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Samuel Zinga April 9, 2019

Spectral Analysis of a Methylamine and Ozone Mixture: A Study to Aid in the Detection of Glycine Precursors in the Interstellar Medium

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Abstract

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By Samuel Zinga

The chemistry in the Interstellar Medium (ISM) is thought to be dominated by reactions on the surfaces of ice-grains that lead to the formation of larger complex-organic molecules. One molecule that is thought to be formed in this way is aminomethanol. Aminomethanol is a predicted precursor to interstellar glycine, the simplest amino acid, and as such its detection in the ISM would provide significant support for models of interstellar chemistry and their connection to prebiotic molecules. Detecting aminomethanol in the ISM is predicated on first collecting its rotational spectrum in the laboratory, which has been attempted by previous researchers in the Widicus Weaver lab. This spectrum was dense with unknown transitions, however, and a new experiment was designed to study the reactants, methylamine and ozone, as a potential source of these unassigned lines. The rotational spectrum of a methylamine and ozone mixture was thus collected, and analysis showed that this spectrum also had a number of unassigned lines. A list of potential products was evaluated as a potential source of these transitions, and none of these products were detected in the spectrum. Further analysis of this spectrum and additional spectra of pure methylamine indicated that these transitions are likely due to a vibrationally-excited state of methylamine, highlighting a gap in the current literature.

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In my culture, the saying, "it takes a village to raise a child" carries a great deal of weight. It highlights the realities of success—that no one can do it alone, that the wellbeing of one person is based on the support of others, and in my case, I owe my success to my village.

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Chapter 1

Introduction

1.1 Aminomethanol and the ISM

The first organic molecule detected in space was formaldehyde (CH_2O) in 1969 [1], and since then, over 200 molecules have been detected in the interstellar medium (ISM) [2]. The majority of these molecules have been detected using radio telescopes and come from a range of sources—interstellar clouds, circumstellar envelopes, and external galaxies [3]. Despite the variety of environments in which these molecules have been observed however, the majority of chemical interactions are thought to occur in one of two settings—in the gas phase or on the surface of icy grains. A model introduced by Garrod et al. in 2008 unified these previously competing ideas and highlighted the importance of gas-grain chemical interactions [4]. This model proposed that cosmic rays, upon striking ices composed largely of water, ammonia (NH_3), methanol (CH_3OH), and formaldehyde (CH_2O), induce radical formation. These radicals then recombine in exothermic processes to form larger complex organic molecules (COMs) which sublimate as the ices warm. Supporting evidence for this model is largely reliant on the detection of these COMs in interstellar environments

using pure rotational spectroscopy in the millimeter/submillimeter spectral range [5]. These searches are guided by laboratory experiments to collect the rotational spectra of the molecules in question. Some of the proposed COMs, however, are often unstable under normal terrestrial conditions, and their rotational spectra are difficult to obtain.

Aminomethanol ($HOCH_2NH_2$) is one terrestrially unstable COM predicted to form via the radical-radical reaction of NH_2 and CH_2OH on interstellar ices [4]. $HOCH_2NH_2$ is also an important prebiotic molecule with implications in astrobiology; the reaction of protonated $HOCH_2NH_2$ with formic acid (HCOOH) is a predicted source of the simplest amino acid glycine (HN_2CH_2COOH) [6–8]. Because both HCOOH and HN_2CH_2COOH have been detected in the ISM, detecting or failing to detect $HOCH_2NH_2$ would assist in determining the validity of this proposed mechanism [8, 9].

Previous work has been done in the Widicus Weaver lab by Hays [10] and McCabe [11] to create aminomethanol and collect its rotational spectrum. In these experiments, $HOCH_2NH_2$ was formed via the insertion of an excited oxygen atom ($O(^1D)$) into the C-H bond of methylamine (CH_3NH_2). The $O(^1D)$ was formed through the photolysis of ozone (O_3) with a UV excimer laser. The reaction was conducted in the gas phase, and subsequent products were cooled via a supersonic expansion into a vacuum chamber. The supersonic expansion was then probed by a millimeter/submillimeter beam in the range 140-302 GHz.

While aminomethanol is predicted to have been formed in these experiments, assigning its spectrum has proven difficult for a number of reasons. First, the laser photolysis method used by Hays and McCabe induced fragmentation of some reactants/products in the experiment. Additionally, any chemistry between the reactants in the experiment prior to photolysis likely produced molecules which added to the

complexity of the measured spectra. Together, these factors resulted in sixteen observed molecules (Table 1.1) beside the reactants and $HOCH_2NH_2$ [10, 11]. Finally, assigning the transitions of $HOCH_2NH_2$ was further complicated by the density and complexity of CH_3NH_2 transitions in this frequency range.

The experiments presented in this work were thus designed to collect the rotational spectra of pure CH_3NH_2 and a CH_3NH_2/O_3 mixture in an attempt to reduce the challenges associated with assigning the spectrum of $HOCH_2NH_2$.

Table 1.1: The products that Hays [10] and McCabe [11] detected in the CH_3NH_2 and O_3 laser photolysis project to measure the spectrum of $HOCH_2NH_2$.

$O_2(^1\Delta)$
NO
HNO
HCN
HNC
HO_2
NO_2
H_2CO
HOOH
H_2CN
HCNO
HNCO
CH_3O
CH_2NH
CH_3OH
$HCONH_2$

1.2 Methylamine Background

CH_3NH_2 is a prototypical non-rigid molecule with two large amplitude motions—the wagging of the NH_2 hydrogen atoms and the rotation of the CH_3 group (Figure 1.1). Because the barriers to this torsion and inversion are so low (8.73 kj/mol and 22.80 kj/mol respectively) [12], these motions in addition to hyperfine splitting caused by

the N nucleus make the microwave/(sub)millimeter spectrum of CH_3NH_2 complex and dense with transitions.

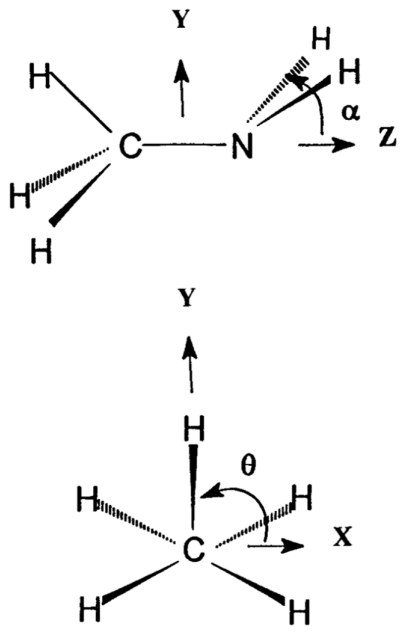

Figure 1.1: The inversion of the NH_2 group is shown on the top, and the torsion of the CH_3 group is shown on the bottom [13].

CH_3NH_2 is also astronomically significant as it was one of the early COMs detected in interstellar space. It was first detected in the ISM toward Sgr B2 [14, 15], then in a cometary sample [16], and most recently in a spiral galaxy [17]. CH_3NH_2 has also been predicted as a potential precursor to glycine, and experiments have shown that glycine can be formed when a water ice containing CH_3NH_2 and CO_2 is bombarded with either high energy electrons [18] or UV radiation [19, 20].

Owing to its relevance in the field of astrochemistry and its complex spectral signatures, CH_3NH_2 has been studied in depth by spectroscopists. The first microwave study of CH_3NH_2 was conducted in 1947 by Hershberger et al. [21], and they measured ten rotational transitions corresponding to the vibrational ground state of CH_3NH_2. Subsequent studies by Shimoda et al. [22, 23] and Lide [24]

extended this spectrum to cover the range 11-38 GHz. In 1956 Itoh et al. [25] introduced a Hamiltonian capable of dealing with semiasymmetric internal rotors, and between 1957 and 1971 a number of studies were published that focused on the effect of internal motion on the spectrum of CH_3NH_2, extending the body of work to 60 GHz [26–29]. In 1978, Kreglewski introduced a new rotational Hamiltonian that addressed the torsional effect of the CH_3 group as well as the inversion of the NH_2 group [30]. Hougen and colleagues further developed this Hamiltonian and used it to assign the spectrum of CH_3NH_2 to 90 GHz [31, 32]. Following this work by Hougen and colleagues, multiple papers were released extending the microwave spectrum up to 500 GHz [33–35] until it was finally extended to 2.6 THz in 2014 by Motiyenko et al. [36].

The vibrational spectrum of CH_3NH_2 has also been studied extensively. The far-infrared spectrum of CH_3NH_2 has been investigated both theoretically and experimentally, and a number of studies have focused on the torsion and inversion of the molecule [13, 37–55]. Specific vibrational modes such as the C-H, N-H, and C-N stretches have also been studied in depth [56–59].

Although the vibrational and ground-state rotational spectra of CH_3NH_2 have been studied extensively, little work has been done to measure the rotational spectrum of excited states of CH_3NH_2. The few studies that have been conducted by Ohashi and colleagues and Gulaczyk et al. have measured the first and second torsionally-excited states, but their data are limited to 90 GHz or focus on the IR spectral region [32, 43, 46, 48, 60]. Additionally, the studies extending into higher frequency ranges have only measured and assigned the transitions of the ground vibrational state of methylamine [35, 36].

The significance of this lack of information is twofold: assigning the spectra of unmeasured molecules, such as $HOCH_2NH_2$, is obstructed when CH_3NH_2 is a reactant

and interstellar detections of these vibrationally-excited transitions is hindered by the lack of laboratory data. Hence it is both necessary and pressing to measure and assign the rotational spectrum of vibrationally excited molecules such as CH_3NH_2.

