GRAINOBLE:
a multiphase gas-grain astrochemical model

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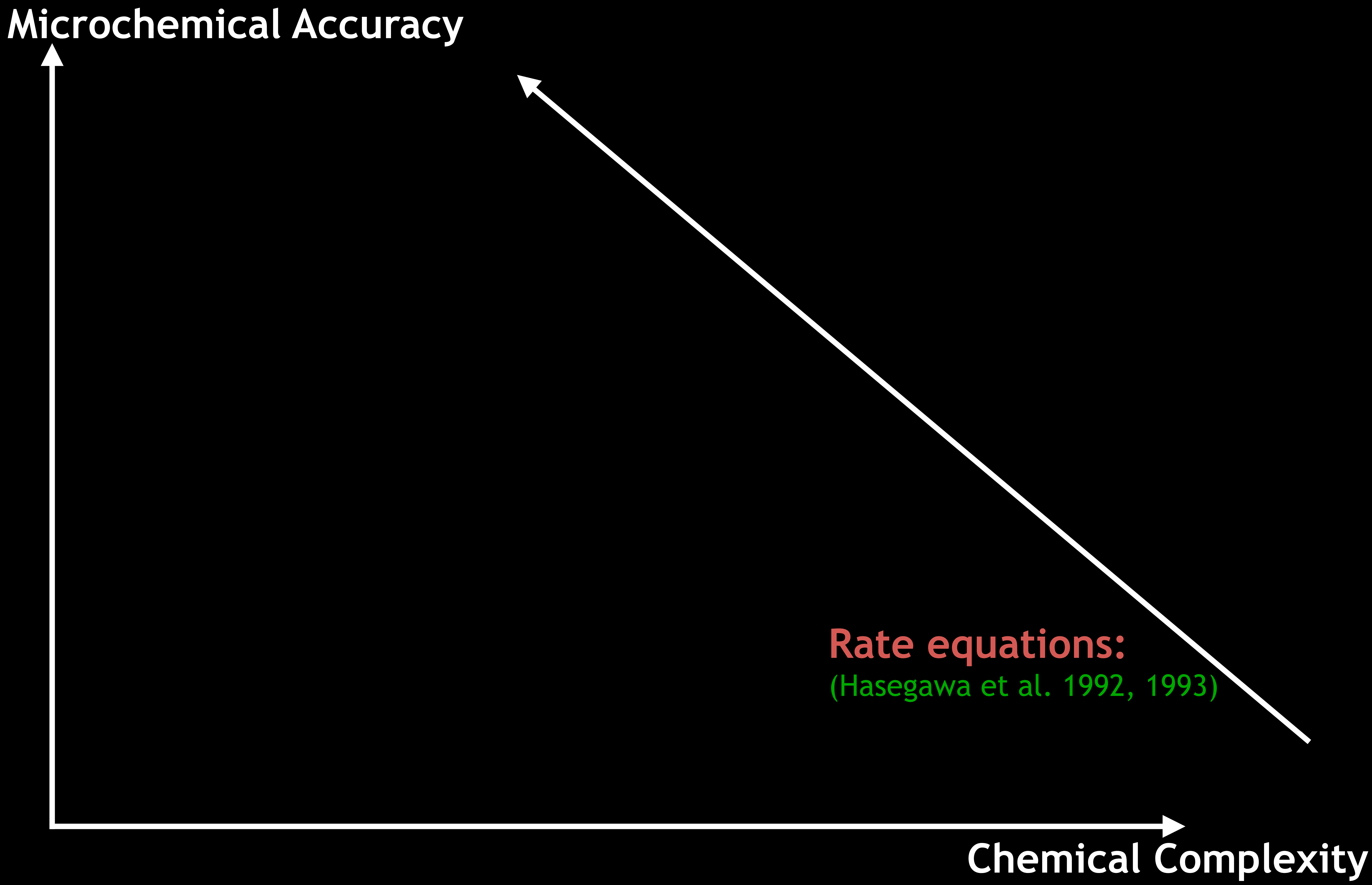
Methods for grain surface modelling



Rate equations

Extended from gas phase models:

Species are produced by the reaction between reactive and mobile species
→ limited by diffusion

$$\frac{dn_{\mathrm{ice}}(i)}{dt} = \underbrace{K_{\mathrm{acc}} n_{\mathrm{gas}}(i)}_{\text{Accretion}} - \underbrace{K_{\mathrm{ev}} n_{\mathrm{ice}}(i)}_{\text{Evaporation}} + \underbrace{\sum_j \sum_k K_{\mathrm{reac,f}} n_{\mathrm{ice}}(j) n_{\mathrm{ice}}(k) - \sum_j \sum_k K_{\mathrm{reac,d}} n_{\mathrm{ice}}(j) n_{\mathrm{ice}}(k)}_{\text{Chemical reactions}}$$

$$K_{\mathrm{reac}} = P_{\mathrm{reac}} \frac{K_{\mathrm{diff}}(j) + K_{\mathrm{diff}}(k)}{N_{\mathrm{s}} n_{\mathrm{d}}}$$

$$K_{\mathrm{diff}}(j) = \nu_0 \exp(-E_{\mathrm{d}}/T_{\mathrm{d}})$$

ν_0: vibrational frequency given by harmonic oscillator relation ~ 10^{12}-10^{13} s^{-1}
E_d: diffusion energy
N_s: number of grain sites

Methods for grain surface modelling

Microchemical Accuracy

Pros: fast
- sophisticated chemical networks
- statistical number of simulations
- can be coupled with dynamics

Cons:
- inaccurate treatment of random walk
- surface structure ignored
- fails in the accretion limit

Rate equations:
(Hasegawa et al. 1992, 1993)

Chemical Complexity

Methods for grain surface modelling

Microchemical Accuracy

Modified Rate equations

+ fast and valid in the accretion limit
- surface structure still ignored

(Caselli et al. 1998, Garrod 2008)

Rate equations

Chemical Complexity

Methods for grain surface modelling

Microchemical Accuracy

Master equation
+ valid in the accretion limit
- few species at a time
- computationally expensive

(Biham et al. 2001, Green et al. 2001)

Modified Rate equations

Rate equations

Chemical Complexity

Methods for grain surface modelling

Microchemical Accuracy

Macroscopic Monte-Carlo models

+ valid in the accretion limit
+ more efficient than the direct Master equation resol.
- more expensive than rate equations

(Tielens & Hagen 1982, Charnley 2001, Vasyunin et al. 2009)

