Tracking the Evolutionary Stage of Protostars through the Abundances of Astrophysical Ices

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Abstract

The physical evolution of young stellar objects (YSOs) is accompanied by an enrichment of the molecular complexity, mainly triggered by the heating and energetic processing of astrophysical ices. In this paper, a study of how the ice column density varies across the protostellar evolution has been performed. Tabulated data of H2O, CO2, CH3OH, and HCOOH observed by ground- and space-based telescopes toward 27 early-stage YSOs were taken from the literature. The observational data show that ice column density and spectral index (α), used to classify the evolutionary stage, are well correlated. A 2D continuum radiative transfer simulation containing bare and ice-covered grains at different levels of cosmic-ray processing were used to calculate the spectral energy distributions in different angle inclinations between face-on and edge-on configurations. The H2O:CO2 ice mixture was used to address the H2O and CO2 column density variation, whereas CH3OH and HCOOH are by-products of the virgin ice after energetic processing. The simulated spectra were used to calculate the ice column densities of YSOs in an evolutionary sequence. As a result, the models show that the ice column density variation of HCOOH with α can be justified by envelope dissipation and energetic processing of ice. On the other hand, the ice column densities are mostly overestimated in the cases of H2O, CO2 and CH3OH, even though the physical and cosmic-ray processing effects are taken into account.

Unified Astronomy Thesaurus concepts: Astrochemistry (75); Infrared sources (793); Interstellar abundances (832); Laboratory astrophysics (2004); Star forming regions (1565)

1. Introduction

Star formation begins in very embedded regions of molecular clouds with no measurable flux in the visible light. Nevertheless, the dust heated by the internal young stellar objects (YSOs) emits radiation in infrared (IR) wavelengths. An IR-based classification of protostars has been proposed by Lada & Wilking (1984), Lada (1987), Andre et al. (1993), and Greene et al. (1994) and takes into account the IR excess in the spectral energy distributions (SEDs). Formally, the method calculates the spectral index between 2 and 24 μm given by

\[ \alpha = \frac{d \log F_\lambda}{d \log \lambda}, \]

where λ is the wavelength and \( F_\lambda \) the flux. The physical structure of each evolutionary stage was reviewed by Williams & Cieza (2011), and the relation with α is also discussed: (i) class I objects are characterized by a large spherical envelope and a disk, with no optical or near-IR emission and \( \alpha_{\text{Class I}} > 0.3 \); (ii) class II are T-Tauri objects with an optically thick disk in the visible, but strong UV emission at short wavelengths and \( -0.3 < \alpha_{\text{Class I}} < -1.6 \); and (iii) the last stage is defined by class III objects characterized by a very low IR excess and \( \alpha_{\text{Class III}} < -1.6 \). Because class 0 YSOs are extremely embedded objects, they are not classified by the spectral index. Its classification is, however, given by the ratio between the submillimeter luminosity (\( L_{\text{submm}} \)) and the bolometric luminosity (\( L_{\text{bol}} \)) as proposed by Andre et al. (1993).

In addition to the physical evolution of YSOs, astrophysical ices play an important role in enriching the chemical complexity in the interstellar medium. As shown in Boogert et al. (2015), given the conditions of large H/CO gas ratio, \( T > 20 \, \text{K}, \, n \geq 10^3 \, \text{cm}^{-3}, \) and \( A_V > 1.5, \) the hydrogenation mechanism leads to the saturation of adsorbed atoms on the dust grain, and the H2O, CH4, and NH3 ices are formed. On the other hand, if the H/CO gas ratio is small, \( T < 20 \, \text{K}, \, n \geq 10^3 \, \text{cm}^{-3}, \) and \( A_V > 3 \, \text{mag}, \) the CO accretion rate is increased. Thereafter, surface reactions between CO and oxygen atoms might lead to the formation of CO2 ice. In low-density regions, however, CO2 ice can be formed via CO + OH reactions (Allamandola et al. 1988). In regions denser than \( 10^4 \, \text{cm}^{-3}, \) the catastrophic CO freeze-out leads to the efficient formation of methanol ice at low temperatures via carbon monoxide hydrogenation (Watanabe et al. 2003; Fuchs et al. 2009).

Besides the adsorption mechanisms, chemical enrichment takes place through the thermal and energetic processing of icy compounds. For instance, at around 30 K, the dust grains are warm enough to enable diffusion and recombination of small molecules on the ice, and complex organic molecules (COMs) are formed (Garrod & Herbst 2006; Herbst & van Dishoeck 2009; Caselli & Ceccarelli 2012). On the other hand, the energetic processing of ices is the main route to form complex species as reviewed by Öberg (2016) for the photochemical-induced processes and particles in Boduch et al. (2015) and Rothard et al. (2017).

In order to investigate how the abundance of ice species is related to the evolutionary sequence, tabulated data of ice column density and spectral index of 27 YSOs were taken from Pontoppidan et al. (2008) and Boogert et al. (2008). Additionally, 2D continuum radiative transfer models of protostars in an evolutionary sequence, containing dust grains covered by ice at different levels of processing, was used to simulate how the ice column density varies with the protostar evolution.
The paper is structured as follows: Section 2 shows the source sample addressed in this paper, and Section 3 shows the ice column density decreasing with the spectral index, i.e., as the protostar evolves. Section 4 characterizes the radiative transfer simulations and the ice-dust model, and shows how the laboratory data were employed in the simulations. In Section 5, the results and discussion are shown, and the conclusions are given in Section 6. The Appendix details how the IR range between 5.5 and 7.5 μm was decomposed using Gaussian profiles.

### 2. Source Sample

Table 1 lists the 27 well-known low-mass YSOs addressed in this paper. Both the evolutionary stage and the column densities for these sources were directly taken from Boogert et al. (2008) and Pontoppidan et al. (2008). The authors used a broadband (2.17−24 μm) classification scheme to calculate the spectral index through Equation (1) and cover class 0/I to class I/II. The spectral index error bar was not provided.

