

AstroChemical Newsletter #80

July 2022

You can access the full abstracts by clicking the paper titles. Submit your abstracts before the 25th of each month for inclusion in the following newsletter.

Abstracts

Detection of Cosmic Fullerenes in the Almahata Sitta Meteorite: Are They an Interstellar Heritage?

H. Sabbah, M. Carlos, P. Jenniskens, M. H. Shaddad, J. Duprat, C. A. Goodrich, C. Joblin

Buckminsterfullerene, C₆₀, is the largest molecule observed to date in interstellar and circumstellar environments. The mechanism of formation of this molecule is actively debated. Despite targeted searches in primitive carbonaceous chondrites, no unambiguous detection of C₆₀ in a meteorite has been reported to date. Here we report the first firm detection of fullerenes, from C₃₀ to at least C₁₀₀, in the Almahata Sitta (AhS) polymict ureilite meteorite. This detection was achieved using highly sensitive laser desorption laser ionization mass spectrometry. Fullerenes have been unambiguously detected in seven clasts of AhS ureilites. Molecular family analysis shows that fullerenes are from a different reservoir compared to the polycyclic aromatic hydrocarbons detected in the same samples. The fullerene family correlates best with carbon clusters, some of which may have been formed by the destruction of solid carbon phases by the impacting laser. We show that the detected fullerenes are not formed in this way. We suggest that fullerenes are an intrinsic component of a specific carbon phase that has yet to be identified. The nondetection of fullerenes in the Murchison and Allende bulk samples, while using the same experimental conditions, suggests that this phase is absent or less abundant in these primitive chondrites. The former case would support the formation of fullerenes by shock-wave processing of carbonaceous phases in the ureilite parent body. However, there are no experimental data to support this scenario. This leaves open the possibility that fullerenes are an interstellar heritage and a messenger of interstellar processes.

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CHEMOUT: CHEMical complexity in star-forming regions of the OUTER Galaxy. II. Methanol formation at low metallicity

Fontani, F., Schmiedeke, A., Sanchez-Monge, A., Colzi, L., Elia, D., Rivilla, V. M., Beltran, M. T., Bizzocchi, L., Caselli, P., Magrini, L., Romano, D.

The outer Galaxy is an environment with metallicity lower than Solar and, because of this, the formation and survival of molecules in star-forming regions located in the inner and outer Galaxy is expected to be different. To gain understanding on how chemistry changes throughout the Milky Way, it is crucial to observe outer Galaxy star-forming regions to constrain models adapted for lower metallicity environments. The project "chemical complexity in star-forming regions of the outer Galaxy" (CHEMOUT) aims to address this problem observing a sample of 35 high-mass star-forming cores at Galactocentric distances up to ~23 kpc with the IRAM 30m telescope in various 3mm and 2mm bands. In this work we analyse observations of methanol (CH₃OH), one of the simplest complex organic molecules crucial for organic chemistry in star-forming regions, and of two chemically related species, HCO and formaldehyde (H₂CO), towards 15 out of the 35 targets of the CHEMOUT sample. We considered only targets previously detected in both HCO and H₂CO, both precursors of methanol. We detected CH₃OH in all 15 targets. Using a Local Thermodynamic Equilibrium approach, we derive CH₃OH excitation temperatures in the range 7 - 16 K and line widths smaller than 4 km/s, consistent with emission from a cold and quiescent envelope. The CH₃OH fractional abundances w.r.t. H₂ are comparable to those found in star-forming regions in the inner and local Galaxy. Our results have important implications in the organic, and possibly pre-biotic, chemistry occurring in the outermost star-forming regions of the Galaxy, and can help setting the frontiers of the Galactic habitable zone.

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A survey of HDCO and D₂CO towards Class 0/I proto-brown dwarfs

Riaz, B. ; Thi, W. -F.