Master equation

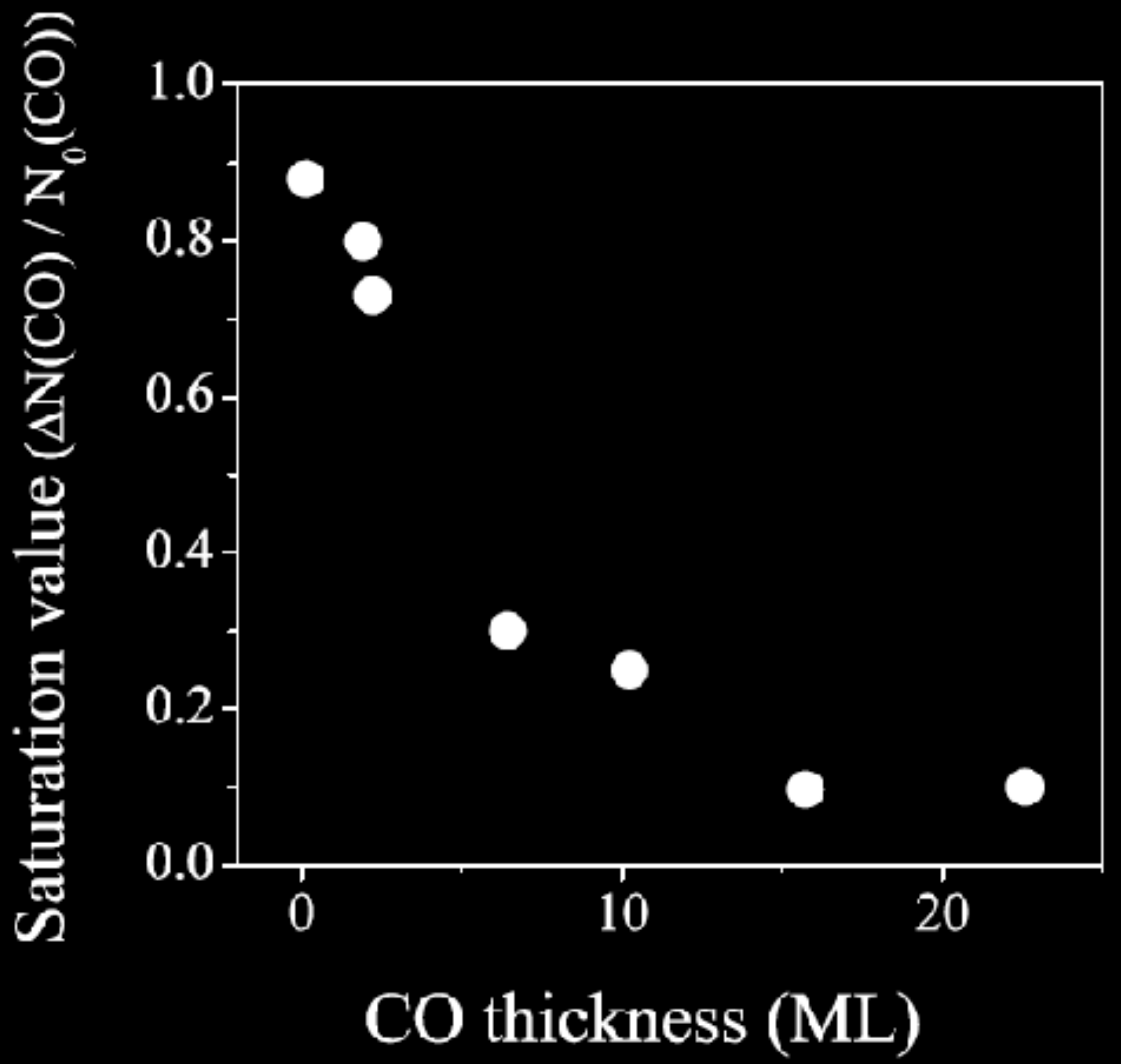
Modified Rate equations

Rate equations

Chemical Complexity

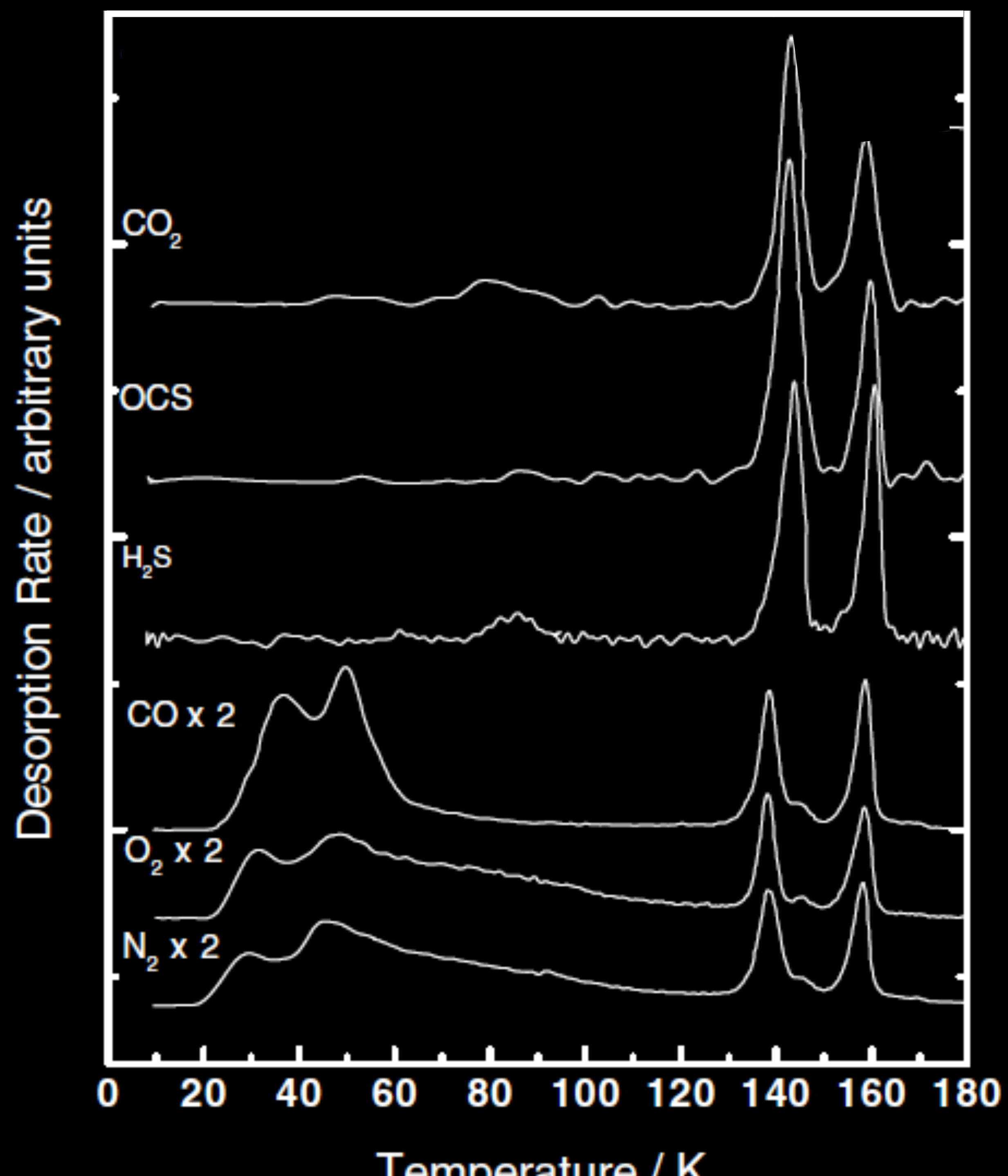
Diffusion of light particles into/from the bulk

Hydrogenation of CO ice



Watanabe et al. (2004)

Desorption of H_2O:X ice mixtures



Collings et al. (2004)

Methods for grain surface modelling

Microchemical Accuracy

Off-Lattice Monte Carlo

+ detailed surface structure
- few species; small network
- gas/grain coupling impossible

(Garrod 2013)

On-Lattice Monte Carlo (a.k.a CTRW, kMC)

+ micro. processes and ice structure
- computationally expensive

(Chang et al. 2005, 2014; Cuppen et al. 2007, 2009)

Rate equations

Chemical Complexity

Ice structure predicted by kinetics MC models

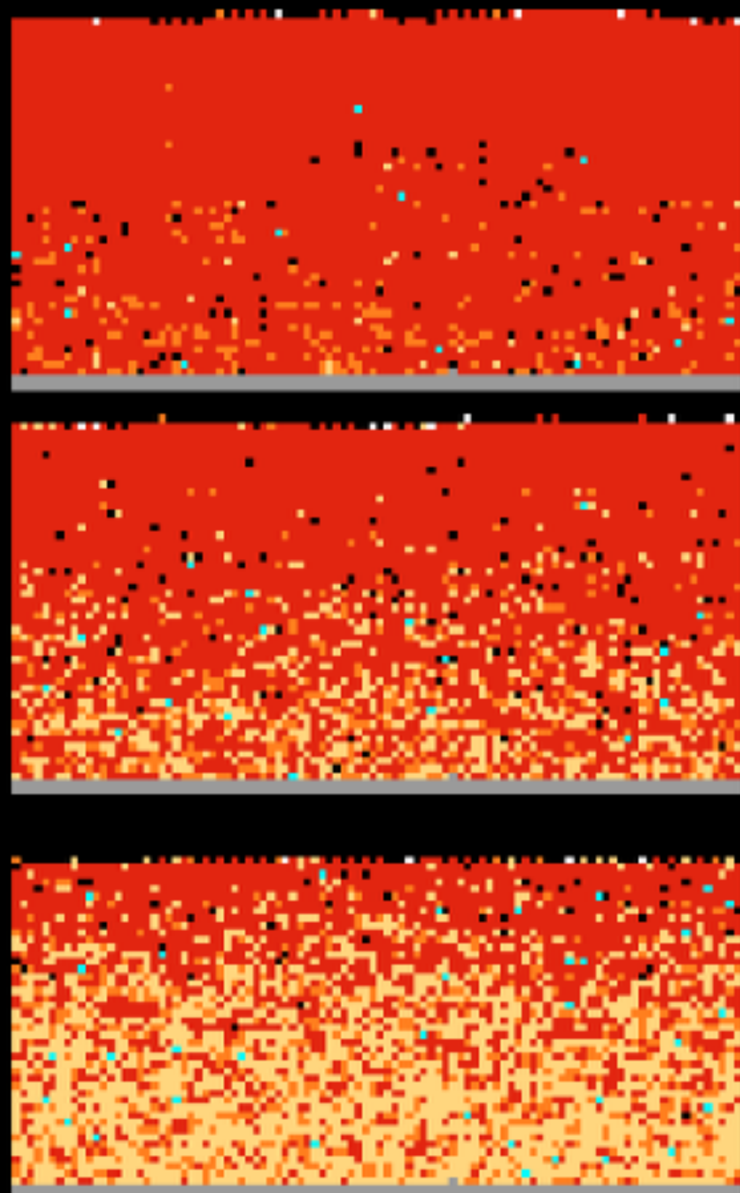
Methanol formation on 2D surface

n_H = 2x10^5 cm^{-3}

T = 12 K

T = 13.5 K

T = 15 K



CH_3OH

H_2CO

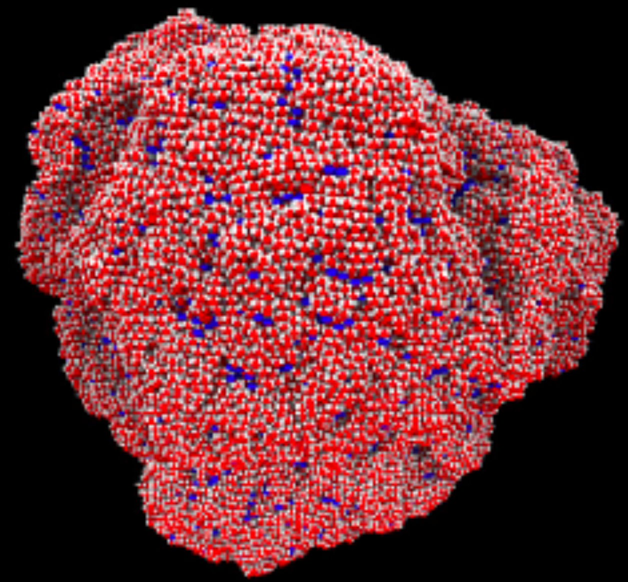
CO

Radicals

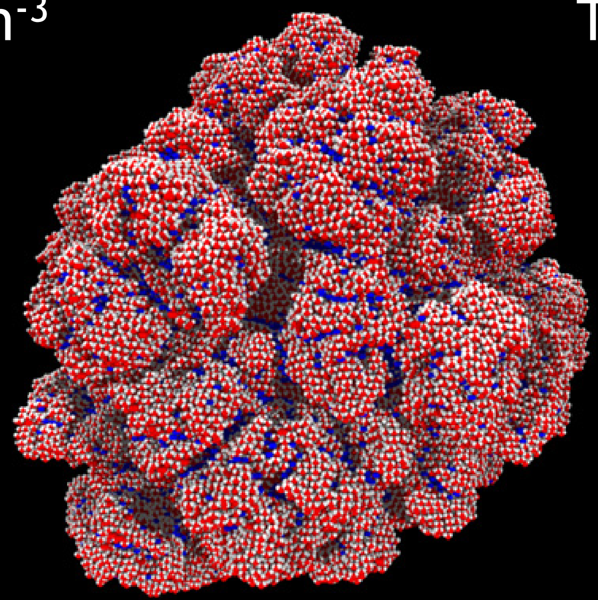
Cuppen et al. (2009)