| Source        | R.A. (J2000) | Decl. (J2000) | Cloud | $\alpha_{obs}$ | Stage      | Telescope     |
|---------------|--------------|---------------|-------|----------------|------------|--------------|
| IRAS 03245+3002 | 03°27′39″03 | +30°12′59″3   | Perseus | 2.70       | Class 0/I | Spitzer      |
| L1455 SMM 1    | 03°27′43″25 | +30°12′28″8   | Perseus | 2.41       | Class 0/I | Spitzer      |
| IRAS 03271+3013 | 03°30′15″16 | +30°23′48″8   | Perseus | 2.06       | Class 0/I | Spitzer, Keck |
| B1-c           | 03°33′17″39 | +31°09′31″70 | Perseus | 2.66       | Class 0/I | Spitzer      |
| HH 46 IRS      | 08°25′33″78 | −51°00′35″6   | HH 46  | 1.70       | Class 0/I | Spitzer, VLT |
| CRBR 2422.8−3423 | 16°27′24″61 | −24°41′03″3   | Oph    | 1.60       | Class 0/I | Spitzer, Keck |
| SSTc2d J171122.2−272602 | 17°11′22″16 | −27°26′02″3 | B9     | 2.26       | Class 0/I | Spitzer      |
| 2MASS J17112317−2724315 | 17°11′23″13 | −27°24′32″6   | B9     | 2.48       | Class 0/I | Spitzer, Keck |
| CrA IRS 7A     | 19°01′55″32 | −36°57′22″0   | CrA    | 2.23       | Class 0/I | Spitzer, VLT |
| CrA IRS 32     | 20°02′58″69 | −37°07′34″5   | CrA    | 2.15       | Class 0/I | Spitzer      |
| L1014 IRS      | 21°24′07″51 | +49°59′09″9   | L1014  | 1.60       | Class 0/I | Spitzer, Keck |
| L1489 IRS      | 04°04′43″07 | +26°18′56″4   | Taurus | 1.10       | Class I   | Spitzer, Keck |
| HH 300         | 04°26′56″30 | +24°43′35″3   | Taurus | 0.79       | Class I   | Spitzer, Keck |
| DG Tau B       | 04°27′02″66 | +26°05′30″5   | Taurus | 1.16       | Class I   | Spitzer, Keck |
| IRAS 12553−7651 | 12°59′06″63 | −77°07′40″0   | Cha    | 0.76       | Class I   | Spitzer      |
| Elias 29       | 16°27′09″42 | −24°37′21″1   | Oph    | 0.53       | Class I   | ISO          |
| IRAS 17081−2721 | 17°11′17″28 | −27°25′08″2   | B9     | 0.55       | Class I   | Spitzer, Keck |
| EC 82          | 18°29′56″89 | +01′14′46″5   | Serpens | 0.38      | Class I   | Spitzer      |
| SVS 4−5        | 18°29′57″59 | +01′13′00″6   | Serpens | 1.26      | Class I   | Spitzer, VLT |
| R CrA IRS 5    | 19°01′48″03 | −36°57′21″6   | CrA    | 0.98       | Class I   | Spitzer, VLT |
| HH 100 IRS     | 19°01′50″56 | −36°58′08″9   | CrA    | 0.80       | Class I   | ISO          |
| RNO 15         | 03°27′47″68 | +30°12′04″3   | Perseus | −0.21      | Class I/II | Spitzer, Keck |
| IRAS 13546−3941 | 13°57′38″94 | −39°56′00″2   | BHR 92 | −0.06      | Class I/II | Spitzer      |
| RNO 91         | 16°34′29″32 | −15°47′01″4   | L43    | 0.03       | Class I/II | Spitzer, VLT |
| EC 74          | 18°29′55″72 | +01′14′31″6   | Serpens | −0.25      | Class I/II | Spitzer, Keck |
| EC 90          | 18°29′57″75 | +01′14′05″9   | Serpens | −0.09      | Class I/II | Spitzer      |
| CK 4           | 18°29′58″21 | +01′15′21″7   | Serpens | −0.25      | Class I/II | Spitzer      |

Thereafter, the ice column density was calculated using

$$N_{ice} = \frac{1}{A} \int_{\nu_{1}}^{\nu_{2}} \tau_{\nu} d\nu,$$

where $\nu$ is the band strength of a specific vibrational mode and $\nu$ is the wavenumber in units of cm$^{-1}$.

In Boogert et al. (2008), the H$_2$O-ice column density was calculated from the O−H stretching mode at 3.0 μm, or the libration mode at 12.3 μm, after the removal of the contribution of silicate absorption. The bending mode at 6.0 μm has been avoided, as such a band cannot be attributed only to water as reported in Gibb et al. (2000, 2004) and Keane et al. (2001). Methanol-ice column density was derived from the absorption features at 3.54 and 9.7 μm. Formic acid (HCOOH) has many vibrational modes in the IR, but its column density was calculated from the band at 7.25 μm, as the other modes are blended with H$_2$O and other alcohols such as ethanol and methanol. The carbon dioxide ice column density in Pontoppidan et al. (2008) was calculated from the bending mode at 15.2 μm. Table 2 shows the column densities calculated for the ices mentioned above.

### 3. Correlations between Ice Column Density and Spectral Index

Figure 1 shows the ice column density and the spectral index ($\alpha$) for class 0/I, class I, and class I/II, given by the black squares, red circles and blue triangles, respectively. H$_2$O is the most abundant ice toward all the sources, followed by CO$_2$, CH$_3$OH, and HCOOH, which agree with the expected abundances shown by Öberg et al. (2011).
Table 2
Ice Column Density of the Source Sample