Chapter 2

Methods

2.1 General Setup

To study the reaction of O_3 and CH_3NH_2 and record the rotational spectrum of CH_3NH_2, two spectrometers were constructed. Both spectrometers operated on a general design: a metal flow cell evacuated to pressures in the mTorr range with a millimeter/submillimeter light source and detector to probe the rotational transitions of molecules. For both spectrometers, a metal cell was fitted with copper gaskets, and silicone o-rings to resist corrosion and degradation due to the reactants. Both flow cells had attachments where gaseous sample could be introduced into the chamber and the pressure of the cell could be monitored during experiments. A schematic of this experimental setup is shown in Figure 2.1. Note that the second spectrometer, used to measure the spectrum of CH_3NH_2 did not have an input gas line for O_3 and was aligned without focusing lenses.

Both cells were pumped down using a high vacuum rotary pump (Edwards E2M-30) capable of evacuating the flow cells to approximately 10-20 mTorr. O_3 was produced by running O_2 (NexAir ultra-high purity) into an ozone generator (Pacific

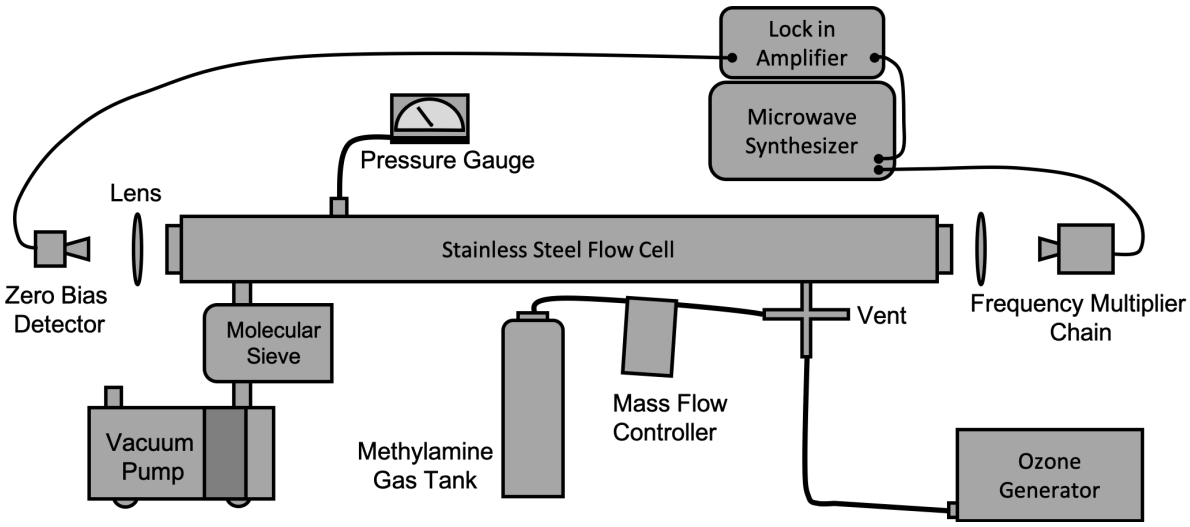

Figure 2.1: Experimental setup. A stainless steel flow cell is pumped down using a rough pump. O_3 and CH_3NH_2 are introduced through individual gas lines that connect at a junction before entering the flow cell. A synthesizer, lock in amplifier, and multiplier chain are used to produce the millimeter light that is detected with a zero bias detector after interacting with the sample.

Ozone L11) at 6-9 psi. The generator was run between 4 and 4.5 volts, with a flow rate of 10 scfh, and an O_3 output of 70%. The resulting $\sim$1% O_3 in O_2 mixture was subsequently mixed with the CH_3NH_2 gas line before entering the flow cell. This mixing scheme contained a line to vent the chamber and two one-way Swagelok checks to prevent back-flow. The mixing scheme can be seen in Figure 2.2. CH_3NH_2 (AirGas, 99%) was introduced directly into the flow cell or into the aforementioned mixing system at a flow rate of 17 sccm via a mass flow controller (MKS Instruments type 247). Due to the safety concerns of working with O_3 and CH_3NH_2, a molecular sieve (Sigma-Aldrich 4$\mathring{A}$ beads) was also added between the flow cell and the vacuum pump to trap excess molecules of both compounds.

An analog signal generator (Agilent Technologies, E8257D PSG with options 1EA, UNU, 550, and UNT) was used to generate light at a base frequency from 20-50 GHz before it was multiplied up by one of two Virginia Diode Inc. (VDI) multiplier chains (Virginia Diodes Inc., AMC-S268 or AMC-S197C). By adding dif-

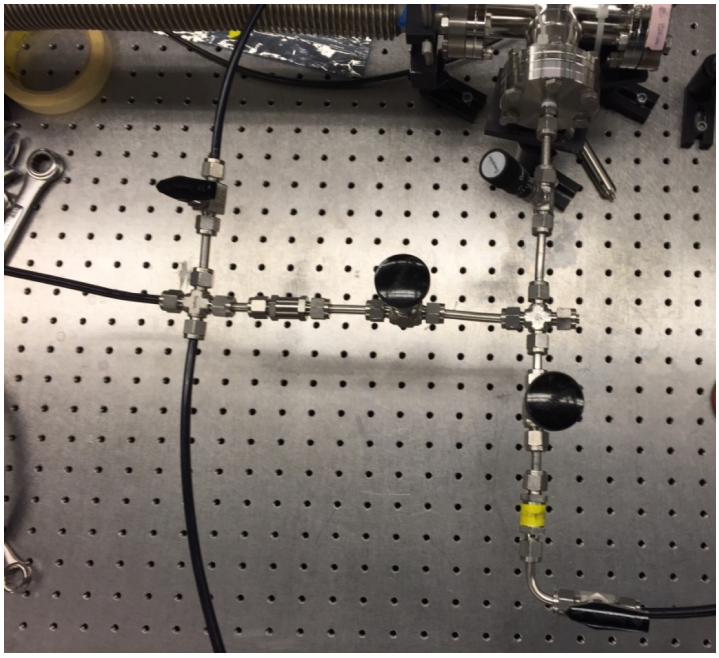

Figure 2.2: A photograph of the mixing scheme used to introduce O_3 and CH_3NH_2 into the flow cell. The left junction contains a line to input the O_2:O_3 mixture and a line to vent the system. This section is connected to the right junction which contains a connection to the CH_3NH_2 line and the flow cell. Two one-way Swagelok checks are included in the setup to prevent back-flow.

ferent multiplier components to the VDI WR10 base amplifier system, various bands corresponding to different frequency ranges could be reached. A diagram of these systems and their frequency ranges is shown in Figure 2.3.

Once the light was multiplied up to the desired frequency, it was directed into the flow cell through a sapphire window and collected on the other side of the cell after passing through a second sapphire window. For the lower frequency ranges up through Band 6 (215-335 GHz), zero bias detectors (WR5.1 S3-23 and WR3.4 R3) were used to collect the emerging light; for frequency ranges in Band 7 (280-460 GHz) and higher, a hot electron bolometer (QMC Ltd., QFI/XBI) was used instead. Signals from the detectors were digitized using a National Instruments digitizer card (PCI-5124) and sent to a computer which controlled the spectrometer via a custom-built data acquisition routine written in LabWindows. To achieve a greater signal

to noise ratio (SNR), a lock-in amplifier (Stanford Research Systems, SR830 DSP) was also used in these experiments. By sending a reference signal from the signal generator to the lock-in amplifier and connecting the detector to the lock-in, 2f, phase-sensitive detection could be achieved, effectively attenuating any out-of-phase noise. In both sets of experiments, the lock-in amplifier was sent a reference frequency of 1.00 kHz with a modulation depth of 75.00 KHz. Additionally, the phase was offset by 154° and the time constant set to 10 ms.

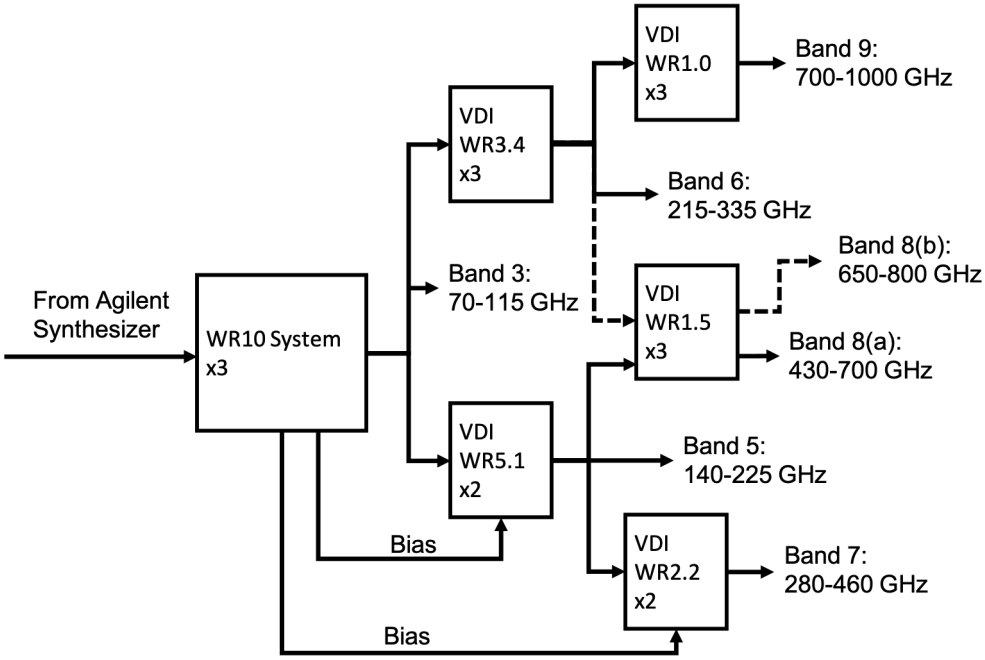

Figure 2.3: Diagram of the VDI-AMC-S268 and VDI-AMC-S197C multiplier chain bands and their corresponding frequency ranges. Adapted from Virginia Diodes Inc. user guide [61].