Water formation on 3D grains

n_H = 2x10^4 cm^{-3} T = 10 K



n_H = 2x10^7 cm^{-3} T = 10 K

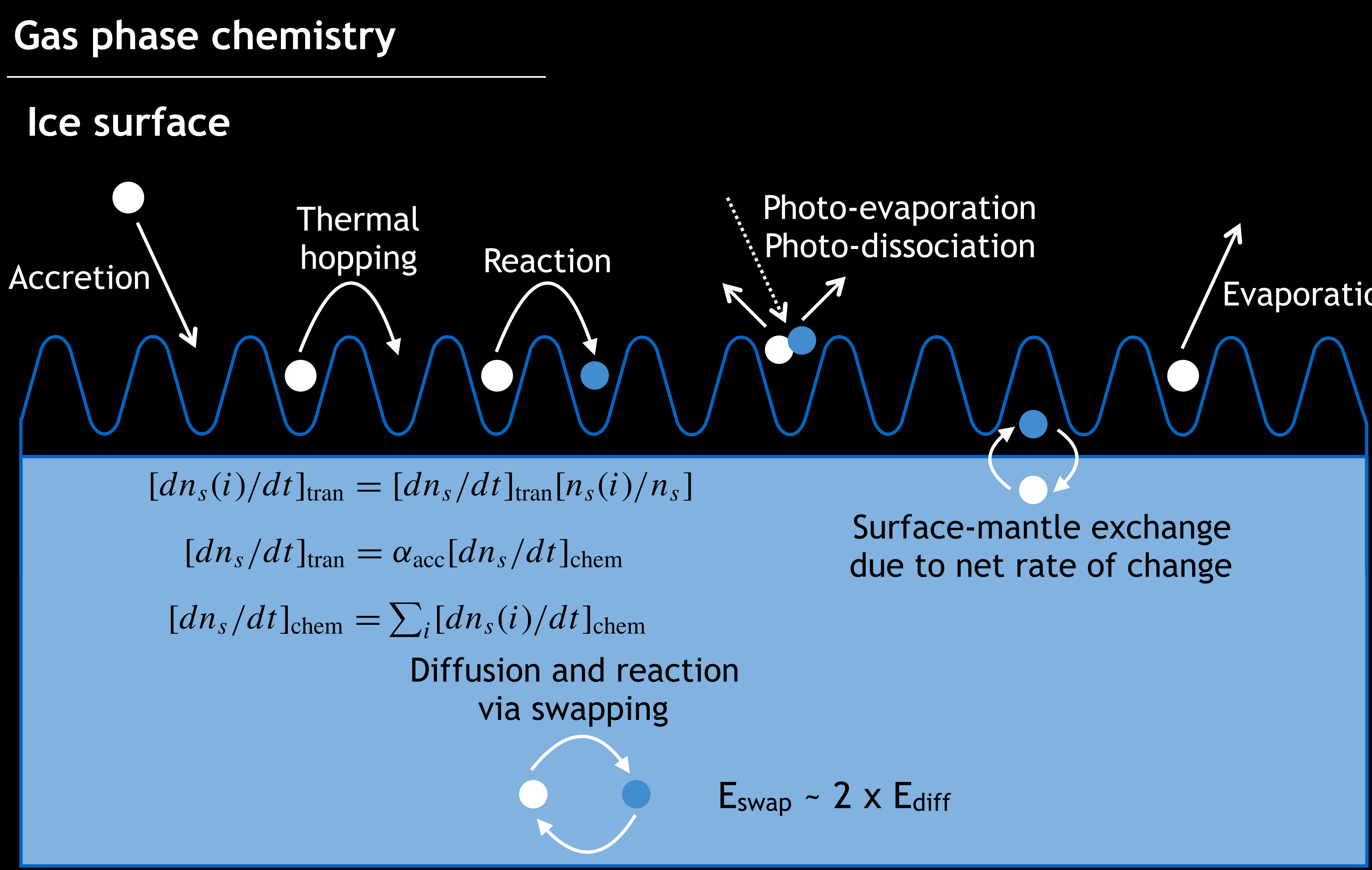


Garrod (2013)

Rate-equations multiphase astrochemical models

Distinction of chemical processes in the bulk and at the surface

(Hasegawa & Herbst 1993, Garrod & Pauly 2011, Taquet et al. 2012, 2014, 2016, Garrod et al. 2013, 2017, Ruaud et al. 2016, Furuya et al. 2017, Vasyunin et al. 2018)



$$[dn_s(i)/dt]_{\rm tran} = [dn_s/dt]_{\rm tran}[n_s(i)/n_s]$$

$$[dn_s/dt]_{\rm tran} = \alpha_{\rm acc}[dn_s/dt]_{\rm chem}$$

$$[dn_s/dt]_{\rm chem} = \sum_i [dn_s(i)/dt]_{\rm chem}$$

$E_{swap} \sim 2 \times E_{diff}$

Rate-equations multiphase astrochemical models

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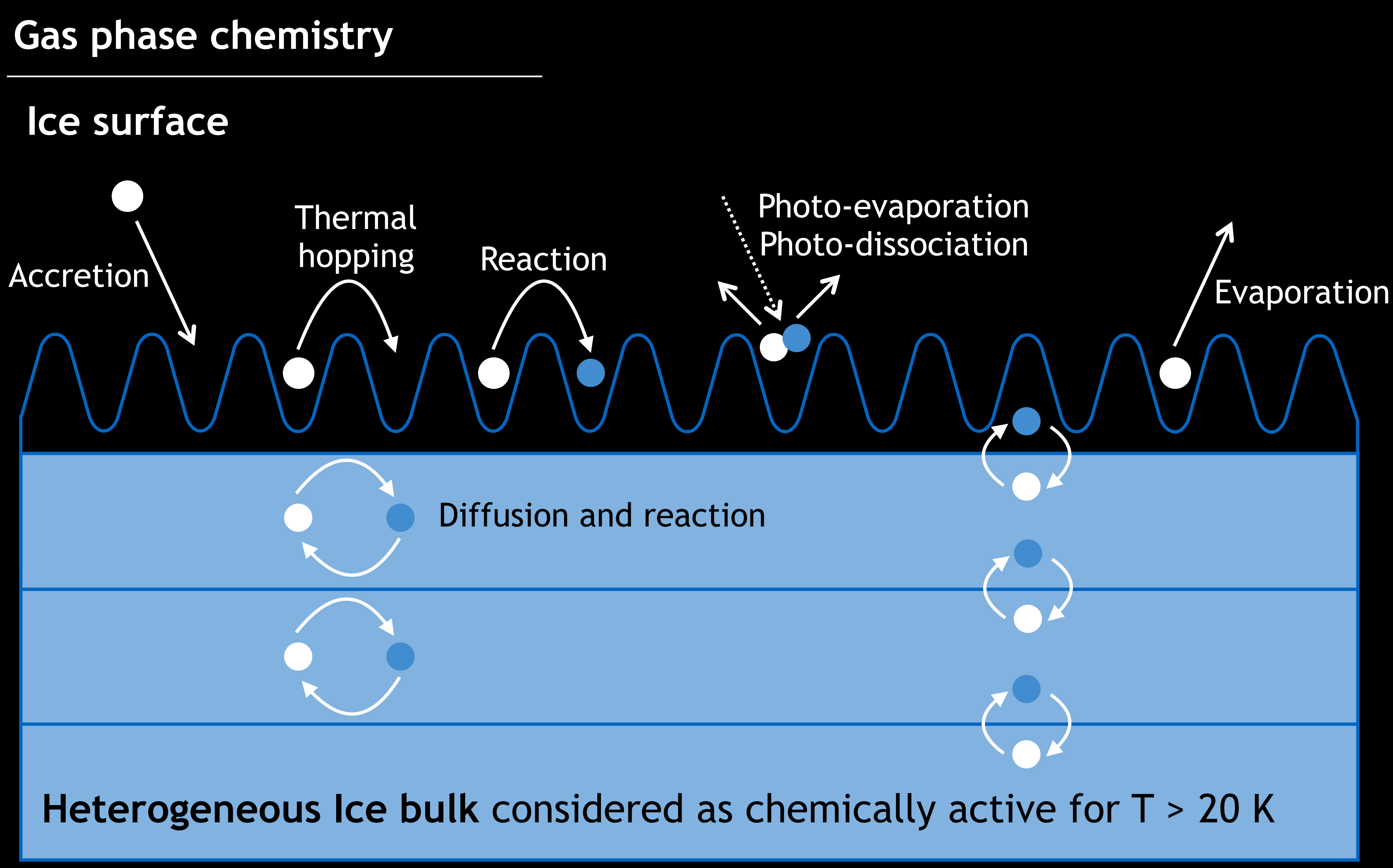
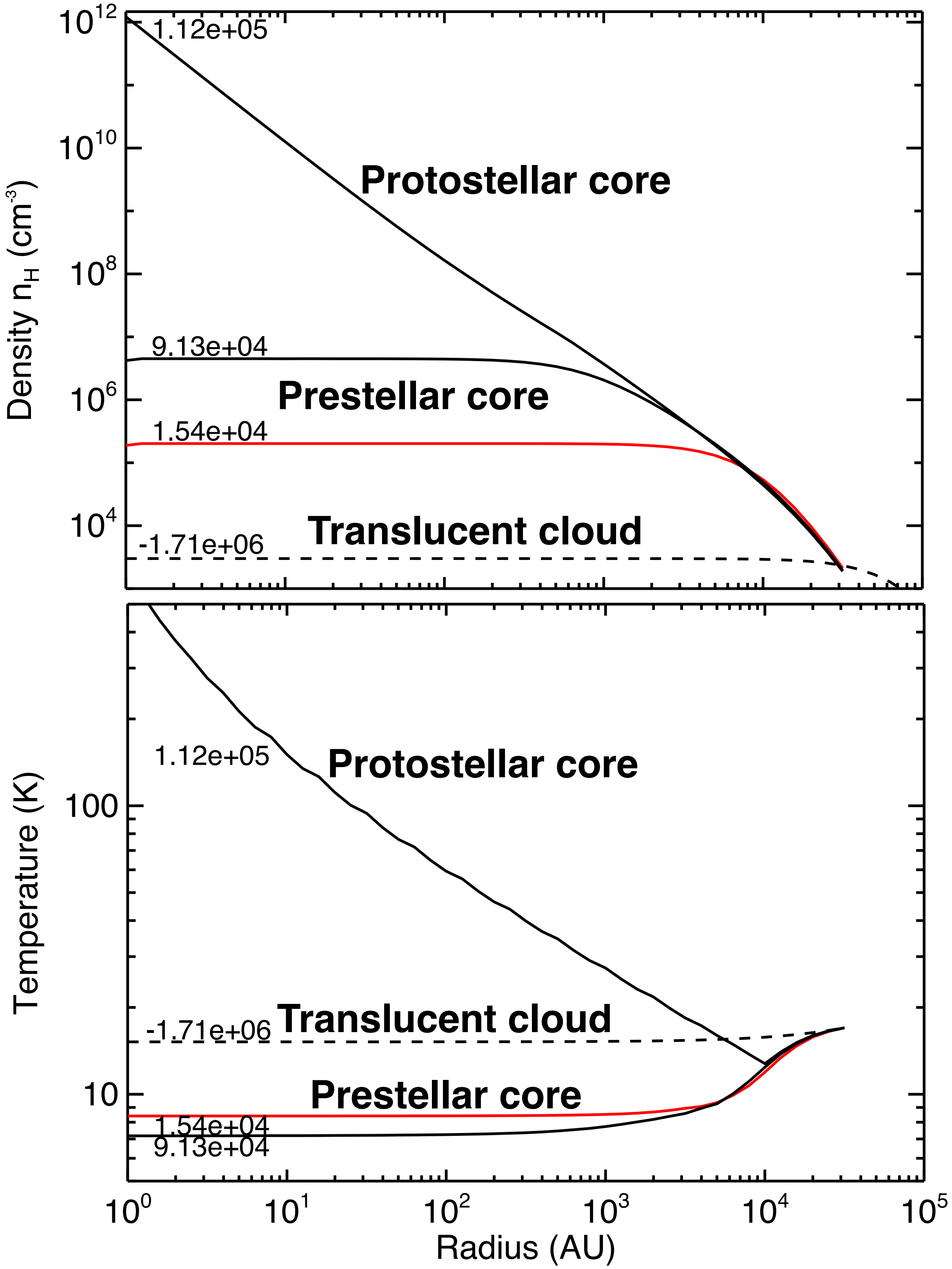
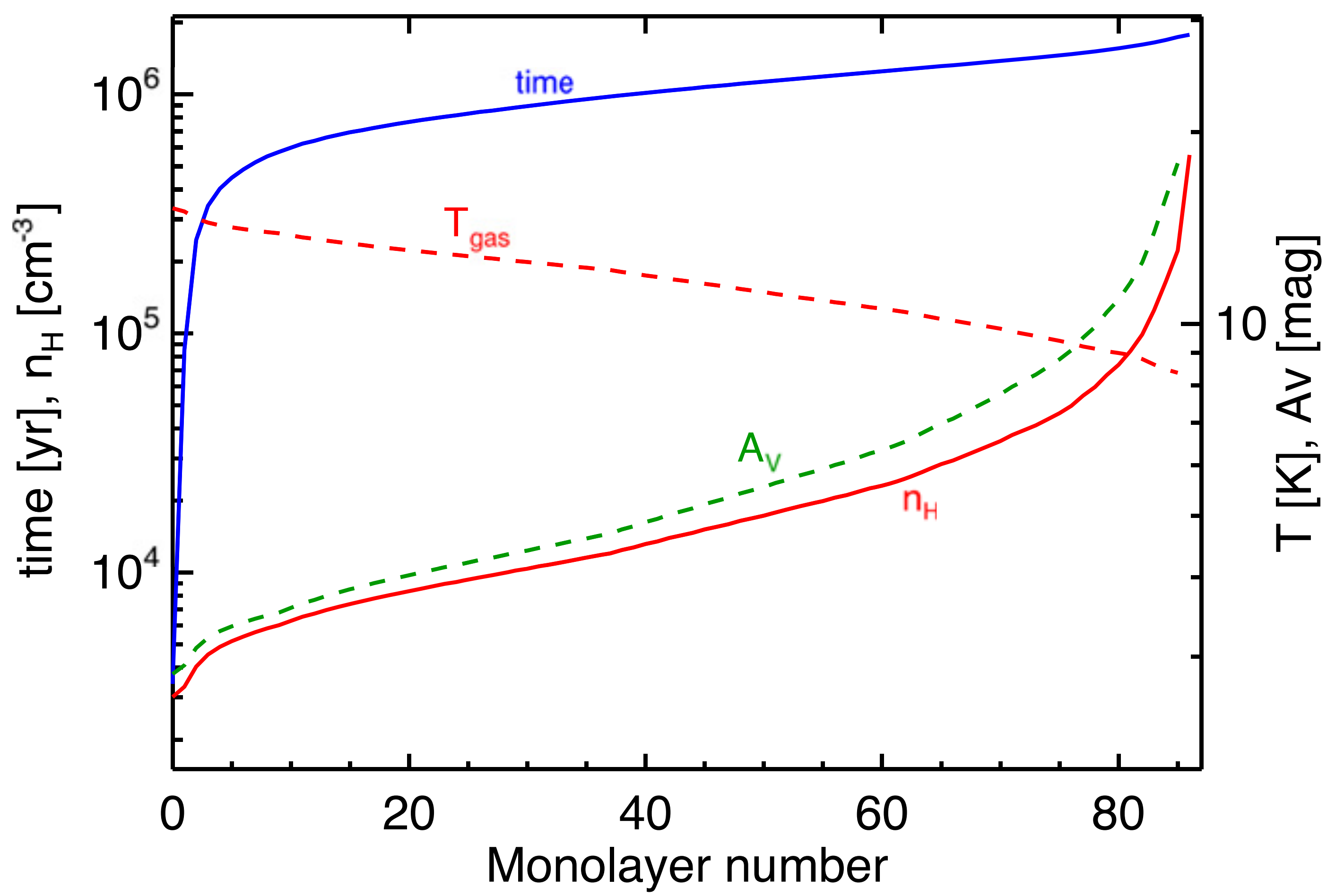