| Source            | \( N_{\text{H}_2\text{O}} \) | \( N_{\text{CO}_2} \) | \( N_{\text{CH}_3\text{OH}} \) | \( N_{\text{HCOOH}} \) |
|-------------------|-----------------|-----------------|-----------------|-----------------|
|                   | \( (10^{18}\text{ cm}^{-2}) \) |     |     |     |
| IRAS 03245+3002   | 39.31 ± 5.65\( ^b \) | ... | 3.85 | 0.47 |
| L1455 SMIM 1      | 18.21 ± 2.82\( ^b \) | 6.34 ± 0.44 | 2.45 | 0.60 ± 0.02 |
| IRAS 03271+3013   | 7.69 ± 1.76\( ^b \) | 1.53 ± 0.09 | 0.43 | 0.19 |
| B1–c              | 29.55 ± 5.65\( ^b \) | 2.1 | 0.35 ± 0.01 |
| HH 46 IRS         | 7.79 ± 0.77 | 2.16 ± 0.01 | 0.42 ± 0.01\( ^d \) | 0.21 ± 0.01 |
| CRBR 2422.8–3423 | 4.19 ± 0.41 | 1.05 ± 0.01 | 0.38 | ... |
| SSTc2d J171122.2–272602 | 13.94 ± 2.92\( ^b \) | ... | 0.18 | 0.41 ± 0.03 |
| 2MASS J17112317–2724315 | 19.49 ± 0.23\( ^b \) | ... | 0.62 | 0.48 ± 0.16 |
| CaA IRS 7A        | 10.89 ± 1.92\( ^b \) | 1.96 ± 0.12 | 0.41 | ... |
| CaA IRAS 32       | 5.26 ± 1.88\( ^b \) | 1.87 ± 0.21 | 0.95 | ... |
| L1014 IRS         | 7.16 ± 0.91\( ^b \) | ... | 0.22 ± 0.05 | ... |
| L1489 IRS         | 4.26 ± 0.51 | 1.62 ± 0.02 | 0.21 ± 0.01 | 0.12 |
| HH 300            | 2.59 ± 0.25 | ... | 0.17 | 0.06 |
| DG Tau B          | 2.29 ± 0.39 | 0.54 | 0.13 | 0.07 |
| IRAS 12553–7651   | 2.98 ± 0.56\( ^b \) | 0.61 ± 0.01 | 0.08 | 0.05 |
| Elias 29          | 3.04 ± 0.30 | 0.84 ± 0.06 | 0.14\( ^d \) | 0.04 |
| IRAS 17081–2721   | 1.31 ± 0.13 | ... | 0.04\( ^d \) | 0.03 |
| EC 82             | 0.39 ± 0.07 | 0.25 ± 0.01 | 0.05 | 0.01 |
| SVS 4–5           | 5.65 ± 1.13 | 1.72 ± 0.05 | 1.41 ± 0.19\( ^d \) | ... |
| R CaA IRS 5       | 3.58 ± 0.26 | 1.42 ± 0.02 | 0.23 ± 0.04 | 0.15 |
| HH 100 IRS        | 2.45 ± 0.24 | ... | 0.23\( ^d \) | 0.06 |
| RNO 15            | 0.69 ± 0.06 | 0.25 ± 0.01 | 0.03\( ^d \) | 0.04 |
| IRAS 13546–3941   | 2.07 ± 0.21\( ^b \) | 0.87 ± 0.02 | 0.08 | ... |
| RNO 91            | 4.25 ± 0.36 | 1.16 ± 0.02 | 0.24 | ... |
| EC 74             | 1.07 ± 0.18 | 0.30 ± 0.05 | 0.1\( ^d \) | 0.03 |
| EC 90             | 1.69 ± 0.16 | 0.54 ± 0.05 | 0.11 ± 0.01 | 0.06 |
| CK 4              | 1.50 ± 0.01 | 0.20 ± 0.01 | ... | ... |

Notes.

\( ^a \) Taken from Boogert et al. (2008).

\( ^b \) \( N_{\text{H}_2\text{O}} \) calculated from the \( \text{H}_2\text{O} \) libration mode at 13.6 \( \mu \text{m} \) using the band strength \( A_{\text{H}_2\text{O}} = 2.8 \times 10^{-17} \text{ cm molecule}^{-1} \). The stretching mode at 3 \( \mu \text{m} \) was used in the other cases.

\( ^c \) Taken from Pontoppidan et al. (2008).

\( ^d \) The column densities were calculated from the absorption features at 3.53 \( \mu \text{m} \), using the band strength of \( A_{\text{CH}_3\text{OH}} = 5.6 \times 10^{-18} \text{ cm molecule}^{-1} \). The absorption at 9.7 \( \mu \text{m} \) was used in the other cases, assuming the band strength of \( A_{\text{CH}_3\text{OH}} = 1.6 \times 10^{-17} \text{ cm molecule}^{-1} \).

\( ^e \) The column densities were calculated from the absorption features at 7.25 \( \mu \text{m} \), using the band strength of \( A_{\text{HCOOH}} = 1.5 \times 10^{-17} \text{ cm molecule}^{-1} \).

Very early stages of the protostellar evolution are dominated by a cold envelope under gravitational collapse. Jørgensen et al. (2009) found evidence of envelope dissipation, estimated from the increase in the disk–envelope mass ratio in 10 class 0 and class I systems. Such a trend has also been observed in Andersen et al. (2019) toward the Perseus Molecular Cloud, where they also found strong evidence of the disk growth at the class 0 stage.

Because the ices are formed onto dust grains in cold regions, the dusty envelope dissipation might lead to the ice column density decreasing, as suggested in Figure 1. In order to make the comparison easier between \( N_{\text{ice}} \) and \( \alpha \) in this paper, an exponential function given by the Equation (3) has been assumed:

\[ N_{\text{ice}} = N_{\text{plateau}}^{\text{ice}} + s \cdot e^{-\alpha t}, \]  

(3)

where \( N_{\text{plateau}}^{\text{ice}} \) is the ice column density plateau toward the line of sight, \( s \) is a scale factor of the amplitude variation in the \( y \)-axis, and \( \alpha \) the decrease rate (positive because the \( x \)-axis is inverted). As the presence of foreground clouds toward star-forming regions has been reported in the literature (Boogert et al. 2002b; Pontoppidan et al. 2005; van Dishoeck et al. 2011; Smith et al. 2015), the term \( N_{\text{plateau}}^{\text{ice}} \) aims to take into account this factor. To compare the goodness of fit among the ice species, the reduced \( \chi^2 \) was used, given by \( \chi^2_n = \chi^2 / (n - m) \), where \( n \) is the number of data points and \( m \) is the degrees of freedom. Acceptable fits require \( \chi^2_n < 1 \).

Table 3 shows the parameters obtained from the exponential fit as well as the \( \chi^2_n \). The \( N_{\text{plateau}}^{\text{ice}} \) and \( N_{\text{plateau}}^{\text{H}_2\text{O}} \) are in agreement with the ice column densities in foreground clouds proposed in the literature. In fact, Boogert et al. (2000) suggest that only 30% of the ice column density observed toward the Elias 29 protostar belongs to the object itself, whereas a plateau of around \( N_{\text{H}_2\text{O}} \approx 2 \times 10^{18} \text{ cm}^{-2} \) is hosted by foreground clouds. Toward CRBR 2422.8–3423, Pontoppidan et al. (2005) estimate that, at least, 50% of the \( \text{H}_2\text{O} \) ice column density lies in foreground clouds, namely, \( N_{\text{H}_2\text{O}} \leq 1.8 \times 10^{19} \text{ cm}^{-2} \). For the face-on class II YSO 2MASSJ 1628137 in Taurus, the estimated ice column density is about one order of magnitude lower than for the previous class I objects, i.e., \( N_{\text{H}_2\text{O}} \approx 0.2 \times 10^{18} \text{ cm}^{-2} \) (Aikawa et al. 2012). In the case of \( \text{CO}_2 \) ice, the estimated column density in foreground clouds are \( N_{\text{CO}_2} \approx 2 \times 10^{17} \text{ cm}^{-2} \) in Elias 29 (Boogert et al. 2000) and \( N_{\text{CO}_2} \approx 6 \times 10^{17} \text{ cm}^{-2} \) (Pontoppidan et al. 2005) in CRBR 2422.8–3423. In the case of \( \text{CH}_3\text{OH} \) and HCOOH, the derived parameters must be used with caution due to the poor fit indicated by the \( \chi^2_n \).
From the decrease rate ($r$), one can observe that CO$_2$ ice decreases slower than H$_2$O with protostellar evolution. Due to the lower thermal desorption of pure CO$_2$ ($\sim 75$ K), the most probable scenario in the interstellar medium is that C-rich molecules are trapped in a H$_2$O-ice matrix. In this case, part of the CO$_2$ ice is only released back to the gas phase at around 150 K, which characterizes the H$_2$O temperature desorption (Collings et al. 2004). In addition, CO$_2$ ice can be formed via a surface reaction between OH and CO as shown by Allamandola et al. (1988). As a consequence, in an environment dominated by radiation, CO$_2$ is slowly destroyed compared to H$_2$O because of its re-formation mechanism from water photoproducts. Regarding formic acid and methanol, although the fits indicate that their

