, this paper needs more emphasis on the new H2S/H2 measurements, with more details about the measurement procedure and tabular reporting of the results.

We thank Reviewer 2 for his/her generous comments and appreciate the suggestions for improving the paper. The reviewer is correct that this is largely a review paper with the objective of summarizing the extensive program of microwave laboratory measurements conducted so as to support remote sensing of water vapor, ammonia, and liquid condensates in the jovian atmosphere from MWR (MicroWave Radiometer) instrument aboard the Juno spacecraft. The is one of a group of papers submitted by Juno instrument and science teams describing the instrument hardware and science studies supporting the upcoming observations which will begin later in 2016. The objective was to have these papers available (at least on-line) by the beginning of the mission operational phase (July 2016).

We agree with the reviewer that it would be good to provide additional data regarding the H2S/H2 measurements (since they are not reported elsewhere), and have included additional descriptions and the additional table detailing the lab results.

The following are specific comments.

Pg. 2

"While more extensive laboratory measurements of the microwave opacity of ammonia under simulated Jovian conditions were conducted (see e.g., Morris and Parsons, 1970; Steffes and Jenkins, 1987; Spilker 1990; Joiner and Steffes 1991), none (except Morris and Parsons) were conducted at pressures above 7 Bars, nor did the measurements carefully track the effects of preferential adsorption of ammonia in the test cells." This statement is not accurate. The Morris & Parsons paper reporting measurements of absorption by NH3 in hydrogen and NH3 in helium (at pressures up to 700 bars) details their method of tracking adsorption of microwave-absorbing species onto the apparatus walls. Spilker made measurements up to 8 atmospheres (as mentioned in section 3.1 of this paper) in mixtures of NH3 in H2 & He, but did not track adsorption.

The reviewer is correct and we have corrected the text to reflect these corrections.

Pg. 3

"Application of these absorption properties to radiative transfer models for the six wavelengths involved provide a valuable tool for planning observations, and will also make possible accurate retrievals of the abundance of these constituents."

Undoubtedly more accurate models of the propagation properties of these species will promote better understanding of the jovian atmosphere. But retrieval of the abundances of multiple intermixed constituents is hardly a straightforward task. If the authors have done work indicating that this retrieval can indeed be done unambiguously and accurately, it should be included here or referenced. If such work has been done by others, including in this journal issue, it should be referenced. Otherwise, this is a rather strong statement to be made without support.

The reviewer makes a very good point. While our preliminary work in this area has shown the importance of accurate models for the microwave opacity for ammonia and water vapor in order obtain accurate results in our simulated retrievals, our work on this is not yet published, nor ready for publication. Thus, we have reworded the statement to indicate the *potential* value of such measurements.

Pg. 15

"As shown in Figure 14, the measured opacity of H2S in a hydrogen atmosphere at 19.86 Bars total pressure and 376 K is consistent with the model of DeBoer and Steffes (1994)."

I have not found a quantitative description of the error bars used in the figures showing results. Are they 1-sigma? 3-sigma? something else? If they are 3-sigma, then the model of DeBoer and Steffes (1994) is not consistent with the data shown. The 5 data at the highest frequencies have the DeBoer and Steffes model well within those error bars, while only 1 of the 5 data at the lowest frequencies has that model within its error bar, and only barely for that one. The frequency dependence of the DeBoer and Steffes model appears to be too shallow. Fortunately, this does not affect the conclusion that H2S opacity is insignificant in Jupiter's lower troposphere at the longest Juno MWR wavelengths.

The error bars shown in Figure 14 are 2-sigma error bars. This is now made clear in both the text and in the table which has been added containing the data from the experiments.

The reviewer is correct in that modifications to the DeBoer and Steffes (1994) model could be employed to provide a better fit to the new, high-temperature/high-pressure data. However, as noted by the reviewer and in the text, for purposes of supporting the Juno mission, it was only necessary to show that the existing model did not *understate* the opacity from H_2S in an H_2/He atmosphere. We have added additional discussion regarding this point.

Fig. 5

This figure appears to be from Hanley & Steffes (2007) and should be attributed as such in the caption.

The reviewer is correct in that Figure 5 is from Hanley and Steffes (2009) and we have corrected the caption.

Fig. 8

The gas conditions specified in this figure are outside of the range of applicability of the Spilker model; the pressure is more than an order of magnitude above the maximum in his data set, and the temperature is more than 120 K above his maximum, so it is inappropriate to include it here.

Corrected!

Fig. 9

Two data near the high-frequency end of this chart are displayed differently from the others and appear to be much more uncertain, with (apparently) upper limits well above 10 dB/km and (unmarked) lower limits of essentially zero. This makes them effectively only upper limits, so standard upper limit symbols should be used for those data.

Corrected!

Figs. 11 & 12

Are these figures from Duong et al. (2014)? If so, they should be attributed as such in their captions.

The reviewer is correct. Figure 11 is from Duong et al. (2014) and has now been so attributed. Figure 12 is an image of the laboratory system (not previously presented) but has been included since the Mission Juno papers being submitted as a group to Space Science Reviews were asked to include information regarding mission instruments and hardware. It was felt appropriate to similarly document the supporting hardware for this laboratory program.

HIGH-PRECISION LABORATORY MEASUREMENTS SUPPORTING RETRIEVAL OF WATER VAPOR, GASEOUS AMMONIA, AND AQUEOUS AMMONIA CLOUDS WITH THE JUNO MICROWAVE RADIOMETER (MWR)

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ABSTRACT

The NASA Juno mission includes a six-channel microwave radiometer system (MWR) operating in the 1.3–50 cm wavelength range in order to retrieve abundances of ammonia and water vapor from the microwave signature of Jupiter (see Janssen et al. [2016]). In order to plan observations and accurately interpret data from such observations, over 6000 laboratory measurements of the microwave absorption properties of gaseous ammonia, water vapor, and aqueous ammonia solution have been conducted under simulated Jovian conditions using new laboratory systems capable of high-precision measurement under the extreme conditions of the deep atmosphere of Jupiter (up to 100 Bars pressure and 505 K temperature). This is one of the most extensive laboratory measurement campaigns ever conducted in support of a microwave remote sensing instrument. New, more precise models for the microwave absorption from these constituents have and are being developed from these measurements. Application of these absorption properties to radiative transfer models for the six wavelengths involved will provide a valuable planning tool for observations, and will also make possible accurate retrievals of the abundance of these constituents during and after observations are conducted.

1. INTRODUCTION

It is well understood that the microwave emission spectrum of Jupiter's troposphere reflects the abundance and distribution of constituents such as ammonia, water vapor, and aqueous ammonia clouds (see, e.g., Janssen et al., 2005), but there are a number of factors that limit the accuracy of this approach for microwave remote sensing of these constituents (de Pater et al., 2005). The most critical of these is the knowledge of the microwave absorption properties of these constituents under Jovian conditions. Previous laboratory measurements of the microwave opacity of water vapor under pressures and temperatures representative of the deep atmosphere of Jupiter were only conducted at one wavelength in a nitrogen atmosphere (Ho et al., 1966), but not in a hydrogen–helium atmosphere. While more extensive laboratory measurements of the microwave opacity of ammonia under simulated Jovian conditions were conducted (see e.g., Morris and Parsons, 1970; Steffes and Jenkins, 1987; Spilker 1990; Joiner and Steffes 1991), none (except Morris and Parsons) were conducted at pressures above 8 Bars, nor did many of the measurements carefully track the effects of preferential adsorption of ammonia in the test cells. Additionally, none of these measurements investigated water vapor's role in broadening ammonia's microwave absorption spectrum. While effects of upper-level crystalline clouds have negligible effect on the centimeter-wavelength emission from Jupiter, putative tropospheric clouds of liquid aqueous ammonia (liquid water with dissolved ammonia) may have detectable influence on its centimeter-wave emission signature (see, e.g., Janssen et al., 2005). To date, nearly all microwave radiative transfer models incorporating effects of aqueous clouds employ the measured properties of pure water to estimate the effects of such clouds. However, the effect of dissolved constituents on the dielectric properties of condensed water can be significant, and no laboratory measurements of the effect of dissolved ammonia on the microwave properties of such condensates had been conducted prior to this study.

In order to enable accurate interpretation of data from high-precision observations of the centimeter-wave emission from Jupiter's troposphere such as will be measured by the Juno Microwave Radiometer (Janssen et al., 2016), over 6000 laboratory measurements of the microwave absorption properties of gaseous ammonia, water vapor, and aqueous ammonia solution have been conducted under simulated Jovian conditions using new laboratory systems capable of high-precision measurement under the extreme conditions of the deep atmosphere of Jupiter (up to 100 bars pressure and 505 K temperature). This is one of the most extensive laboratory measurement campaigns ever conducted in support of a microwave remote sensing instrument. New, more precise models for the microwave absorption from these constituents have been developed from these measurements. Application of these absorption properties to radiative transfer models for the six wavelengths involved provide a valuable tool for planning observations, and will also assist in potential retrievals of the abundance of these constituents.

The laboratory program described in this paper makes use of three different laboratory systems. The first, referred to as the "medium-pressure" system, is described in Section 3.1 and was the used for measurements of the 1.1-20 cm wavelength (1.5-6 GHz) opacity of ammonia in a hydrogen/helium atmosphere at pressures up to 12 bars and temperatures from 185-450 K (results discussed in Section 3.1 and in Hanley et al., 2009). The second system, referred to as the "high-pressure" system, is described in Section 2.1 and was the used for measurements of the 5-21 cm wavelength (1.4-6 GHz) opacity of water vapor in a hydrogen/helium atmosphere at pressures up to 101 bars and temperatures up to 505 K (results discussed in Section 2.1 and in Karpowicz et al.