Ice structure predicted by multiphase models

Ices form and evolve from translucent dark clouds to dense prestellar and protostellar cores



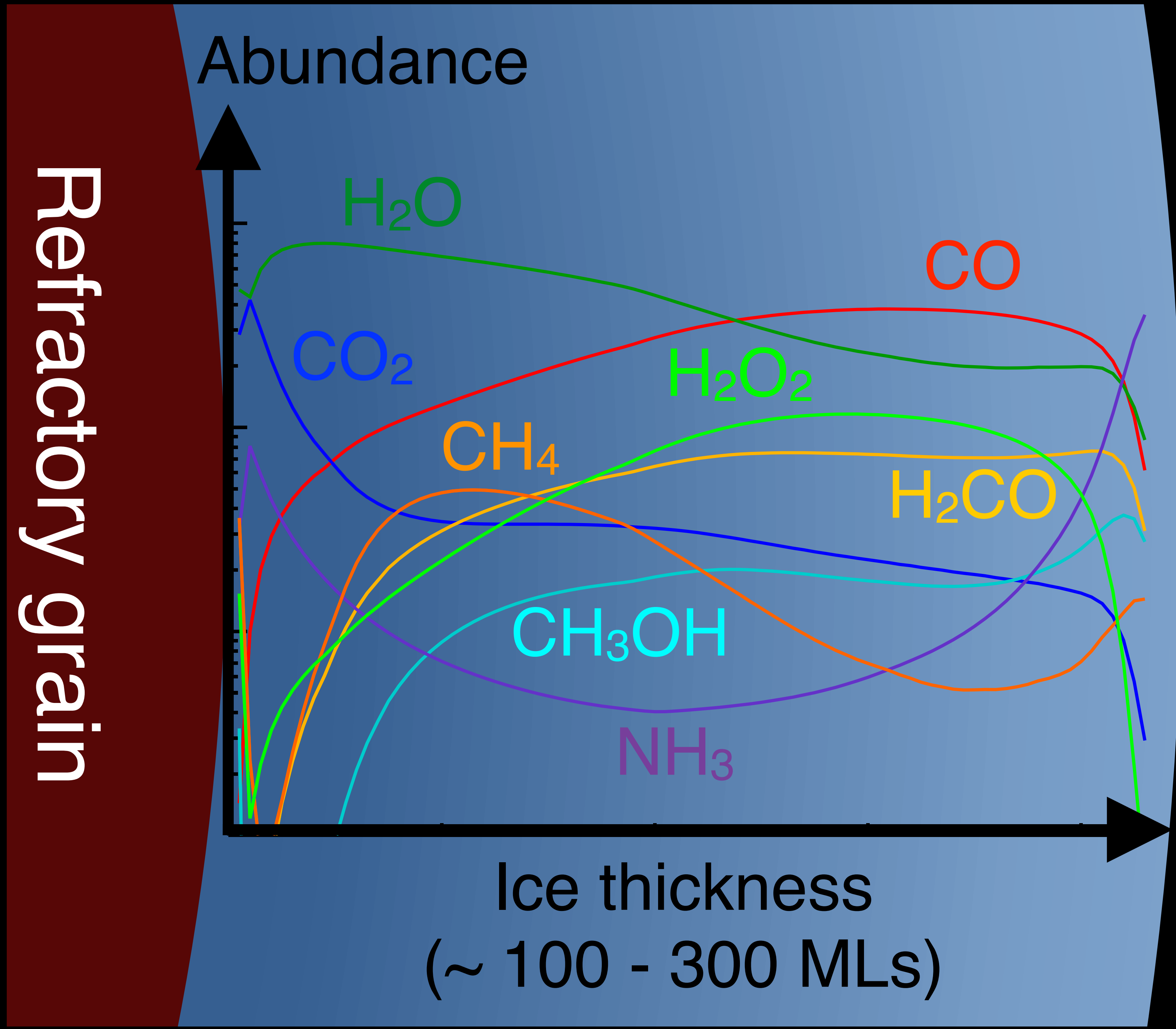
Physical conditions and ice thickness



Taquet et al. (2014)

Ice structure predicted by multiphase models

Chemical heterogeneity of ices induced by the gas phase abundance and physical evolution in dark clouds