![Figure 1](image)

**Figure 1.** Column density correlation of the frozen molecules H$_2$O, CO$_2$, CH$_3$OH, and HCOOH with the spectral index ($\alpha$). Panel (a) shows a schematic representation of the structure of a YSO for each evolutionary stage (Persson 2014). The colors and symbols in panels (b)–(e) represent the evolutionary stages of protostars, and the green shaded area corresponds to the 99% confidence interval. The exponential fit is shown by the solid black line. Panel (f) compares all fits for the normalized data.

| Ice Specie | $N_{\text{H}_2\text{O}}^{\text{plateau}} \times 10^{18}$ (cm$^{-2}$) | Scale Factor ($s$) $\times 10^7$ (cm$^2$) | Decrease Rate ($r$) ($\alpha$) | $\chi_r^2$ |
|------------|-------------------------------------------------|----------------------------------|-------------------------------|----------|
| H$_2$O     | 2.9 $\pm$ 0.4                                   | 9.7 $\pm$ 2.7                   | 3.0 $\pm$ 0.2                 | 4.26     |
| CO$_2$     | 0.3 $\pm$ 0.1                                   | 1.3 $\pm$ 0.4                   | 1.5 $\pm$ 0.1                 | 0.15     |
| CH$_3$OH   | 0.2                                             | 4.6                              | 4.2 $\pm$ 0.1                 | 0.08     |
| HCOOH      | $-0.03$                                         | 0.1                              | 0.64 $\pm$ 0.01               | 0.01     |

**Note.**

* Due to the poor fitting with the exponential function, the error bars of the derived parameters are not shown.

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respective ice column densities drop slower and faster with the spectral index, this result can be called into question due to the poor exponential fit. If, however, this is a real effect, a likely explanation is the efficient HCOOH formation in both gas and solid phases compared to CH$_3$OH (Agündez & Wakelam 2013). To allow easier comparison between all fits, Figure 1(f) shows the normalized ice column densities for the four ices addressed in this paper.

In order to address if the envelope dissipation, viewing angle, and energetic processing of ices are able to provide a likely explanation for the ice column density variation shown in Figure 1, a computational model of YSOs at different stages surrounded by dust and ice was employed in this paper. To mimic the energetic processing of the ices across the protostellar evolution, laboratory data of H$_2$O:CO$_2$ taken from Pilling et al. (2010) and Rocha et al. (2017) have also been used.

4. Models of YSOs Surrounded by Dust and Ice

Whitney et al. (2003) describes the physical evolution of YSOs in an evolutionary sequence using the 2D radiative transfer code HO-CHUNK. In this paper, however, the models used by Whitney et al. are employed as a template, and laboratory data of energetically processed ice are included in the simulations to address how the ice column density changes during the protostellar evolution. The physical parameters adopted in these simulations and the dust-ice model are described in the next sections.

4.1. Template Models

All models share the same parameters of the central star, namely, $R(R_\odot) = 2.09$, $T(K) = 4000$ K, $M(M_\odot) = 0.5$, and $L(L_\odot) = 1.0$. A flared disk in hydrostatic equilibrium is set by the density profile below:

$$\rho_{\text{disk}} = \rho_0 \left(1 - \frac{R_e}{\bar{w}}\right)^3 \left(\frac{R_e}{\bar{w}}\right)^{\alpha} \exp \left\{ -\frac{1}{2} \left(\frac{z}{h(\bar{w})}\right)^2 \right\}, \quad (4)$$

where $\bar{w}$ is the radial coordinate in the disk midplane, and $h = h_0(\bar{w}/R_e)^\beta$ is the scale height. It assumed in all models that $\alpha = 2.25$, $\beta = 1.25$, and an inner disk scale height $h_0 = 0.01$. The infalling envelope is characterized by Ulrich’s density structure (Ulrich 1976), given by

$$\rho_{\text{env}} = \frac{M_{\text{env}}}{4\pi} \left(\frac{GM_e}{R_e^2}\right)^{-1/2} \left(\frac{r}{R_e}\right)^{-3/2} \times \left(1 - \frac{\mu}{\mu_0}\right)^{-1/2} \left(\frac{\mu}{\mu_0} + \frac{2\mu_0^2 R_e}{r}\right)^{-1}, \quad (5)$$

where $M_{\text{env}}$ is the envelope mass infall rate, $R_e$ the centrifugal radius, and $\mu = \cos(\theta)$. $\theta$ is the angle from the axis of symmetry and $\mu_0$ is the cosine polar angle of a streamline of infalling particles for $r \rightarrow \infty$ given by

$$\mu_0^3 + \mu_0 \left(\frac{r}{R_e} - 1\right) - \mu \left(\frac{r}{R_e}\right) = 0. \quad (6)$$

The evolutionary sequence is simulated by varying the parameters in Equations (4)–(6) for the disk and envelope, as shown in Table 4. The values used for each evolutionary stage were constrained from previous observational and theoretical works as pointed out by Whitney et al. (2003). Briefly, these models start with the central protostar surrounded by a massive envelope with a high infall rate, which decreases across the evolution. The disk mass remains constant from class 0/1 to class II, and decreases by six orders of magnitude in class III. Both inner and outer disk and envelope radius are the same as in Whitney et al. (2003). As expected from the images of molecular outflows, the cavity angle increases with age, whereas the cavity density decreases (Padgett et al. 1999; Mottram et al. 2017; de Valon et al. 2020).

4.2. Dust and Ice Properties

The dust properties in the models change between the envelope, outflow cavity, upper disk layers, and midplane regions. Because physical processes such as settling and coagulation (Henning & Semenov 2013) might take place at the midplane, millimeter-size grains (~1 mm) were placed in dense regions given by $n_H > 10^8$ cm$^{-3}$. The upper disk layers ($n_H < 10^8$ cm$^{-3}$) were populated by grains with size between 1 and 10 $\mu m$. For the envelope, a size distribution between 0.5 and 1.0 $\mu m$ was adopted, whereas a fixed size of 0.1 $\mu m$ was used for the cavity region. It should be mentioned that all these dust models are already available in the HO-CHUNK code.