2011a,b). Subsequently the same high-pressure system was used to conduct measurements of the 5-21 cm wavelength (1.4-6 GHz) opacity of gaseous ammonia in a hydrogen/helium atmosphere at pressures up to 98 bars and temperatures up to 503 K (results discussed in Section 3.2 and in Devaraj et al. 2014). Of special note was the use of the high-pressure system to conduct the first measurements of the effects of water vapor broadening on the centimeter-wavelength absorption spectrum of ammonia (results discussed in Section 3.3 and in Devaraj et al. 2014). A final measurement using the high-pressure system was conducted of the 5-21 cm wavelength (1.4-6 GHz) opacity of gaseous hydrogen sulfide in a hydrogen/helium atmosphere at pressures up to 20 bars and temperatures up to 376 K (results discussed in Section 5). The third system is described in Section 4 and was the used for measurements of the 3.5-15 cm wavelength (2-8.5 GHz) complex dielectric properties of aqueous ammonia, the putative liquid cloud constituent in the jovian atmosphere (results discussed in section 4 and in Duong et al. 2014).

2. WATER VAPOR

In the laboratory measurement campaign for water vapor, over 2000 laboratory measurements of the microwave opacity of water vapor in a hydrogen/helium atmosphere were conducted in the 5-21 cm wavelength range (1.4-6 GHz) under conditions representative of the altitude ranges in the Jovian atmosphere where water vapor exists in detectable quantities. The wavelength range measured corresponds to the channels of the Juno microwave radiometer (MWR) most sensitive to the altitudes where water exists in an uncondensed state (Janssen et al., 2005). The environmental conditions measured included pressures from 30 mbars to 101 bars and temperatures from 330-505 K. The mole fraction of water vapor ranged from 0.19% to 3.6% with some additional measurements of pure H_2O. In order to conduct experiments under the extreme conditions of the deep Jovian atmosphere, a new laboratory measurement system was developed that is the first to provide such flexibility in temperature, pressure and wavelength (Karpowicz and Steffes, 2011a). The method used to measure the microwave absorptivity of a gas is based on the lessening in the quality factor (Q) of a resonant mode of a cylindrical cavity in the presence of a lossy gas. This technique involves monitoring the changes in 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. Described at length by Hanley and Steffes (2007), it has been successfully utilized for over one half of a century. The cavity resonator technique is also used for measuring refractivity based on the shift of the resonant frequency and is similarly described by Hanley and Steffes (2007).

2.1 Laboratory Configuration

Shown in Figure 1 is a block diagram of the high-pressure measurement system used for these measurements. Figure 1a shows the gas handling system used to create the gas mixtures under simulated Jovian conditions. Figure 1b shows the data acquisition system necessary to monitor the environmental conditions of the gases under test and the microwave system used to measure their microwave properties. The heart of the gas handling system is the pressure vessel that contains the microwave resonator used to characterize the microwave properties of the gases under test. Figure 2 shows a photograph of the pressure vessel and water reservoir located within the temperature chamber (oven).

The pressure vessel was custom-built by Hays Fabrication and Welding (Springfield, Ohio). The vessel is constructed from a 30.48 cm section of schedule 100 pipe that is 35.56 cm (14 inches) in diameter (outer dimension). An elliptical head is welded to the bottom giving the vessel a maximum interior height of 46.04 cm (19 inches). The top includes an ANSI (American National Standards Institute) class 900 flange 10.16 cm (4 inches) thick, with a top plate that is 9.2 cm (3-5/8 inches) thick. The vessel has an internal volume of 32.75 liters, and weighs approximately 544.3 kg.

The water reservoir is made of a section of T-304 stainless steel pipe 46 cm long and 3.8 cm in diameter. In addition, the system includes a Grieve industrial oven model AB-650 (maximum temperature 332 C), two Matheson R regulators (Model 3030-580 for Ar/He, and 3030-350 for H_2), two Omega RDPG7000 pressure gauges (one rated from 0-2 bars absolute, the other rated to 20 bar), an Omega RPX1009L0-1.5KAV pressure transducer capable of measuring up to 103 bars at 315 C, and a temperature sensor, which was initially an Omega R thermocouple probe (TC-T-NPT-G-72). Valves rated for high temperature and pressure were used throughout the system.

The pressure vessel with all input flanges and microwave cable feedthroughs was tested by the manufacturer at pressures from 13 to 100 bars. In place of a standard rubber or viton O-ring, a composite (glass fiber/NBR) KLINGERsil C-4430 is used to seal the pressure vessel along with 20 nuts 6 cm (2-3/8 inches) in diameter torqued to 62 Newton-meters (1300 lb-ft) . The weights of the pressure vessel (544.3 kg) and of the shipping weight oven (739 kg) far exceeded the load capacity of our laboratory floor. As an alternative, the system was placed upon an outdoor concrete pad on which a decommissioned crane once stood. Thus, all system components except for the microwave network analyzer, sensor monitors, and the control computer are placed outdoors, protected by a metallic shed.

Over the course of the measurement campaign, some additions were made to the system described above. First, after initial experiments at 375 K, the thermocouple probe was replaced by a high temperature thermometer / hygrometer (JLC international EE33-MFTI-9205-HA07-D05-AB6-T52) which also provided an independent, secondary measure of water vapor density. Since the thermometer/hygrometer had limited temperature range (only up to 475 K), a high precision Omega Resistance Temperature Detector (RTD) (PRTF-10-2-100-1/4-9-E-SP) was used above 475 K.

The microwave resonator included in Figure 1b has been used in several studies. Its most recent version was described in Hanley and Steffes (2007). The resonator is a cylindrical cavity resonator with an interior height of 25.75 cm, and an interior radius of 13.12 cm. The resonator is connected to the network analyzer via high temperature coaxial cables and via Ceramtec (16545-01-CF) coaxial bulkhead feedthroughs capable of maintaining pressures up to 103 bars at temperatures up to 350 C. Outside of the oven, two sections of Andrews RCNT 600 microwave cable (each 24 meters in length) connect to the Agilent R E5071C network analyzer, located in a stable, indoor environment. The S parameters measured by the network analyzer are read in via GPIB to the data acquisition computer.

The measured pressure and temperature conditions for each experiment are delivered to the data acquisition computer via USB buses leading to the laboratory from outdoors. After initially using pressure sensors that reported pressure relative to ambient, new absolute pressure gauges (rather than pressure relative to ambient) were installed, with the same precision as the Omega DPG7000 series (GE Sensing /Druck DPI-104). A detailed list of all instrumentation used and their associated precisions are presented in Karpowicz and Steffes (2011a).

2.2 Results of Laboratory Measurements

The 2000-plus measurements conducted enabled development of the first model for the opacity of gaseous H_2O in a H_2/He atmosphere under Jovian conditions developed from laboratory data of H_2/He/H_2O mixtures. As shown in Figure 3, the environmental conditions of the laboratory data bracket the putative temperature/pressure profile for the deep Jovian atmosphere. The new model is based on a model for the microwave opacity of water vapor in a terrestrial atmosphere from Rosenkranz (1998), with substantial modifications to reflect the effects of Jovian conditions, as described in detail by Karpowicz and Steffes (2011a,b). Shown in Figure 4 is an example of the processed data collected versus two pre-existing, non-laboratory based models for the opacity from water vapor (Goodman, 1969, and DeBoer, contained in dePater et al., 2005), and the new model from Karpowicz and Steffes (2011a,b). The new model from these measurements will play a key role in the detection and measurement of the water vapor abundance at Jupiter.

A key aspect of developing an accurate model for the microwave opacity of water vapor includes understanding the effects of non-ideality in the relationships between pressure, temperature, and density in the gas mixtures under test. As shown in Figure 1a, we employ a flow meter, which when combined with measurements of temperature and pressure allows us to characterize the compressibility of the gas mixture so as to more accurately determine the actual densities of each constituent. A detailed study of the equation-of-state for a Jovian atmospheric system resulting from these measurements is described by Karpowicz and Steffes (2013).

3. AMMONIA

Gaseous ammonia (NH_3) has long been known to be the largest source of centimeter wavelength absorption in the Jovian troposphere. (See, e.g., Law and Staelin, 1968.) As a result, all six channels of the Juno MWR (1.3-50 cm) will measure effects from the presence of ammonia, at different altitudes in the Jovian troposphere (Janssen et al., 2005). The microwave opacity of gaseous ammonia dominates the microwave emission spectrum of Jupiter. Knowledge of ammonia's microwave properties is critical, since retrieval of the residual effects on microwave emission from water vapor requires precise knowledge of the ammonia absorption spectrum. Accuracies better than +/-6% under conditions for the altitude ranges where water vapor exists have been achieved, so as to allow reliable detection of the residual effects of water vapor on the Jovian microwave emission spectrum, which is a key objective for the Juno Microwave Radiometer (MWR).

Since the environmental conditions in altitude ranges probed by the six MWR channels are quite different, two separate measurement campaigns were conducted. The first focused on conditions in the upper and middle troposphere, which affect the highest frequency channels. Over 1400 measurements of the microwave absorption and refraction of ammonia in a hydrogen/helium atmosphere were conducted from 1.1 to 20 cm at temperatures from 184-450K and at pressures from 30 mbar to 12 bars, using a system described in Hanley and Steffes (2007). Subsequently, measurements of the microwave absorption and refraction of ammonia in a hydrogen/helium atmosphere were conducted (with the system described above for measurements of water vapor) from 5-21 cm at temperatures from 323 to 503 K and pressures from 66 mbar to 98 Bars (Devaraj et al., 2014).

