Interstellar Complex Organic Molecules in SiO-traced Massive Outflows

O. S. Rojas-García1, A. I. Gómez-Ruiz2, A. Palau2, M. T. Orozco-Aguilera1, M. Chavez Dagostino1, and S. E. Kurtz2

1 Instituto Nacional de Astrofísica, Óptica y Electrónica, Luis E. Erro 1, Tonantzintla, Puebla, C.P. 72840, Mexico; sergio.rojas@inaoep.mx
2 Instituto de Radioastronomía y Astrofísica, Universidad Nacional Autónoma de México, Antigua Carretera a Pátzcuaro # 8701, Ex-Hda. San José de la Huerta, Morelia, Michoacán, C.P. 58089, Mexico

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Abstract

The interstellar medium contains dust and gas, in which molecules can proliferate at high densities and in cold conditions. Interstellar complex organic molecules (iCOMs) are C-bearing species that contain at least six atoms. As they are detected in young stellar objects, iCOMs are expected to inhabit early stages of star formation evolution. In this study, we try to determine which iCOMs are present in the outflow component of massive protostars. To do this, we analyzed the morphological extension of blue- and redshifted iCOM emission in a sample of 11 massive protostars employing mapping observations at 1 mm within a ~1 GHz bandwidth for both the IRAM-30 m and APEX telescopes. We modeled the iCOM emission of the central pointing spectra of our objects using the XCLASS local thermal equilibrium radiative transfer code. We detected the presence of several iCOMs such as CH$_3$OH, $^{13}$CH$_3$OH, CH$_3$OCHO, C$_2$H$_5$C$_1$5N, and (c-C$_3$H$_2$)CH$_2$. In G034.41+0.24, G327.29-0.58, G228.81+0.63, G333.13-0.43, G340.97-1.02, G351.45+0.66, and G351.77-0.54, the iCOM lines show a faint broad-line profile. Due to the offset peak positions of the blue- and redshifted emission, covering from ~0.1 to 0.5 pc, these wings are possibly related to movements external to the compact core, such as large-scale low-velocity outflows. We have also established a correlation between the parent iCOM molecule CH$_3$OH and the shock tracer SiO, reinforcing the hypothesis that shock environments provide the conditions to boost the formation of iCOMs via gas-phase reactions.

Unified Astronomy Thesaurus concepts: Star forming regions (1565); Massive stars (732); Interstellar molecules (849)

1. Introduction

Interstellar clouds, star formation regions, circumstellar envelopes of AGB stars, and planetary nebulae have been proven to hold the physical conditions needed to harbor a rich molecular content. Among these molecules, many are organic in nature. This fact is especially remarkable for the larger molecules, of which those with at least six atoms have been defined as interstellar complex organic molecules (iCOMs; Herbst & van Dishoeck 2009; Ceccarelli et al. 2017).

Recent efforts have been conducted to establish the mechanism that explains the abundance of iCOMs in several interstellar environments. The dominant scenarios are dust-grain chemistry and warm gas-phase reactions (Garrod & Herbst 2006; Taquet et al. 2012). These two scenarios could work simultaneously, and are thought to take place as follows. In a first cold prestellar phase, atoms and small molecules are stuck into dust grains and are subsequently hydrogenated and oxidized, forming mainly ice mantles of H$_2$O, CO$_2$, NH$_3$, and CH$_4$. In a subsequent phase, triggered by a temperature increase due to the star formation process or by nonthermal processes, the mantle components are released to the gas phase where additional reactions form the iCOMs. Alternatively, there is also the possibility that the increase in temperature triggers further reactions within processed (irradiated by UV photons or cosmic-rays) grain mantles, forming the iCOMs in the dust-grain surfaces, which will be later desorbed (see the review by Ceccarelli et al. 2022 for further details).

The shock waves produced by outflow movements during the star formation process are one of the environments that can facilitate chemical reactions, leading to a more complex chemistry. This phenomenon is present during the early phases of the formation of low- and high-mass stars (Arce et al. 2007).

The supersonic material ejected from the inner regions of a protostellar system generates shock waves along its path, causing an increase in density and temperature in the surrounding material. Such conditions could boost chemical reactions that otherwise would not take place in the cold and quiescent interstellar medium (ISM).

The increase in the abundance of certain molecules within the shocked regions of low-mass outflows has unveiled a particular shock-induced chemistry, the prototype of which has been the low-mass protostar L1157-mm (Bachiller et al. 2001). Other examples are BHR71 (Bourke et al. 1997, low mass), HH 114-MMS (Tafalla & Hacar 2013, low mass), IRAS 16547-4247 (Garay et al. 2007b; De Simone et al. 2020, high mass), and IRAS 17233-3606 (Leurini et al. 2011, 2013c, 2013, high mass).

The extensive studies in the archetype outflow L1157-mm have revealed the rich chemistry induced by shocks. One of the remarkable discoveries was the detection of iCOMs within these shocked regions (Bachiller et al. 1995), which suggests a relation between the shocks and iCOM formation (i.e., Lefloch et al. 2017).

