Abstract

We present the first determination of the abundance ratios of $^{13}$C substitutions of cyanoacetylene (HC$_3$N), [H$^{13}$CCCN]:[HC$^{13}$CCN]:[HCC$^{13}$CN], in Titan’s atmosphere measured using millimeter-wave spectra obtained by the Atacama Large Millimeter/submillimeter Array. To compare the line intensities precisely, data sets that include multiple molecular lines were extracted to suppress effects of Titan’s environmental conditions and observation settings. The [HC$^{13}$CCN]:[HCC$^{13}$CN] and [H$^{13}$CCCN]:[HCC$^{13}$CN] ratios were obtained from 12 and 1 selected data sets, respectively. As a result, nearly the uniform [H$^{13}$CCCN]:[HC$^{13}$CCN]:[HCC$^{13}$CN] abundance ratios of 1.17 ($\pm$0.20) : 1.09 ($\pm$0.25) : 1 (1$\sigma$) were derived, whereas previously reported ratios for the interstellar medium (ISM) have shown large anomalies that may be caused by $^{13}$C concentrations in precursors. The result obtained here suggests that $^{13}$C concentration processes suggested in the ISM studies do not work effectively on precursors of HC$_3$N and HC$_3$N themselves owing to Titan’s high atmospheric temperature and/or depletion of both $^{13}$C and $^{13}$C$^+$. 

Unified Astronomy Thesaurus concepts: Planetary atmospheres (1244); Radio astronomy (1338); Submillimeter astronomy (1647)

1. Introduction

The $^{13}$C substituted species of cyanoacetylene (HC$_3$N), namely, H$^{13}$CCCN, HC$^{13}$CCN, and HCC$^{13}$CN, have been discovered in various interstellar media (ISMs) and are known to exhibit large isotopic anomalies such that HCC$^{13}$CN and/or H$^{13}$CCCN show high $^{13}$C concentrations (Takano et al. 1998; Araki et al. 2016; Taniguchi et al. 2016, 2017). Such anomalies are considered to be due to the $^{13}$C concentrations on the precursors of HC$_3$N, such as CN and C$_2$H (Furuya et al. 2011; Taniguchi et al. 2019), and give us important information on the environments and possible chemical reactions in the ISM.

HC$_3$N is also present on Saturn's largest moon, Titan, and was first detected in the atmosphere by the Voyager 1 spacecraft (Kunde et al. 1981). The main production pathway of HC$_3$N was expected as follows: \[ C_2H_2 + CN \rightarrow HC_3N + H. \] Since then, a number of in situ, ground- and space-based observations have been performed to illustrate the spatial and time variation of HC$_3$N along with other nitriles and hydrocarbons (Hidayat et al. 1997; Coustenis et al. 1998, 2003, 2007, 2010, 2016, 2018; Gurwell 2004; Cordiner et al. 2014; Thelen et al. 2019).

Adding to Reaction (1), a following reaction of the C$_2$H radical with HNC (Loison et al. 2015) possibly produces a portion of HC$_3$N because HNC is present in the upper stratosphere (Moreno et al. 2011; Cordiner et al. 2014).

\[ C_2H + HNC \rightarrow HC_3N + H. \] In turn, due to the extremely high reaction barrier, the reaction of C$_2$H with HCN, an isomer of HNC, does not work effectively. Thus, Reaction (2) is the only reaction to produce HC$_3$N from the C$_2$H radical.

Two precursor radicals of HC$_3$N, namely, CN and C$_2$H, are important for the HC$_3$N production, along with the reaction counterparts, C$_2$H$_2$ and HNC. They are easily produced by the photodissociation HCN and C$_2$H$_2$ molecules, and their expected abundances are $\sim$ 10 ppb at 1000 km (Lavvas et al. 2008).