3.1 Laboratory Measurements for the Upper and Middle Jovian Troposphere

A large amount of work on the modeling of the microwave absorption properties of ammonia has been conducted for many decades. (See, e.g., Townes and Schawlow, 1955.) However, most of the early models for ammonia opacity were based on laboratory measurements that were limited to the pressures and temperatures that could be readily produced in the laboratory, usually on the order of a few bars. Morris and Parsons (1970), however, were able to measure the broadening effects of H_2, He, N_2, and Ar on NH_3 up to pressures of nearly 700 bars by using a high-pressure vessel and gas compressor. Their measurements were only performed at room temperature and at one frequency (9.58 GHz) in a tunable resonant cavity. Measurements were conducted over a much wider frequency range (up to 6 bars pressure) by Steffes and Jenkins (1987) and by Spilker (1990) up to 8 bars. However, neither accounted for effects of adsorption of gaseous ammonia onto the metal surfaces in the pressure vessels and resonators, limiting their precision. In the first measurement campaign conducted by our team (Hanley et al., 2009) in support of the Juno mission, 1440 laboratory measurements were conducted of the microwave opacity of NH3 in an H2/He atmosphere in the 1.1-20 cm wavelength range (1.5-27 GHz) across a wide range of temperatures and pressures, characteristic of those found in the middle and upper tropospheres of Jupiter and Saturn (Temperatures from 184-450 K and pressures from 30 mBar to 12 Bar).

The system used to conduct these measurements is shown in Figure 5, and is described at length in Hanley and Steffes (2007). Unlike the system described in the previous section used to conduct measurements of water vapor under conditions simulating the deeper troposphere of Jupiter, this system contained two resonators and an additional low-temperature chamber (see Figure 6), so as to also provide measurements in the wavelength range and under simulated conditions for the altitude ranges probed by the 1.3 cm, 3 cm, and 5.7 cm channels of the Juno Microwave Radiometer (MWR). Results of these measurements are described in Hanley et al. (2009), which also contains a new model for opacity of ammonia based on these laboratory measurements. As shown in Figure 7, the new model for ammonia opacity better fits the extraordinarily precise data obtained from this measurement system than previous models.

3.2 Laboratory Measurements for the Lower Jovian Troposphere

While the model for ammonia opacity in a hydrogen/helium atmosphere described in Hanley et al. (2009) performs well at pressures up to 50 Bars and at frequencies up to 30 GHz, further improvements to the model were considered important to reflect several aspects of the deep Jovian troposphere. First, under conditions of the deep Jovian atmosphere such as those sensed by the longest 2 wavelengths of the Juno Microwave Radiometer (MWR), the effects of compressibility (non-ideality) of the gaseous constituents will change their microwave absorption properties as a function of temperature and pressure. Second, the measurements of Hanley et al. (2009) were conducted with a fixed ratio of hydrogen and helium (86.4% hydrogen and 13.6% helium by mole fraction) as the broadening gas. Thus the model developed from those laboratory measurements assumed a fixed ratio for the pressure broadening effects of hydrogen and helium. While this assumption was valid for pressures below 50 Bars, at higher pressures the differential compressibility of hydrogen and helium could change the relative abundances of each constituent, requiring a more detailed knowledge of the pressure broadening behavior of each. Finally, at higher pressures, the effects of the submillimeter-wave rotational lines of ammonia play a more significant role in the centimeter-wavelength absorption spectrum. While this was noted in Hanley and Steffes (2009), the actual effects of such contributions were too small to measure at pressures of 12 Bars or less.

As a result, an extensive program of over 1100 laboratory measurements of the microwave opacity of ammonia in a hydrogen/helium atmosphere were conducted in the 5-21 cm wavelength range (1.4-6 GHz) under conditions representative of the altitude ranges in the Jovian atmosphere sensed by the 4 longest wavelengths of the Juno MWR. The measurement system developed by Karpowicz and Steffes (2011a), described above, was used for these measurements, and the environmental conditions measured included pressures from 66 mbars to 98 bars and temperatures from 323-503 K.

These centimeter-wavelength measurements, plus those from Hanley and Steffes (2009), Morris and Parsons (1970), and millimeter-wavelength measurements from Devaraj et al. (2011) were used by Devaraj et al. (2014) to develop a more robust model that performs well even at millimeter-wavelengths and at high pressure. The new model developed by Devaraj et al. (2014) accounts for compressibility and also incorporates the effects of the sub-millimeter wavelength lines on the high pressure centimeter-wavelength absorption spectrum. As shown in Figure 8, the new model for ammonia opacity better fits the extraordinarily precise data obtained from this measurement system than previous models.

3.3 Laboratory Measurements of Water Vapor's Effects on the *Ammonia* Microwave Absorption Spectrum

Detailed understanding of the microwave absorption properties of *ammonia* is necessary for the reliable detection of *water vapor* in the deep Jovian atmosphere since ammonia opacity dominates at centimeter-wavelengths, and the signature of water vapor is measured as a residual effect. In the original conception of MWR, described by Janssen et al. (2005), detection of water vapor was based only on the measurable effect of the intrinsic opacity from water vapor on centimeter-wavelength limb darkening at Jupiter. However, since our measurements of the self-broadening of water vapor indicated that water vapor had a remarkably large self-broadening cross-section (Karpowicz and Steffes, 2011a,b), we conducted further measurements which showed that water vapor is also an extremely strong source of broadening of the ammonia absorption spectrum. Depending on its abundance, water vapor's effect on the *ammonia* absorption spectrum may even compare with its own intrinsic opacity in affecting the centimeter-wavelength Jovian emission spectrum. In order to accurately characterize this effect, we have completed over 850 measurements of the effects of water vapor broadening on the ammonia absorption spectrum. These data were taken at temperatures from 373-503 K, under Jovian conditions (H_2/He atmosphere with pressures up to 97 Bars).

These measurements of the effects of water vapor broadening of the microwave absorption spectrum of ammonia in a hydrogen/helium atmosphere indicate that water vapor broadens the centimeter-wavelength absorption spectrum about 5 times more than the equivalent amount (by volume) of molecular hydrogen, and about 9 times as much as helium. As shown in Figure 9, when 2.3 Bars of water vapor is added to 99 milliBars of ammonia vapor, the increase in microwave absorption due to water vapor's broadening of the ammonia spectrum far exceeds the added intrinsic opacity of the water vapor itself. Thus, in a very water-rich environment (10x solar+) the effect of water vapor on the ammonia absorption spectrum may compare with its intrinsic opacity in its effect on centimeter wavelength Jovian emission. The new model developed by Devaraj et al. (2014) for the centimeter- and millimeter-wavelength opacity of ammonia also allows for addition of linewidth and coupling parameters for foreign gas broadening by water vapor The resulting fit to the laboratory data is shown in Figure 9.

3.4 Summary of Laboratory Measurements Involving Gaseous Ammonia

In aggregate, over 3300 measurements of the microwave properties of ammonia under a wide range of conditions characteristic of the portions of the Jovian atmosphere to be sensed by the Juno Microwave Radiometer (MWR) were conducted. The range of conditions tested, together with a plot of the nominal Jovian temperature-pressure profile is shown in Figure 10. The models for microwave opacity resulting from these measurements will enable the reliable retrieval of ammonia abundances from MWR data and will also enable reliable detection of the residual microwave opacity from water vapor.

4. AQUEOUS AMMONIA

Depending on the local abundance of water vapor, liquid aqueous clouds with dissolved ammonia likely form near the 6-10 Bar level of the Jovian atmosphere. (See e.g. Roos-Serote *al*. 2005.) While the actual bulk densities of such clouds are not known, the maximum possible values (corresponding to the amounts of each condensate exceeding the saturation vapor pressure at each altitude) are significant in that they could potentially be dense enough to affect the atmospheric microwave emission spectrum (see e.g., Janssen *et al.* 2005 or de Pater *et al.* 2005). In previous radiative transfer models of the microwave emission from Jovian atmospheres, the complex dielectric constant of the cloud liquid was assumed to be approximately that of water (see e.g., Janssen *et al.* 2005 or de Pater *et al.* 2005) since the dissolved ammonia concentration is expected to be relatively low (approximately 2-3%) due to the relatively low abundance of ammonia (see e.g. Atreya et al., 1999). This assumption was made since no model existed for the complex dielectric constant for aqueous ammonia.

In this work, a model for the complex dielectric constant of aqueous ammonia (NH_4OH) has been developed based on several thousand new laboratory measurements in the frequency range between 2 and 8.5 GHz and at temperatures from 274-297 K using a dielectric probe measurement system (Figures 11 and 12). This new model, described in Duong et al. (2014), is a significant step in better understanding the microwave properties of aqueous ammonia and is useful for characterizing cloud opacity of aqueous ammonia clouds under Jovian conditions. Shown in Figure 13 are the results from the new model showing how dissolved ammonia in the range between 0.85% to 8.5% (by volume) enhances the microwave opacity of an aqueous cloud.

5. OTHER GASEOUS CONSTITUENTS

While a number of additional microwave absorbing constituents exist in the Jovian atmosphere (e.g., H_2S and PH_3), their putative abundances are low enough so that their opacities will not significantly affect the modeled centimeter-wavelength emission from Jupiter. (See, e.g., DeBoer and Steffes, 1994, Hoffman et al., 2001 or Hanley et al., 2009.) Since the opacity models for such constituents were determined based on laboratory measurements conducted under conditions characteristic of the middle to upper troposphere of the outer planets, it was felt that a test should be conducted on at least one such constituent so as to verify the reasonability of extrapolating such models to higher temperatures and pressures.