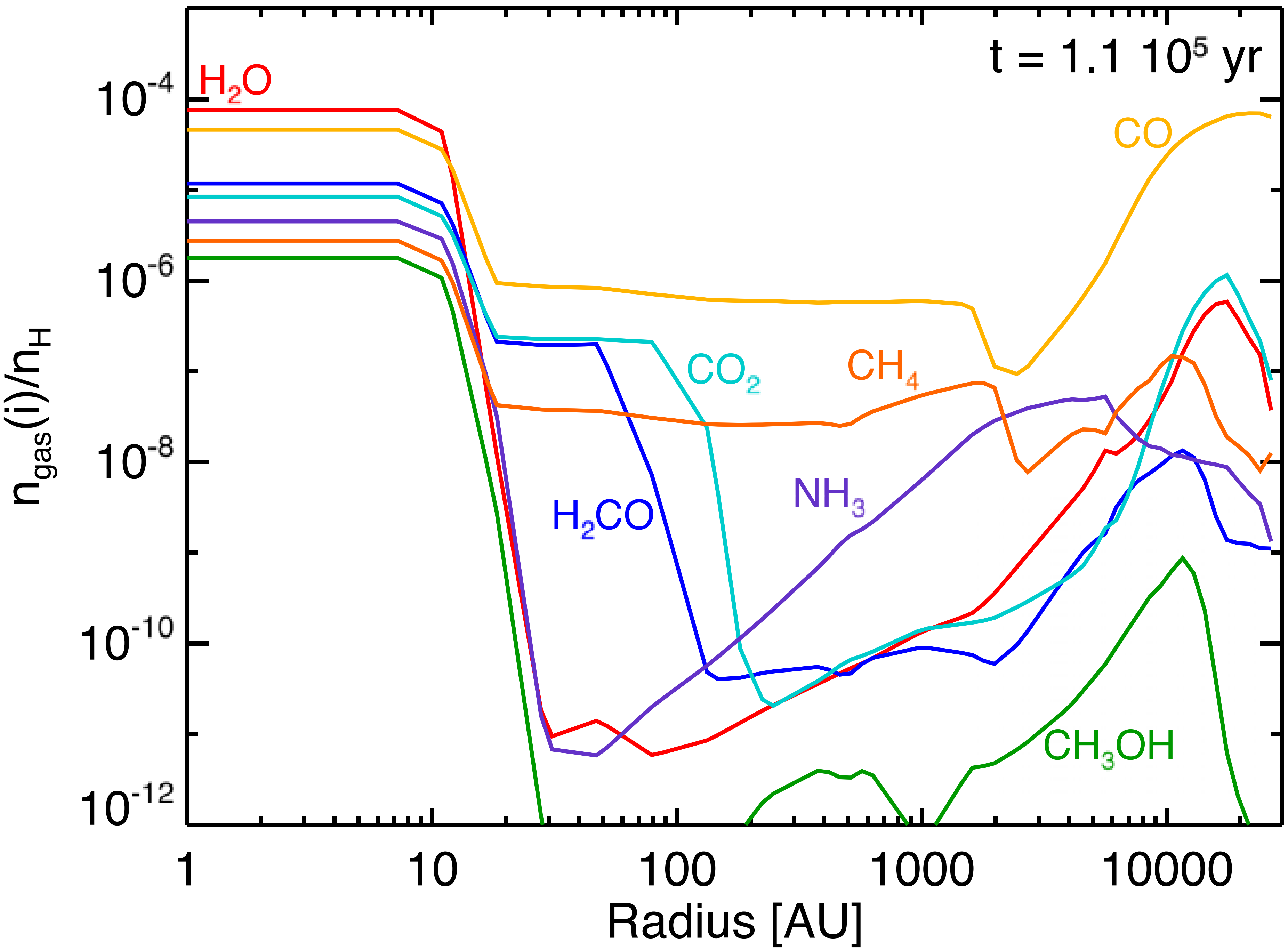


Taquet et al. (2014)

see Charnley & Rodgers (2009), Garrod & Pauly (2011), Vasyunin & Herbst (2013), Furuya et al. (2016)

Abundance profiles of “icy” species around protostars

Multiphase models predict double abundance jump profiles around protostars, due to the complex evaporation of ice mixtures



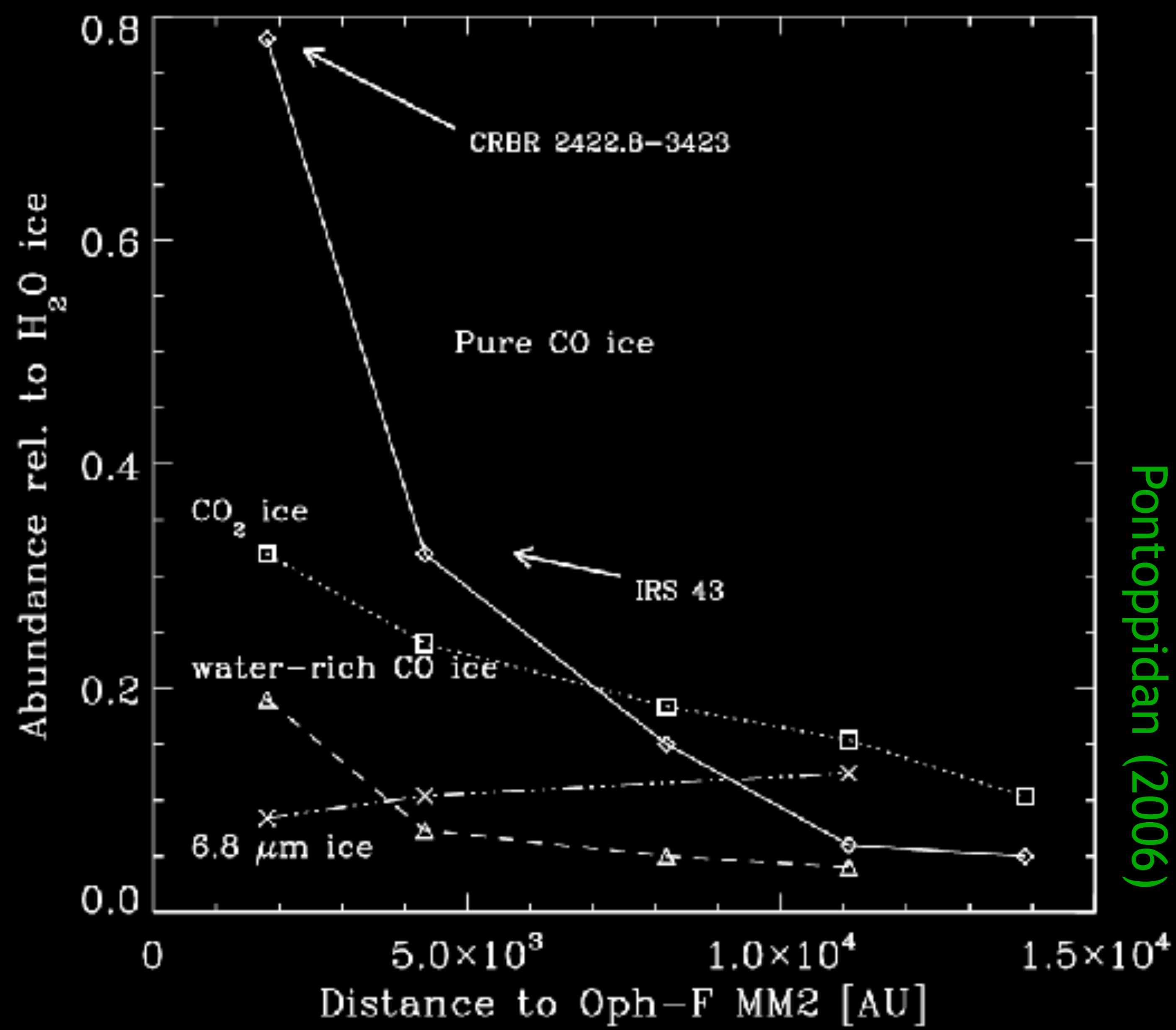
Taquet et al. (2014), See also Vasyunin et al. (2013)

Coupling with radiative transfer models ?

Prediction of synthetic spectra to prepare JWST (and SPHEREx) ?

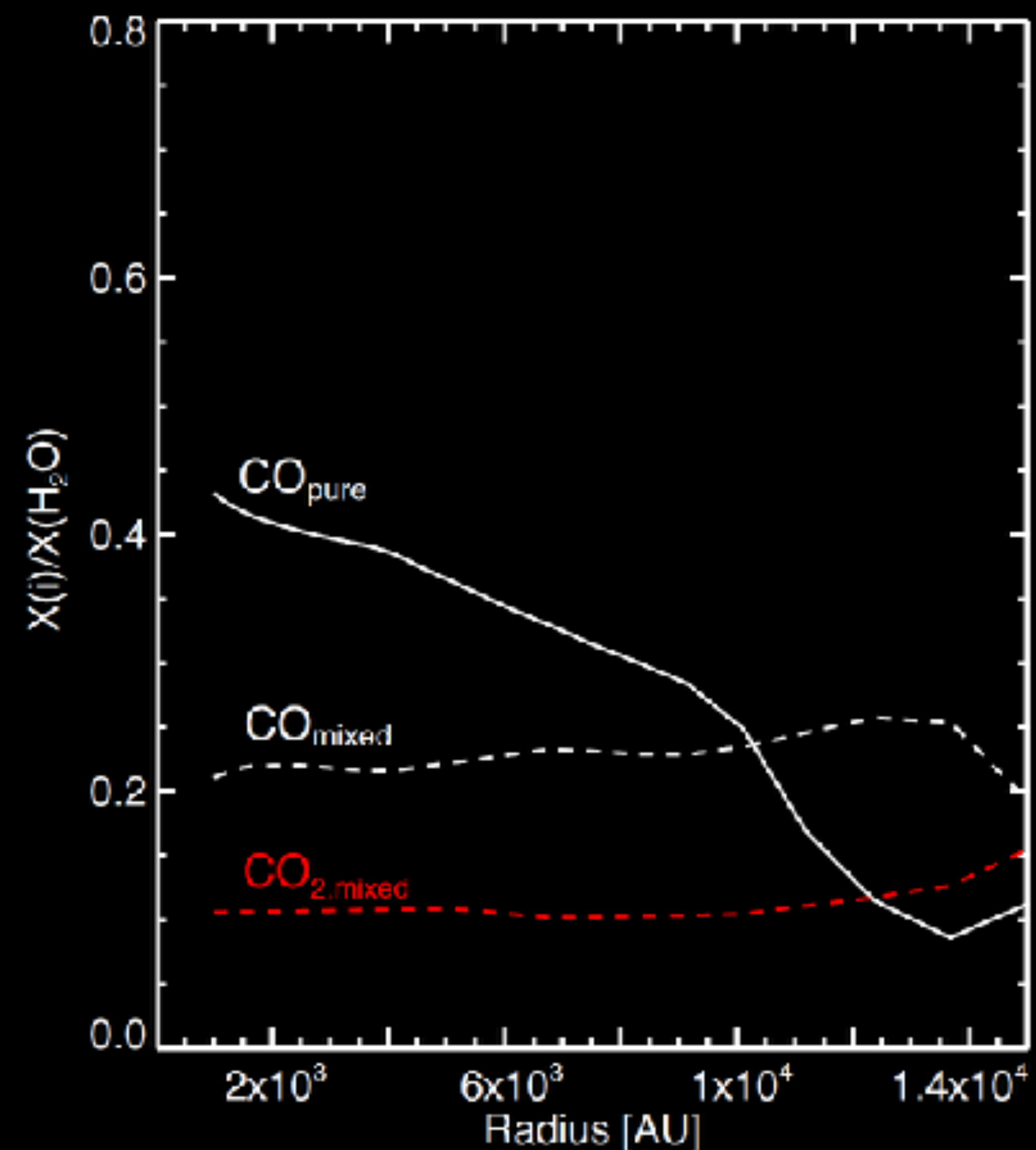
→ Band profiles highly depend on ice composition and mixture: can models guide observations ?