Deuterium fractionation can constrain the physical and chemical conditions at the early stage of brown dwarf formation. We present IRAM 30m observations over a wide frequency range of 213-279 GHz of singly and doubly deuterated species of formaldehyde (HDCO and D₂CO) towards Class 0/I proto-brown dwarfs (proto-BDs). Multiple low-excitation HDCO and D₂CO transition lines with upper energy level ≤ 40 K are detected. The D₂CO/HDCO, HDCO/H₂CO, D₂CO/H₂CO abundance ratios range between 0.01 and 2.5 for the proto-BDs, similar to the range seen in low-mass protostars. The

highest ratios of D2CO/HDCO \sim 1.3-2.5 are measured for two Stage 0 proto-BDs. These objects could possess a warm corino, similar to the few hot corino cases reported among Class 0 protostars. The mean D2CO/HDCO, D2CO/H2CO, and HDCO/H2CO ratios for the proto-BDs are comparatively higher than the range predicted by the current gas-grain chemical models, indicating that HDCO and D2CO are formed via grain surface reactions in the dense and cold interiors of the proto-BDs at an early formation stage.

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Discovery of the cyclic C5H radical in TMC-1

C. Cabezas, M. Agundez, R. Fuentetaja, Y. Endo, N. Marcelino, B. Tercero, J. R. Pardo, P. de Vicente and J. Cernicharo

Cyclic C5H (c-C5H), the radical formed by substituting an ethynyl group CCH for the hydrogen atom in the c-C3H radical, has been detected for the first time in the space. The c-C5H species is an isomer of the well-known linear radical l-C5H and is 6 kcal/mol less stable. A total of 17 rotational transitions were detected for the c-C5H species in TMC-1 within the 31.0-50.3 GHz range using the Yebes 40m radio telescope. We derive a column density of $(9.0 \pm 0.9)e10$ cm⁻² for c-C5H. Additionally, we observed 12 lines for l-C5H and derive a column density for it of $(1.3 \pm 0.3)e12$ cm⁻², which results in an abundance ratio c-C5H/l-C5H of 0.069. This is in sharp contrast with the value found for the analogue system c-C3H/l-C3H, whose ratio is 5.5 in TMC-1. We discuss the formation of c-C5H and conclude that this radical is probably formed in the reaction of atomic carbon with diacetylene.

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Theoretical Distribution of the Ammonia Binding Energy at Interstellar Icy Grains: A New Computational Framework

L. Tinacci, A. Germain, S. Pantaleone, S. Ferrero, C. Ceccarelli, P. Ugliengo

The binding energies (BE) of molecules on the interstellar grains are crucial in the chemical evolution of the interstellar medium (ISM). Both temperature-programmed desorption (TPD) laboratory experiments and quantum chemistry computations have often provided, so far, only single values of the BE for each molecule. This is a severe limitation, as the ices enveloping the grain mantles are structurally amorphous, giving rise to a manifold of possible adsorption sites, each with different BEs. However, the amorphous ice nature prevents the knowledge of structural details, hindering the development of a common accepted atomistic icy model. In this work, we propose a computational framework that closely mimics the formation of the interstellar grain mantle through a water by water accretion. On that grain, an unbiased random (but well reproducible) positioning of the studied molecule is then carried out. Here we present the test case of NH₃, a ubiquitous species in the molecular ISM. We provide the BE distribution computed by a hierarchy approach, using the semiempirical xTB-GFN2 as a low-level method to describe the whole icy cluster in combination with the B97D3 DFT functional as a high-level method on the local zone of the NH₃ interaction. The final ZPE-corrected BE is computed at the ONIOM(DLPNO-CCSD(T)/B97D3:xTB-GFN2) level, ensuring the best cost/accuracy ratio. The main peak of the predicted NH₃ BE distribution is in agreement with experimental TPD and computed data in the literature. A second broad peak at very low BE values is also present, which has never been detected before. It may provide the solution to a longstanding puzzle about the presence of gaseous NH₃ also observed in cold ISM objects.

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Unsaturated Dinitriles Formation Routes in Extraterrestrial Environments: A Combined Experimental and Theoretical Investigation of the Reaction between Cyano Radicals and Cyanoethene (C2H3CN)

Demian Marchione, Luca Mancini, Pengxiao Liang, Gianmarco Vanuzzo, Fernando Pirani, Dimitrios Skouteris, Marzio Rosi, Piergiorgio Casavecchia, and Nadia Balucani