Using the “high pressure” system described above (Section 2.1), and using the techniques described in Hanley et al. (2007), measurements of the microwave opacity of hydrogen sulfide in a hydrogen atmosphere were conducted. A method was employed to compensate for any possible adsorption effect by saturating the surface of the gas handling system with a layer of hydrogen sulfide before the measurements are taken. (A similar approach was employed by Hanley et al., 2009 and Devaraj et al., 2014 for their measurements of ammonia.) Since only a limited number of adsorbate layers can form, any additional gas added would not adsorb after the substrate surface is fully saturated. The adsorption of hydrogen sulfide is monitored by measuring changes in the quality factors of the resonances with time. Once the quality factors stabilize, the internal surface is said to be fully saturated, at which point, the rate of adsorption and desorption is equal. Only after the hydrogen sulfide abundance is stable are measurements taken, and then the pressure-broadening hydrogen is added to the system. As with our previous absorptivity measurements, the first set of measurements of the available resonances is taken under vacuum conditions at the desired temperature. The next step is to add the primary test gas (hydrogen sulfide). Once the system is thermally stabilized and the necessary measurements are taken, hydrogen is added as the broadening gas. The frequency shifted resonances are then again measured. Afterwards, a second vacuum is drawn. After venting down to atmospheric pressure, the vacuum pump is run for at least 12 hours to ensure that the remaining test gases have been evacuated. A second set of vacuum measurements are taken at the desired temperature. The dielectric matching process consists of shifting the resonances by the same amount as that of the test gas mixture using pure argon. Measurements of the quality factors of each resonance are recorded as dielectric matches are made with the reference gas (argon).

As shown in Table I and Figure 14, the measured opacity of H_2S in a hydrogen atmosphere at 19.86 Bars total pressure and 376 K is generally consistent with the model of DeBoer and Steffes (1994). Note that since the radiative transfer studies indicated that the opacity from H_2S does not affect the Jovian centimeter-wavelength emission, it was only necessary to demonstrate that the opacity did not *exceed* that predicted by the DeBoer and Steffes (1994) model. (However, the new data may be used to further refine the DeBoer and Steffes (1994) model for the centimeter-wavelength absorption of hydrogen sulfide under jovian conditions.)

6. FUTURE WORK

Our results for ammonia opacity described by Hanley et al. (2009) and by Devaraj et al. (2014) and our water vapor results from Karpowicz and Steffes (2011a, 2011b) included models for the opacity of these constituents valid over the pressure and temperature ranges measured in the laboratory experiments (temperatures up to 500 K and pressures up to 100 bars). However, our studies of the microwave emission made using these models indicate that significant contributions to the emission at the 24-cm and 50-cm wavelengths to be measured by the Juno MWR will be made by layers of the atmosphere with temperatures at or exceeding 600 K. While the ammonia centimeter-wavelength opacity models described by Hanley et al. (2009) and Devaraj et al. (2014) give consistent results at temperatures up to 500 K (within 6 %), they diverge more significantly when extrapolated to temperatures and pressures exceeding 600 K and 80 bars (approximately 15 % divergence at the 600 K and 80 bars). Similarly, at temperatures above 550 K, the model for water vapor opacity developed by Karpowicz and Steffes (2011a and 2011b) exhibits non-physical attributes. (That is, the opacity model shows no temperature dependence for temperatures above 550 K.) Such inaccuracies can increase the uncertainty of ammonia and water vapor retrievals, which are key products of the Juno MWR experiment. Although our pressure vessel is unable to maintain high pressures at temperatures exceeding 565 K, laboratory measurements of the centimeter-wavelength opacity of pure ammonia and pure water vapor have recently been conducted at 600 K and lower pressures. Since one of the major differences between the Hanley et al. (2009) and Devaraj et al. (2014) models involves the temperature dependence terms for the self-coupling of ammonia and the broadening gases, these lab results will allow correcting these terms in the current models, and then enable re-fitting the remaining terms so as to best match the entirety of our data set for the centimeter-wavelength opacity of ammonia. This refinement will assure a reliable estimate of the centimeter-wavelength opacity from ammonia under high-pressure conditions at temperatures up to 600 K or higher. Additionally, since the major issue with the water-vapor model at high temperatures involves the self-continuum term of the microwave opacity, the measurements of pure water vapor opacity at 600 K will allow us to develop a correction to the temperature dependence of this term, providing a reliable opacity model at higher temperatures.

7. SUMMARY

The extensive laboratory measurements campaign conducted in support of the Juno Microwave Radiometer (MWR) instrument has provided high-accuracy models for the microwave absorptive properties of gaseous ammonia and water vapor, and for aqueous ammonia cloud condensates, based on thousands of laboratory measurements. Application of these absorption properties to radiative transfer models for the six wavelengths used by the Juno MWR provide a valuable tool for planning observations, and will also make possible accurate retrievals of the abundance of these constituents during and after observations are conducted.

Acknowledgements

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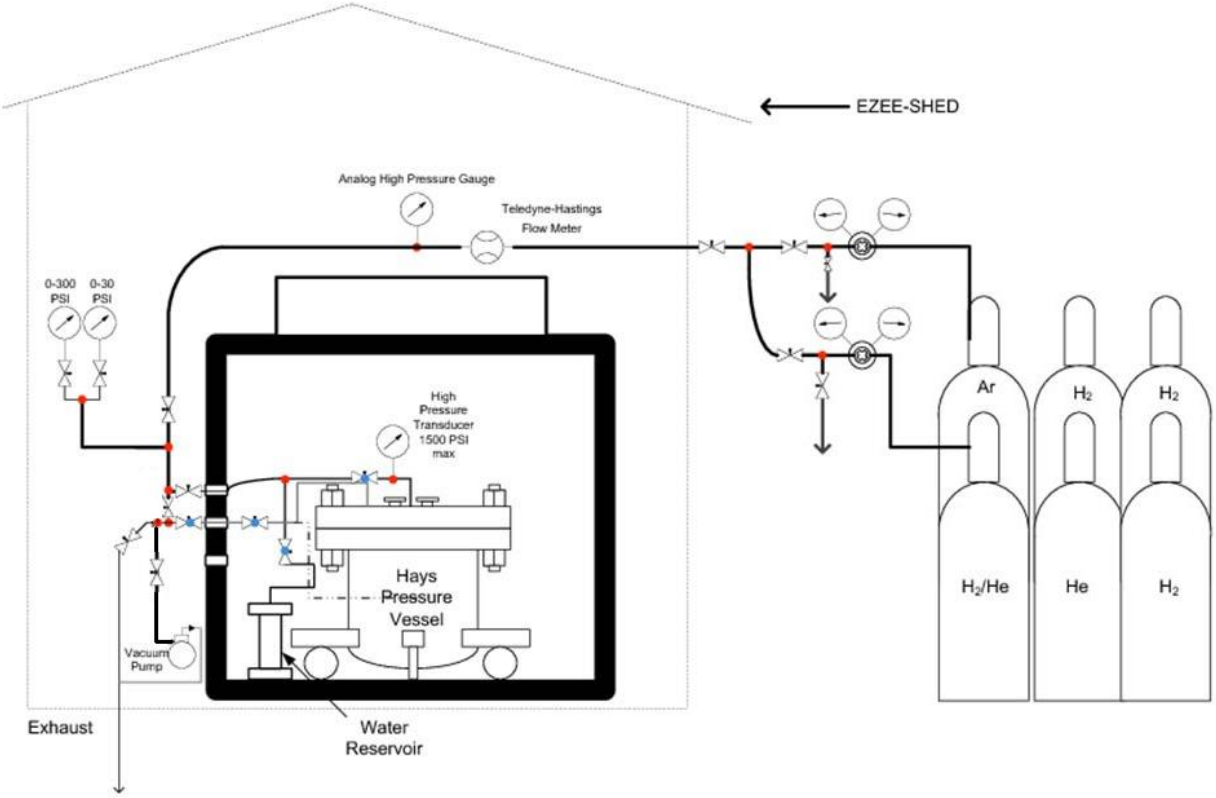

Figure 1a: The Georgia Tech high-pressure system used for measurement of the centimeter-wavelength properties of ammonia and water vapor under simulated Jovian conditions (described in Karpowicz and Steffes, 2011).

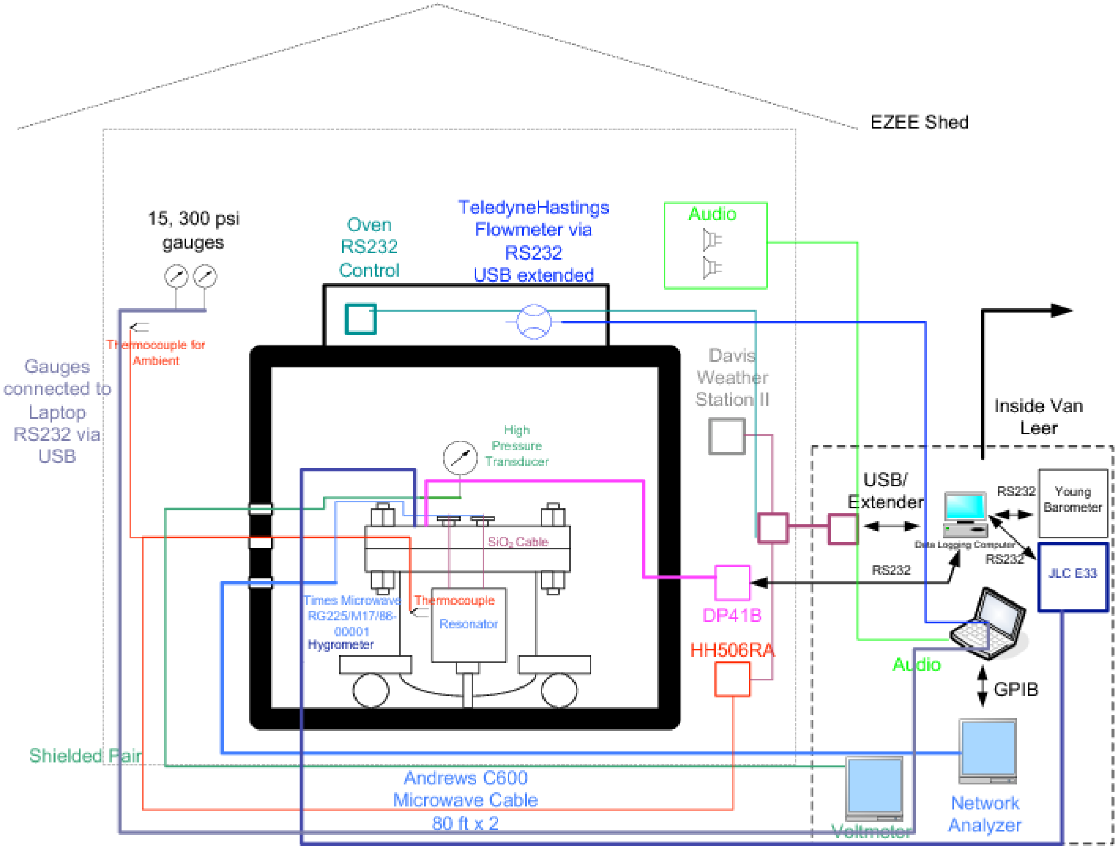

Figure 1b: The microwave measurement and data acquisition system used for measurement of the centimeter-wave properties of water vapor and ammonia under simulated Jovian conditions (from Karpowicz and Steffes, 2011).

