National Aeronautics and Space Administration

FY23 Innovative Spontaneous Concepts Research and Technology Development (ISC)

Ice Desorption of ^{14}N- and ^{15}N-bearing molecules relevant to protoplanetary disks.

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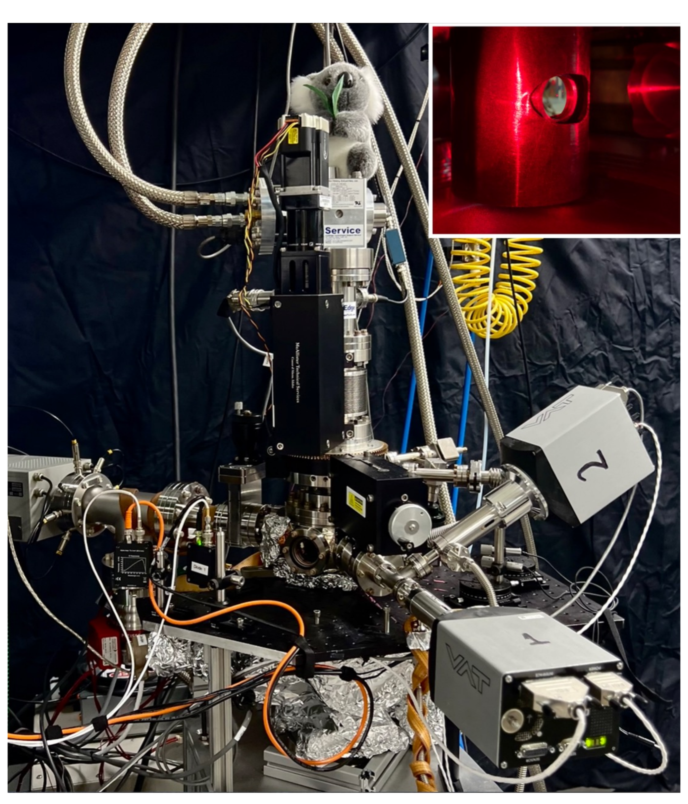
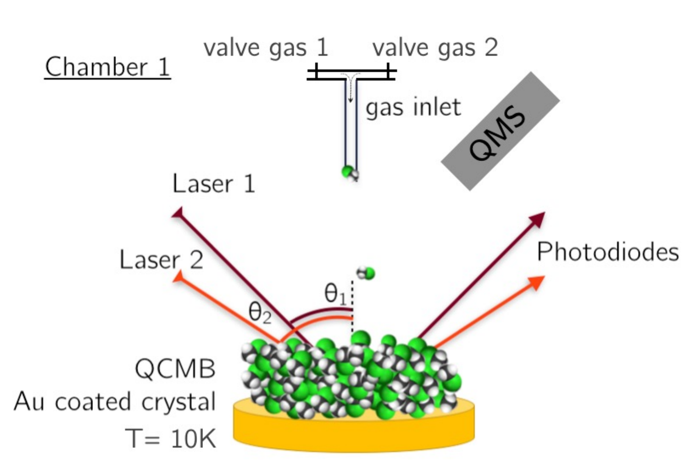
Objectives: The objective was to perform a preliminary experimental investigation in order to find out whether a difference in sublimation energy between isotopologues of N-bearing molecules relevant to protoplanetary disks exist and could lead to significant isotopic fractionation.

Background: The isotopic ratios in molecules measured throughout star-forming and planetary environments are some of the most powerful tools to understand the origin of chemicals from molecular clouds where young stars form all the way to mature planetary systems. In our Solar System, the high D/H ratio measures in Jupiter family comets suggests that these objects do not entirely have the same chemical origin than Earth's oceans[1]. A spread in $^{14}N/^{15}N$ ratio has been observed in various Solar System objects[2] and differences in protoplanetary disk emission regions between $HC^{15}N$ and $HC^{14}N$ have been found[3]. Understanding Solar System formation through isotopic measurements relies, however, on a robust knowledge of the fractionation processes at the various stages of the Protosolar Nebulae formation.

Given the temperature and densities found in planet forming regions, molecules can either be found in the gas phase or condensed on small grains with the adsorption and desorption processes regulate this partionning. Thus differences in sublimation efficiencies for between N-isotopologues of a similar species may lead to fractionation.