The ice mantle is the new part in these models, compared to the original paper. Although the authors have also used H$_2$O ice in their models, this paper employs an ice composition given by the mixture of H$_2$O:CO$_2$ (1:1) processed by $^{58}$Ni$^{13+}$ ions from its pristine composition: (i) fluence 0 (virgin ices), (ii) fluence $1 \times 10^{12}$ ions cm$^{-2}$ (first processing), and (iii) fluence $1 \times 10^{13}$ ions cm$^{-2}$ (second processing) obtained from Pilling et al. (2010; see Figure 2(a)). As calculated by Drury et al. (2000) and Shen et al. (2004), the $^{58}$Ni flux is around $6.4 \times 10^{-9}$ cm$^{-2}$ s$^{-1}$, which corresponds to a relative time to the virgin ice of $5 \times 10^6$ yr and $5 \times 10^7$ yr for fluences 1 and 2, respectively. These two irradiated ice spectra were selected from the whole range of experiments because they cover the timescales of class I and class II YSOs. The inner panels in Figure 2(a) show the ice features that will be used to calculate the ice column densities, except the range between 14 and 17 $\mu m$. It is important to note from the shape of the O–H stretching mode that no water crystallization is induced by cosmic-ray (CR) processing, although the ice segregation is evident from the formation of the double peak of the CO$_2$ feature at 15 $\mu m$.

Although the CO$_2$/H$_2$O ratio is expected to be around 30% toward low-mass YSOs (Oberg et al. 2011), a higher ratio is used in this paper. Even though this high CO$_2$ fraction is unlikely for the star-forming regions, it was used in the experiments to maximize the formation of C-bearing products. However, as a product of the ice processing in the experiments, carbon monoxide ice was only detectable in the laboratory spectrum after the first irradiation dose, thereby being destroyed at high fluences. Given this unique detection of CO ice in the selected experiments used in this paper, its chemical evolution with the evolutionary stage of YSOs was not addressed in this paper. Another sample containing CO in the initial mixture could also be explored. The effective interaction limit of other ionizing agents such as UV and
X-rays is much lower compared to CRs (Indriolo & McCall 2013) in dense regions such as the protostellar envelope. CR processing, on the other hand, might remove electrons from the inner shells of atoms or excite H2 molecules, leading to an induced X-ray and/or UV radiation field.

The optical constants of the H2O:CO2 ices were taken from Rocha et al. (2017) (Figures 2(b)–(c)). In order to create an ice-covered dust model for the envelope and midplane, the Maxwell–Garnett effective medium theory (Bohren & Huffman 1983) and the Mie theory were employed to calculate the absorption opacity, albedo (ω), and anisotropy parameter (g), as shown in Figures 2(d)–(a) for the outflow cavity, envelope, disk upper layer, and disk midplane.

The position of the ices in the disk and envelope was defined via an interactive procedure by calculating the dust temperature using the Monte Carlo method. As a first approach, the dust temperature was calculated for all models, without ice components. The calculated temperature was used to replace bare grains by covered grains, using the water desorption temperature. Because the new temperature distribution is different compared to the models using bare grains, the simulations were repeated until the temperature converges for values around 5% of the difference between the prior and posterior calculations, as also done in Pontoppidan et al. (2005).

Three models of YSOs including ices are addressed in this paper. The virgin-ice model describes a physical evolution without chemical evolution because the ices are not processed during the evolutionary sequence. The first-processing model assumes an ice mantle slightly processed by external CRs, whereas the second-processing Model 3 is given by ices highly processed by CRs.

### 4.3. Density and Temperature Profiles

Figure 3 show the temperature and density distribution for the evolutionary sequence between class 0/I and class III. The left panels in each box is a zoom-in of the right ones. The left box displays the density distribution that ranges between $10^{-18.5}$ and $10^{-19.5}$ g cm$^{-3}$, i.e., $10^4$–$10^5$ cm$^{-3}$ in class 0/I and I, and is extended to large scales ($R_{\text{out}} = 5000$ au). In class II, this density variation is distributed in a disk scale until 300 au. The right box shows the dust temperature distribution in each model, ranging from $T \sim 10000$ K near the central star to $T \sim 10–30$ K at the envelope region. At such a low temperature, the outer envelope is still cold enough to host ice-covered dust grains, which can be processed by the interstellar radiation field (ISRF). For very embedded YSOs, the UV field might not dominate the synthesis of molecules, unless it is 40 times higher than the typical ISRF of the interstellar medium (Rocha & Pilling 2018). In that case, the chemistry is dominated by a low-temperature reaction, such as CR-induced processes.

In the disk region, the temperature in the midplane remains at around 15–20 K in all evolutionary stages due to UV shielding caused by the dust grains. In the upper disk layers, on the other hand, the temperature increases as the envelope mass decreases because of the accretion process and the temperature is around 100 K for class I and 300 K for class II at a radius of 100 au. As a consequence, the major part of the class I disk might host ices because the H2O freeze-out occurs at temperatures below 150 K (Collings et al. 2004). The ice reservoir in class II disks is therefore reduced compared to the previous evolutionary stage.

Figure 4 shows the dust temperature across the midplane for all the stages, and the H2O freeze-out limit is indicated. The models show that the water snow line moves inward until class II, and outwards for class III, which is in agreement with previous works (Kennedy & Kenyon 2008; Baillié et al. 2015), and would benefit the formation of giant planets at around 5 au without significant migration. Quantitatively, Kennedy & Kenyon (2008) and Baillié et al. (2015) suggest that the water snow line changes from 3.5 au (class 0/I) to 1.7 au (class II) and from 2.3 to 1.5 au (same stages), respectively. In this work, the snow line is closer to the central star compared to previous models, because it changes from 2 au in class 0/I to 0.9 au in class II. Despite the number difference, the same trend and the reduction by a factor of 2 are still observed. It is worth noting, however, that snow lines might temporarily move outward in the disk in the scenario of inside-out collapse (Zhang & Jin 2015; Cieza et al. 2016).