Figure 2: Ultra-high pressure vessel capable of maintaining a 100 Bar pressure Jovian environment, contained within the temperature vessel (oven). The microwave resonator is contained within the pressure vessel. The water reservoir is the large metallic tube at left.

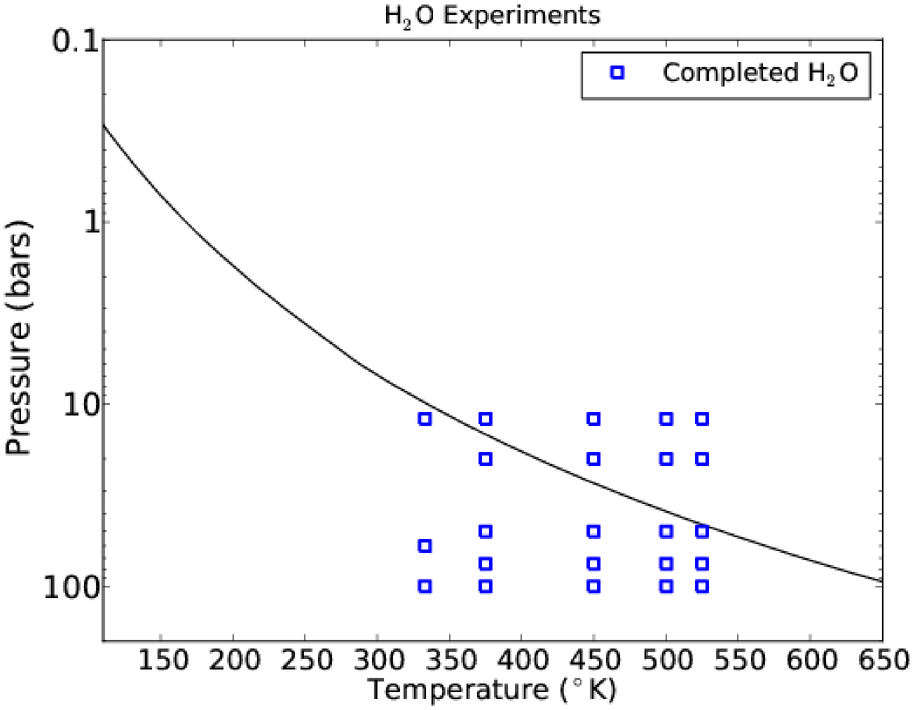

Figure 3: Range of temperatures and pressures over which measurements of microwave opacity of water vapor were conducted. A reference dry Jovian adiabatic temperature-pressure profile is shown for reference (from Karpowicz and Steffes, 2011).

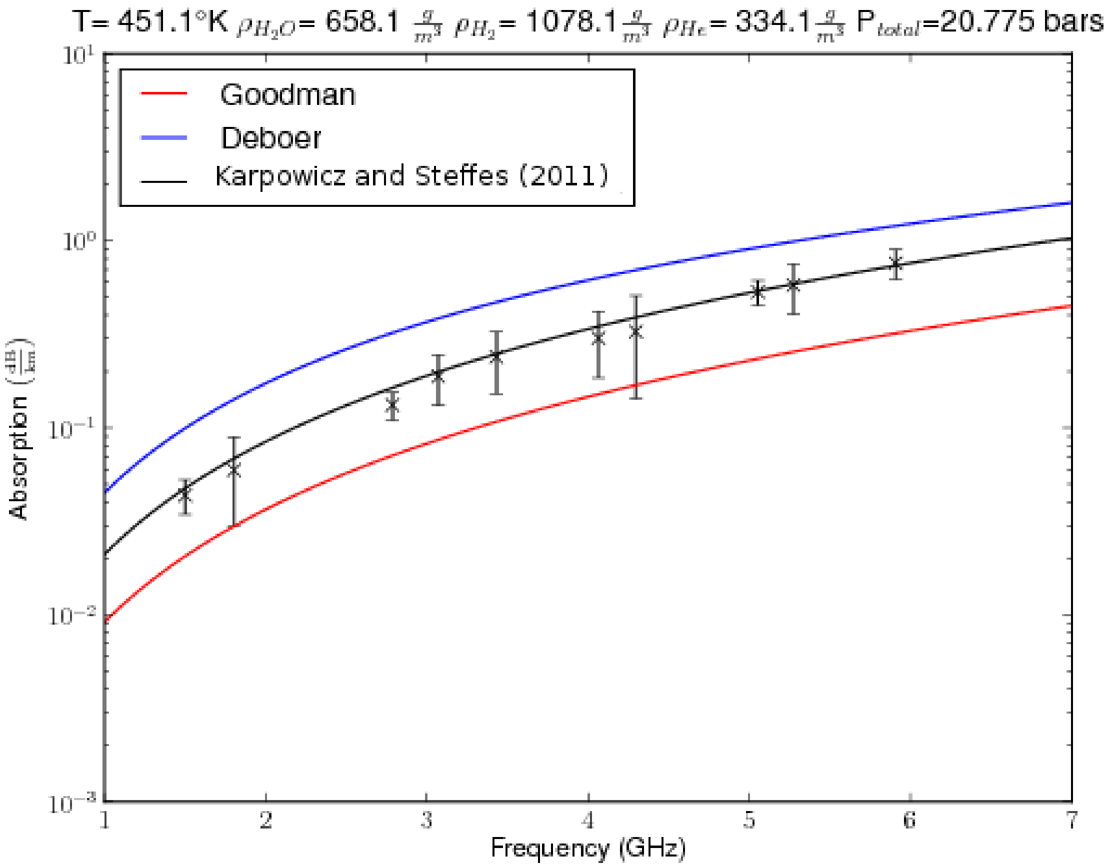

Figure 4: Example laboratory measurements of the microwave absorption from water vapor in an H_2/He atmosphere at 20.8 Bars pressure, along with models from Goodman (1969), DeBoer (described in de Pater et al., 2005), and this work (described in Karpowicz and Steffes, 2011). Displayed error bars are 2-sigma.

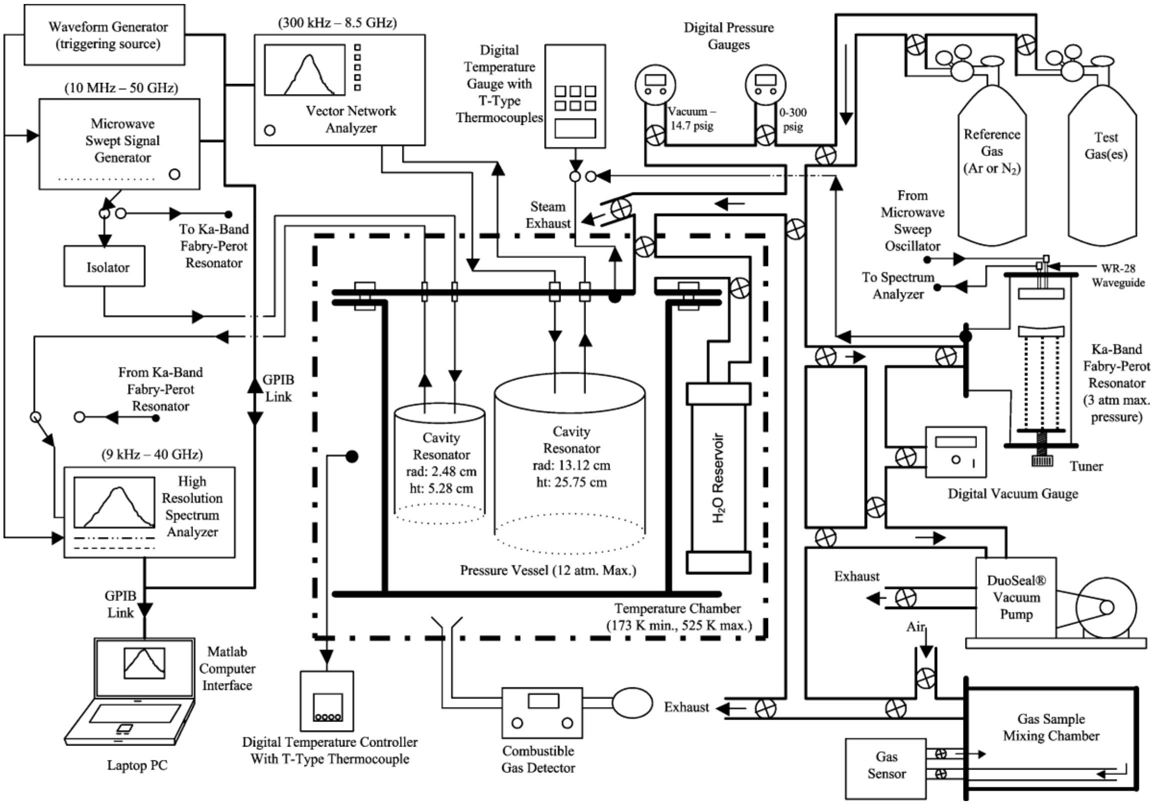

Figure 5: Laboratory system used for measurement of 1.3-21 cm opacity and refractive indices of gases under simulated conditions for the upper and middle Jovian troposphere (from Hanley et al., 2009).