In the high-mass regime, a few outflows have also demonstrated a peculiar chemical richness, however, an unambiguous analysis of the high-velocity emission has been difficult given the outflow multiplicity usually found in these environments, together with the usual low-angular-resolution observations. This leads to confusion about whether the different spectral components belong to the same outflow
2. Observations

2.1. Sample Selection

Our data set consists of 11 massive protostars associated with Extended Green Objects (EGOs), and APEX Telescope Large Area Survey of the Galaxy (ATLASGAL) sources (Cyganski et al. 2008; Csengeri et al. 2014), three of them (the northern ones) belong to the EGO survey, whereas eight of them (the southern ones) come from the ATLASGAL survey. These specific sources were selected based on a previous survey focused on the detection of strong emitters of SiO 2–1 and 1–0 transitions. As it is well known, SiO is a selective tracer of shocked environments, among these, the outflow phase of the star formation process (Tafalla et al. 2010; Tafalla & Bachiller 2011; Duarte-Cabral et al. 2014; Csengeri et al. 2016).

2.2. Data

Our sample was observed by means of mapping observations covering a 100’ square area in each high-mass star-forming region (HMSFR). Eight of the sources belong to the southern sky and were observed by the APEX telescope with a spectral coverage from 216.681 to 217.680 GHz ($\Delta v = 0.34$ km s$^{-1}$) at $\theta_{\text{HPBW}} \sim 30''$ and with a positional accuracy of 4''. These observations were taken in two observation sessions, the first one during 2010 May 21, 22, 24, and 26, and the second on 2010 September 22 and 26. The remaining three sources belong to the northern sky and were observed with the IRAM-30 m telescope. In this case, the spectral coverage ranges from 216.925 to 217.791 GHz ($\Delta v = 2.76$ km s$^{-1}$) at $\theta_{\text{HPBW}} \sim 11.94''$ with a positional accuracy of 2''. These observations were taken on 2011 February 19 and 2011 March 21 (O. S. Rojas-García et al. 2022, in preparation).

The complete list of these objects is shown in Table 1.

2.3. Reduction and Observational Results

The data cubes were smoothed to a nominal spectral resolution of 2.76 km s$^{-1}$ (our lower spectral resolution), to improve the signal-to-noise ratio and to make a consistent analysis throughout the sample. We identified and avoided the damaged observations along each data cube for all the sources. Then, a baseline was subtracted (in most of the sources the baseline was fitted with a linear curve), and the resulting spectra were converted from $T_A^*$ to $T_{\text{mb}}$ according to their corresponding efficiencies, for IRAM $n_{\text{mb}} = 0.915$ and $n_{\text{mb}} = 0.57$ and for APEX $n_{\text{mb}} = 0.970$ and $n_{\text{mb}} = 0.75$.

We then analyzed the 12 strongest lines observed in most of the objects (common lines among the sample) and did a preliminary identification by visual inspection based on the coincidence of the line emission peak frequency and the nominal frequency expected for molecules in the Splatalogue catalogue. This line identification was later confirmed by means of a local thermal equilibrium (LTE) synthetic model and, after this procedure, we obtained the molecular species listed in Table 2.

The integrated area of the lines was calculated within their full width at zero power (FWZP) widths. Their corresponding uncertainties were computed according to standard formalism: $\sigma_{\text{area}} = \sqrt{N \times \Delta v \times \sigma}$, where $\Delta v$ is the velocity resolution, $\sigma$ is the rms noise level per channel, and $N$ is the number of channels within the integrated velocity range. The resulting values are reported in Table 3.

3. Analysis Method

To propose molecular transition candidates, we have taken the currently accepted procedure for line identification described by Herbst & van Dishoeck (2009) to unequivocally identify new molecules: (i) Rest frequencies are accurately known. To have accurately known frequencies, we only used molecular transitions in consolidated astronomical catalogs employing the spectroscopic database Splatalogue, which is a web-based platform that compiles information of several catalogs such as the Cologne Database for Molecular Spectroscopy (CDMS; Müller et al. 2005), Jet Propulsion Laboratory (Pearson et al. 2005) and the catalog of the National Institute of Standards and Technology (NIST; Lovas 2004), among others. (ii) Observed frequencies of nonblended lines

| Table 1 |
|---|---|---|
| Sources | Coordinates | Distance |
| G034.26+0.15 | 18:53:16.40 | 01:15:07.00 | 3.64 ± 0.36 |
| G034.41+0.24 | 18:53:17.90 | 01:25:25.00 | 3.59 ± 0.36 |
| G035.13−0.74 | 18:58:06.40 | 01:37:01.00 | 2.34 ± 0.38 |
| G237.29−0.58 | 15:53:08.56 | −54:37:05.50 | 3.03 ± 0.65 |
| G328.81−0.63 | 15:55:48.56 | −52:43:08.40 | 2.83 ± 0.65 |
| G333.13−0.43 | 16:21:02.67 | −50:33:13.30 | 3.54 ± 0.65 |
| G337.92−0.48 | 16:41:10.72 | −47:08:07.20 | 3.10 ± 0.65 |
| G340.97−1.02 | 16:54:57.30 | −45:09:04.00 | 2.27 ± 0.65 |
| G351.45+0.66 | 17:20:54.34 | −35:45:03.80 | 0.84 ± 0.65 |
| G351.77−0.54 | 17:26:42.93 | −36:09:20.00 | 0.59 ± 0.65 |
| G353.41−0.36 | 17:30:26.86 | −34:41:50.50 | 3.50 ± 0.65 |

Note. Sources at positive declinations were observed with IRAM whereas at negative declinations, sources were observed with APEX. The distances were obtained from He et al. (2012), Wienen et al. (2012), and Miettinen et al. (2006).