As for the ISM, $^{13}$C substitutions of HC$_3$N have also been detected on Titan. The first observational result was reported by Jennings et al. (2008) using infrared spectra obtained with the Composite Infrared Spectrometer on board the Cassini spacecraft. Spectral emissions of the three isotopologues were clearly detected, whereas the H$^{13}$CCCN and HCC$^{13}$CN lines were blended. Using the H$^{13}$CCCN and HC$_3$N lines, the $^{12}$C/$^{13}$C ratio was measured to be 79 $\pm$ 17, which is consistent with that measured in HCN (70-120, Hidayat et al. 1997; 132 $\pm$ 25 or 108 $\pm$ 20, Gurwell 2004; 89.8 $\pm$ 2.8, Molter et al. 2016) and C$_2$H$_2$ (84.8 $\pm$ 3.2; Nixon et al. 2012). A recent submillimeter spectroscopy using ALMA succeeded in the detection of H$^{13}$CCCN with a high signal-to-noise ratio (S/N), along with HCCC$^{13}$N (Cordiner et al. 2018), although the $^{12}$C/$^{13}$C value was not determined for HC$_3$N.

In this study, we report the first observational determination of the relative $^{13}$C carbon isotopic ratios of three isotopologues of HC$_3$N, namely, [H$^{13}$CCCN]:[HC$^{13}$CCN]:[HCC$^{13}$CN], on Titan by analyzing a large data set obtained by ALMA. The result enables us to compare the chemical environment of Titan with that of the ISM.

2. Analysis

2.1. Data Selection

Since Titan is often used as a calibrator of ALMA, a large amount of observation data of Titan is available in the ALMA archive. We calibrated and imaged all of the archived observational
data of Titan that were available as of 2020 January. The details of the calibration and imaging procedure were as described in our previous paper that analyzed the nitrogen isotopic ratio of CH$_3$CN on Titan (Iino et al. 2020).

Spectral lines of the isotopologues of HC$_3$N are often observed by ALMA by chance because their pure rotational transitions appear every ∼10 GHz. To measure the line intensities precisely, we have chosen spectral windows (SPWs) that observed multiple isotopologues simultaneously. The usage of the data in the same SPW suppresses the systematic uncertainties arising from the differences in the observation configurations, such as the synthesized beam size and absolute flux calibration, and Titan’s environmental conditions, such as the horizontal and vertical distribution of HC$_3$N and the atmospheric structure. To measure the line intensities that have a narrow (∼1.5 MHz) line width, SPWs that have a high-frequency resolution of <2 MHz were chosen. The frequency difference between HC$^{13}$CCN and HCC$^{13}$CN that share the same rotational state $J$ is no more than 20 MHz because they have a very similar rotational constant $B$. Thus, in most cases, they are observed in the same SPW. In turn, since the rotational constant of H$^{13}$CCCN is ∼2% smaller than that of HC$^{13}$CCN and HCC$^{13}$CN, the number of SPWs including three lines was smaller than that including the HC$^{13}$CCN–HCC$^{13}$CN pair.

As an important phenomenon, Titan’s trace gases, including HC$_3$N and its precursors, C$_2$H$_2$ and HCN, are known to exhibit large spatial and time variations. The analyzed period, from 2012 to 2015, is a season of northern summer, when an increase and a decrease of trace species have been observed by Cassini and ALMA for southern and northern hemispheres, respectively (Cordiner et al. 2015; Coustenis et al. 2018; Thelen et al. 2019).

To decrease the effect of such data-to-data variability of the HC$_3$N spatial distribution, disk-averaged spectra were extracted from the imaged cube fits with a 0"34 radius circle, which is large enough to cover the entire disk of Titan for all of the analyzed data sets.

We attempted to remove the baseline structure of the spectra using the polynomial fitting method, while the effect was very limited. For the line intensity measurement, spectral intensities within a range of ±1 MHz from the line center were integrated. The noise level was measured in the line-free region and multiplied by $\sqrt{n}$, where $n$ is the number of averaged channels. After extracting 29 SPWs that include multiple emission lines, SPWs that exhibit high S/N (>4) were chosen for the frequency ratio measurement analysis.