### 4.4. SEDs

Figure 5 shows the SEDs of the simulated protostars at 10 inclination angles ($i$). Because we are looking at the ice features between 2.5 and 8.0 μm, a small range of wavelengths is shown, although the radiative transfer simulation was performed for $0.1 \mu$m $\leq \lambda \leq 1000 \mu$m. In order to get the best signal-to-noise ratio due to the Monte Carlo noise in the simulation of dense regions, $4 \times 10^7$ photons were used in the models. The SEDs show strong absorption bands which are associated with the H2O ice (3 and 6 μm), CO2 ice (4.27 μm), silicate (9.8 μm), and less noticeable in this scale shown, the contribution of complex molecules between 5.5 and 8.0 μm in the first- and second-processing panels (see Figure 9). The silicate feature is seen in
Figure 2. Laboratory data and opacity models. Panels (a)–(c) show the absorbance and optical constants $n$ and $k$ at different fluences. The inner figures in panel (a) show different regions of the absorbance spectrum. Panels (d)–(o) show the dust parameters used in the models where the temperature is below 150 K and for different regions in the YSOs (see details in the text).
absorption for all inclinations in the class 0/I stage, whereas it turns to an emission profile at a pole-on inclination in class I due to the low optical depth to the source of emission. Such an emission profile is more pronounced for $i \leq 60^\circ$ at class I/II because the envelope mass decreases by a factor of 5 from the previous stage to the new stage in these models. The ice profiles in
class 0/I and class I are always observed in absorption at all inclinations. In class I/II, on the other hand, the ice absorption is only seen between \( i = 20^\circ \) and \( i = 60^\circ \). In extremely edge-on (\( i = 80^\circ \) and \( i = 90^\circ \)) and face-on (\( i = 20^\circ \) and \( i = 10^\circ \)) inclinations, the ice features are weak. It is worth noting that the intensity of the ice absorption does not vary monotonically with the viewing angle, namely, from edge-on to face-on inclination as will be discussed in Section 5.

In addition to the SEDs, the continuum emission was also calculated for all 90 spectra described here. As discussed in Boogert et al. (2008), determining the best baseline is not trivial in real objects, and very accurate modeling is required to determine how the different regions of the disk or disk envelope contribute to the continuum emission. In this paper, however, all SED components are known and were used to calculate the correct baseline for each model. For instance, Figure 6 shows the continuum given by the dashed line for SEDs in the model of virgin ices in Figure 5. The same procedure was employed for the models of first and second processing.

5. Results and Discussion

5.1. Ice Optical Depths

The optical depth (\( \tau \)) for the models of virgin and processed ices, relative to H2O ice (\( \sim 3 \mu m \)), CO2 ice (\( \sim 4.27 \mu m \)), and the complex molecules between 5.5 and 7.5 \( \mu m \), was calculated at 10 inclinations between face-on and edge-on angles using the equation described in Section 2. Figures 7–9 show the ice features for each case. The optical depth variation with inclination is due to the relation between optical depth and density along the line of sight. One can note from Equation (5) that \( \rho_{\text{env}} \) decreases with radius \( r \), but also with \( \cos \theta \), namely, from the midplane toward the cavity region. Because the optical depth is given by \( d\tau = -\kappa \rho \cos \theta ds \), where \( \kappa \) is the opacity, \( \rho \) the density, and \( ds \) the optical path, \( d\tau \) decreases if \( \rho_{\text{env}} \) also does. Furthermore, the small variation observed in class 0/I compared to class I and class I/II is due to the negligible envelope density variation with the polar angle for small cavity apertures. In class I and class I/II, however, the ice optical depth does not vary monotonically between \( i = 90^\circ \) and \( i = 60^\circ \). It is evident that the inclination of \( i = 70^\circ \) shows the deepest ice bands due to a geometric effect as previously shown by Pontoppidan et al. (2005). Due to the large optical depth through the midplane, the IR source at edge-on inclination is dominated by a small fraction of scattered IR photons toward high angles. However, at an inclination above the disk-opening angle, the stellar emission itself dominates and the entire envelope is probed, leading to the deepest ice bands in the spectrum. Below \( i = 70^\circ \) in the models shown in this paper, the ice column density decreases monotonically until pole-on inclinations.

The H–O stretching vibrational mode seen in Figure 7 between 2.7 and 3.6 \( \mu m \) is usually reported in YSOs as containing a red wing at longer wavelengths caused by scattering on large ice-coated grains (Boogert et al. 2000) and the absorption by ammonia hydrates (Hagen et al. 1983). The absence of this effect in the synthetic spectra does not argue in favor of any of these two cases as neither large grains nor N-containing species were included in the ice-dust model for the envelope. It is also true that this water vibrational mode shows evidence of crystallization if the ice is heated above 100 K. However, as noticed in Figure 2(a), CR processing is not enough to induce discernible structural changes in the ice matrix as seen from the IR spectrum.

In Figure 8, the C–O stretching mode of CO2 ice at around 4.27 \( \mu m \) is shown. As discussed in Boogert et al. (2002a) and Ehrenfreund et al. (2001), the position and width of this peak change with the grain geometry and its fraction in the ice matrix. As pointed out by Ehrenfreund et al., this vibrational mode presents a narrow profile if CO2 ice is less abundant in a polar matrix compared to pure ice. For instance, the fraction of 14% of CO2 ice in a H2O matrix fits the CO2 band at 4.27 \( \mu m \) better for the class I YSO Elias 29 compared to the pure CO2 ice adopted by Rocha & Pilling (2015). The CO2 analysis at \( \sim 15 \mu m \) from Spitzer observations (Pontoppidan et al. 2008) has strongly suggested that two-thirds of CO2 absorption features is due to carbon dioxide ice diluted in H2O ice, whereas one-third is likely due to CO:CO2 mixture. Ehrenfreund et al. (1997) show a narrowing of the CO2 absorption feature if mixed in CO ice at fractions below 26%. As a consequence, if the band strength (\( A \)) is kept constant, this effect would lead to lower column density values as it depends on the integrated optical depth as seen in Equation (2). Nevertheless, this is hard to verify because the \( A \) for CO2 diluted in CO ice is unknown.

Figure 9 shows the optical depth between 5.5 and 7.5 \( \mu m \) that is usually associated with the presence of COMs such as HCOOH, CH3OH, CH3CHO, and CH3CH2OH (Schutte et al. 1996; Gibb et al. 2000; Keane et al. 2001; Boogert et al. 2008, 2015). In this work, we highlight the contribution of methanol and formic acid at 6.75 \( \mu m \) and 7.24 \( \mu m \), respectively, as a result of the ice processing by CRs. Addressing the ice components in this spectral range is rather difficult as the IR profile of complex molecules changes with the chemical environment inside the ice matrix. Boogert et al. (2008) suggest a decomposition method by removing the pure water contribution and fitting the residual with five independent absorption components obtained from the combination of different YSO spectra. Each component might have multiple carriers due to blended vibrational modes, such as, for example, CH3OH and NH4+ at 6.85 \( \mu m \). In this paper, however, a simple
Figure 5. SED of the YSOs simulated in this paper for the class 0/I–I/II at 3 levels of ice processing and 10 inclinations.
Gaussian decomposition of the spectrum by using five components is employed in order to isolate the contribution of CH$_3$OH and HCOOH, whose position in the spectra is shown in Pilling et al. (2010). The Appendix shows the Gaussian decomposition method, where the components due to CH$_3$OH and HCOOH ice are shown.