Figure 6: Medium-pressure vessel containing dual resonators placed in ultra-cold temperature chamber operating at 185 K.

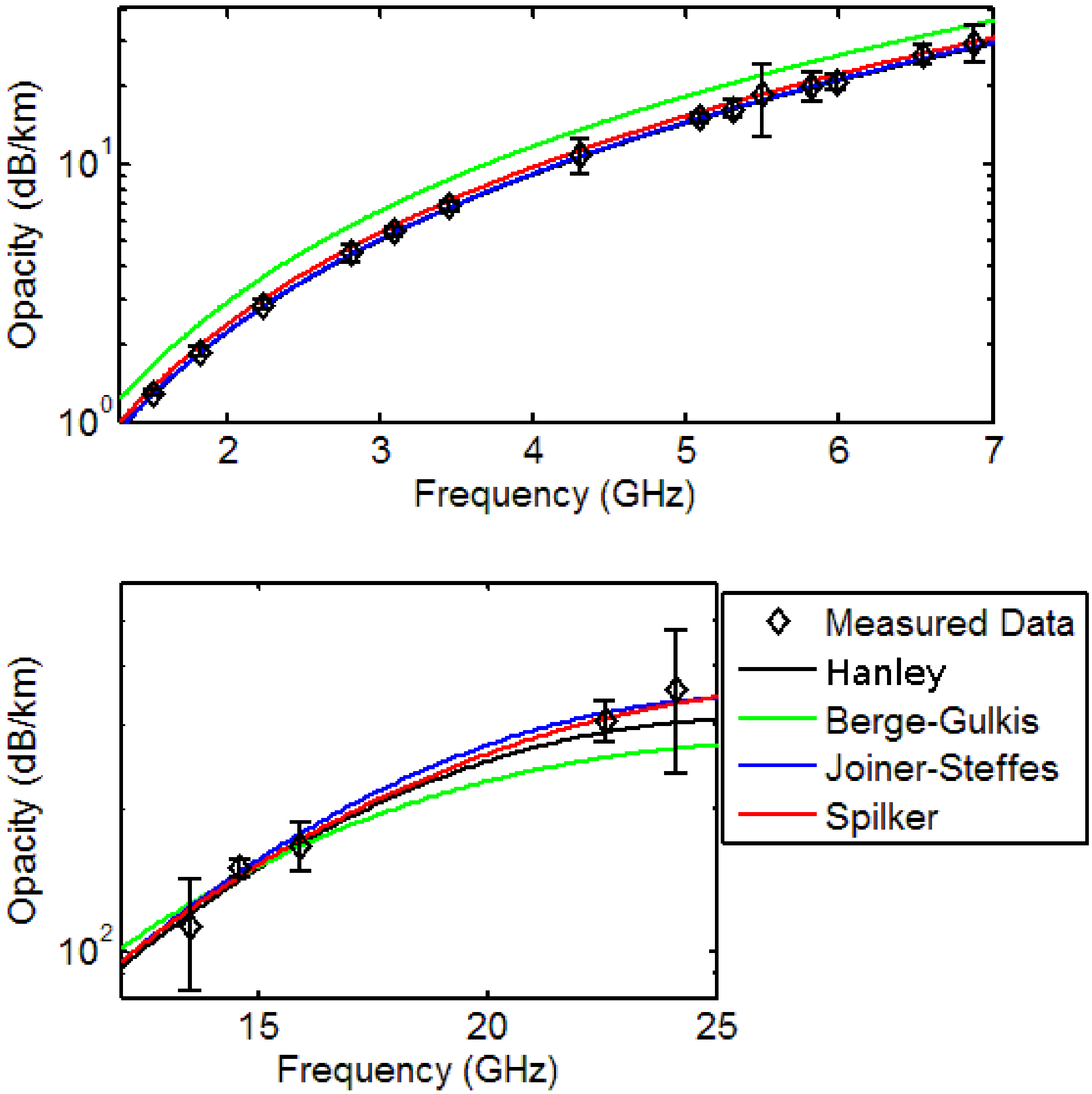

Figure 7: Opacity data measured using the large cavity resonator (above) and small cavity resonator (below) for a mixture of NH_3 = 0.95%, He = 13.47%, H_2 = 85.58% at a pressure of 8.0 bars and temperature of 295.5 K compared to models from Berge and Gulkis (1976), Joiner and Steffes (1991), Spilker (1990) and this work (described in Hanley et al., 2009). Displayed error bars are 2-sigma.

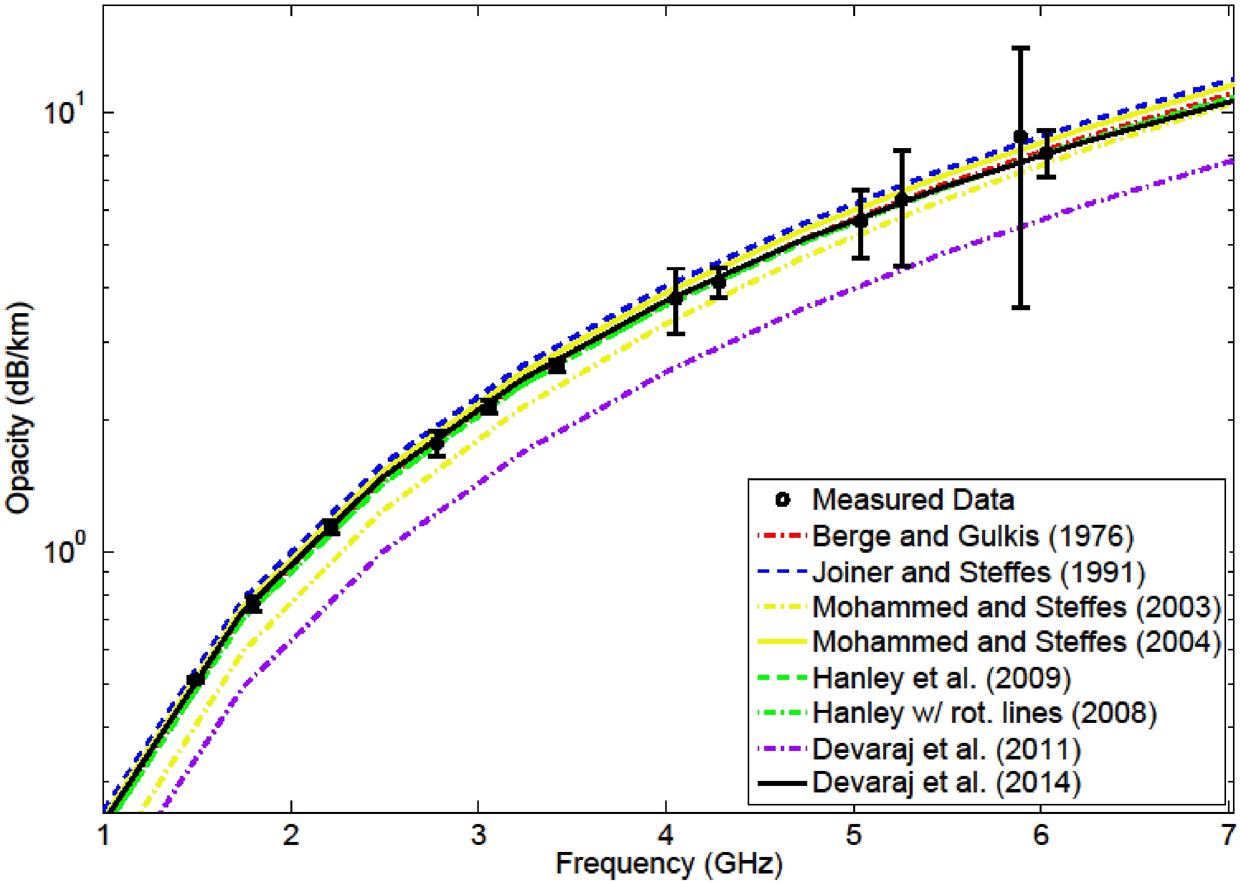

Figure 8: Opacity data measured using the high-pressure centimeter-wavelength system for a mixture of NH_3 = 0.09%, H_2 = 99.91% at a pressure of 93.545 bar and temperature of 446.9 K compared to various models (described in Devaraj et al., 2014). Displayed error bars are 2-sigma.

