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## Molecular reactivity of ices in the protosolar nebula

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#### **Abstract**

Using laboratory experiments, we probe, at the molecular level, the evolution within the protosolar nebula of the composition of organic refractory materials obtained from astrophysical ices.

#### 1. Introduction

While ices and their organic content are observed in wide range of objects, spanning from dense molecular clouds to comets, the information on the composition of their organic content remains sparse and difficult to obtain. Laboratory experiments have been designed over the past decades to overcome this difficulty. This allows obtaining information on the chemistry and types of organic molecules transformed during the accretion and evolution of the early solar system bodies. In the present work, we investigate, using laboratory experiment, the complexification of organic matter originating from astrophysical ices under thermal and irradiation processing. The modification in the organic matter are monitored using very high resolution Orbitrap mass spectrometry.

#### 2. Methods

The setup used to produce our laboratory analogues has been described in detail in [1]. The samples analysed have been previously used to investigate the relationship between cometary materials and interstellar ices at a macromolecular level [2]. Briefly, to produce an organic residue, water, ammonia and methanol ices are co-deposited on a MgF2 substrate under vacuum and at 77K while being irradiated with VUV photons from a Hydrogen discharge lamp (2 x  $10^{14}$  photons.cm<sup>-2</sup>.s<sup>-1</sup>) for 48h. After which, the sample is slowly warmed up back to room temperature. To investigate the effect of UV dosage

received by the organic residue during its lifetime inside proto-planetary disks, one sample was over-irradiated at room temperature for an extra 48 hours without further deposition. For this study we synthetized 5 different samples from different ice composition or VUV alteration processing, summarized in Table 1.

Table 1: List of samples produced.

H <sub>2</sub> 0:CH <sub>3</sub> OH:NH <sub>3</sub>	Irradiation
ratio for ice	time (h)
formation	
3:1:1	48
3:1:1	96
3:1:5	48
3:1:0.2	48
10:1:1	48

Samples were analyzed using very high-resolution mass spectrometry (HRMS) with a LTQ Orbitrap XL. Residues were first dissolved in methanol. The soluble fraction was then analyzed by HRMS with equipped with ESI ionization in positive and negative mode. The very high resolution of the Orbitrap allows to determine exactly the formula of the several hundreds of molecules constituting the sample between m/z 200 and m/z 400 [3].

#### 3. Results and discussion

By determining precisely, the molecular composition of the samples we were able to retrieve part of their complexification history. Figure 1 presents a ternary space diagram corresponding to the relative proportion of carbon, nitrogen and oxygen atom in each individual molecules detected in the 3:1:5 and 3:1:0.2 samples. Each circle in this diagram corresponds to a molecule detected in the sample,

while the coloured stars correspond to the position of the original ices in this representation.

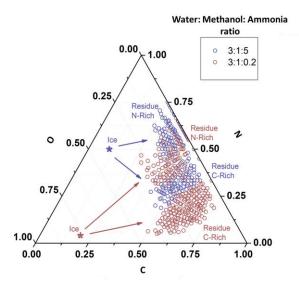


Figure 1: Ternary diagram of the molecules detected in the 3:1:5 (blue circles) and 3:1:0.2 (red circles) samples. The red and blue stars represent the position in the ternary space of the ice used to form the residues, respectively the 3:1:0.2 and 3:1:5 samples. Coloured arrows show the chemical complexification pathway underwent by the organic matter from the ice to the formation of the residue.

We see that, for both samples, the evolution of the ices leads to two distinct fraction of organic matter in the residue, one rich in nitrogen and one rich in carbon. This means that nitrogen incorporation, being in competition with the carbon, strongly drives the final composition of the residue. On one hand, ammonia rich ices lead to the formation of a group of unsaturated molecules in the final residue, while on the other hand  $H_2O$  rich ices lead to more saturated ones.

When looking at the effect of over VUV irradiation on the residue, we observe that this irradiation strongly drives the decarboxylation of organic matter [4]. The total dosage in our experiment would represent ~200 kyr of particle evolution in the protosolar nebula, which is enough to induce a complete decarboxylation of the organic matter in our sample. If organic rich grains spend around 1 Myr in the protosolar nebula as commonly admitted, our result show that they should be completely depleted in such functional groups. This is not the

case; as carboxylic acids are one of the main component of the soluble organic matter in CM meteorites. This mismatch could have three possible explanations. First, the organic grains could be shielded in the nebula, either by silicates or by enough prevent complete themselves, to decarboxylation. Second, the timescale of their residence in the irradiated zone could be shorter than previously suspected, either due to a shorter lifetime of the protosolar nebula, or a more effective convection in the disk. Finally, the carboxylic functions present in today meteorites, may not be inherited from interstellar ices, but from later reprocessing of the organic matter in the parent bodies accretion.

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