**Figure 6.** SED for the model of virgin ices, where the continuum line is also indicated by the dashed lines. The colors indicate the inclinations.
Figure 7. Optical depth of H$_2$O ice at around 3 μm shown for class 0/I–I/II at 3 levels of ice processing and 10 inclinations as indicated by the color bar.
Figure 8. Optical depth of CO$_2$ ice at around 4.27 μm shown for class 0/I–I/II at 3 levels of ice processing and 10 inclinations as indicated by the color bar.
In order to avoid blending effects, the H$_2$O and CO$_2$ column densities were calculated from the bands at 3 and 4.27 μm shown in Figures 7–9, using Equation (2). In the case of CH$_3$OH (6.85 μm) and HCOOH (7.25 μm), the spectral decomposition method described in the Appendix was used. The band strengths adopted for each vibrational mode were $A_{H_2O} = 2.6 \times 10^{-16}$ cm molecule$^{-1}$, (Hagen et al. 1981), $A_{CO_2} = 7.6 \times 10^{-17}$ cm molecule$^{-1}$ (Gerakines et al. 1995), $A_{HCOOH} = 1.5 \times 10^{-17}$ cm molecule$^{-1}$ (Park & Woon 2006), and $A_{CH_3OH} = 1.2 \times 10^{-17}$ cm molecule$^{-1}$ (Hudgins et al. 1993). This paper assumes a constant $A$ for the three models, although the band strength is a sensitive physicochemical parameter to the ice composition. As shown by Öberg et al. (2007), the band strength of the H$_2$O bulk stretch at 3 μm drops linearly in the H$_2$O:CO$_2$ ice mixture, compared to pure H$_2$O. However, in the scenario of energetic processing, where several species are formed, determining an accurate band strength is still an open problem in astrochemistry.

Figure 10 shows the synthetic column density (N) of the ices X: H$_2$O, CO$_2$, CH$_3$OH, and HCOOH as a function of the spectral index calculated for the model of virgin ices, first processing, and second processing, shown by the filled circles, squares, and triangles, respectively. The colors indicate the inclination angle as given by the color bar at the bottom of the figure. The virgin-ice model represents a scenario where the ice column density varies only due to the envelope dissipation and angle inclination. The two processing models, on the other hand, simulate a case where the ice column density decreases due to the physical effects in the virgin-ice model, as well as due to energetic processing of ice. Due to the ambiguity in the envelope mass with the spectral index shown by Crapsi et al. (2008), the spectral index ($\alpha$) to
characterize the evolutionary stage of YSOs in this paper is kept constant for each evolutionary stage and processing level. For class 0/I, the adopted $\alpha$ values are 2.8, 2.6, and 2.4 for the virgin ice, and first and second processing, respectively. The small offset is only to allow better readability. At class I, the $\alpha$ values are 1.4, 1.1, and 0.9, whereas for class I/II, it is assumed to be 0.2, 0, and $-0.2$. In this way, Figure 10 shows the comparison between model and observation from the perspective of the inclination angle and ice-processing level. As pointed out in Section 4.4, the ice column density does not vary monotonically with the 10 inclination angles shown in Figure 10. The maximum $N_X$ occurs for $60^\circ \leq i \leq 70^\circ$, whereas the minimum $N_X$ corresponds to $0^\circ$. The observational fit and confidence intervals taken from Figure 1 are shown by the solid and dashed green lines, respectively. The gray, red, and blue shaded regions cover the column density dispersion for all inclinations according to the spectral index.

Figure 10(a) shows that, except for the pole-on inclination angle, the H$_2$O column densities predicted by the three models lie inside the confidence interval of the observational fit at the stage of class 0/I objects. However, the inclination effect and ice processing cannot explain the vertical spread of two orders of magnitude observed at this stage. Even though the pole-on column densities are 50%–70% lower than the highest inclinations, they are still unable to explain the entire vertical variation. A likely cause of this wide vertical spread in the same evolutionary stage is the initial envelope mass variation associated with protostars at early stages, because, at the same physical conditions, that lower envelope mass leads to a lower ice column density. At the class I stage, all three models overestimate the water-ice column density, except for the pole-on inclination, which suggests that H$_2$O is destroyed by mechanisms other than CR radiolysis ($H_2O + CR \rightarrow OH + H$), such as photolysis induced by UV and X-rays. At the class I/II

Figure 10. Plot containing the spectral index against the column density of the ices H$_2$O, CO$_2$, CH$_3$OH, and HCOOH calculated from the models of virgin and processed ices. The three levels of ice processing are indicated by the filled circles, squares, and triangles, respectively. The inclination effect is shown by the color scheme given by the color bar. Each evolutionary stage is indicated by the top labels. The solid and dashed green lines correspond to the exponential fit and confidence intervals in Figure 1. The gray, red, and blue shaded regions cover the column density dispersion for all inclinations according to the spectral index.
stage, on the other hand, only the virgin-ice model overestimates the H2O-ice column density, whereas the first- and second-processing models are in agreement with the observation. Nevertheless, it is inconclusive that such a result is related to the physical and chemical evolution of the protostar because only one stage of evolution fits the data.

The trends shown by the shaded areas in Figure 10(a) indicate a slow $N_{\text{ice}}$ decrease at early stages and the absence of the plateau for late stages when compared to the observational data. While the fast decrease observed at early stages for water ice is likely related to other destruction mechanisms not included in this paper, the absence of the plateau at late stages can suggest that at least 30% of the observed ices lying outside the protostar itself, and therefore, not affected by the accretion process, might contribute to keeping the observed ice column density fraction roughly constant, as shown by Pontoppidan et al. (2005), Boogert et al. (2000), and Aikawa et al. (2012).

In the case of the $N_{\text{CO}_2}$ variation shown in Figure 10(b), the virgin-ice and the first-processing models provide column densities inside the observational confidence interval in class 0/I. The CO2 destruction in the second-processing model, on the other hand, underestimates $N_{\text{CO}_2}$. Nevertheless, in the more evolved stages, namely, class I and class I/II, the second-processing model predicts CO2 ice column densities inside the confidence interval, whereas the virgin-ice and the first-processing models generally overestimate the column densities. As in the case of H2O ice, the effect of the physical and chemical evolution of protostars cannot be confirmed. The decreasing trends shown by the shaded areas also indicate the absence of the plateau at late stages. For the class I YSOs Elias 29 and CRBR 2422.8–3423, the estimated column densities in foreground clouds are $N_{\text{CO}_2} \approx 2 \times 10^{17}$ cm$^{-2}$ (Boogert et al. 2000) and $N_{\text{CO}_2} \lesssim 6 \times 10^{17}$ cm$^{-2}$ (Pontoppidan et al. 2005), respectively.