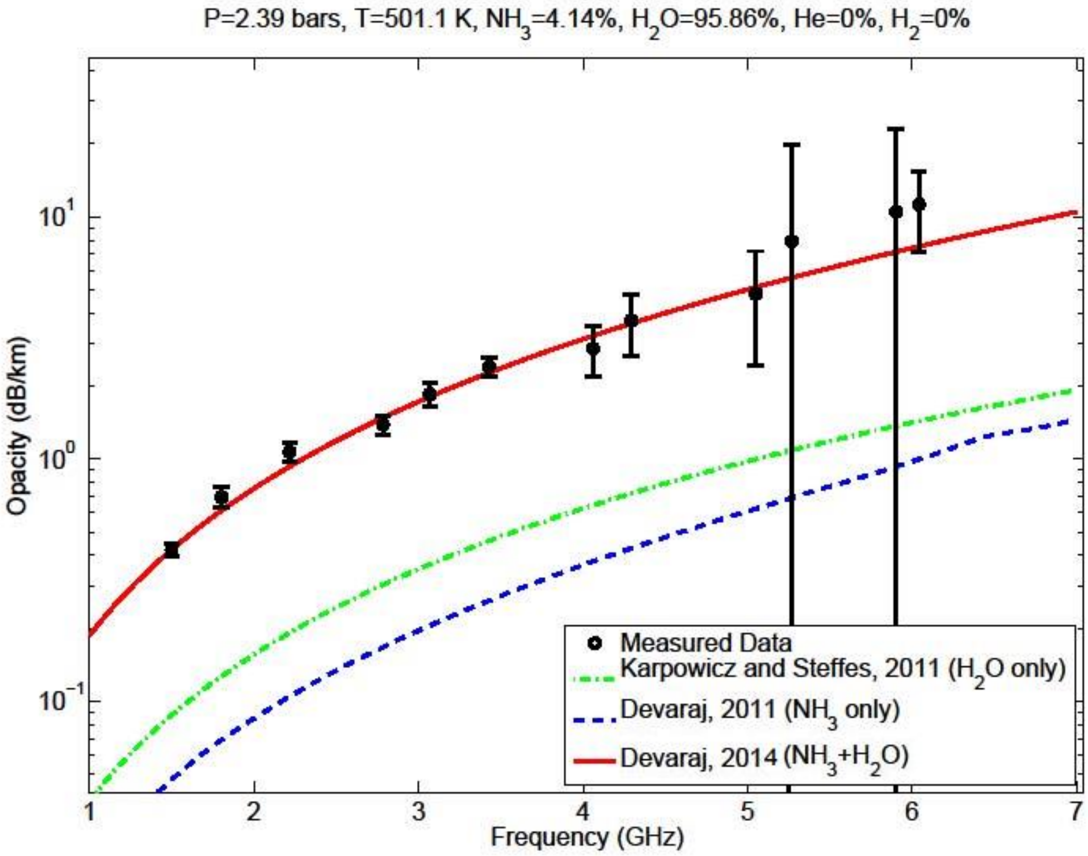

Figure 9: Opacity from 99 milliBars of ammonia vapor broadened by 2.3 Bars of water vapor.. Note that the aggregate absorption (top line) far exceeds the sum of the intrinsic opacity of each constituent, since the broadening of the ammonia spectrum by the water vapor is so strong. (Note that because of its low pressure, ammonia has little effect on the water vapor spectrum.) Displayed error bars are 2-sigma. (from Devaraj et al., 2014)

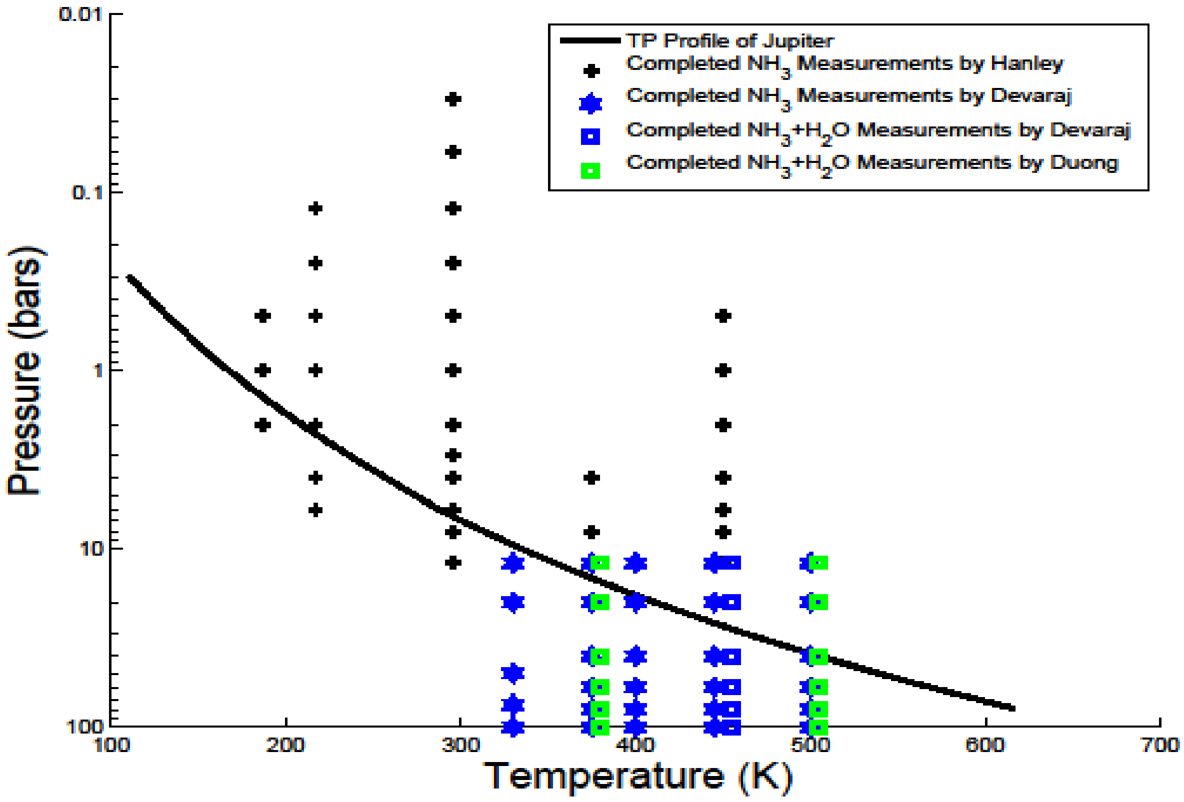

Figure 10: Summary of measurements made of ammonia under Jovian conditions, and those of ammonia and water vapor under simulated Jovian conditions. A plot of the nominal Jovian temperature-pressure profile is shown for reference.

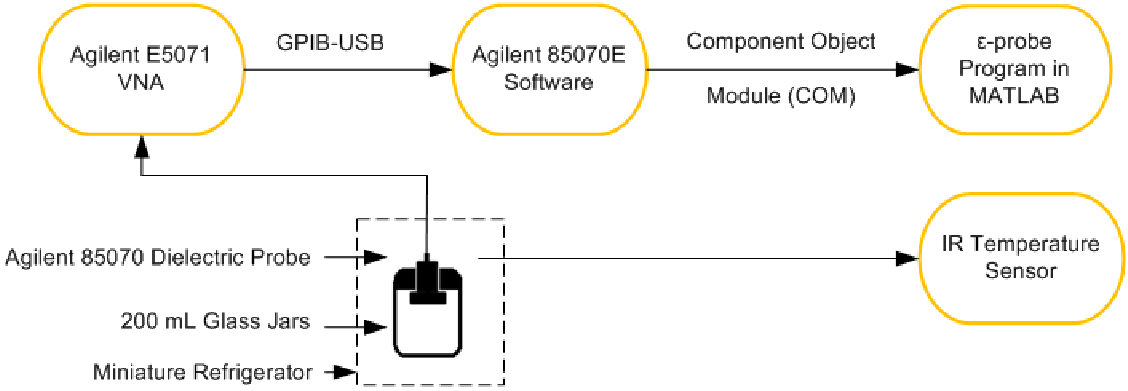

Figure 11: Block diagram of dielectric probe measurement system used for measurement of the complex dielectric properties of aqueous ammonia from 274-297 K (from Duong et al., 2014).