2.2 Methylamine and Ozone Setup

The spectrometer used to investigate the O_3 and CH_3NH_2 mixture, had a path length of 114.3 cm and an inner diameter of approximately 40 mm. Additionally, two Teflon focusing lenses with focal lengths of 10 cm were used to collimate the submilimeter

light entering the cell and focus the light exiting the cell onto the detector.

During these experiments, the flow cell was constantly open to the pump which produced a baseline pressure of 20 mTorr before the introduction of any sample. The CH_3NH_2 gas line was then opened and throttled with a needle valve to stabilize the total chamber pressure at 50 mTorr. Finally, the gas line for the O_2:O_3 mixture was opened to bring the cell pressure to about 100 mTorr. While the partial pressure of CH_3NH_2 was held constant with the use of the mass flow controller, the partial pressure of the O_2:O_3 mixture varied such that the total pressure was maintained between 90-120 mTorr. The spectra for this mixture were collected at room temperature in Band 5 (140-225 GHz) and part of Band 6 (225-316.7 GHz).

2.3 Methylamine Setup

To collect the spectrum of CH_3NH_2, a spectrometer with a path length of $\sim$200 cm and an inner diameter of $\sim$40 mm was constructed. Initial power scans indicated that there was no substantial increase in power when using focusing lenses, so the spectrometer was designed without focusing lenses. When evacuated, the cell reached a pressure of $\sim$13 mTorr. During scans, the chamber was filled to a pressure of 65 mTorr with CH_3NH_2 and sealed off to both the vacuum and the sample line. The pressure inside the cell increased over the course of scanning, so the pressure was effectively maintained between 65 mTorr and 120 mTorr during data collection. Spectra for CH_3NH_2 were collected at room temperature in parts of Band 5 (140-225 GHz).

Chapter 3

Methylamine and Ozone Results

The spectra for the CH_3NH_2 and O_3 gas mixture were recorded from 140-316.7 GHz. Band 5 (140-220 GHz) was divided into two scans and Band 6 (220-316.7 GHz) was divided into four scans. A sample of these scans showing the second half of Band 5 is shown in Figure 3.1. A smaller window of this scan (186274-186424 MHz) is shown in Figure 3.2.

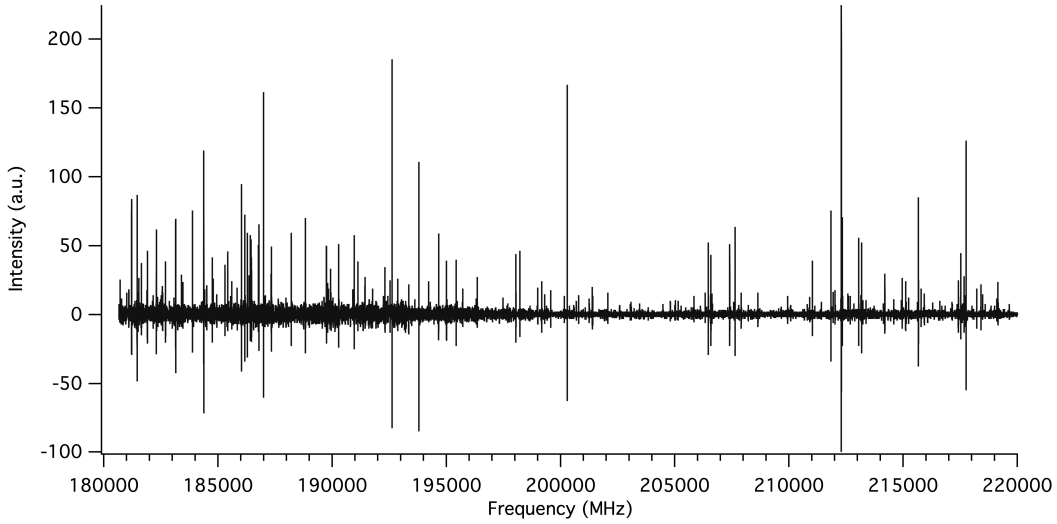

Figure 3.1: A millimeter/submillimeter scan of the O_3 and CH_3NH_2 mixture in the range 180-220 GHz.

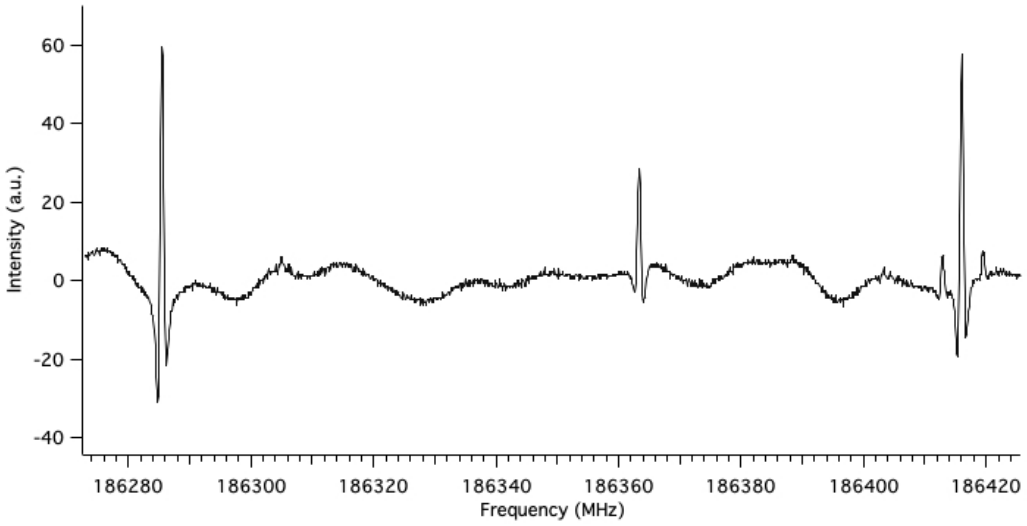

Figure 3.2: A millimeter/submillimeter scan of the O_3 and CH_3NH_2 mixture in the range 186274-186424 MHz. Five clear transitions can be seen at this scale.

In order to clearly isolate transitions corresponding to new products, the known transitions of CH_3NH_2, O_3, and O_2 first had to be identified. Several files containing the measured and predicted rotational transitions of the reactants were downloaded from the JPL Molecular Spectroscopy Database [62] and the Cologne Database for Molecular Spectroscopy [63, 64]. Among these catalog files were files corresponding to various vibrationally-excited states and isotopologues of O_3 and O_2. A list of the different species which were investigated can be found in Table 3.1.

The catalog files were visualized using Drouin's SubMillimeter Analysis Program (SMAP) and overlaid onto the experimental data. With the catalog files indicating the transitions that corresponded to the reactants, it was evident that the spectra contained additional, unassigned transitions (u-lines). Figure 3.3 illustrates how these u-lines were identified—the catalog files were used to demarcate known transitions in color, and any transitions not marked by the catalog were considered u-lines. By systematically parsing through the spectra, several u-lines were found. A list of these u-lines was assembled by choosing the unassigned transitions with a SNR

Table 3.1: These ground states, vibrationally-excited states, and isotopologues of CH_3NH_2, O_3, and O_2 were investigated as potential reactants in the CH_3NH_2 and O_3 mixture.

CH_3NH_2
O_3
O_3, ν_2
$^{18}O_2$
$^{16}O_2$
$^{17}O^{16}O$
$^{18}O^{16}O$
$O_2\,^1\Delta_g$
O_2, v=1
O_3, $2\,\nu_2$
O_3 asymmetric ^{18}O, ν_2=1
O_3 symmetric ^{18}O, ν_2=1
O_3, asymmetric ^{17}O isotope
O_3, symmetric ^{17}O isotope
O_3, asymmetric ^{18}O isotope
O_3, symmetric ^{18}O isotope
O_3, ν_1, ν_3
O_3, Coriolis coupled states $\nu_1 + \nu_2$ and $\nu_2 + \nu_3$

greater than approximately ten as a preliminary pass. A list of these transitions can be found in Appendix A.