This investigation will be presented in a separate paper, but the results of this survey are already published in a master’s thesis that can be accessed at the following link: https://inaoe.repositorioinstitucional.mx/jspui/handle/1009/851.
Table 2
Full List of the Identified iCOMs after the XCLASS Modeling

| Molecule          | Name                | Frequency (GHz) | Transition Line | Log10 (Afi) | Ei/κ (K) | gE |
|-------------------|---------------------|-----------------|-----------------|-------------|----------|----|
| CH3OCHO           | Methyl formate      | 216.830197      | 18(2.16)–17(2.15) E | −3.8297     | 105.677  | 74 |
| CH3OCHO           | Methyl formate      | 216.838889      | 18(2.16)–17(2.15) A | −3.8297     | 105.666  | 37 |
| CH3OCHO           | Methanol            | 216.945559      | 5(1)–4(2.2)      | −4.9158     | 55.871   | 11 |
| CH3OCHO           | Methyl formate      | 216.965900      | 20(1.20)–19(1.19) A | −3.8149     | 111.481  | 82 |
| CH3OCHO           | Methyl formate      | 216.967994      | 20(1.20)–19(1.19) E | −4.6118     | 111.498  | 82 |
| C2H3C15N          | Ethyl cyanide       | 217.190187      | 12(3.10)–11(2.9) | −4.5314     | 31.953   | ...|
| CH3OCHO           | Methyl formate      | 217.237558      | 7(3.5)–6(1.6)    | −6.5284     | 22.527   | 30 |
| CH3OCHO           | Methyl formate      | 217.298353      | 8(5.4)–8(3.5)    | −6.7361     | 37.837   | 34 |
| CH3OCHO           | Methyl formate      | 217.301326      | 7(3.5)–6(1.6)    A | −6.5259     | 22.511   | 30 |
| 13CH3OH           | Methanol            | 217.399550      | 10(2.8)–9(3.7)   ++ | −4.8153     | 162.413  | 21 |
| (c-C3H7)CH2       | Methylene cyclopentene | 217.420065  | 16(4.13)–16(2.14) | −5.8861     | 102.323  | 165|

Note. The main properties for each detected species are displayed. From left to right each column shows: (1) chemical formula, (2) molecule name, (3) rest frequency, (4) resolved quantum number transition, (5) Einstein emission coefficient $A_{fi}$, (6) upper energy level, and (7) upper degeneracy level.

Table 3
Integrated Area over the Strongest 12 Molecular Emission Lines Detected in Most of the Sources

| Molecule        | Frequency (GHz) | G327.29–0.58 (K km s$^{-1}$) | G328.81+0.63 (K km s$^{-1}$) | G333.13–0.43 (K km s$^{-1}$) | G337.92–0.48 (K km s$^{-1}$) | G340.97–1.02 (K km s$^{-1}$) | G351.45+0.66 (K km s$^{-1}$) |
|-----------------|-----------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|
| CH3OCHO         | 216.830197      | 1.5 ± 0.2                     | ...                           | ...                           | ...                           | ...                           | ...                           |
| CH3OCHO         | 216.838889      | 3.2 ± 0.3                     | ...                           | ...                           | ...                           | ...                           | ...                           |
| CH3OCHO         | 216.945559      | 6.7 ± 0.6                     | 3.9 ± 0.2                     | 2.0 ± 0.1                     | 2.1 ± 0.3                     | 1.5 ± 0.4                     | 4.1 ± 0.1                     |
| CH3OCHO         | 216.965900      | 5.5 ± 0.4                     | 0.8 ± 0.1                     | ...                           | ...                           | ...                           | ...                           |
| CH3OCHO         | 216.967994      | ...                           | 1.0 ± 0.2                     | 0.4 ± 0.1                     | ...                           | ...                           | 1.1 ± 0.2                     |
| SiO              | 217.104984      | 7.0 ± 0.2                     | 13.1 ± 0.4                   | 5.9 ± 0.2                     | 8.8 ± 0.4                     | 4.1 ± 0.3                     | 14.2 ± 0.5                     |
| C2H3C15N         | 217.190187      | 2.4 ± 0.2                     | ...                           | ...                           | ...                           | ...                           | ...                           |
| CH3OCHO         | 217.298353      | 3.2 ± 0.2                     | ...                           | ...                           | ...                           | ...                           | 0.7 ± 0.1                     |
| CH3OCHO         | 217.301326      | ...                           | 1.5 ± 0.2                     | 0.3 ± 0.1                     | ...                           | ...                           | ...                           |
| +13CH3OH         | 217.399550      | ...                           | ...                           | ...                           | ...                           | ...                           | ...                           |
| CH3OCHO         | 217.418711      | ...                           | ...                           | ...                           | ...                           | ...                           | ...                           |
| (c-C3H7)CH2      | 217.420065      | ...                           | ...                           | ...                           | ...                           | ...                           | 0.4 ± 0.1                     |

Note. The labeled iCOMs were added after their confirmation by LTE modeling. Column (1) contains the chemical formula of the corresponding emitter. Column (2) lists their corresponding rest frequency. Columns (3)–(8) display the integrated area $f_i d\nu$ in [K km s$^{-1}$] over the FWZP width for all the sources labeled.