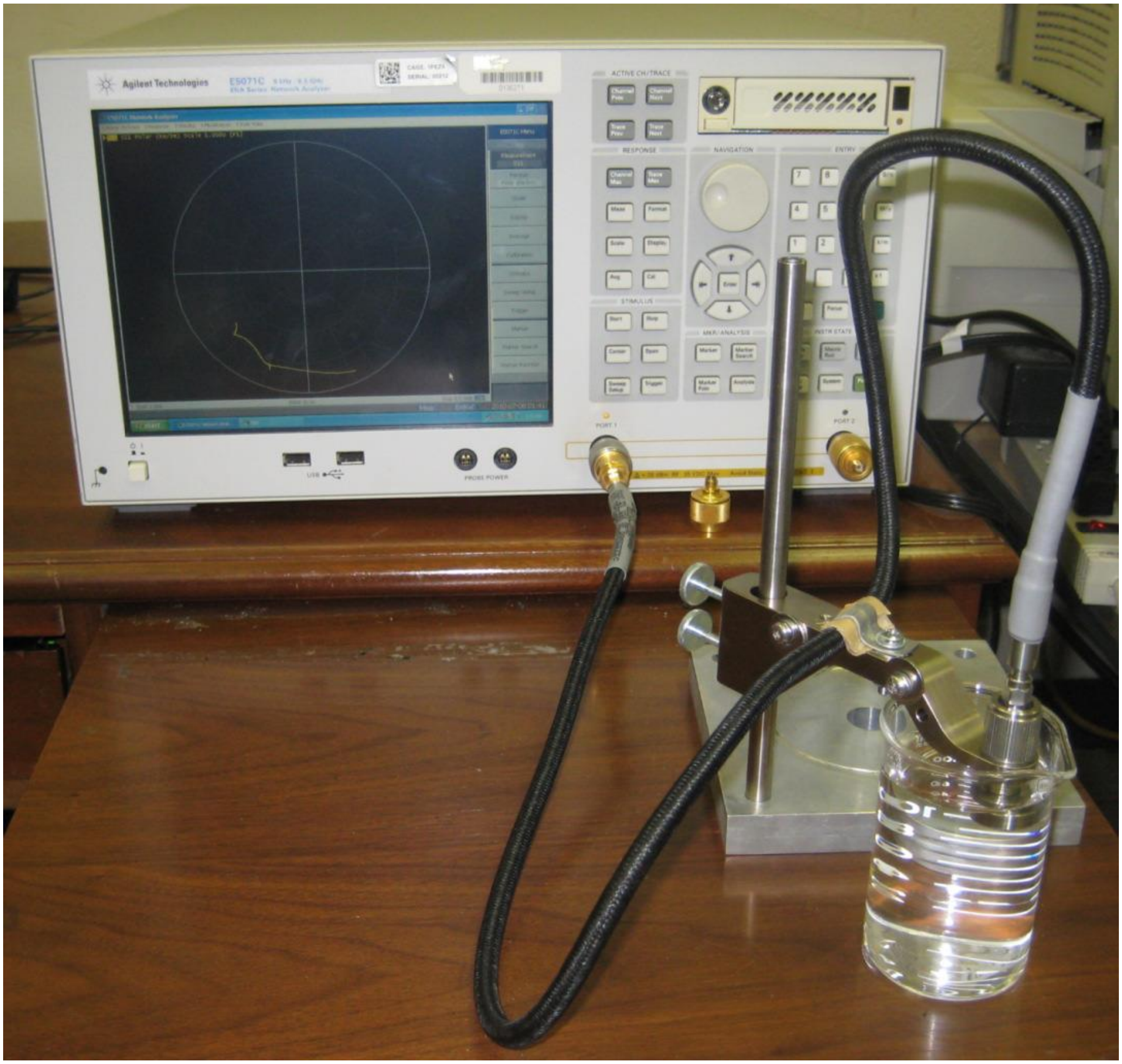

Figure 12: The dielectric probe measurement system as configured for room temperature measurements.

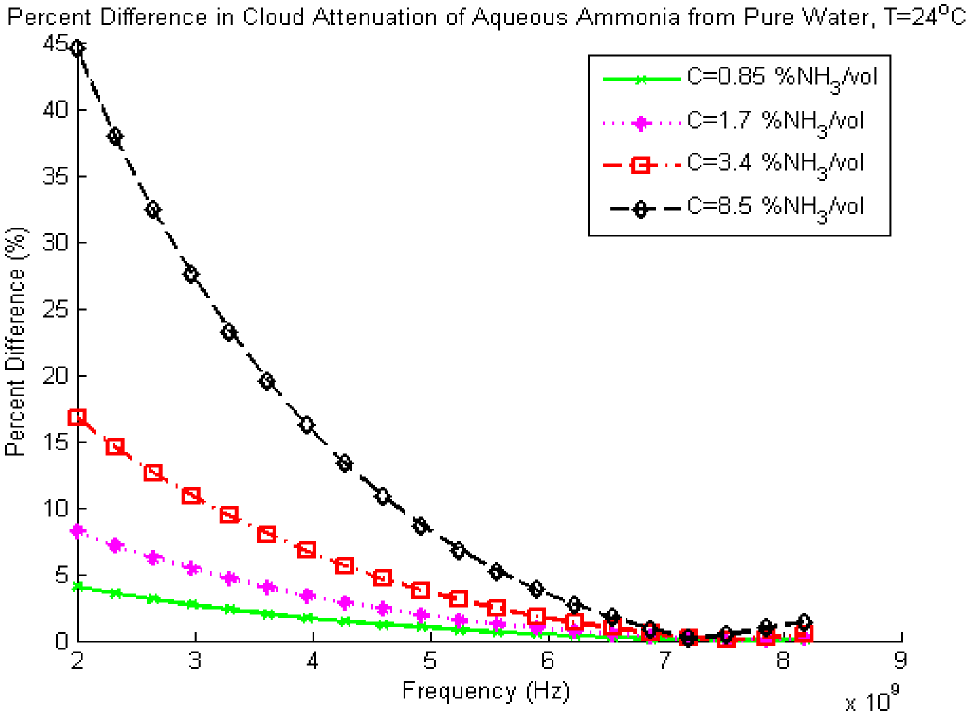

Figure 13: Percent difference in cloud opacity due to dissolved ammonia, developed using the new model for complex dielectric properties of aqueous ammonia (Duong et al., 2014) and assuming a fixed value for cloud bulk density.

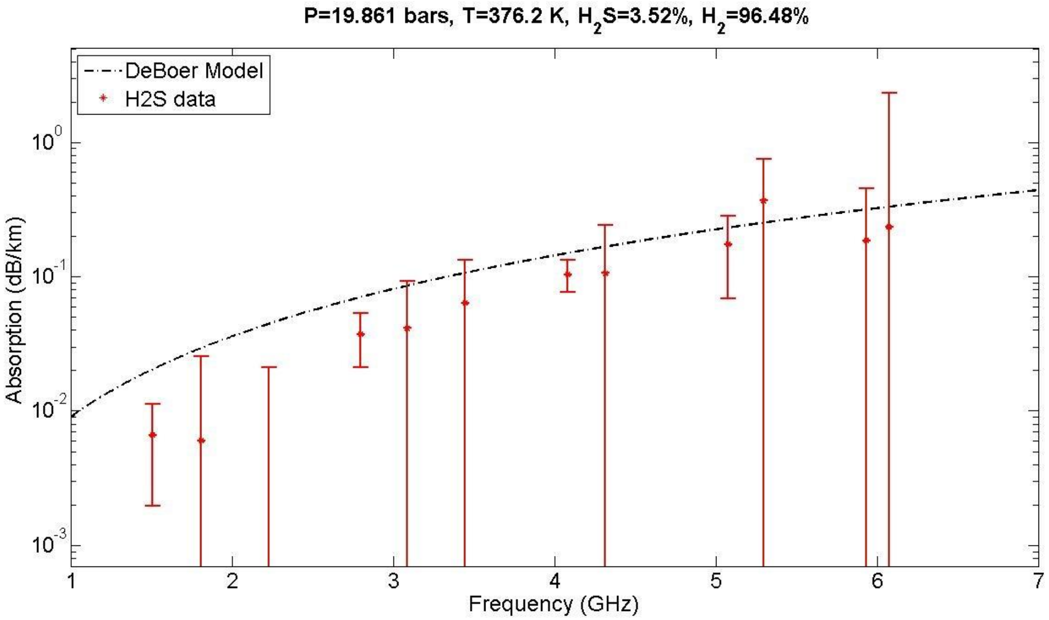

Figure 14: Measured centimeter-wavelength opacity of H_2S in a hydrogen atmosphere at 19.86 Bars total pressure and 376 K. Displayed error bars are 2-sigma. The modeled values (dashed line) are from DeBoer and Steffes (1994).

Temperature (K)	Pressure (bars)	Mixing ratio (% of H2S)	Frequency (GHz)	Measured Opacity (dB/km)	Measured 2σ (dB/km)	Modeled Opacity (dB/km)
377.56	0.7	100	1.507	-0.0018	0.0092	0.0020
377.56	0.7	100	1.812	-0.0330	0.0204	0.0028
377.56	0.7	100	2.229	-0.0873	0.0345	0.0043
377.56	0.7	100	2.799	-0.0115	0.0173	0.0068
377.56	0.7	100	3.085	-0.0169	0.0588	0.0082
377.56	0.7	100	3.447	-0.0392	0.0843	0.0103
377.56	0.7	100	4.083	0.0166	0.0360	0.0144
377.56	0.7	100	4.316	-0.0908	0.1923	0.0161
377.56	0.7	100	5.077	-0.0023	0.1018	0.0223
377.56	0.7	100	5.298	-0.2945	0.3997	0.0243
377.56	0.7	100	5.935	-0.0952	0.2242	0.0305
377.56	0.7	100	6.075	-0.7059	1.8561	0.0319
375.55	20	3.5	1.504	0.0063	0.0059	0.0208
375.55	20	3.5	1.809	-0.0139	0.0229	0.0301
375.55	20	3.5	2.225	-0.0533	0.0335	0.0455
375.55	20	3.5	2.794	0.0267	0.0164	0.0717
375.55	20	3.5	3.080	0.0171	0.0518	0.0872
375.55	20	3.5	3.440	0.0449	0.0715	0.1088
375.55	20	3.5	4.075	0.0807	0.0249	0.1527
375.55	20	3.5	4.307	0.0720	0.1379	0.1706
375.55	20	3.5	5.067	0.1598	0.1049	0.2361
375.55	20	3.5	5.288	0.2931	0.4627	0.2571
375.55	20	3.5	5.923	0.2926	0.3498	0.3227
375.605	20	3.5	6.064	-0.0440	2.1168	0.3380
376.16	20	3.5	1.504	0.0077	0.0047	0.0207
376.16	20	3.5	1.809	0.0042	0.0196	0.0299
376.16	20	3.5	2.225	-0.0142	0.0336	0.0452
376.16	20	3.5	2.794	0.0346	0.0162	0.0713
376.16	20	3.5	3.080	0.0373	0.0515	0.0867
376.21	20	3.5	3.440	0.0663	0.0689	0.1081
376.26	20	3.5	4.076	0.0943	0.0278	0.1516
376.26	20	3.5	4.308	0.1055	0.1359	0.1694
376.26	20	3.5	5.068	0.1519	0.1061	0.2345
376.26	20	3.5	5.288	0.2515	0.3880	0.2553
376.26	20	3.5	5.923	0.2732	0.2694	0.3205
376.31	20	3.5	6.064	0.1986	2.1091	0.3357

Table I: Measured centimeter-wavelength opacity of H_2S in a hydrogen atmosphere at 19.86 Bars total pressure and 376 K. Displayed error bars are 2-sigma. The modeled values are from DeBoer and Steffes (1994).