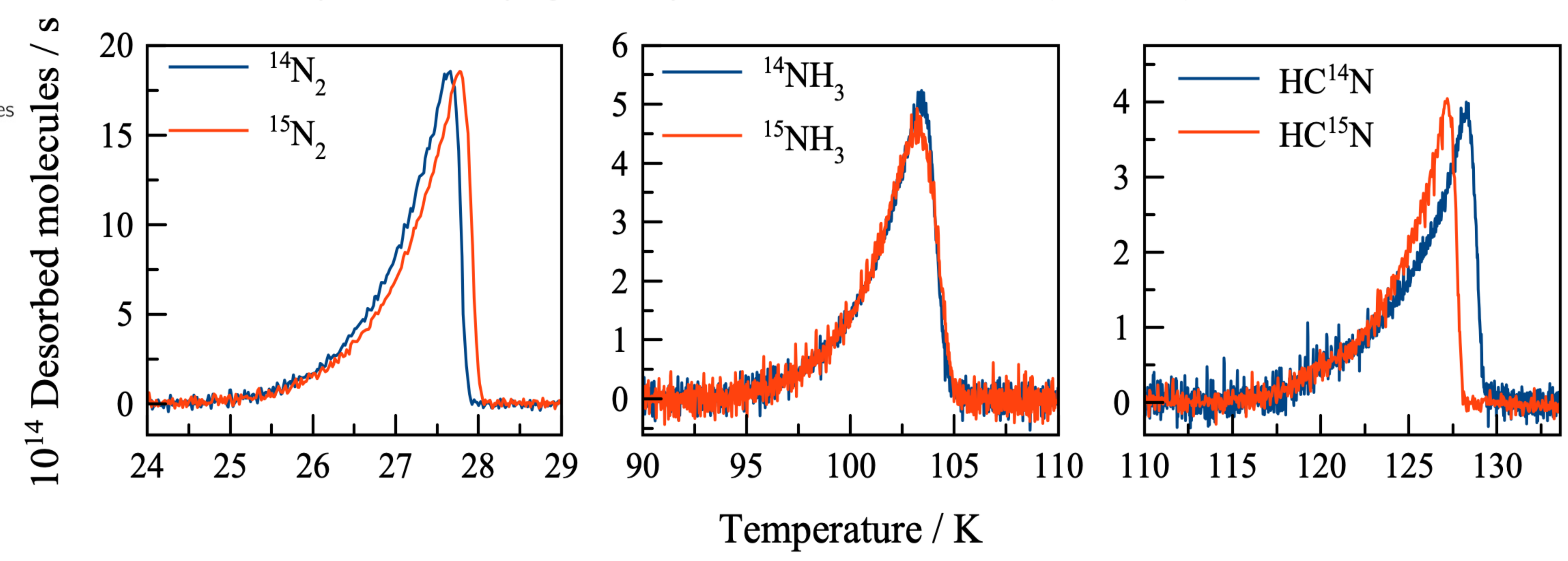
Approach and Results:
We measured the sublimation efficiencies of $^{14}N_2$ &$^{15}N_2$, $^{14}NH_3$ & $^{15}NH_3$, and $HC^{14}N$ and HC15N as these small molecules are the main N-carriers in protoplanetary disks. We then condensed the pure gaseous samples onto a quartz crystal microbalance cooled down to low temperatures (15K or 30K) in UHV. We deposited thin ice amorphous films (typically 50 monolayers equivalent) and slowly warm them at linear rates (0.5, 1, 2K.min^{-1}) until complete sublimation. The results can be seen in the figure for the sublimation at 2K.min^{-1}.





The temperature of the QCMB is increased linearly and the mass loss recorded

Desorption rate of 50x10^{15} molecules of ^{14}N- and ^{15}N-bearing molecules deposited on the crystal of a cryogenically cooled microbalance (0.53cm^2) heated at 2 K.min^{-1}



The sublimation of multilayer ice films typically occurs at a constant temperature dependent rate, so we was fit with zeroth-order desorption kinetics for the three sublimation rates in the form of d(ice loss)/dt = n.exp(-Edes/T(t)).

The comparison between the desorption of ^{14}N- and ^{15}N-isotopologue showed a range of behavior.
The N_2 isotopologue desorption followed the simple case where the heavier isotopologue go from ice to gas and desorb at higher temperature. This was not the case of NH_3, for which both isotopologues displayed similar desorption rates versus temperature.
In case of HCN, we observed that the lighter isotopologue $HC^{14}N$ sublimated at higher temperature than the $HC^{15}N$.

We are now working on a physical-chemical rational for our observations through, for e.g., a different description of the desorption process through transition state theory, which allows to include translational and rotational contribution to the desorption attempt frequency, which are not considered in the simple Arrhenius description.

Significance/Benefits to JPL and NASA:
This work should lead to a publishable letter, which will doubtlessly be of interest to the astrochemical community.
Both **deeper experimental investigations** (ice mixture desorption, desorption of different surfaces, fractionation inheritance into organics) coupled to state-of-the-art **disk chemistry simulations** would allow:
i) a quantification of the importance of N-ice fraction versus other fractionation mechanisms at the protoplanetary disk scale,
ii) predictions of ^{15}N enrichment during the Solar Nebulae evolution that could be compared to observed $^{15}N/^{14}N$ ratio throughout our Solar System.
This could be proposed to the Emerging Worlds ROSES program and subsequently provide rationales for planetary mission observables related to unveil the origin of our Solar System.

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Clearance Number: CL#00-0000
Poster Number: RPC#

Refs: [1]Altwegg et al, Science 2015, [2]Füri & Marty 2015, [3]Guzmán et al, ApJ 2017

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