agree with rest frequencies for a single well-determined velocity of the source. To do this, we took the nonblended lines over the 3σ level and measured the central frequency of their peak intensity, and tested if their velocity coincides with the $v_{LSR}$ of the source. (iii) All predicted lines of a molecule based on an LTE spectrum at a well-defined rotational temperature and appropriately corrected for beam dilution are present in the observed spectrum at roughly their predicted relative intensities. This requirement was satisfied by computing an XCLASS model (Möller et al. 2017). In this approach, we tested the proposed iCOM candidates that fit the frequencies and simulated their emission spectra to compare with the observed spectra. Among the proposed iCOMs, the extra species were discarded by their discordance with the observed spectra, e.g., when a molecule generates artificial lines that were not observed in experimental data (i.e., CH2DOH, (CH3OH)$_2$, C$_2$H$_5$CN, CHOCOOH, or HOCHCH-CHO). The final chemical diversity was tested by a classical
nonparametric $\chi^2$ test whose results are presented in Section 3.1. (iv) Simulate the entire spectrum of a source using all identified molecules to check consistency and separate contaminating blended features. This step was also fulfilled with our XCLASS LTE synthetic spectrum, given that after the refinement of the species catalog we recompute the entire synthetic spectrum to compare it to the observed one. Throughout this specific tuning, we also considered the blended species in the observed spectra, taking into consideration the proportions of the dominant and secondary iCOM emitters according to their relative abundances to set up the modeling parameters.

The data cube of each source was centered at these nonblended lines, and their adjacent emission velocity channels were analyzed for congruent spatial distribution, which could be tracing the outflow lobes. To constrain the spatial distribution of the possible outflow component, we also computed maps of the integrated intensity in the blue- and redshifted emission of the line profiles wider than FWHM > 8 km s$^{-1}$. These maps were plotted as intensity isocontours over a background image composed using the color code of red, green, and blue, for the 3.6, 4.5, and 8.0 $\mu$m bands of the IRAC camera of the Spitzer Space Telescope (Cyganowski et al. 2008). This allowed us to see the spatial correlation between the 4.5 $\mu$m band, the green fuzzy emission which gives EGOs their name (Cyganowski et al. 2008), and the iCOM spatial distribution. These maps are described and shown in Section 4.2.

### 3.1. XCLASS LTE Spectral Modeling

To test our line identification method, we modeled the observed averaged spectra within the central 10$''$ of each source utilizing the XCLASS software. The computed grid was run with the following values as free parameters: $T_{\text{rot}}$ (rotational temperature in kelvin), $N_{\text{tot}}$ (total column density in cm$^{-2}$), $v_{\text{width}}$ (velocity width in kilometers per second) and $v_{\text{off}}$ (velocity offset in kilometers per second).

The $T_{\text{rot}}$ was limited from 10 to 500 K, the $v_{\text{width}}$ from 1 to 7 km s$^{-1}$ for the narrow emission lines, and from 5 up to 15 km s$^{-1}$ for the wider ones. The $v_{\text{off}}$ range was from −2 to 2 km s$^{-1}$ away from the $v_{\text{LSR}}$, and the $N_{\text{tot}}$ values vary depending on the species, with the lowest and highest limits going from $1 \times 10^{13}$ to $1 \times 10^{16}$ cm$^{-2}$, in agreement with the typical values reported in other works (Csengeri et al. 2019; De Simone et al. 2020). The size parameter was fixed to 25$''$ as a conservative spatial extension covered by the iCOMs in our maps. The modeling of other simpler molecules was approached similarly, and their details will be discussed in a separate paper.

To establish the chemical diversity present in our bandwidth we took as our archetype the source with more emission lines: G327.29-0.58. A sample plot of our resulting synthetic spectrum as well as the observed one is shown in Figure 1. The complete collection of our XCLASS modeled spectra are displayed in Appendix A.

The resulting physical parameters were computed using an algorithm chain, consisting of a particle swarm optimization method, named Bees (Pham et al. 2005), linked to the Levenberg–Marquardt method (Marquardt 1963), an optimization of the Gauss–Newton and descendant gradient methods (Möller et al. 2017). Additionally, the error calculation was computed employing the Interval Nested Sampling algorithm (Ichida & Fujii 1979).

The goodness of the fit in our LTE models was evaluated by a classic $\chi^2$ test. Using the respective channels in each bandwidth as our degrees of freedom, we were able to compute a critical $\chi^2$ value to reach a p-value of 0.05 significance level in our entire sample. We ensured that our modeled spectra successfully reached this critical $\chi^2$ value, therefore reinforcing the species identification.

### 4. Results: iCOM Emission toward Massive Protostars

#### 4.1. XCLASS Results

Employing the LTE modeling, we were able to identify iCOM emission in our sample within the frequency range 216.681–217.791 GHz. Our numerical approximation allowed us to compute $T_{\text{rot}}$ [K], $N_{\text{tot}}$ [cm$^{-2}$], and $v_{\text{width}}$ [km s$^{-1}$], for the detected iCOMs along our sample. We identified three complex molecules, from CH$_3$OH up to the more complex CH$_3$OCHO and (c-C$_3$H$_2$)CH$_2$, along with the two isotopologues, $^{13}$CH$_3$OH and C$_2$H$_4^{13}$N, associated with the outflow stage in our sample. These findings are reported in Table 4.
4.2. Integrated Maps Results

In order to elucidate the nature of the emission, we inspected the integrated maps of the shifted emission in the broadest iCOMs, looking for high-velocity spatial components. For this, we computed the integrated maps of the blue- and redshifted emissions for iCOMs with an FWHM wider than 8 km s\(^{-1}\) (\(\nu(\text{min})_{\text{wing}} - \nu(\text{min})_{\text{core}} \geq 4.0\) km s\(^{-1}\)). This criterion, corresponding to FWHM \(\sim 3\) km s\(^{-1}\) (for a Gaussian profile assuming FWHM \(\sim 6\sigma\)), is appropriate as it would include previously observed iCOMs associated with outflow environments (with FWHM \(\geq 10\) km s\(^{-1}\), or FWHM \(\geq 3\) km s\(^{-1}\), e.g., Palau et al. 2007; Codella et al. 2015; Burkhardt et al. 2016). This procedure tries to avoid excitation mechanisms related to the central core (Walmsley et al. 1999; Schilke et al. 2001) and reinforces that the emission is produced as a result of outflow activity (Tafalla & Hacar 2013; Li et al. 2019). Thus, looking for elongated distributions along adjacent channels in the data cube could diagnose the emission related to large-scale outflows. This elongation should be more extended than the \(\theta_{\text{HPBW}}\) for each telescope (\(\sim 30.5''\) for APEX and \(\sim 11.0''\) for IRAM) and the minimum detectable offset should be greater than 3 times the pointing accuracy (12'' for APEX and 6'' for IRAM).