The observed u-lines were initially suspected to be products forming as a result of the reaction of CH_3NH_2 with O_3. Because Hays and McCabe had observed sixteen molecules in their experiments that were not one of the reactants or the desired product $HOCH_2NH_2$, these were the first molecules considered. Nitromethane (CH_3NO_2) and nitrosomethane (CH_3NO) were also suspected as potential products from the reaction of CH_3NH_2 and O_3. The fragmentation of CH_3NH_2 and the subsequent reaction of these fragments with each other and with reactants was also considered as a possible formation route to new products. Based on this reasoning, various small molecule configurations that contained the atoms H, C, N, and/or O were considered as potential products. Finally, the recombination of any of these molecules to

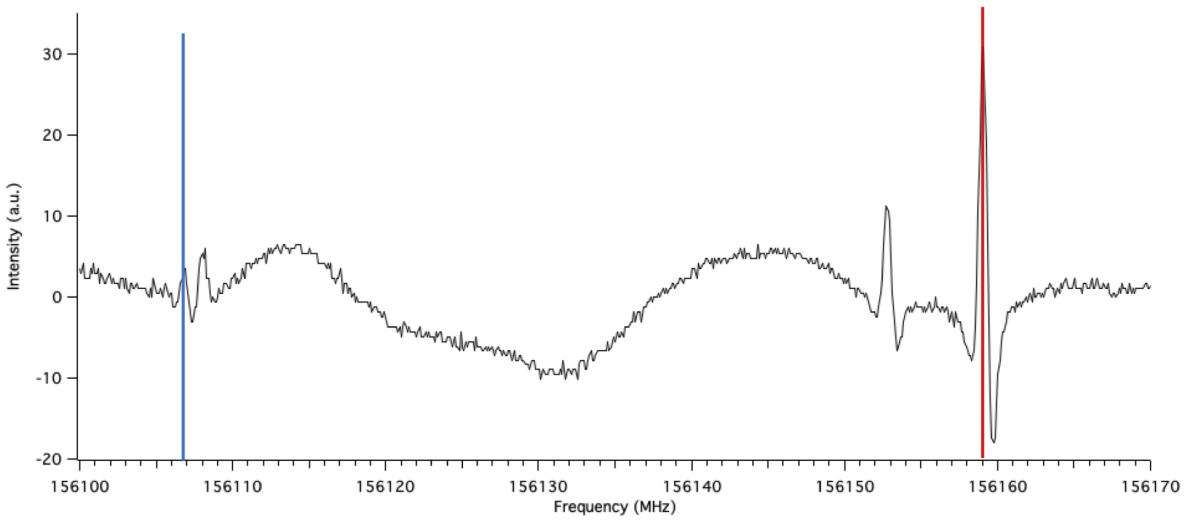

Figure 3.3: A scan of the O_3 and CH_3NH_2 reaction mixture illustrating how unassigned transitions were identified. The black trace shows the experimental data that were collected with the spectrometer. The blue line indicates a O_3 transition at 156106.8 MHz and the red line indicates a CH_3NH_2 transition at 156159 MHz. The transition at 156153 MHz was classified as a u-line.

produce larger structures was also considered. On the whole, this resulted in twenty potential molecules. A list of these proposed products is given in Table 3.2 and their structures are given in Figure 3.4.

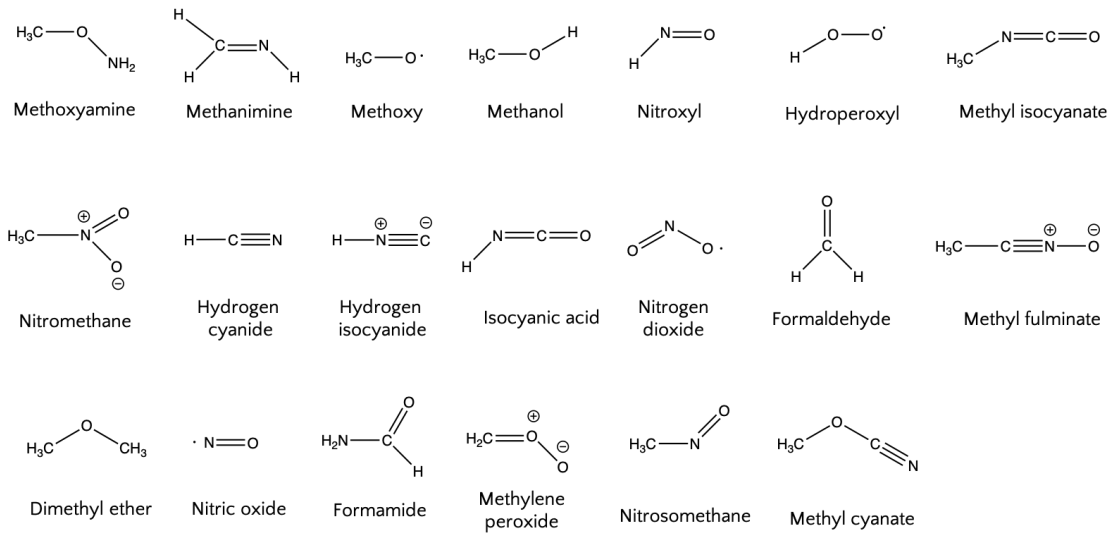

Figure 3.4: Structures of the potential products which may have formed in the O_3 and CH_3NH_2 mixture.

Table 3.2: A list of the potential products that were investigated as potential sources of unassigned transitions in the O_3 and CH_3NH_2 mixture. None of these molecules were detected in the spectra.

Methanimine	CH_2NH
Methoxy	CH_3O
Methanol	CH_3OH
Nitroxyl	HNO
Hydroperoxyl	HOO
Dimethyl ether	CH_3OCH_3
Nitric oxide	NO
Hydrogen cyanide	HCN
Hydrogen isocyanide	HNC
Isocyanic acid	HNCO
Nitrogen dioxide	NO_2
Formaldehyde	H_2CO
Formamide	$HCONH_2$
Methylene peroxide	H_2COO
Methyl fulminate	CH_3CNO
Methyl cyanate	CH_3OCN
Methyl isocyanate	CH_3NCO
Methoxyamine	CH_3ONH_2
Nitromethane	CH_3NO_2
Nitrosomethane	CH_3NO

For most of the proposed products, catalog files were downloaded from the JPL Molecular Spectroscopy Database and the Cologne Database for Molecular Spectroscopy. A catalog for CH_3NO_2 was obtained from recent work by Illyushin et al. [65].

For methoxyamine (CH_3ONH_2) and CH_3NO, however, catalog files had to be generated based on the published rotational constants and dipole moments of the molecules. The rotational spectrum of CH_3ONH_2 was recently recorded up to 480 GHz by Kolesniková et al. [66], and in their study, the A and E states of CH_3ONH_2 were fitted separately using Picket's SPFIT and SPCAT suite of molecular fitting programs [67]. The microwave spectrum of CH_3NO has also been studied and assigned up to 37 GHz by Coffrey et al. and provides A, B, and C constants as well

as dipole moments for the molecule [68].

For both (CH_3ONH_2) and CH_3NO, estimates of the rotational partition function at 300 K were calculated using an approximation for asymmetric tops as described by Gordy & Cook [69]. The equation is given by Equation 3.1 where T is temperature in K; A, B, and C, are rotational constants in MHz; and σ is the symmetry number.

$$Q_r = 5.3311 \times 10^6 (T^3/(ABC))^{1/2}/\sigma \tag{3.1}$$

Using the constants published in these papers and the calculated partition functions, catalog files were generated for CH_3NO and the A state of CH_3ONH_2 in SPCAT. The program outputted recalculated values for the partition functions, and these new values were updated in the files before rerunning them to obtain final catalog files. The program was run using a standard Watson S-reduced Hamiltonian [70]. The constants used to generate the catalogs are given in Table 3.3 and the catalog files can be found in Appendix B and Appendix C.

Using the downloaded and generated catalog files for the predicted products, expected rotational transitions for each product were compared to the experimental data. The strongest transitions for each of the products were investigated first followed by a systematic check at the frequency of each of the u-lines that had been compiled. Using this methodology, none of the proposed products were determined to be a likely source of the unassigned transitions.

Table 3.3: Fitting constants

	CH_3ONH_2 A species	CH_3NO
A (MHz)	42 487.2256 (67)	60 938.2 (99)
B (MHz)	10 049.6972 (13)	11 459.32 (38)
C (MHz)	8962.8910 (14)	10 246.58 (37)
D_J (kHz)	9.7862 (55)	...
D_{JK} (kHz)	3.575 (20)	...
D_K (kHz)	471.46 (38)	...
d_1 (kHz)	-1.5834 (15)	...
d_2 (kHz)	0.33027 (33)	...
H_J (Hz)	0.0200 (78)	...
H_{JK} (Hz)	-0.163 (35)	...
H_{KJ} (Hz)	1.88 (88)	...
H_K (Hz)	124.4 (92)	...
h_1 (Hz)	0.0030 (15)	...
h_2 (Hz)	-0.0039 (11)	...
h_3 (Hz)	-0.00153 (33)	...
μ_a (D)	0.5	2.240
μ_b (D)	0.1	0.522
μ_c (D)	0.0	0.0
Q_r initial	14160.18668	10355.83230
Q_r returned	3328.9716	10361.3984

Chapter 4

Methylamine Results

A low-resolution power scan was collected in Band 5 (140-225 GHz) prior to collecting experimental data (Figure 4.1). Based on the results of this scan, it was evident that there were large discrepancies in the amount of power reaching the detector at various frequencies. This was in part due to the variable amount of power produced by the synthesizer and multiplier chains at each frequency, but also by the absorption of the sapphire windows at different frequencies. Because of the large differences in power over different regions, scans were collected in the regions with the highest amounts of power. Spectra for CH_3NH_2 were thus collected in the ranges 162-176 GHz, 190-202 GHz, and 211-225 GHz.