Ice mapping in the Ophiuchus-F core

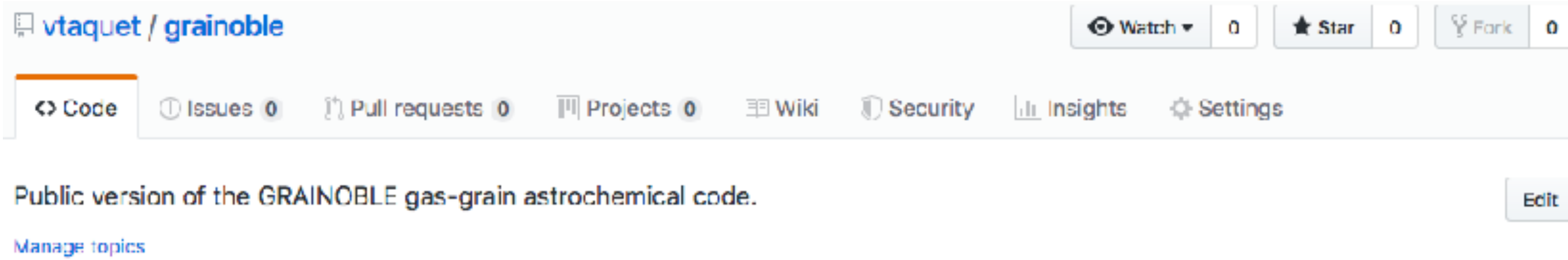


Pontoppidan (2006)

Predicted mixtures



GRAINOBLE on GitHub



Fortran90 code with two Python scripts to run and analyse the simulations:

Three networks:

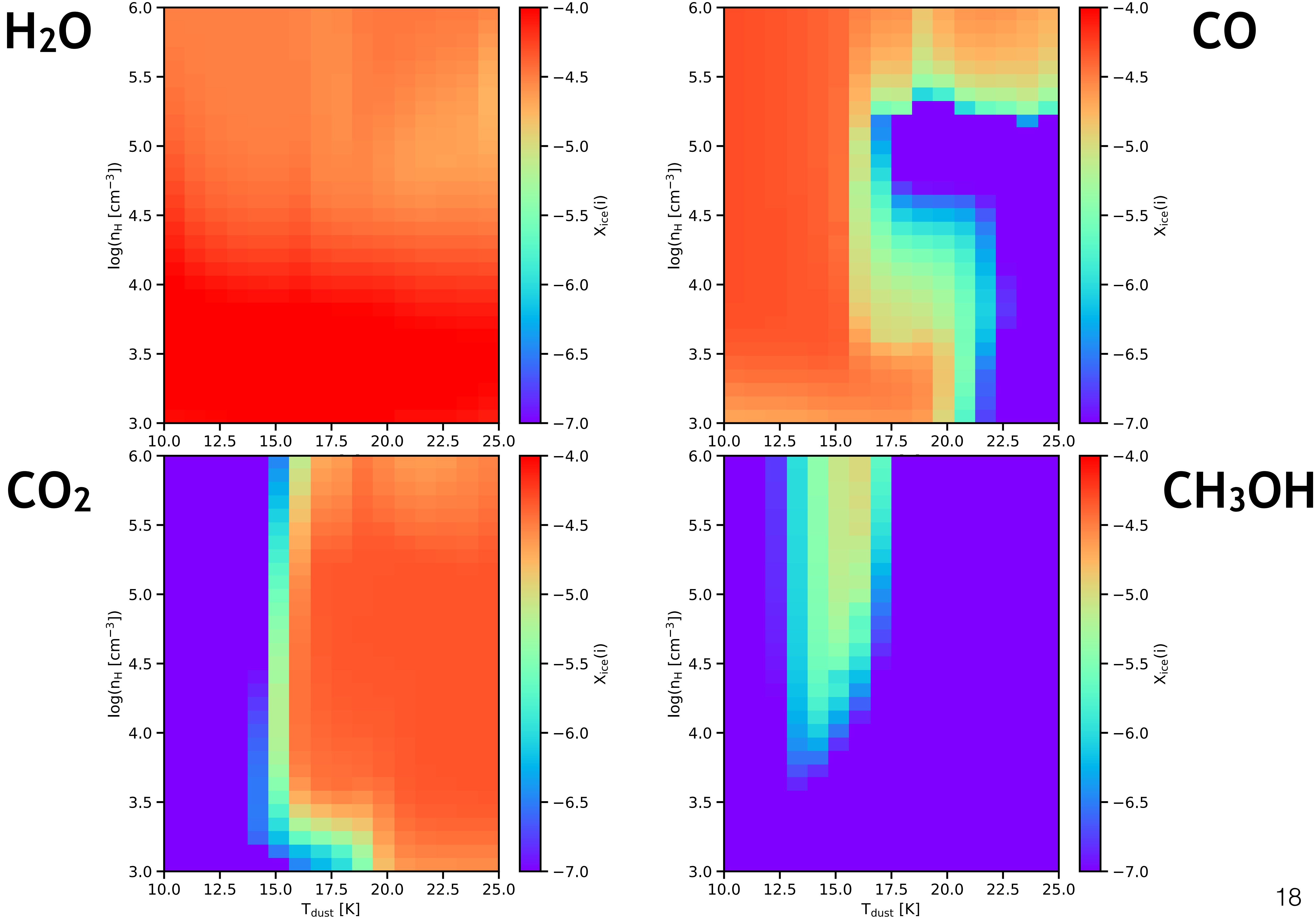
- Network 1: H_2O, CO_2, and CH_3OH formation without gas phase chemistry
- Network 2: ice deuteration
- Network 3: extended surface + KIDA gas phase networks

Four options:

1) Invididual simulations with constant physical conditions
2) Spatial evolution with evolving physical conditions
3) Model grid to explore the impact of physical conditions on ice chemistry
4) Sensitivity analysis to evaluate the impact of surface and chemical parameter uncertainties on ice chemistry

“Astronomical” and “experimental” versions, if requested

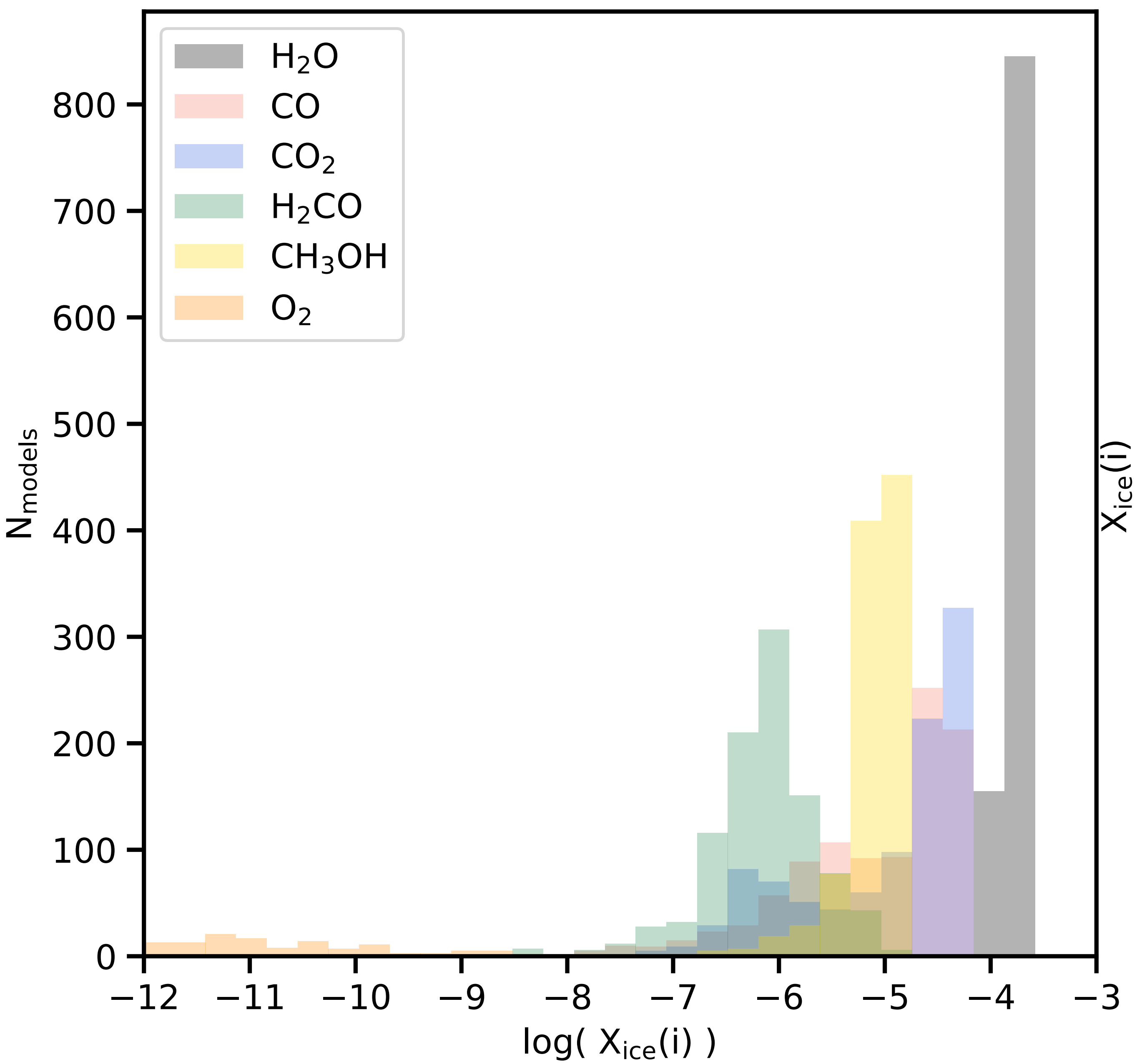
Impact of physical conditions on ice composition