After this analysis, we have not found collimated outflow structures traced by the iCOMs. However, we point out that this could be a resolution problem, as the angular resolution reached in our mapping observations is still poor and beam dilution could be affecting the compact and faint structures expected from outflows (Csengeri et al. 2019). Nevertheless, we have found several iCOM maps with the blue- and redshifted emission peaking at offset positions (away from the near-IR peak). This spread emission could be tracing large-scale low-velocity movements, possibly related to bipolar or multipolar outflows. The most prominent example of this behavior is the methyl formate (MF) emission at 217.301 GHz toward G351.77-0.54, which shows an east–west elongated emission; a spatial distribution also observed in its methanol emission (see Figure 2(a)). Nonetheless, we should recall that iCOM emission does not strictly follow the EGO emission, but this association could be plausible considering our low resolution and the spatially spread emission at 4.5 \(\mu\)m shown by this object.

The MF emission at 216.968 GHz is also interesting toward G351.45+0.66, since the blue- and redshifted contour maps show an offset of their peaks, following a north–south direction (see Figure 2(b)). This behavior is also followed by the methanol emission at 216.945 GHz and the MF at 217.298 GHz.

The object G328.81+0.63 also shows an offset of the blue- and redshifted methanol emission at 216.945 GHz in the north–south direction (Figure 3(a)).

The blueshifted emission traced by methanol at 216.945 GHz toward G340.97-1.02 shows a peak following the southeast–northwest direction of the EGO, but at a larger
scale (Figure 3(b)). These morphological features are potential candidates for high-angular-resolution interferometric studies, to avoid the beam dilution and therefore tightly constrain the extended emission of this iCOM.

Other iCOM emission lines are mainly tracing the massive protostar but their closest intensity contour peaks, for both blue- and redshifted emission, are commonly overlapped on the inner parts of the core, and therefore related to the extended envelope. This behavior is appreciated in G327.29-0.58, as its iCOM contour map shows a compact emission that is related to the core and extends up to outer envelope (Figure 4). Even when the contours show a faint east-to-west elongation this appreciation is marginal at our angular resolution.

The whole set of previously mentioned integrated maps are displayed in Figure 10.

### 4.3. iCOM Correlations

The most prominent and nonblended observed iCOM in our bandwidth was the methanol transition at 216.945 GHz, which was observed in all except G035.13-0.74 and G353.41-0.36. From this, we have proposed to test the correlation between their integrated emission and the shock tracer SiO at 217.105 GHz (also present within our bandwidth). We tested their correlation by employing two nonparametric tests, the Pearson and Spearman’s rank correlations, in order to evaluate a linear or a monotonic dependence, respectively. The analysis showed a Spearman rank correlation factor $r_s$ of 0.76, with a corresponding $p$-value of 0.016, and a Pearson coefficient $r_P$ of 0.58, with a $p$-value of 0.10. These two tests have confirmed the relation between these emission lines with a good significance level. And, given that these two lines were observed simultaneously, the systematic errors are expected...
to similarly affect both emission lines, reinforcing this correlation. We plotted this distribution and used a linear regression in log–log space to illustrate this trend, obtaining a slope close to unity, i.e., $y = 0.91x - 0.40$, as is displayed in Figure 5. The only outlier is G327.29-0.58, an ATLASGAL object that differs from the other regions of the sample as it is not an extended object at 4.5 μm, but rather a compact near-IR bright object. This together with a higher chemical activity suggests a late evolutionary stage for G327.29-0.58.

The observed correspondence suggests the enhancement of CH$_3$OH within the shocks. This relation could also be explained because shock conditions boost the desorption of iCOMs, previously synthesized onto the dust and ice mantles. The efficient release of the CH$_3$OH parent molecule to the gas phase could ease other gas-phase reactions and therefore lead to secondary molecules as MF and dimethyl ether (Balucani et al. 2015), iCOMs that are feasibly formed at the temperatures we have measured toward our sample, $\sim$30 K (Minissale et al. 2016; Ceccarelli et al. 2017). The injection of kinetic energy into the grains not only boosts the evaporation through thermal desorption, but also the mobility of radicals in the grains, stimulating movements of the radicals in the dust through the Langmuir–Hinshelwood mechanism (Herbst & van Dishoeck 2009).

Due to the essential role of the evolution of methanol as a precursor of several iCOMs, we analyzed the CH$_3$OH/SiO ratio against the popular time indicator ratio $L_{bol}/M_{dust}$, expecting a decreasing trend as the source evolves (López-Sepulcre et al. 2011; Sánchez-Monge et al. 2013; Csengeri et al. 2016). In this case, we take the values of $L_{bol}$ and $M_{dust}$ from literature (Miettinen et al. 2006; Urquhart et al. 2014; Wienen et al. 2015; Csengeri et al. 2016); but, as $L_{bol}$ was not
reported for all our sources, this results in a sample size of six sources, therefore, we were unable to establish any conclusive result. Nevertheless, we think this relation is a way to measure the proficiency of the shocks releasing the CH$_3$OH into the gas phase as time evolves.