The numbers of selected SPWs including HC$^{13}$CCN–HCC$^{13}$CN and H$^{13}$CCCN–HCC$^{13}$CN pairs obtained with high S/N were 12 and 1, respectively. Observation parameters of the selected SPWs are summarized in Table 1. The rotational state transitions corresponding to 217, 226, 235, 244, and 271 GHz bands were chosen. The frequency difference (Δ$J$) is small, particularly 1, 2, and 3 for H$^{13}$CCCN and HCC$^{13}$CN, respectively. Most of the bands are of ALMA Band 6, except for 308 GHz of Band 7. The project code 2015.1.00512.S data have a long observation time by concatenating short observation time data to improve the S/N.

Figures 1 and 2 show the obtained spectra for each pair. The HC$^{13}$CCN and HCC$^{13}$CN lines are plotted in the same panels as in Figure 1 owing to small differences in frequency, while the lines for H$^{13}$CCCN and HCC$^{13}$CN are overplotted in Figure 2. For H$^{13}$CCCN including data, as shown in Figure 2, since the HC$^{13}$CCN line was blended with an ethyl cyanide (C$_2$H$_2$CN(36,36–35,35)) line, only the HCC$^{13}$CN line was used for the intensity comparison. Note that the detection of C$_2$H$_2$CN with ALMA was reported previously (Cordiner et al. 2015).

In Titan’s atmosphere, the chemical processes associated with HC$_3$N vary with altitude, with ion chemistry being dominant at high altitudes and neutral chemistry being dominant at low altitudes. A previous ALMA observation study (Thelen et al. 2019) derived an altitude range where HC$_3$N ($J = 35–34$) is sensitive by the radiative transfer analysis of the disk-averaged spectra. The optically thick line core region of ±2 MHz from the line center probes at the ∼800 km high-altitude region, whereas wings have sensitivity at 150 km, where HC$_3$N shows abundance peaks in the high-latitude regions. Because the obtained intensities

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**Table 1** Summary of Observation Parameters and Result with Error

| Project Code | Obs. Date | Obs. Time | Beam Shape (arcsec) | $\Delta f$ (GHz) | Peak Intensity (Jy beam$^{-1}$ km s$^{-1}$) | Integrated Intensity (Jy beam$^{-1}$ km s$^{-1}$) | Abundance Ratio |
|--------------|-----------|-----------|---------------------|-----------------|----------------------------------------|-----------------------------------------------|----------------|
| (a) 2011.0.00735 | 2012-01-13 | 17:1.68 | 2.00 x 1.30 | 488 | 226.4541 | 0.0614(7.1) | 226.4761 | 0.0712(8.2) | 0.86(0.16) |
| (b) 2011.0.00735 | 2012-01-13 | 17:1.68 | 1.87 x 1.21 | 488 | 244.5644 | 0.0451(4.2) | 244.5882 | 0.0726(6.8) | 0.62(0.17) |
| (c) 2012.1.00932 | 2014-03-11 | 157.344 | 1.15 x 0.75 | 244 | 217.3985 | 0.0254(6.8) | 217.4196 | 0.0196(5.4) | 1.44(0.32) |
| (d) 2012.1.00248 | 2014-09-29 | 158.304 | 0.81 x 0.59 | 488 | 235.5094 | 0.0185(6.5) | 235.5323 | 0.0199(6.2) | 0.95(0.22) |
| (e) 2012.1.00248 | 2014-09-29 | 158.304 | 0.88 x 0.65 | 488 | 217.3985 | 0.0345(12.1) | 217.4196 | 0.0251(8.8) | 1.37(0.19) |
| (f) 2012.1.00248 | 2014-09-29 | 158.304 | 0.95 x 0.58 | 488 | 235.5094 | 0.0255(6.8) | 235.5323 | 0.0216(5.8) | 1.18(0.27) |
| (g) 2012.1.00453 | 2014-07-07 | 157.344 | 0.48 x 0.44 | 488 | 244.5644 | 0.0150(5.3) | 244.5882 | 0.0125(4.1) | 1.19(0.35) |
| (h) 2013.1.00271 | 2015-04-05 | 157.344 | 1.45 x 1.01 | 488 | 226.4541 | 0.0214(6.3) | 226.4761 | 0.0161(4.7) | 1.34(0.35) |
| (i) 2012.1.00453 | 2015-05-13 | 157.344 | 0.66 x 0.47 | 488 | 271.7271 | 0.0124(5.1) | 271.7536 | 0.0104(4.3) | 1.20(0.37) |
| (j) 2015.1.01312 | 2016-03-21 | 151.2 | 0.90 x 0.72 | 976 | 226.4541 | 0.0191(5.1) | 226.4761 | 0.0172(5.3) | 1.11(0.32) |
| (k) 2015.1.00512 | 2016-03-31 | 604.8 | 1.07 x 0.84 | 1953 | 217.3985 | 0.0105(4.6) | 217.4196 | 0.0143(6.3) | 0.73(0.20) |
| (l) 2015.1.00315 | 2016-04-02 | 755.0 | 1.09 x 0.84 | 976 | 217.3985 | 0.0199(4.3) | 217.4196 | 0.0201(4.4) | 1.00(0.32) |