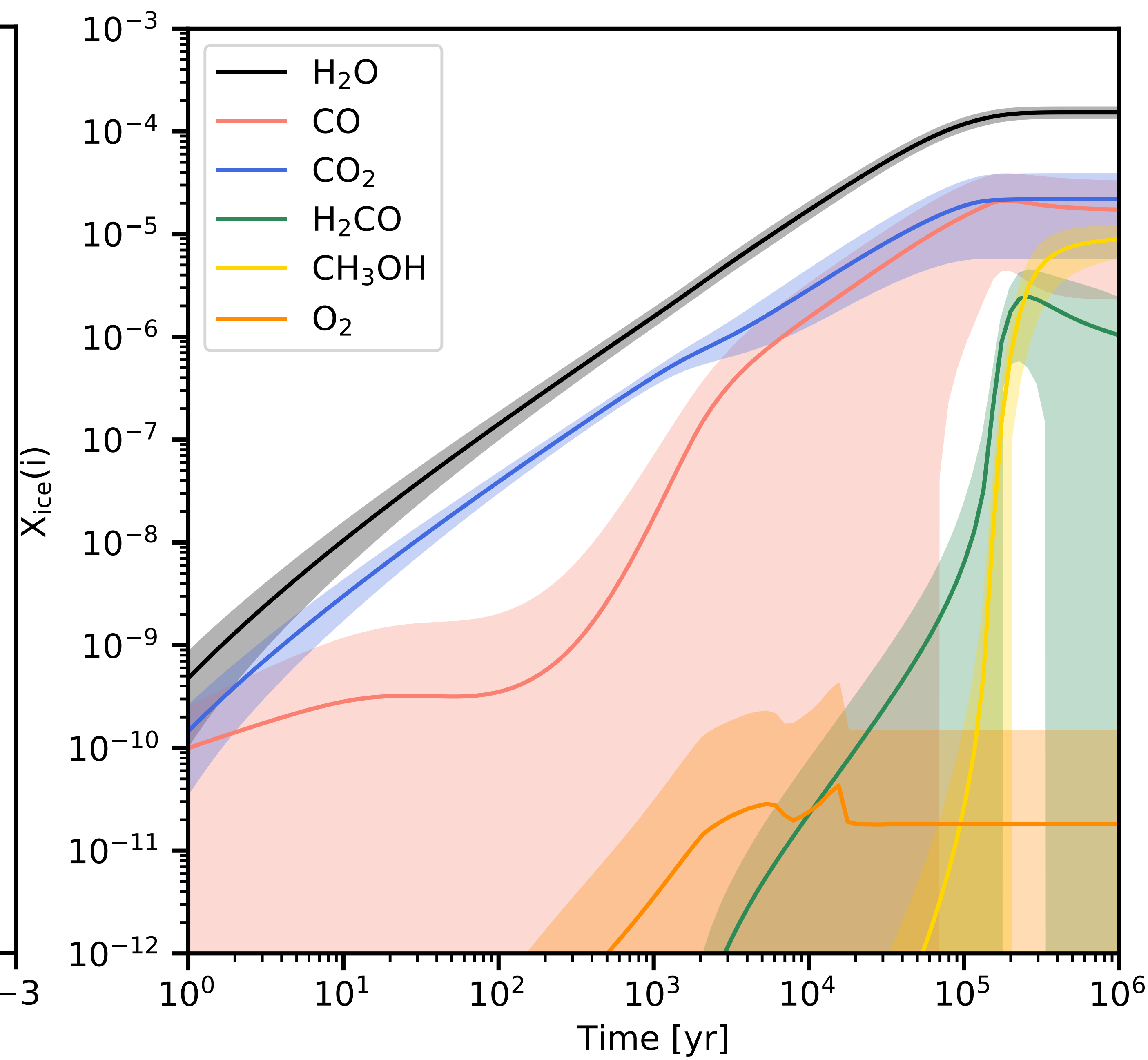


Impact of uncertainties on ice composition

10% uncertainty on binding energies, diffusion-to-binding energy ratio, and activation energies

$n_H = 2x10^4\ cm^{-3}$, T = 10 K





See recent works by Penteado et al. (2017), Holdship et al. (2018)

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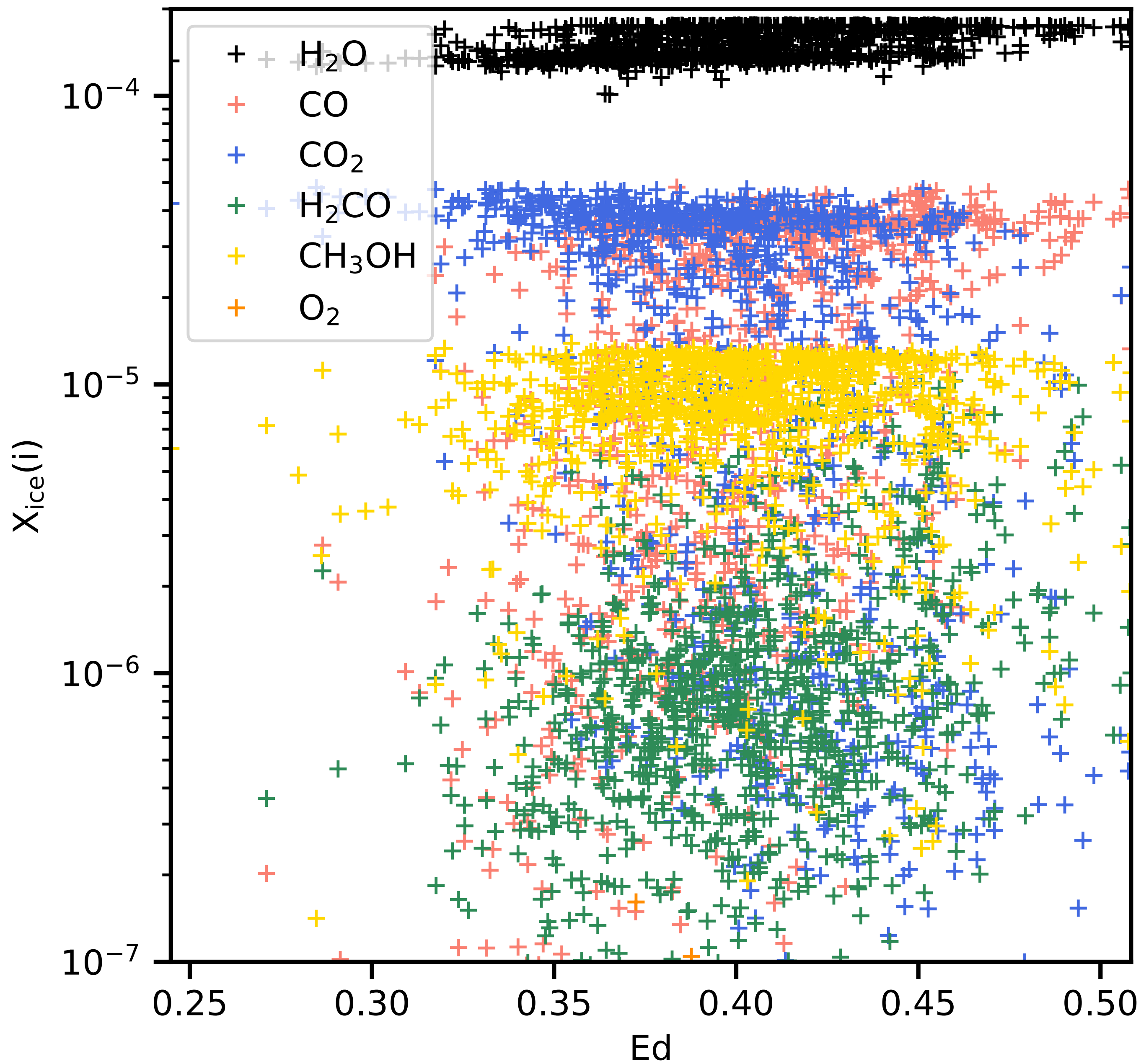