Similarly, we also evaluated the CH$_3$OH/SiO ratio versus the dust mass of the object using the same tests. In this case, we found dust mass data for nine of our sources, which traces an increasing trend with a corresponding correlation values of $r_S = 0.19, p$-value $= 0.61$, and $r_p = 0.19, p$-value $= 0.62$. Both of these correlation coefficients are low, but due to our low-significance $p$-values, we cannot fully establish this null relation.

5. Discussion

5.1. Methanol

The methanol molecule was found in 100% of our sample. This parent molecule in its gas phase is the precursor of several iCOMs (Ceccarelli et al. 2017). It has been proved to be formed on grain surfaces by the hydrogenation of frozen CO by the successive addition of H atoms (Tielens & Hagen 1982; Watanabe & Kouchi 2002; Boogert et al. 2015). The main proposed mechanisms to release this iCOM into the gas phase are cosmic-rays or thermal desorption (Ceccarelli et al. 2017). In our sample, we expect that the shocks themselves could transfer the needed amount of energy into the dust grains to induce evaporation.

Our observed methanol emission lines suggest a high temperature in the gas induced by the shocks, with values going from 126 to 362 K and an average value of 234 ± 78 K. This temperature is close to the dust temperature reported by Fu & Lin (2016) of 250 K toward G34.26+0.15.

These high temperatures along with the observed iCOM chemical richness in our sample are congruent with a C-type shock (Arce et al. 2007; Palau et al. 2017).

To test the shock impact into the CH$_3$OH enhancement, we examined the relation between the $T_{mb}$ integrated emission of CH$_3$OH at 216.945 GHz and SiO at 217.105 GHz. In our sample, this shows an increasing behavior described by a linear Pearson correlation with $r_p$ of 0.78 and a $p$-value of 0.013. This relation suggests that shocks are efficient at boosting methanol to their gas phase. Considering the importance of grain-surface chemistry in methanol formation (Garrod & Herbst 2006; Balucani et al. 2015), we could speculate that the observed methanol enhancement in our sample implies an increase in the dust mass (a property of ATLASGAL sources), which allows the shocks to more easily inject the energy needed to sublimate the methanol from the dust grains. This reinforces the hypothesis that the radiation process is not the sole mechanism needed to generate complex molecules near to forming stars (Arce et al. 2008; Zeng et al. 2018).

Alternatively, this could suggest that SiO and CH$_3$OH are being affected by a common excitation mechanism such as photoevaporation due to an external UV field (Schilke et al. 2001; Palau et al. 2007). In that case, we should also consider and further discriminate if the central integrated line of SiO is under the influence of another excitation mechanism different to shocks, but this topic is beyond the scope of this study.

Methanol abundance enhancement has been observed in several outflows (Bachiller et al. 1995; Palau et al. 2007), and their line-profile wings are significantly lower than that of SiO. This could be due to CH$_3$OH not surviving at velocities as high as those reached by the outflow, or to the fact that high-velocity shocks are required to boost the injection of Si into the gas phase (Guillet et al. 2011).

According to our maps, the spatial distribution of methanol in our sample follows the central 4.5 $\mu$m peak emission, and for most of the cases sustained a nearly circular morphology. Their extension is wide and seems to be related to the central massive protostar, tracing the central ∼20″ away from the central pointing and up to ∼35″ for the more extended sources. This represents a physical scale going from ∼0.1 to 0.5 pc according to the distance of the objects.

In G328.81+0.63, the methanol redshifted emission displays an elongated southeast–northwest morphology, which covers ∼35″ (∼98 × 10$^3$ au) (Figure 3(a)). On the other hand, the blueshifted emission is located along the north–south axis and has a more compact structure. The emission center of these two components shows an offset, which suggests a relationship with an extended component. Further observations with improved angular resolution are necessary to confirm the outflow origin of this molecule.

In G340.97-1.02, the methanol blueshifted emission at 216.945 GHz is elongated in the southeast–northwest direction for more than ∼30″ (Figure 3(b)).

Toward G351.77–0.54, the methanol redshifted emission at 217.418 GHz spreads along the east–west direction for ∼80″ (∼24 × 10$^3$ au), following the central EGO emission (Figure 6(a)). Also, the redshifted emission extends to the south, but this emission is probably related to the compact nearest 4.5 $\mu$m source located just 35″ to the south.

In G351.45+0.66, the methanol blueshifted emission at 216.945 GHz is directed to the southern part from the central pointing for more than ∼40″ (Figure 6(b)). This emission is poorly collimated and it could be more related to the dust continuum emission.
Figure 6. Contour maps of the blue- and redshifted emission of methanol at (a) G351.77-0.54 and (b) G351.45+0.66. The central frequency and physical scale are labeled. The contours show the emission up from 66% + 10% peak emission; the blue and red contours stand for the blue- and redshifted emissions, respectively. The shifted emissions were integrated outward from the central 8 km s\(^{-1}\) to reach the FWZP in the emission line.

5.2. Methyl Formate

MF was found in ~82% of our sample. This molecule is expected to be formed on ices by the reaction of CH\(_3\)O with HCO and O (Ceccarelli et al. 2017), or in the gas-phase reactions starting from dimethyl ether (Balucani et al. 2015). Even at our high detection rate, the correlation of this molecule versus the previously mentioned tracer was avoided as their emission lines among the sources belong to different transitions, and this implies a nonhomogeneous correlation bias.