A sample of the CH_3NH_2 spectrum from 162-176 GHz is shown in Figure 4.2. The black trace shows the spectrum that was measured and the vertical red lines indicate the center frequencies for the previously observed unassigned lines. In Figure 4.2, it is clear that some of the u-lines observed in the spectrum of the CH_3NH_2 and O_3 mixture were not observed in the spectrum of pure CH_3NH_2. After comparing the u-lines to the three scans from 162-176 GHz, 190-202 GHz, and 211-225 GHz, most of the u-lines in fact, were not observed. For example, of the 50 u-lines observed in

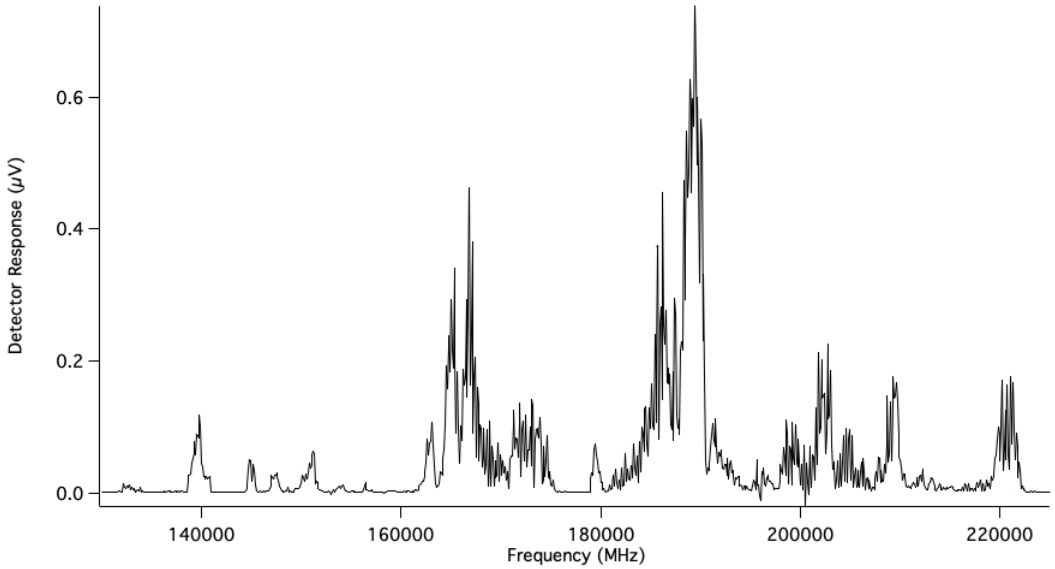

Figure 4.1: A powerscan of the spectrometer over the Band 5 (140-225 GHz) range.

CH_3NH_2 and O_3 mixture between 162-176 GHz, only 13 were definitively observed, and of the 21 u-lines observed between 190-202 GHz, only 7 were observed in these scans. One of the detections that was made in this region is shown in Figure 4.3

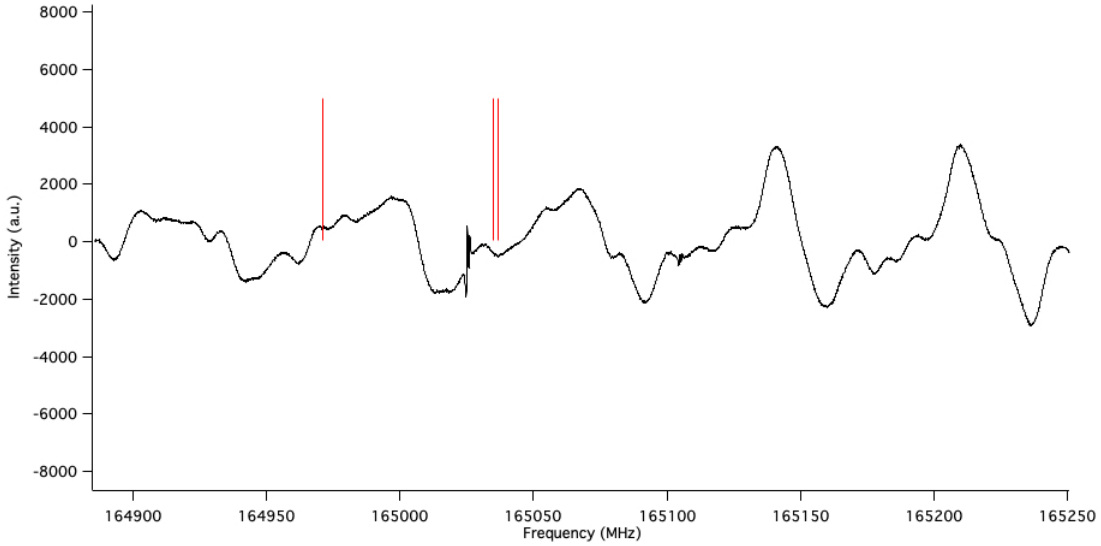

Figure 4.2: A sample region from 164900-165250 MHz in the scan of CH_3NH_2 from 162-176 GHz is shown as the black trace. The plot of the data in this region shows the rolling baseline. The vertical red lines are frequencies at which u-lines were previously observed.

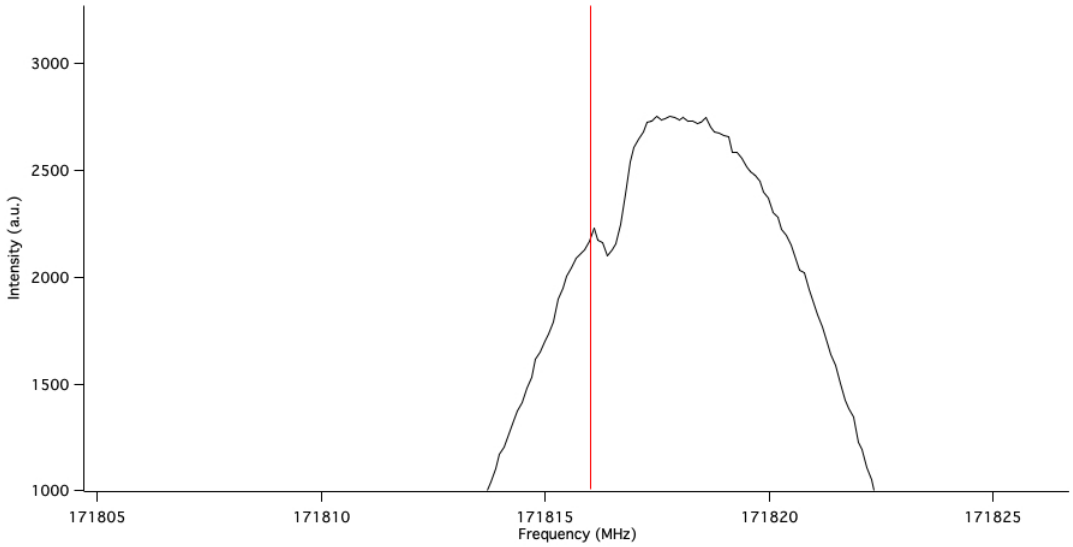

Figure 4.3: A magnified image of the scan of CH_3NH_2 showing a detected u-line.

Chapter 5

Discussion and Conclusion

The results of both experiments (measuring the millimeter/submillimeter spectra of a CH_3NH_2/O_3 mixture and pure CH_3NH_2) provide some evidence that the observed u-lines are due to the transitions of vibrationally-excited CH_3NH_2.

Although the majority of the u-lines tested in the ranges 162-176 GHz, 190-202 GHz, and 211-225 GHz were not observed in the CH_3NH_2 data, it is still possible that they correspond to transitions of CH_3NH_2. Given optimum conditions, it is likely that many more of the u-lines can be detected in the spectrum of pure CH_3NH_2. As evident in Figure 4.3, even the observed u-lines were relatively weak and difficult to detect. As these were some of the most intense u-lines previously observed, the weaker u-lines were probably too weak to detect with the given experimental parameters. This could be due to one or a combination of several reasons. First, the spectra in these experiments contained a very prominent rolling baseline—a manifestation of a standing wave in the spectrometer. While completely removing a standing wave is difficult, the effects of such a standing wave can be minimized by manipulating the placement of the source and detector, physically changing the dimensions of the chamber, or smoothing and taking the derivative of the data. This third approach

was attempted, but no additional u-lines were definitively brought up above the noise. Additionally, the pressure within the static cell gradually rose during scanning, requiring that the scan be paused and the cell be reset to 65 mTorr before continuing scans. During this process, the cell was reset when the pressure reached 120 mTorr, but it is possible that at this pressure, the weaker lines are pressure broadened and rendered undetectable. One solution to this problem is to reset the pressure more frequently during scans, or implement fittings that allow for a tighter sealed static cell.

Although many of the u-lines were not detected in the spectrum of pure CH_3NH_2, the few u-line transitions which were almost certainly correspond to some transition of CH_3NH_2. When the center frequencies of these and the other unassigned transitions are compared to the literature on CH_3NH_2 in the ground state however, it is clear that the u-lines do not belong to the ground state of CH_3NH_2. This is illustrated in Figure 5.1. Here, u-line frequencies are compared to the expected ground state frequencies for CH_3NH_2 based on the experiments and fit of Ilyushin et al.[35]. The u-lines with the strongest intensity were compared to the ground state transitions that were immediately higher in frequency and lower in frequency. As is evident from the comparison in Table 5.1, the u-lines do not correspond to the ground state of CH_3NH_2.

While still unconfirmed, it is very likely that the unassigned transitions observed in this work correspond to the rotational spectrum of vibrationally-excited CH_3NH_2. Because the barriers to methylamine torsion and inversion are so low (8.73 kj/mol and 22.80 kj/mol respectively) [12], it is feasible that excited vibrational states of CH_3NH_2 could exist at room temperature. Additionally, throughout the measured spectra but especially at the lower frequency ranges, some of the unassigned transitions displayed characteristic hyperfine splitting for CH_3NH_2 as described by Motiyenko et al. [36].

Table 5.1: A comparison of the unassigned transition frequencies of CH_3NH_2 observed in this work and the predicted frequencies based on the spectrum collected by Ilyushin et al. from 1-500 GHz. The frequencies on the left represent a sample of the strongest unassigned transitions that were found in the CH_3NH_2 and O_3 spectra. The frequencies on the right are the calculated transitions for the ground state of CH_3NH_2 that are nearest the corresponding u-line. The frequencies are reported in MHz and the work by Illyushin et al. is reported with uncertainties.