**Note.** Integrated intensities and abundance ratios are followed by 1σ S/N in parentheses. Abundance ratios are expressed as [HC$^{13}$CCN]/[HCC$^{13}$CN] or [H$^{13}$CCCN]/[HCC$^{13}$CN].
of isotopomer lines analyzed in our study are much weaker than that of the HC$_3$N line, we infer that the isotopomer lines have sensitivity at the lower stratosphere.

2.2. Abundance Ratio Calculation

To calculate the abundance ratios for the two pairs of isotopologues, as described in the methods below, we compared the measured integrated line intensities instead of retrieving the vertical abundance using the radiative transfer method. Since they share similar vertical abundance and optical depth and the same temperature profile, their relative abundance can be derived with a proper consideration of the difference of the rotational transitions between the two isotopologues. In addition, optically thin molecular lines enable us to assume that the measured intensity is proportional to the optical depth. This method was also applied to the previous [H$^{13}$CCCN]/[HCC$^{15}$N] measurement using ALMA observation results (Cordiner et al. 2018). Because we did not need to consider the effect of the difference of beam size, temperature profile, and three-dimensional distribution of HC$_3$N, the only effect on the line intensity that was estimated and applied was the difference in spectroscopic parameters with respect to the stratospheric temperature. For the calculation of the parameters relating to opacity, such as the partition function and population, the equations used were those listed in the appendix of Turner (1991) and Iino et al. (2014). Line parameters such as the Einstein coefficient $A_{ul}$, the lower-state energy $E_u$, and the rotational constant $B$ were obtained from the NASA JPL catalog (Picket et al. 1998). The considered excitation temperatures were from 140 to 180 K under the assumption of the local temperature equilibrium condition.

For the HC$_{13}$CCN–HCC$_{13}$CN pair, the evaluation was simple because they share the same rotational state $J$. For the range of analyzed rotational transitions, the line intensity difference between HC$_{13}$CCN and HCC$_{13}$CN was estimated to be less than 0.02% for the modeled temperature range. The only exception was the J = 24–23 transition, where HCC$_{13}$CN has three hyperfine splitting lines. In this case, the line intensities of three transitions were simply integrated. As a result, the line intensity difference for the J = 24–23 pair was determined to be $\sim$0.2%. Thus, considering the line intensity difference between two isotopologues and optically thin line intensities, for all the HC$_{13}$CCN–HCC$_{13}$CN pairs, we used the integrated line intensity ratios as the abundance ratio. The derived [HC$_{13}$CCN]/[HCC$_{15}$CN] ratios are shown in the rightmost column of Table 1. Figure 3 shows a histogram of the obtained [HC$_{13}$CCN]/[HCC$_{15}$CN] ratio. Any fractionation relating to the rotational transition was found. The averaged mean value is 1.09 with a standard deviation of 0.25. Since the time variations of the isotopic ratios are beyond the scope of this paper,
we simply average the data taken from the epoch from 2012 to 2015.