The lower limit of the computed range of rotational temperatures (40–404 K) is close to the temperatures required by gas-grain chemical models such as in Garrod & Herbst (2006, \(T\)\(_{\text{rot}}\) \(\sim\) 30 K), which explains the formation of iCOMs such as CH\(_3\)OCH\(_3\), CH\(_3\)CHO, and CH\(_3\)CHO. This temperature is needed for the radicals to have sufficient energy to ensure their mobility (Bacmann et al. 2012). Even when this model does not explain the presence of these iCOMs in their gas phase, as previously mentioned in our sample the shocks could be a major contributor to this release.

It is noteworthy that the XCLASS fit gives an average MF \(T\)\(_{\text{rot}}\) value of 184 K, i.e., more than the temperature needed to trigger the gas-phase reactions (Minissale et al. 2016; Ceccarelli et al. 2017).

In ~45% of the sample, the MF emission presents a FWZP ~6 km s\(^{-1}\), not broad enough to overpass our limit to define an outflow origin of the emission. Actually, the integrated emission shows a circular morphology centered on the central massive protostar with an average coverage of 15″.

In contrast, the following examples have shown an outstanding spatial distribution in the MF emission.

In G351.77-0.54 the redshifted emission is extended and seems to follow the eastern emission of the EGO (Figure 2(a)); however, the presence of nearby sources could affect the emission and produce the observed broadening.

The MF emission in G351.45+0.66 at 216.968 GHz shows an offset location of their blue- and redshifted peaks. These peaks are located in a north–south direction through a ~35″ extension (Figure 2(b)), and, in contrast to the other sources in our sample, they do not have a strong 4.5 μm emission, possibly being the less evolved source in our sample (note, however, that this source was selected from the ATLASGAL catalog). The blueshifted emission traced by MF is more compact and elongated than the blueshifted emission of methanol (Figure 6(b)).

In G333.13-0.43, the blue- and redshifted emission is completely offset from our central pointing (see Figure 7). The western emission has an elongated bipolar morphology, which could be tracing an outflow. Due to the fact that this emission is located ~15″ away from the central object and the fact that the near-IR emission is very complex, this MF emission could be associated with nearby sources different to the massive protostar studied here.

5.3. Acetone

Acetone was detected in G034.26+015 and G327.29-0.58. Their emission lines were confirmed by the XCLASS model with three unblended transitions at 216.972, 217.021, and 217.070 GHz, and a faint contribution at 217.191 GHz. In all cases the line intensity is faint and is tracing the central source. The contour maps show a circular extension over ~35″.

Its presence in our XCLASS modeling is justified as it contributes to the optimal fit for faint emission lines in the chemical-rich object G327.29-0.58, improving the significance of the \(\chi^2\) test.

5.4. Ethyl Cyanide

The ethyl cyanide (C\(_2\)H\(_5\)C\(_{15}\)N) isotopologue has been detected in G034.41+0.24 and G327.29-0.58, with two transitions within our bandwidth: one at 216.880 GHz (S/N ~ 3\(\sigma\) in G327.29-0.58, not present in G034.41+0.24), and another at 217.190 GHz (S/N ~ 4\(\sigma\) in G034.41+0.24 and S/N ~ 6\(\sigma\) in G327.29-0.58). Note, however, that the last emission line is blended with a faint CH\(_3\)COCH\(_3\) signature, but this is just a minor contributor to the total observed intensity.

The blue- and redshifted components of the 217.191 GHz transition overlap toward the central region in a spherical
morphology, which relates this iCOM to the central massive protostar (see Figure 8).

5.5. Vinyl Cyanide

We have two sources that show a contribution of C2H3CN: G327.29-0.58 and G351.77-0.54. In both cases the 216.936 GHz line is the only transition within our bandwidth, with this identification being tentative. Nevertheless, this molecule has been already reported toward massive protostars (Codella et al. 2013; Fu & Lin 2016), and is therefore expected. The emission-line profile in both sources is narrow and their contour maps are concentrated to the central massive protostar and exhibit a circular morphology for ∼40″. Their column density is about one order of magnitude lower than the O-bearing iCOMs that have been also reported in other investigations (Csengeri et al. 2019).

5.6. Methyleneencyclopropene

The proposed transition of methyleneencyclopropene (c-C3H2CH2) at 217.428 GHz was only observed toward the most chemically rich source, G327.29-0.58. Since we identified just one transition, this reduces its reliability. The observed emission line is close to 217.428 GHz, and therefore could be related to the more common isotopologue of 13CN, but this species was discarded as its species was discarded as its abundance is too low for a Δv = 1 transition within our bandwidth. Note that this iCOM has not been detected in the ISM5 but it is already listed in CDMS (Müller et al. 2005).

6. Summary and Conclusions

We have reported the detection and identification of six iCOMs toward 11 HMSFRs selected from the ATLASGAL and EGOs catalogs, previously related to outflow activity, enlarging the existing outflow sample associated with iCOMs (van Dishoeck et al. 1993; Garay et al. 2007a; Leurini et al. 2013; Lefloch et al. 2017; Palau et al. 2017). The identified iCOMs are methanol (CH3OH), methyl formate (CH3OCHO), acetone (CH3COCH3), vinyl cyanide (C2H3CN), and ethyl cyanide (C3H7CN).