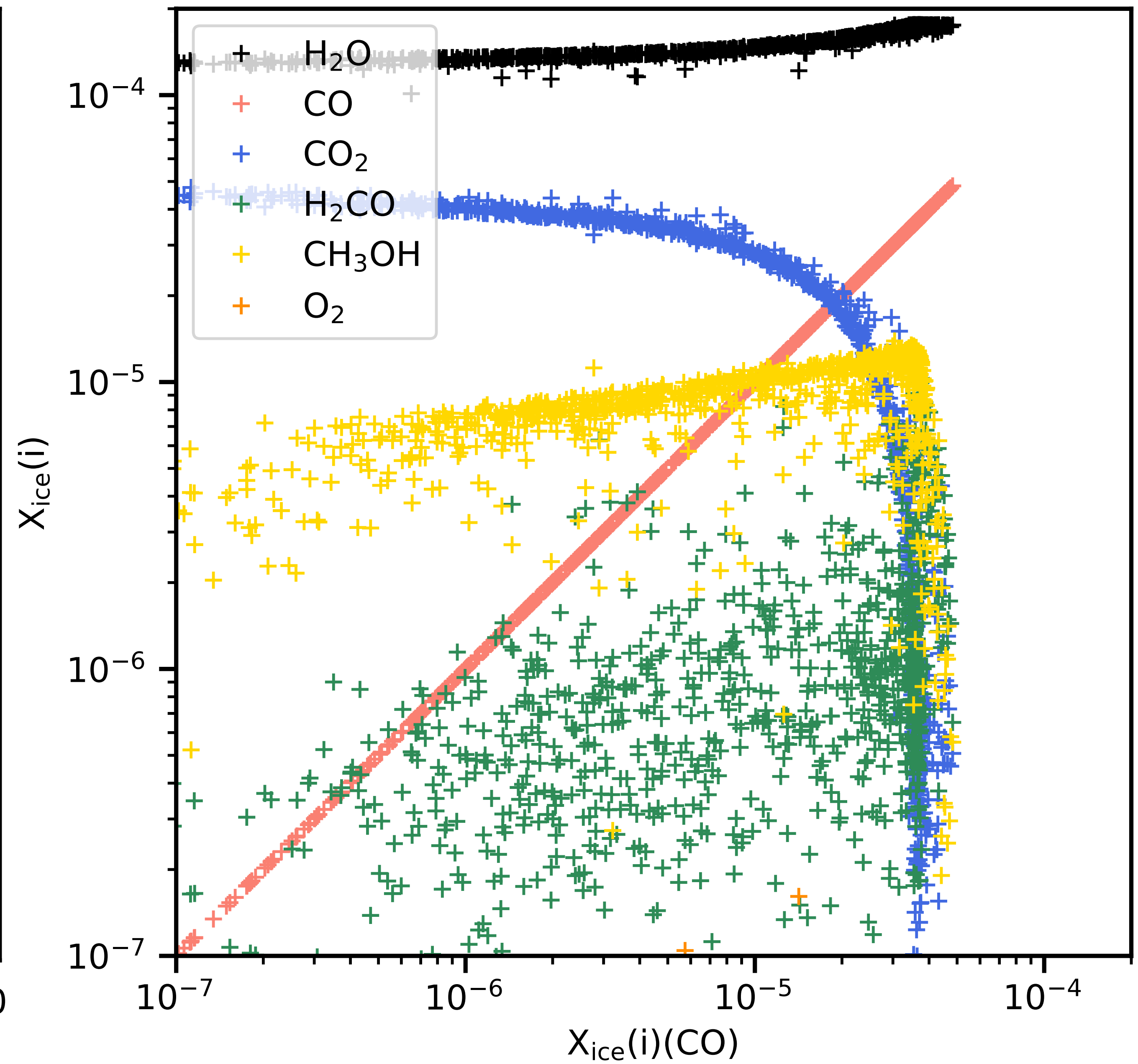

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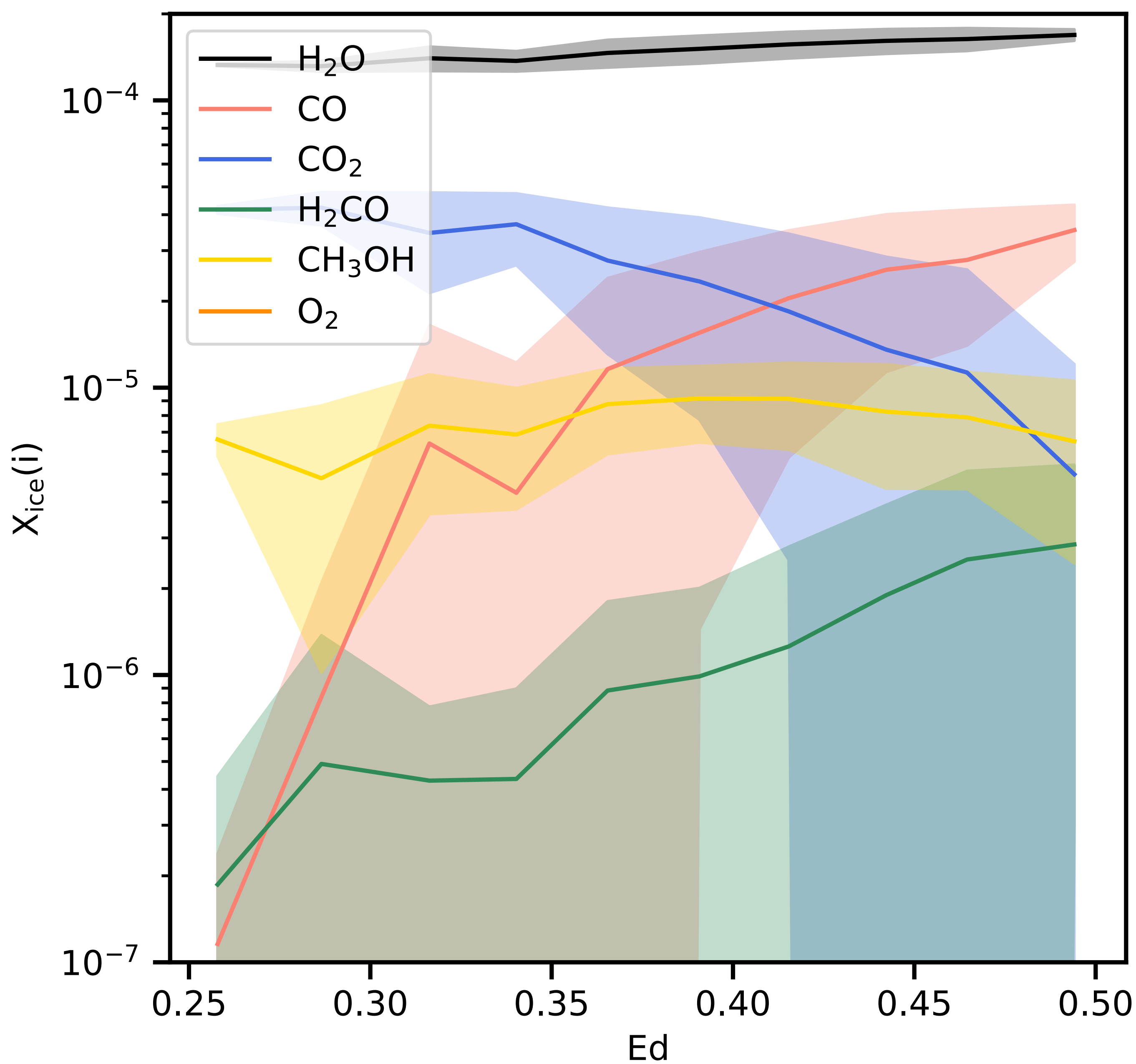
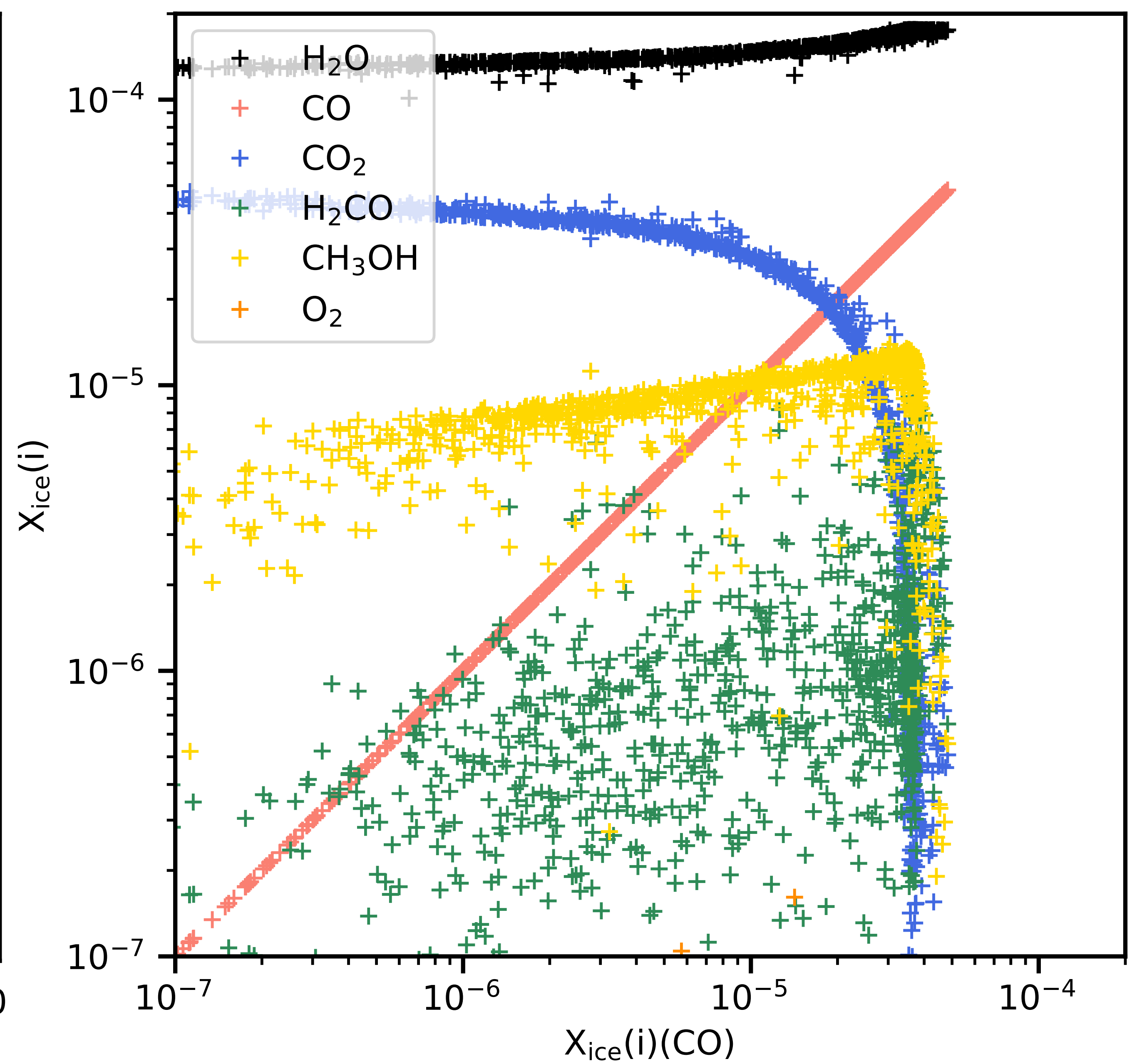




See recent works by Penteado et al. (2017), Holdship et al. (2018)

Constraining the models with experiments

Chemical modelling of laboratory experiments:

→ validation of the formalism **and** constraints on surface/chemical parameters

Example for CO hydrogenation (**Watanabe** et al. 2004 and **Fuchs** et al. 2009)

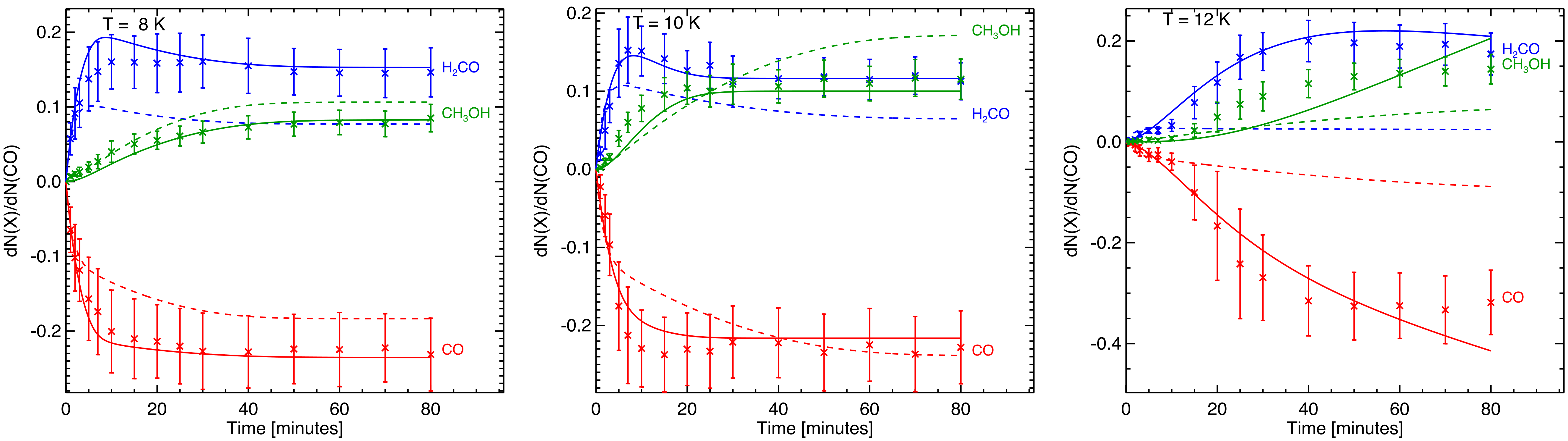
$E_d/E_b = 0.45 \pm 0.05$

$E_a(CO+H \rightarrow HCO) = 4000 \pm 500$ K

$E_a(H_2CO+H \rightarrow HCO+H_2) = 4000 \pm 250$ K

$E_a(H_2CO+H \rightarrow CH_3O) = 4750 \pm 250$ K

Flux = 5×10^{14} cm^{-2} s^{-1}



Experiments from Watanabe et al. (2004)

Conclusions

Multiphase rate equations offer a good compromise between chemical complexity, computational efficiency, and microchemical accuracy:

✔ **Predictions of the ice structure and ice evolution from dark clouds to protostars, disks, and planetary systems**

- Coupling with radiative transfer models to prepare and interpret IR spectra ?

✔ **High number of simulations to estimate uncertainties of predictions**

- Statistical analysis to evaluate the influence of input parameters
- Apply the model to laboratory experiments to constrain the models and understand the processes at work ?

Please contact me if you wish to use the code and/or if you have suggestions !
(taquet@arcetri.astro.it)