For the H$^{13}$CCCN–HCC$^{13}$CN pair, taking into account the different rotational states, it was determined that HCC$^{13}$CN($J = 34–33$) has a 6.4% higher intensity than H$^{13}$CCCN($J = 35–34$) on average under the assumed excitation temperature range. With this correction, we found the abundance ratio of [H$^{13}$CCCN]/[HCC$^{13}$CN] to be 1.17 ± 0.20. Note that this error was determined from the noise level of a single spectrum.

3. Discussion

The measured [H$^{13}$CCCN]:[HCC$^{13}$CN]:[HCC$^{13}$CN] ratios on Titan are nearly uniform within their errors. The ratios do not show the large anomalies as reported by the ISM observations that HCC$^{13}$CN and/or H$^{13}$CCCN show 35–110 and 20% higher abundance than the others, respectively (Takano et al. 1998; Araki et al. 2016; Taniguchi et al. 2016, 2017). Assuming that no time variation of the $^{12}$C/$^{13}$C ratio is present since the previous observation, the result indicates that the $^{12}$C/$^{13}$C ratios on three isotopologues are the same as the value of 79 ± 17 measured on H$^{13}$CCCN (Jennings et al. 2008). Below, the derived results are discussed from two viewpoints: $^{13}$C concentration on HCC$^{13}$CN and H$^{13}$CCCN. The obtained absence of the carbon fractionation process on Titan is possibly explained by the environmental differences between Titan and the ISM, such as atmospheric temperature and chemical composition. The differences may constrain chemical reactions present in Titan’s middle and upper atmosphere.

3.1. HCC$^{13}$CN Concentration

Assuming the main HC$_3$N production as in Reaction (1), $^{13}$C concentrations on CN and/or its precursor are greater than that in reaction counterparts, C$_2$H$_2$, whose $^{12}$C/$^{13}$C ratio was determined to be 84.8 ± 3.2 using the infrared spectra obtained by the Cassini spacecraft (Nixon et al. 2008). A main production pathway of CN is a photodissociation of HCN (Loison et al. 2015). The other pathway, a photodissociation of C$_2$N$_2$, is negligible because the abundance is below 1%–0.1% of HCN. The most recent ALMA observation reported that no significant $^{13}$C concentration in HCN (89.8 ± 2.8; Molter et al. 2016) is relative to C$_2$H$_2$. Thus, if it exists, $^{13}$C concentration on CN occurs after the photolysis of HCN.

An ion–molecule isotope exchange process between $^{13}$C$^+$ and CN has been proposed for the exothermic $^{13}$CN concentration process for interstellar clouds as follows (Colzi et al. 2020):

$$^{13}\text{C}^+ + \text{CN} \leftrightarrow ^{13}\text{C}^+ + ^{13}\text{CN} + \Delta E(31.1 \text{ K}).$$ (3)

Reaction (3) likely causes HCC$^{13}$CN enrichment in some ISMs, especially in low-temperature conditions (~10 K; Takano et al. 1998). However, Reaction (3) does not seem to be effective on Titan’s relatively higher atmospheric temperature (140–180 K in the stratosphere) compared to the interstellar one, because the backward reaction of Reaction (3) can proceed and suppress the isotopic fractionation of CN in such a high-temperature environment.

The other scenario is related to abundance of the $^{13}$C$^+$ ion. Vuitton et al. (2007) calculated $^{13}$C$^+$ number density as 1.4 × 10$^{-2}$ cm$^{-3}$ considering mas spectral measurement results. The calculated density is smaller than other major ions such as CH$_2^+$, CH$_3^+$, CH$_2^+$, N$^+$, and so on; thus, Reaction (3) may not be effective in concentrating $^{13}$C on CN and subsequently HC$_3$N.