Figure 10(c) shows the variation of the methanol-ice column density against the spectral index as calculated from the band at the 6.85 μm spectral feature. One can note that the methanol column densities calculated from the first- and second-processing models mostly lie inside the observational range for class 0/I, although the vertical spread is not explained, whereas it is overestimated in the later stages. The reason for the overestimation is likely due to (i) the high efficiency of CH3OH production because of the large CO2 fraction in the experiments and (ii) due to the decomposition method itself. In the latest case, in Boogert et al. (2008), the component attributed to CH3OH ice has an FWHM 50% lower than that assumed in this paper, which would lead to a lower column density calculation. Taking this difference into account, if the FWHM Gaussian component associated with methanol in this paper is reduced by 50%, it would represent an ice column density 20% lower. Such a reduction, however, would not provide a better agreement between observation and model. In summary, the model fails to reproduce the methanol-ice column density variation and the fast decrease compared to the other molecules as pointed out at the beginning of the paper. Further investigation must be carried out for this case, and other laboratory ice samples can be used instead, as well as how the methanol formation is affected during the protostellar collapse.

HCOOH ice column density variation is shown in Figure 10(d) as calculated from the absorption feature at 7.24 μm. Both first- and second-processing models provide $N_{\text{HCOOH}}$ inside the observational confidence interval in most of the cases. The good agreement between observation and the two models suggests that the lower decrease rate of formic acid is mainly due to envelope dissipation by accretion than due to the energetic processing of the ice, i.e., chemical evolution. The absence of the plateau at late stages, on the other hand, indicates that if it is caused by ices in foreground clouds, then its abundance in quiescent molecular clouds is very low.

In summary, the evolutionary models along with the ice samples used in this paper, in general, do not explain the observed trends of ice column density with the evolutionary stage, with the exception of HCOOH. Further investigations addressing different physical parameters and ice mixtures must be used in order to explain the observational data.

6. Conclusions

This paper shows the correlation between the ice column densities of H2O, CO2, CH3OH, and HCOOH and the spectral index ($\alpha$) of 27 early-stage YSOs. A computational simulation combining the 2D radiative transfer model and laboratory data of virgin and processed ices was carried out to study the ice column density decreasing from early to late stages of the protostellar evolution. The conclusions are summarized below.

1. The observational data suggest that ice column densities are correlated with the spectral index from class 0/I to class I/II. Additionally, it is observed that H2O and CH3OH ices reach an ice column density plateau at class I, whereas for CO2 ice, the plateau is reached at class I/II. In the case of HCOOH, however, it is not observed. If an exponential function is assumed to fit the data, it is found that CH3OH ice decreases faster then other ices, whereas HCOOH decreases slowly.
2. In agreement with previous works, the models simulated in this paper show that the H2O snow line moves inward from 2 au at class 0/I to 0.7 au at class II. During the evolution to class III, the snow line moves outward to 20 au. Nevertheless, the region where the ices are located is optically thick in the mid-IR, and therefore the absorption seen in the spectra are due to the ices located in the envelope. As a consequence of the density distribution in class 0/I to class I/II, the ice column density does not vary monotonically with the inclination angle ($i$). The highest column density is, generally, seen for $i \approx 20^\circ$ at class 0/I and $i \approx 60^\circ$ at class I.
3. The computational models show that the combination of physical evolution (envelope dissipation), chemical evolution (ice processing) and inclination angle effect is able to reproduce the $N_{\text{HCOOH}}$ decrease with the spectral index. However, in the case of H2O, CO2, and CH3OH, the models fail to reproduce the observations in all evolutionary stages. Additionally, other destruction pathways not included in the current method must be addressed to understand why water and methanol-ice column density decrease faster than CO2 and HCOOH.
4. The absence of the ice column density plateau in the models suggests that there is a fraction of ice absorption located in foreground clouds in order to explain the observations. More accurate models including other physical and chemical effects not addressed in this paper could confirm or reject this hypothesis. From the observational perspective, ices in foreground clouds have...
been already presented in the literature indicating values close to the ice column density plateau estimated in this paper for H$_2$O and CO$_2$.

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Appendix

Deconvolution of the Profile Between 5.5 and 7.5 $\mu$m

The spectral region between 5.5 and 7.5 $\mu$m contains several O–H and C–H vibrational modes that might be associated with alcohols, ketone, and aliphatic ethers (Schutte et al. 1996; Gibb et al. 2000; Keane et al. 2001; Boogert et al. 2008, 2015). In order to determine the contribution of CH$_3$OH and HCOOH to the spectral profile between 5.5 and 7.5 $\mu$m, a Gaussian decomposition of this spectral range using five components was employed and is given by

$$G(\lambda; A, \sigma) = \frac{A}{\sigma \sqrt{2\pi}} \exp \left[-\frac{(\lambda - \lambda_0)^2}{2\sigma^2}\right]$$  \hspace{1cm} (A1a)

$$\tau_\lambda = \sum_{i=0}^{m-1} G(\lambda; A_i, \sigma_i),$$  \hspace{1cm} (A1b)

where $\lambda$ is the wavelength, $A$ is the integrated area, and $\sigma$ is the FWHM of the Gaussian profile. Equation (A1a) is the Gaussian function of one component and Equation (A1b) is the sum of all components, assumed equal to five in this paper. Figures A1–A6 show the Gaussian decomposition of the spectral range between 5.5 and 7.5 $\mu$m according to the evolutionary stage variation and ice-processing level. The two components around 6.0 $\mu$m are associated with H$_2$O and daughter species formed from the ice processing. The blue shaded component at 6.8 $\mu$m is attributed to CH$_3$OH whereas the yellow shaded component at 7.24 $\mu$m refers to HCOOH. The component in between ($\sim$7.0 $\mu$m) has been attributed to CH$_3$CHO in Pilling et al. (2010).

Figure A1. Decomposition of the optical depth between 5.5 and 7.5 $\mu$m for the first-processing model and class 0/I. The black line shows the modeled data, whereas the green lines indicate the Gaussian profiles. The contributions of CH$_3$OH and HCOOH are given by the blue and orange areas, respectively.
Figure A2. Same as Figure A1, but for class I.
Figure A3. Same as Figure A1, but for class I/II.
Figure A4. Same as Figure A1, but for the second-processing model and class 0/I.
Figure A5. Same as Figure A1, but for the second-processing model and class I.
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Figure A6. Same as Figure A1, but for the second-processing model and class I/II.
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