This work	Ilyushin et al.[35]
140 245	140 143.577 (0.012)
...	140 254.570 (0.010)
141 451	141 415.946 (0.014)
...	141 478.620 (0.460)
156 272	156 159.130 (0.014)
...	156 681.635 (0.013)
162 272	161 194.432 (0.016)
...	162 314.378 (0.050)
163 647	163 242.638 (0.017)
...	164 303.480 (0.015)
178 055	178 011.303 (0.009)
...	178 199.016 (0.655)
181 645	181 521.718 (0.025)
...	182 071.953 (0.017)
186 030	185 425.290 (0.013)
...	186 769.707 (0.014)
188 416	188 256.359 (0.030)
...	188 830.715 (0.016)
192 874	192 622.389 (0.016)
...	192 997.356 (0.012)
206 629	206 584.764 (0.020)
...	207 317.291 (0.024)
217 521	217 152.337 (0.023)
...	217 625.145 (0.027)

This pattern of a doublet with an approximately 2:1 ratio of intensities indicates that the transitions belong to CH_3NH_2 or another species with this kind of hyperfine splitting. An example of the pattern can be seen in Figure 5.1.

Although the possibility remains that the observed u-lines are due to some contaminant in the spectrometer, this explanation is unlikely. Additionally, because many of the transitions show characteristic hyperfine splitting, any potential con-

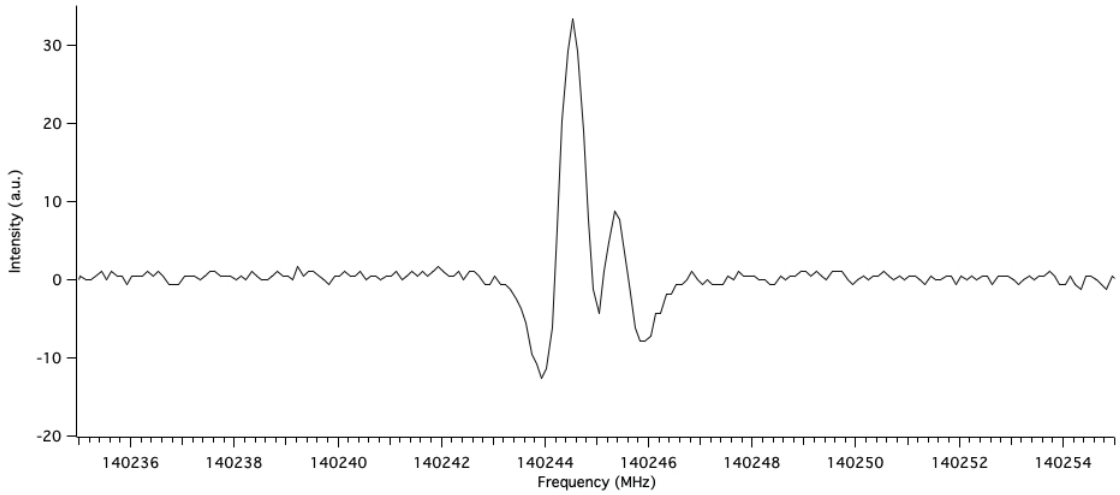

Figure 5.1: A sample unassigned transition showing the characteristic hyperfine splitting that Motiyenko et al. report in their study of CH_3NH_2 [36]

taminant is limited to a species that shows this same spectral signature. It must also be noted, however, that the methods used to rule out potential products do not absolutely disprove their presence in the flow cell. The possibility remains that one of the products was in fact produced in the CH_3NH_2 and O_3 mixture if the product was produced in low enough concentrations or if the catalog files used in the comparisons are inaccurate. The first situation applies to all products while the seconds is of particular concern for nitrosomethane CH_3NO because the fit used to generate the catalog file was based only on the A, B, and C rotational constants and not higher-order distortion constants. More studies are needed on this molecule before it can be definitively ruled out as a potential product.

While a definitive assignment was not made regarding the unassigned transitions in the measured spectra, significant progress was made to eliminate other potential molecular sources and to propose that the u-lines may arise from vibrationally-excited CH_3NH_2. Further work is needed to assign these transitions, and this will likely require a fitting program like ERHAM [71] designed to better handle internal motion and hyperfine splitting of CH_3NH_2.

Appendix A

Appendix A

A list of the center frequencies for the unassigned transitions observed in the spectrum of the CH_3NH_2 and O_3 mixture.

140077 140095 140245 140505 141451 141837 141845 142101 142736 145282 147124 147681 149644 150086 151977 152247 152260 153063 153395 153396 153399 153446 153448 153587 153715 153876 154735 154737 154781 155645 155864 156040 156108 156153 156272 156357 156358 156359 156367 156377 156378 156425 156601 156603 156929 157018 157077 157082 157217 157401 158222 158478 158611 159051 159309 159605 159607 159737 159842 159857 160263 160407 160565 160785 160923 161561 161787 161800 161802 161862 161990 162577 162672 162711 162723 162749 162784 163352 163609 163610 163647 163920 163920 164385 164757 164971 165035 165037 165763 166046 166090 166214 166612 167232 167526 167528 167734 167735 169296 169546 171588 171816 171923 172460 172601 173023 173073 173658 173661 174118 174330 174481 174678 174758 174861 174950 174952 175073 175230 175397 175407 175913 176202 176279 176687 176692 176693 176742 176812 176869 177593 178055 178234 178854 178882 178936 179199 179240 179359 179930 179970 179971 179991 181007 181097 181538 181645 181658 181890 181910 182160 182190 182355 182365

182446 182514 182554 182577 182704 182866 182994 183398 183446 184275 184421
184509 184850 184943 185287 185289 185304 185601 185762 185840 186030 186176
186285 186363 186412 188416 186419 186439 186448 186453 186461 186465 186468
186470 186499 186512 186557 186937 187074 187979 188336 189005 189007 189837
189932 189942 190037 190900 191760 191771 191919 192242 192370 192440 192536
192874 193127 194220 195025 195477 196324 197482 197690 198661 200166 201248
201260 203085 203459 205007 205031 205840 206121 206330 206474 206575 206595
206626 206629 206647 206650 211417 212343 212581 212606 212607 213237 213373
215133 215624 215633 216605 217362 217415 217423 217425 217521 217589 217591
217600 217708 218483 218593 219102 219163

Appendix B

Appendix B

A sample of the catalog file that was generated for the A-state of Methoxyamine CH_3ONH_2 in Band 5 (140-225 GHz) and Band 6 (225-317 GHz).