The reaction between cyano radicals (CN, X²Σ⁺) and cyanoethene (C₂H₃CN) has been investigated by a combined approach coupling crossed molecular beam (CMB) experiments with mass spectrometric detection and time-of-flight analysis at a collision energy of 44.6 kJ mol⁻¹ and electronic structure calculations to determine the relevant potential energy surface. The experimental results can be interpreted by assuming the occurrence of a dominant reaction pathway leading to the two but-2-enedinitrile (1,2-dicyanoethene) isomers (E- and Z-NC-CH=CH-CN) in a H-displacement channel and, to a much minor extent, to 1,1-dicyanoethene, CH₂C(CN)₂. In order to derive the product branching ratios under the conditions of the CMB experiments and at colder temperatures, including those relevant to Titan and to cold interstellar clouds, we have carried out RRKM statistical calculations using the relevant potential energy surface of the investigated reaction. We have also estimated the rate coefficient at very low temperatures by employing a semiempirical method for the treatment of long-range interactions. The reaction has been found to be barrierless and fast also under the low temperature

conditions of cold interstellar clouds and the atmosphere of Titan. Astrophysical implications and comparison with literature data are also presented. On the basis of the present work, 1,2-dicyanoethene and 1,1-dicyanoethene are excellent candidates for the search of dinitriles in the interstellar medium.

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Modeling snowline locations in protostars: The impact of the structure of protostellar cloud cores

Nadia M. Murillo, Tien-Hao Hsieh, Catherine Walsh

Abridged Context: Snowlines during star and disk formation are responsible for a range of effects during the evolution of protostars, such as setting the chemical composition of the envelope and disk. This in turn influences the formation of planets by changing the elemental compositions of solids and affecting the collisional properties and outcomes of dust grains. Snowlines can also reveal accretion bursts, providing insight into the formation process of stars. **Methods:** A numerical chemical network coupled with a grid of cylindrical-symmetric physical models was used to identify what physical parameters alter the CO and H₂O snowline locations. The investigated parameters are the initial molecular abundances, binding energies of CO and H₂O, heating source, cloud core density, outflow cavity opening angle, and disk geometry. Simulated molecular line emission maps were used to quantify the change in the snowline location with each parameter. **Conclusions:** The models presented in this work show that the CO and H₂O snowline locations do not occur at a single, well-defined temperature as is commonly assumed. Instead, the snowline position depends on luminosity, cloud core density, and whether a disk is present or not. Inclination and spatial resolution affect the observability and successful measurement of snowline locations. We note that N₂H⁺ and HCO⁺ emission serve as good observational tracers of CO and H₂O snowline locations. However, constraints on whether or not a disk is present, the observation of additional molecular tracers, and estimating envelope density will help in accurately determining the cause of the observed snowline position. Plots of the N₂H⁺ and HCO⁺ peak emission radius versus luminosity are provided to compare the models with observations of deeply embedded protostars aiming to measure the CO and H₂O snowline locations.

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Diffusion activation energy and desorption activation energy for astrochemically relevant species on water ice show no clear relation

Kenji Furuya, Tetsuya Hama, Yasuhiro Oba, Akira Kouchi, Naoki Watanabe, Yuri Aikawa

The activation energy for desorption (*E_{des}*) and that for surface diffusion (*E_{sd}*) of adsorbed molecules on dust grains are two of the most important parameters for the chemistry in the interstellar medium. Although *E_{des}* is often measured by laboratory experiments, the measurement of *E_{sd}* is sparse. Due to the lack of data, astrochemical models usually assume a simple scaling relation, *E_{sd}* = *fE_{des}*, where *f* is a constant, irrespective of adsorbed species. Here, we experimentally measure *E_{sd}* for CH₄, H₂S, OCS, CH₃OH, and CH₃CN on water-ice surfaces using an ultra-high-vacuum transmission electron microscope (UHV-TEM). Compiling the measured *E_{sd}* values and *E_{des}* values from the literature, we find that the value of *f* ranges from ~0.2 to ~0.7, depending on the species. Unless *f* (or *E_{sd}*) for the majority of species is available, a natural alternative approach for astrochemical models is running multiple simulations, varying *f* for each species randomly. In this approach, ranges of molecular abundances predicted by multiple simulations, rather than abundances predicted by each simulation, are important. We here run 10,000 simulations of astrochemical models of molecular clouds and protostellar envelopes, randomly assigning a value of *f* for each species. In the former case, we identify several key species whose *E_{sd}* most strongly affects the uncertainties of the model predictions; *E_{sd}* for those species should be investigated in future laboratory and quantum chemical studies. In the latter case, uncertainties in the *E_{sd}* of many species contribute to the uncertainties in the model predictions.