Our identification of iCOMs has been tested by LTE modeling using the XCLASS software, from which we obtained the physical parameters $N_{tot}$, $T_{kin}$, $v_{width}$, and $v_{offset}$. The agreement between the modeled synthetic spectra and the observed spectra was evaluated by a classical $\chi^2$ test. This test showed a statistically significant concordance as the critical $\chi^2_{crit}$ for a 0.05 $p$-value was underpassed ($\chi^2_{crit} < \chi^2_{obs}$) in all but one of our sources (G340.97-1.02). The observed high abundance of O-bearing molecules points out the expected desorption from the ice mantles due to outflow-induced shocks in these HMSFRs. This premise was evaluated using the ubiquitously observed iCOM transition of methanol at 216.945 GHz. For this, we tested the correlation between the integrated emission of this line versus the integrated emission of the shock tracer SiO at 217.105 GHz. The correlation has shown an increasing linear behavior with a good significance level, which reinforces the premise that shocks could release the iCOMs trapped in the dust and mantles. This desorption could help the proliferation of second-generation iCOMs, since, as the mother precursor, methanol has been proven to trigger the production of other CH3-bearing molecules (Balucani et al. 2015), such as CH3OCHO and CH3COCH3, that were observed in our sample.

The proportional relation between methanol and SiO has been observed in the collection of our spectra (Figure 9). The peak of the methanol line is observed to be smaller than the SiO one, in all but G327.29-0.58, and the integrated line area is always larger for SiO than methanol. This relation could be a key ratio to describe the energetic processes that are taking place in this outflow stage of HMSFRs.

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5 Set of detected molecules in space according to CDMS: https://cdms.astro.uni-koeln.de/classic/molecules.
Other N-bearing species such as vinyl cyanide (C$_2$H$_3$CN) and an ethyl cyanide isotopologue (C$_2$H$_5$C$^{15}$N), have been observed only in three objects: G327.29-0.58, G351.77-0.54, and G034.41+0.24. The identification of vinyl cyanide is not unambiguously confirmed as it only shows one transition in our bandwidth. The column densities of N-bearing species are one to two orders of magnitude lower than the O-bearing CH$_3$OH and CH$_3$OCHO. The column density of vinyl cyanide goes from $3.7 \times 10^{14}$ to $1.7 \times 10^{15}$ and that of ethyl cyanide goes from $8.2 \times 10^{14}$ to $6.4 \times 10^{15}$, whereas the column density of methanol goes from $2.4 \times 10^{15}$ to $4.7 \times 10^{17}$ and that of MF goes from $5.7 \times 10^{13}$ to $3.6 \times 10^{16}$.

Acetone has been confirmed through four emission lines toward G034.26+015 and G327.29-0.58, with column density

![Figure 9](image-url)
Figure 9. (Continued.)
values of $1.5 \times 10^{13}$ and $1.7 \times 10^{15}$, respectively; its abundances are one to two orders of magnitude lower than methanol.

We have proposed a faint line emission produced by methylenecyclopropene. This emission line is observed toward G327.29-0.58 at 217.428 GHz, and could not be modeled by other simpler molecules, since they would produce other high-intensity lines within our bandwidth that we did not detect.

We made maps of the blue- and redshifted emission of the widest iCOM lines. In general, this emission is mainly tracing the central core of the massive protostars. Nevertheless, some of the sources have shown an iCOM extended emission with offset peak positions of the blue- and redshifted emission, covering from 20,000 to 100,000 au; hence, this could be related to movements external to the compact core, such as large-scale low-velocity outflows. Unfortunately, due to our angular resolution, these outflows are strongly affected by beam dilution.

In this context, our sample provides an excellent set of candidates to carry out further studies toward the high-mass-outflow regime. Observations at high angular resolution are highly desirable to avoid the beam dilution of these faint iCOM features. Since iCOMs have been proven to better trace the compact cores of HMSFRs, high-angular-resolution observations could give hints for the innermost mechanisms at work in the formation of massive stars (as in Palau et al. 2017 and Csengeri et al. 2019).

Further studies with a wider bandwidth coverage are desirable to observe more transitions and better constrain the modeling parameters of the identified iCOMs.

A bigger sample is also convenient to further constrain the correlation of methanol toward SiO shocks in HMSFRs.

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Appendix A
XCLASS Spectra

In the following, we present the corresponding spectra toward all of our sample and their corresponding XCLASS best fit. The physical parameters to get the synthesized spectra are shown in Table 4.

Appendix B
Integrated Maps

In Figure 10, we present the complete integrated emission maps over each source at which we have detected wider iCOM lines. The images consist of a three-band composition of the IRAC camera of 3.6, 4.5, and 8.0 μm bands, color coded in blue, green, and red, respectively (Cyganowski et al. 2008) and overplotted contours of the integrated emission of the iCOMs. The contours go from 66% to 90% of the peak emission with a spacing of 10%. The shifted emission was integrated avoiding the central 8 km s$^{-1}$ until the emission reaches its FWZP. Considering that our sample has an average distance of 3.7 kpc, 10″ would correspond to 0.18 pc (a more precise physical scale is labeled for each source).
Figure 10. Integrated maps for the high-intensity iCOM lines on the different transitions.
Figure 10. (Continued.)
Figure 10. (Continued.)
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ORCID iDs
O. S. Rojas-García https://orcid.org/0000-0003-1054-4637
A. I. Gómez-Ruiz https://orcid.org/0000-0001-9395-1670
A. Palau https://orcid.org/0000-0002-9569-9234
S. E. Kurtz https://orcid.org/0000-0002-1084-6275

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Figure 10. (Continued.)