140003.8250999.9999 -5.8231 3 87.4673 29 47 30314 213 14 114

140686.7768999.9999 -5.8044 3 147.9938 35 47 30317 216 17 215

141633.4332999.9999 -6.1800 3 36.5443 21 47 30310 1 9 9 2 8

142146.6931999.9999 -6.5039 3 274.3018 43 47 30321 219 20 416

142292.6147999.9999 -6.7680 3 203.9787 37 47 30318 414 19 217

145001.5822999.9999 -5.6732 3 249.1783 41 47 30320 318 20 219

146380.0813809.5203 -4.1697 3 14.7615 15 47 303 7 1 7 6 1 6

147686.4898594.7922 -8.2685 3 1.9137 5 47 303 2 2 1 2 0 2

147937.2452612.4972 -7.7600 3 3.8498 7 47 303 3 2 2 3 0 3

148045.3966793.3997 -4.1496 3 13.9100 15 47 303 7 0 7 6 0 6

148128.0781999.9999 -6.8774 3 48.9711 23 47 30311 210 11 011

148181.1240845.5663 -4.1957 3 18.8739 15 47 303 7 2 6 6 2 5

148240.5650654.1291 -7.4440 3 6.4684 9 47 303 4 2 3 4 0 4

148339.8293999.9999 -6.8706 3 39.6209 21 47 30310 2 9 10 010

148396.0369999.9999 -6.8858 3 100.4990 31 47 30315 114 14 311

148459.9339999.9999 -6.8858 3 59.8271 25 47 30312 211 12 012

148553.1700733.9537 -7.2271 3 9.8053 11 47 303 5 2 4 5 0 5

148584.3189389.6018 -5.8113 3 9.8053 13 47 303 6 1 6 5 0 5

148673.3845999.9999 -6.8786 3 31.5851 19 47 303 9 2 8 9 0 9

148798.1846845.0336 -4.1921 3 18.9258 15 47 303 7 2 5 6 2 4

148812.6827869.7999 -7.0759 3 13.9100 13 47 303 6 2 5 6 0 6

148824.7361999.9999 -8.9984 3 1347.9598 63 47 30331 428 32 231

148904.3467999.9999 -6.9105 3 24.7046 17 47 303 8 2 7 8 0 8

148948.4101999.9999 -6.9737 3 18.8483 15 47 303 7 2 6 7 0 7

149157.6768999.9999 -8.8151 3 1046.7572 61 47 30330 228 30 129

149186.1028999.9999 -4.2607 3 28.3199 15 47 303 7 3 5 6 3 4

149203.9181999.9999 -4.2606 3 28.3208 15 47 303 7 3 4 6 3 3

149751.0910999.9999 -9.1259 3 1051.7326 59 47 30329 525 30 228

150034.1740999.9999 -6.8750 3 72.4157 27 47 30313 212 13 013

150127.0056999.9999 -6.3149 3 44.5881 23 47 30311 111 10 2 8

150234.2275806.4358 -4.1485 3 15.3504 15 47 303 7 1 6 6 1 5

151366.4975999.9999 -8.3715 3 48.9711 21 47 30310 3 8 11 011

151543.4428999.9999 -5.8000 3 104.2358 31 47 30315 214 15 115

151705.4538999.9999 -5.7578 3 19.6443 17 47 303 8 0 8 7 1 7

151882.4809585.5238 -6.1688 3 1.7738 5 47 303 2 2 1 1 1 0

152082.7747999.9999 -5.7374 3 292.5052 43 47 30321 319 21 220

152514.6690999.9999 -5.5745 3 211.9283 39 47 30319 317 19 218

152534.7548999.9999 -4.3769 3 53.1701 15 47 303 7 4 4 6 4 3

152534.8918999.9999 -4.3769 3 53.1701 15 47 303 7 4 3 6 4 2

152993.7583585.4338 -6.1687 3 1.7379 5 47 303 2 2 0 1 1 1

153969.5358999.9999 -6.8117 3 87.0015 29 47 30314 213 14 014

156897.8524999.9999 -8.2170 3 789.9623 55 47 30327 523 28 226

157121.0407999.9999 -6.5345 3 875.3504 57 47 30328 425 28 326

157150.7228999.9999 -6.6635 3 18.9258 11 47 303 5 3 3 6 2 4

158719.1220999.9999 -6.6569 3 18.8739 11 47 303 5 3 2 6 2 5

159273.6086999.9999 -7.6661 3 268.9890 41 47 30320 416 21 121

160665.6663999.9999 -4.6046 3 117.6438 15 47 303 7 5 3 6 5 2

160665.6666999.9999 -4.6046 3 117.6438 15 47 303 7 5 2 6 5 1

161800.9569999.9999 -6.7453 3 268.9047 41 47 30320 416 21 021

162137.0525999.9999 -6.6462 3 103.8825 31 47 30315 214 15 015

163916.8681999.9999 -7.3253 3 808.1626 55 47 30327 522 28 325

167491.9027999.9999 -8.4672 3 1046.1456 61 47 30330 228 30 229

167851.4867999.9999 -8.0332 3 912.1545 59 47 30329 228 29 227

168172.5173999.9999 -6.1650 3 136.5129 31 47 30315 411 16 314

169754.3242999.9999 -5.4624 3 180.0073 37 47 30318 316 18 217

170418.5245999.9999 -5.3974 3 310.6748 43 47 30321 417 21 418

170780.7474602.7104 -6.0245 3 3.0879 7 47 303 3 2 2 2 1 1

170965.3614999.9999 -6.7479 3 310.6566 43 47 30321 417 22 022

171907.4555838.9795 -5.6269 3 13.9100 15 47 303 7 1 7 6 0 6

172461.6647999.9999 -5.7666 3 123.5546 33 47 30316 215 16 116

172586.4291999.9999 -8.8788 3 40.2824 19 47 303 9 3 7 10 110

172876.8752999.9999 -7.9497 3 310.5928 43 47 30321 417 22 122

174056.7713999.9999 -3.9703 3 19.6443 17 47 303 8 1 8 7 1 7

174100.3936602.2806 -6.0260 3 2.9829 7 47 303 3 2 1 2 1 2

175523.4493999.9999 -3.9930 3 23.8167 17 47 303 8 2 7 7 2 6

175537.6443999.9999 -6.1283 3 136.2031 31 47 30315 412 16 313

175567.5126999.9999 -3.9548 3 18.8483 17 47 303 8 0 8 7 0 7

175873.5228999.9999 -3.9912 3 23.8892 17 47 303 8 2 6 7 2 5

176124.3477999.9999 -4.0475 3 33.2962 17 47 303 8 3 6 7 3 5

176128.4566999.9999 -4.0475 3 33.2977 17 47 303 8 3 5 7 3 4

176410.4714999.9999 -5.1092 3 270.7783 15 47 303 7 6 1 6 6 0

176410.4714999.9999 -5.1092 3 270.7783 15 47 303 7 6 2 6 6 1

176706.9783999.9999 -6.0070 3 86.2430 29 47 30314 213 13 310

177447.2977999.9999 -3.9552 3 20.3617 17 47 303 8 1 7 7 1 6

178121.6054999.9999 -6.3218 3 123.3658 33 47 30316 215 16 016

179401.0499999.9999 -4.1427 3 58.2582 17 47 303 8 4 4 7 4 3

179401.0851999.9999 -4.1427 3 58.2581 17 47 303 8 4 5 7 4 4

179594.2300999.9999 -6.7583 3 14.7918 9 47 303 4 3 2 5 2 3

180219.4523999.9999 -6.2911 3 342.6756 45 47 30322 320 22 319

180442.3116999.9999 -8.2110 3 53.8082 25 47 30312 012 11 2 9

180537.2390999.9999 -6.7549 3 14.7605 9 47 303 4 3 1 5 2 4

181474.3344999.9999 -5.9327 3 44.5690 23 47 30311 110 10 2 9

183921.7858999.9999 -5.5394 3 25.4502 19 47 303 9 0 9 8 1 8

184535.5919999.9999 -5.8708 3 608.6390 51 47 30325 520 25 521

184920.8697999.9999 -7.4784 3 117.1975 33 47 30316 016 15 313

185850.5281999.9999 -5.7697 3 342.4878 45 47 30322 320 22 221

186043.7143594.7945 -8.2885 3 0.6355 5 47 303 2 2 0 1 0 1

186073.4675999.9999 -5.8443 3 173.8006 37 47 30318 217 18 216

187991.0626999.9999 -4.3274 3 123.0030 17 47 303 8 5 3 7 5 2

187991.0627999.9999 -4.3274 3 123.0030 17 47 303 8 5 4 7 5 3

189993.0617643.8683 -5.8845 3 5.0757 9 47 303 4 2 3 3 1 2

190580.8104999.9999 -6.3369 3 117.1975 33 47 30316 116 15 313

192418.8648999.9999 -8.2608 3 39.6209 19 47 303 9 3 7 10 010

192442.4410999.9999 -5.3594 3 152.6866 35 47 30317 315 17 216

196051.5160999.9999 -6.3436 3 232.0051 41 47 30320 119 20 020

196531.4875642.6491 -5.8903 3 4.8731 9 47 303 4 2 2 3 1 3

196607.6479999.9999 -6.0139 3 100.6057 31 47 30315 213 14 312

197918.8301999.9999 -5.4558 3 18.8483 17 47 303 8 1 8 7 0 7

198006.8205999.9999 -7.2954 3 232.2092 39 47 30319 415 20 120

198008.7514999.9999 -6.0475 3 53.8082 25 47 30312 112 11 2 9

200612.6262999.9999 -7.0331 3 11.4287 7 47 303 3 3 1 4 2 2

201077.2156999.9999 -7.0316 3 11.4132 7 47 303 3 3 0 4 2 3

201105.4558999.9999 -8.0181 3 1159.7190 59 47 30329 623 30 328

204124.3180999.9999 -6.5877 3 232.0051 39 47 30319 415 20 020

204888.4158999.9999 -8.5941 3 783.1280 57 47 30328 226 27 423

204937.4888999.9999 -3.7891 3 25.4502 19 47 303 9 1 9 8 1 8

205124.8874999.9999 -4.7162 3 276.6627 17 47 303 8 6 2 7 6 1

205124.8874999.9999 -4.7162 3 276.6627 17 47 303 8 6 3 7 6 2

205707.2194999.9999 -3.8111 3 29.7557 19 47 303 9 2 7 8 2 6

205862.8804999.9999 -3.8594 3 39.1727 19 47 303 9 3 6 8 3 5

205904.1762999.9999 -3.8593 3 39.1711 19 47 303 9 3 7 8 3 6

206042.1409999.9999 -3.8098 3 29.6715 19 47 303 9 2 8 8 2 7

206152.5529612.5219 -7.7296 3 1.9137 7 47 303 3 2 1 2 0 2

206273.1032999.9999 -3.7769 3 24.7046 19 47 303 9 0 9 8 0 8

206404.5077999.9999 -8.9326 3 32.2861 17 47 303 8 3 6 9 1 9

207660.1639999.9999 -3.7794 3 26.2807 19 47 303 9 1 8 8 1 7

208451.4175999.9999 -9.4181 3 1345.9708 63 47 30331 428 32 131

208881.1483999.9999 -3.9435 3 64.2423 19 47 303 9 4 5 8 4 4

208881.4577999.9999 -3.9435 3 64.2423 19 47 303 9 4 6 8 4 5

208959.1651999.9999 -5.7251 3 145.7165 35 47 30317 216 17 117

209404.9931999.9999 -6.4847 3 1039.0480 57 47 30328 623 28 622

209902.2444731.7000 -5.7528 3 7.7589 11 47 303 5 2 4 4 1 3

210030.4947999.9999 -5.8465 3 145.6807 35 47 30317 216 17 017

210366.5175999.9999 -9.0471 3 26.2807 15 47 303 7 3 4 8 1 7

211487.2628999.9999 -9.1267 3 2255.5722 69 47 30334 628 34 529

211870.4311999.9999 -7.4028 3 934.1135 57 47 30328 523 27 622

215174.3846999.9999 -7.1543 3 531.7270 49 47 30324 519 25 224

216019.5758999.9999 -5.2776 3 129.3073 33 47 30316 314 16 215

216109.3484999.9999 -6.6424 3 271.1335 43 47 30321 120 20 417

217651.6257999.9999 -4.1083 3 129.2737 19 47 303 9 5 4 8 5 3

217651.6260999.9999 -4.1083 3 129.2737 19 47 303 9 5 5 8 5 4

219889.8191999.9999 -5.3439 3 32.2861 21 47 30310 010 9 1 9

220486.5739729.1215 -5.7661 3 7.4372 11 47 303 5 2 3 4 1 4

223884.7879999.9999 -6.0130 3 207.9250 39 47 30319 316 18 415

226329.6897999.9999 -5.7232 3 53.9121 25 47 30312 111 11 210

227207.1553655.5356 -7.3726 3 3.8498 9 47 303 4 2 2 3 0 3

227288.8062999.9999 -5.2951 3 24.7046 19 47 303 9 1 9 8 0 8

227420.2107999.9999 -8.2457 3 31.5851 17 47 303 8 3 6 9 0 9

230982.0214918.6910 -5.6288 3 11.1692 13 47 303 6 2 5 5 1 4

234123.3073999.9999 -8.2825 3 470.1573 49 47 30324 322 25 025

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234764.6575999.9999 -5.5585 3 606.6754 17 47 303 8 7 2 7 7 1