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A New Method for Direct Measurement of Isotopologue Ratios in Protoplanetary Disks: A Case Study of the 12CO/13CO Ratio in the TW Hya Disk

T. Yoshida, H. Nomura, K. Furuya, T. Tsukagoshi, S. Lee

Planetary systems are thought to be born in protoplanetary disks. Isotope ratios are a powerful tool for investigating the material origin and evolution from molecular clouds to planetary systems via protoplanetary disks. However, it is challenging to measure the isotope (isotopologue) ratios, especially in protoplanetary disks, because the emission lines of major species are saturated. We developed a new method to overcome these challenges by using optically thin line wings induced by thermal broadening. As a first application of the method, we analyzed two carbon monoxide isotopologue lines, 12CO 3–2 and 13CO 3–2, from archival observations of a protoplanetary disk around TW Hya with the Atacama Large Millimeter/submillimeter Array. The 12CO/13CO ratio was estimated to be 20±5 at disk radii of 70–110 au, which is significantly smaller than the value observed in the local interstellar medium, ~69. It implies that an isotope exchange reaction occurs in a low-

temperature environment with $C/O > 1$. In contrast, it is suggested that $^{12}CO/^{13}CO$ is higher than ~ 84 in the outer disk ($r > 130$ au), which can be explained by the difference in the binding energy of the isotopologues on dust grains and the CO gas depletion processes. Our results imply that the gas-phase $^{12}CO/^{13}CO$ can vary by a factor of > 4 even inside a protoplanetary disk, and therefore, can be used to trace material evolution in disks.

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Experimental study of the removal of excited state phosphorus atoms by H₂O and H₂: implications for the formation of PO in stellar winds

Kevin M. Douglas, David Gobrecht, John M. C. Plane

The reactions of the low-lying metastable states of atomic phosphorus, P(2D) and P(2P), with H₂O and H₂ were studied by the pulsed laser photolysis at 248 nm of PCI₃, combined with laser induced fluorescence detection of P(2D), P(2P) and PO. Rate coefficients between 291 and 740 K were measured, along with a yield for the production of PO from P(2D or 2P) + H₂O of (35±15)%. H₂ reacts with both excited P states relatively efficiently; physical (i.e. collisional) quenching, rather than chemical reaction to produce PH + H, is shown to be the more likely pathway. A comprehensive phosphorus chemistry network is then developed using a combination of electronic structure theory calculations and a Master Equation treatment of reactions taking place over complex potential energy surfaces. The resulting model shows that at the high temperatures within two stellar radii of a MIRA variable AGB star in oxygen-rich conditions, collisional excitation of ground-state P(4S) to P(2D), followed by reaction with H₂O, is a significant pathway for producing PO (in addition to the reaction between P(4S) and OH). The model also demonstrates that the PN fractional abundance in a steady (non-pulsating) outflow is under-predicted by about 2 orders of magnitude. However, under shocked conditions where sufficient thermal dissociation of N₂ occurs at temperatures above 4000 K, the resulting N atoms convert a substantial fraction of PO to PN.

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Resolving desorption of complex organic molecules in a hot core: Transition from non-thermal to thermal desorption or two-step thermal desorption?

L. A. Busch, A. Belloche, R. T. Garrod, H. S. P. Müller, and K. M. Menten

Using the high angular resolution provided by the ALMA interferometre we want to resolve the COM emission in the hot molecular core Sagittarius B2(N1) and thereby shed light on the desorption process of Complex Organic Molecules (COMs) in hot cores. We use data taken as part of the 3 mm spectral line survey Re-exploring Molecular Complexity with ALMA (ReMoCA) to investigate the morphology of COM emission in Sagittarius B2(N1). Spectra of ten COMs are modelled under the assumption of LTE and population diagrams are derived for positions at various distances to the south and west from the continuum peak. Based on this analysis, resolved COM rotation temperature and COM abundance profiles are derived. Based on the morphology, a rough separation into O- and N-bearing COMs can be done. Temperature profiles are in agreement with expectations of protostellar heating of an envelope with optically thick dust. Abundance profiles reflect a similar trend as seen in the morphology and, to a great extent, agree with results of astrochemical models that, besides the co-desorption with water, predict that O-bearing COMs are mainly formed on dust grain surfaces at low temperatures while at least some N-bearing COMs and CH₃CHO are substantially formed in the gas phase at higher temperatures. Our observational results, in comparison with model predictions, suggest that COMs that are exclusively or to a great extent formed on dust grains desorb thermally at ~ 100 K from the grain surface likely alongside water. Non-zero abundance values below ~ 100 K suggest that another desorption process is at work at these low temperatures: either non-thermal desorption or partial thermal desorption related to lower binding energies experienced by COMs in the outer, water-poor ice layers. In either case, this is the first time that the transition between two regimes of COM desorption has been resolved in a hot core.