3.2. H$^{13}$CCCN Concentration

Reaction (2) is the only pathway to produce HC$_3$N from the C$_2$H radical. Because C$_2$H$_2$, a precursor of C$_2$H, is a symmetric carbon molecule, an anomaly between H$^{13}$CCCN and HC$_3$CCCN is caused by the abundance difference between C$_{15}$CH and $^{13}$CCH. In the ISM, the anomaly is possibly due to the isotope exchange reaction to achieve $^{13}$C concentration on C$_{15}$CH as follows (Furuya et al. 2011):

$$^{13}\text{CCH} + \text{H} \leftrightarrow ^{13}\text{CCH} + \text{H} + \Delta E(8.1 \text{ K}).$$ (4)

Similar to the case of Reaction (3), the forward reaction of Reaction (4) is considered to be active only in a low-temperature environment such as the starless cores. In the case
of Titan, Reaction (4) is not expected to work for concentration of C13CH owing to the high-temperature condition. In addition, because of low HNC abundance (Moreno et al. 2011; Cordiner et al. 2014), the contribution of Reaction (2) for HC3N production may be negligible.

4. Summary and Future Prospects

We have detected the presence of all three 13C substituted species of HC3N, namely, H13CCCN, HC13CCN, and HCC13CN, in Titan’s atmosphere using observational data from the ALMA archive. The statistically derived [HC13CCN]/[HCC13CN] value was determined to be 1.09 ± 0.25, whereas those measured in starless dark clouds and low-mass star-forming regions were previously reported to exceed the present error. Additionally, [H13CCCN]/[HC13CCN] was found to be 1.17 ± 0.20, although this result is less reliable than that for [HC13CCN]/[HCC13CN] because of a single pair detection. For both cases, no significant 13C concentration in HC3N was detected, which differs from most of the ISM cases.

The large environmental difference with ISM is the high atmospheric temperature environment of Titan. A recent ALMA temperature measurement revealed that the measured stratospheric temperature above 100 km is at least 130 K, reaching 180 K at 300 km (Thelen et al. 2018). These temperatures are much higher than that expected in the ISM (10 K; Taniguchi et al. 2019). In such a cold region with temperatures around 10 K, the barrierless and exothermic isotopic exchange reactions, Reactions (3) and (4), which have been considered to cause fractionation on HC3N, are driven by the differences in the zero-point energy. On the other hand, in a high-temperature environment, such as Titan’s stratosphere and mesosphere, the backward reactions of Reactions (3) and (4) can proceed to suppress the isotopic fractionation in the precursors of HC3N and HC3N themselves.

Recently, similar to the case of Titan, Taniguchi et al. (2021) reported uniform carbon isotopic ratios of HC3N around massive young stellar objects. They proposed that HC3N is mainly produced via the HC3NH+ ion, which has a complicated formation pathway, which introduces more complicated pathways of HC3N formation, and thus all the HC3N isotopic ratios would be affected by reactions other than Reactions (3) and (4). In addition to Reactions (1) and (2), a recent study suggested that the photodissociation of C2H3CN and the H-atom addition to HC3N2 may produce HC3N (Vuitton et al. 2019). For the total understanding of Titan’s isotopic fractionation processes, such ion-relating reactions and newly proposed neutral reactions should be investigated.

Similar to Reaction (3), new exchange reactions of 13C and 13C+ with C-bearing species are proposed by recent publications (Colzi et al. 2020; Loison et al. 2020). These newly proposed reactions may induce 13C concentrations in C-bearing species, in particular if 13C and 13C+ are abundant, which is unlikely in the Titan atmosphere. Our result, together with the previously reported nonconcentration of 13C in C-bearing species on Titan such as CH3, C2H2, HCN, HC3N, and CO, may be interpreted as the consequence of 13C and 13C+ depletion.

This study makes use of the ALMA data listed in Table 1. ALMA is a partnership of ESO (representing its member states), NSF (USA), and NINS (Japan), together with NRC (Canada), MOST and ASIAA (Taiwan), and KASI (Republic of Korea), in cooperation with the Republic of Chile. The Joint ALMA Observatory is operated by ESO, AUI/NSF, and NAOJ. This work was supported by grants from the Telecommunications Advancement Foundation (TI), the Japan Society for the Promotion of Science (JSPS) Kakenhi (17K14420, 19K14782, 20K14523, 20K04046, and 20K04017), and the Astrobiology Center Program of National Institutes of Natural Sciences (NINS).

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