235217.6488999.9999 -8.9039 3 25.4502 15 47 303 7 3 5 8 1 8

235831.2489999.9999 -4.4555 3 283.5050 19 47 303 9 6 3 8 6 2

235831.2489999.9999 -4.4555 3 283.5050 19 47 303 9 6 4 8 6 3

237037.0418999.9999 -5.2188 3 109.2908 31 47 30315 313 15 214

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240571.9670999.9999 -3.6407 3 36.5443 21 47 30310 2 9 9 2 8

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248546.9331999.9999 -5.8790 3 208.7251 39 47 30319 317 18 414

249529.7419747.5832 -7.1289 3 6.4684 11 47 303 5 2 3 4 0 4

250188.1487999.9999 -5.4022 3 348.6871 45 47 30322 419 22 320

250307.0136999.9999 -3.9220 3 136.5338 21 47 30310 5 6 9 5 5

250307.0223999.9999 -3.9220 3 136.5338 21 47 30310 5 5 9 5 4

253813.6842999.9999 -5.5108 3 15.3504 15 47 303 7 2 6 6 1 5

253876.8703999.9999 -5.1804 3 92.1373 29 47 30314 312 14 213

254973.2066999.9999 -5.8016 3 64.4480 27 47 30313 113 12 210

256041.7143999.9999 -7.2390 3 912.1545 59 47 30329 326 29 227

256500.2149999.9999 -5.8038 3 117.1975 29 47 30314 410 15 313

256824.5225999.9999 -6.5577 3 357.7279 45 47 30322 418 23 023

257568.9662999.9999 -8.3114 3 24.7046 15 47 303 7 3 5 8 0 8

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260775.8809999.9999 -8.1726 3 357.5960 45 47 30322 418 23 123

261220.8104999.9999 -5.7877 3 117.0165 29 47 30314 411 15 312

263483.5949999.9999 -6.0221 3 316.3594 45 47 30322 220 21 417

263570.6186999.9999 -5.6218 3 100.4990 31 47 30315 214 14 311

264351.6823999.9999 -8.7420 3 15.3504 11 47 303 5 3 2 6 1 5

264763.2990999.9999 -5.8096 3 117.1975 33 47 30316 214 15 313

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275904.2991999.9999 -6.2364 3 412.3212 45 47 30322 517 23 420

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277754.4378999.9999 -6.5273 3 173.8006 35 47 30317 413 18 216

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280305.0877999.9999 -5.1605 3 53.9121 23 47 30311 3 9 11 210

280312.3845999.9999 -3.4569 3 39.6209 23 47 30311 011 10 010

280400.0948999.9999 -7.0065 3 268.9890 43 47 30321 120 21 121

280412.8682999.9999 -3.4633 3 41.2687 23 47 30311 110 10 1 9

280423.5030999.9999 -5.6316 3 11.4287 9 47 303 4 3 1 4 2 2

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284651.0025999.9999 -5.2114 3 36.5443 19 47 303 9 3 7 9 2 8

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285686.6057999.9999 -5.1566 3 76.7135 27 47 30313 310 13 211

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286747.9204999.9999 -3.7568 3 144.8831 23 47 30311 5 6 10 5 5

288202.8029999.9999 -8.8840 3 11.1692 9 47 303 4 3 1 5 1 4

289202.7903999.9999 -5.1715 3 90.8522 29 47 30314 311 14 212

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306725.0470999.9999 -4.9994 3 49.5958 25 47 30312 012 11 111

307154.5005999.9999 -9.0903 3 1210.7815 61 47 30330 526 31 229

307169.9324999.9999 -9.1897 3 1391.5820 63 47 30331 427 32 230

307518.9603999.9999 -8.7035 3 13.9100 11 47 303 5 3 3 6 0 6

307697.7492999.9999 -5.2865 3 26.2807 19 47 303 9 2 8 8 1 7

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308447.5993999.9999 -5.8453 3 238.8896 41 47 30320 219 20 218

310629.2813999.9999 -9.2786 3 7.7589 7 47 303 3 3 0 4 1 3

316824.0202999.9999 -5.5613 3 147.9938 35 47 30317 314 17 215

317047.8658999.9999 -8.6136 3 803.0547 55 47 30327 522 28 227

Appendix C

Appendix C

A sample of the catalog file that was generated for Nitrosomethane CH_3NO in Band 5 (140-225 GHz) and Band 6 (225-317 GHz).

140056.2681421.0381 -8.6753 3 1239.9786115 45 30357 553 58 356

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140162.8585215.5716 -7.1281 3 1240.5052109 45 303541045 531142

140165.5399117.5129 -5.6030 3 397.6320 63 45 30331 526 30 625

140171.3785215.5895 -7.1280 3 1240.5052109 45 303541044 531143

140257.2791595.9304 -8.2445 3 1344.8684121 45 30360 457 59 554

140308.3818420.3676 -8.3254 3 1239.9702115 45 30357 553 58 256

140317.1958167.6031 -5.6360 3 565.5234 77 45 30338 434 38 435

140651.5095180.3133 -6.8742 3 284.5166 55 45 30327 325 28 028

140668.4127104.8996 -6.6289 3 134.4236 37 45 30318 316 19 019

140830.2525332.9722 -9.0122 3 2179.3077137 45 303681751 691654

140830.2525332.9722 -9.0122 3 2179.3077137 45 303681752 691653

140946.1485100.3936 -6.9253 3 121.3441 35 45 30317 315 18 118

141080.8664 58.0041 -5.2149 3 85.7718 31 45 30315 114 15 115

141185.7543 98.3067 -6.1828 3 234.5641 51 45 30325 124 24 321

141251.7079 49.7904 -6.0168 3 17.5665 9 45 303 4 3 2 5 2 3

141295.9571168.6465 -6.4468 3 313.1560 59 45 30329 128 28 325

141336.0987174.5641 -6.6723 3 474.6894 71 45 30335 332 34 529

141546.9566 30.5499 -4.5663 3 127.8476 37 45 30318 216 18 117

141863.8353404.4621 -7.4780 3 788.6380 93 45 30346 244 45 441

142019.8716 29.7190 -5.0157 3 12.8293 11 45 303 5 2 3 5 1 4

142021.5047 49.6347 -6.0129 3 17.5409 9 45 303 4 3 1 5 2 4

142037.6007191.1041 -6.0982 3 645.1551 73 45 303361027 37 928

142037.7828191.1038 -6.0982 3 645.1551 73 45 303361026 37 929

142250.8812 88.5888 -5.1370 3 188.6495 45 45 30322 220 22 221

142653.8038 68.4343 -5.4570 3 168.1616 43 45 30321 220 20 317

143415.8615175.1749 -6.3780 3 853.7388 91 45 30345 838 44 935

143502.3380 67.0774 -5.3398 3 210.5296 47 45 30323 321 22 418

143541.6446175.3843 -6.3773 3 853.7386 91 45 30345 837 44 936

143861.2330199.3514 -6.1417 3 334.6913 61 45 30330 229 29 326

144089.0610316.8670 -9.6842 3 2550.6733155 45 303771563 761660

144089.0705316.8670 -9.6842 3 2550.6733155 45 303771562 761661

144333.3123 89.9807 -5.4275 3 296.5922 55 45 30327 424 26 521

144443.1073186.0440 -7.2286 3 593.8401 77 45 30338 633 39 436

144551.2256748.8472 -9.4958 3 1994.4208147 45 30373 569 72 666

144563.4411 29.7301 -5.1128 3 9.1113 9 45 303 4 2 2 4 1 3

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144992.0112113.8096 -6.2130 3 253.0730 53 45 30326 125 25 322

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145155.5504306.3068 -7.7431 3 709.3333 87 45 30343 440 44 243

145210.5126190.9988 -7.2466 3 304.6187 57 45 30328 326 29 129

145318.7821 89.9854 -5.4460 3 102.8572 27 45 30313 5 9 14 410

145421.5545 89.9218 -5.4454 3 102.8538 27 45 30313 5 8 14 411

145486.8794305.6605 -7.3918 3 709.3222 87 45 30343 440 44 143

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146219.8309131.0497 -6.2686 3 272.3432 55 45 30327 126 26 323

146250.3525509.5635 -8.5784 3 1366.9753121 45 30360 456 61 359

146268.2076128.9536 -5.2859 3 352.0945 61 45 30330 327 30 328

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146987.1351 95.7687 -6.8934 3 108.7981 33 45 30316 314 17 117

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147062.5059276.4198 -8.5304 3 1980.3311137 45 303681355 671454

147119.5805129.0780 -6.5752 3 257.9095 51 45 30325 422 26 125

147201.7631219.9929 -6.1267 3 863.1386 95 45 30347 542 47 543

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147809.1213272.0853 -7.4505 3 1399.2574109 45 303541440 551343

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148127.3405136.2674 -5.7866 3 539.0945 73 45 30336 630 35 729

148391.7830121.5758 -6.7722 3 222.2007 47 45 30323 420 24 223

148425.5787292.6150 -7.2114 3 439.4858 69 45 30334 232 35 135

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148498.1076402.8495 -7.6802 3 839.6310 95 45 30347 344 48 247

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