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Laboratory Experiments on the Radiation Astrochemistry of Water Ice Phases

Duncan V. Mifsud, Perry A. Hailey, Péter Herczku, Zoltán Juhász, Sándor T. S. Kovács, Béla Sulik, Sergio Ioppolo, Zuzana Kaňuchová, Robert W. McCullough, Béla Paripás, Nigel J. Mason

Water (H₂O) ice is ubiquitous component of the universe, having been detected in a variety of interstellar and Solar System environments where radiation plays an important role in its physico-chemical transformations. Although the radiation chemistry of H₂O astrophysical ice analogues has been well studied, direct and systematic comparisons of different solid phases are scarce and are typically limited to just two phases. In this article, we describe the results of an in-depth study of the 2 keV electron irradiation of amorphous solid water (ASW), restrained amorphous ice (RAI) and the cubic (Ic) and hexagonal (Ih) crystalline phases at 20 K so as to further uncover any potential dependence of the radiation physics and chemistry on the solid phase of the ice. Mid-infrared spectroscopic analysis of the four investigated H₂O ice phases revealed that electron irradiation of the RAI, Ic, and Ih phases resulted in their amorphization (with the latter undergoing the process more slowly) while ASW underwent compaction. The abundance of hydrogen peroxide (H₂O₂) produced as a result

of the irradiation was also found to vary between phases, with yields being highest in irradiated ASW. This observation is the cumulative result of several factors including the increased porosity and quantity of lattice defects in ASW, as well as its less extensive hydrogen-bonding network. Our results have astrophysical implications, particularly with regards to H₂O-rich icy interstellar and Solar System bodies exposed to both radiation fields and temperature gradients.

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The role of highly vibrationally excited H₂ initiating the N chemistry: Quantum study and 3-sigma detection of NH emission in the Orion Bar PDR

Javier R. Goicoechea, Octavio Roncero

The formation of hydrides by gas-phase reactions between H₂ and a heavy element atom is a very selective process. Reactions with ground-state neutral carbon, oxygen, nitrogen, and sulfur atoms are very endoergic and have high energy barriers because the H₂ molecule has to be fragmented before a hydride bond is formed. In cold interstellar clouds, these barriers exclude the formation of CH, OH, NH, and SH radicals through hydrogen abstraction reactions. Here we study a very energetically unfavorable process, the reaction of N(4S) atoms with H₂ molecules. We calculated the reaction rate coefficient for H₂ in different vibrational levels, using quantum methods for v=0-7 and quasi-classical methods up to v=12. Owing to the high energy barrier, these rate coefficients increase with v and also with the gas temperature. We implemented the new rates in the Meudon PDR code and studied their effect on models with different ultraviolet (UV) illumination conditions. In strongly UV-irradiated dense gas (Orion Bar conditions), the presence of H₂ in highly vibrationally excited levels (v>7) enhances the NH abundance by two orders of magnitude (at the PDR surface) compared to models that use the thermal rate coefficient for reaction N(4S) + H₂ -> NH + H. The increase in NH column density across the PDR is a factor of ~25. We explore existing Herschel/HIFI observations of the Orion Bar and Horsehead PDRs. We report a 3-sigma emission feature at the ~974 GHz frequency of the NH N_{J=1_2-0_1} line toward the Bar. The emission level implies N(NH)~10¹³ cm⁻², which is consistent with PDR models using the new rate coefficients for reactions between N and UV-pumped H₂. This formation route dominates over hydrogenation reactions involving the less abundant N⁺ ion. JWST observations will soon quantify the amount and reactivity of UV-pumped H₂ in many interstellar and circumstellar environments.

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Announcements

ngVLA Astrochemistry Science Working Group: mailing list and first meetings

In recognition that astrochemistry is expected to be a key science driver for the ngVLA, the project team has formed a new, dedicated astrochemistry science working group to interface with the community and solicit feedback critical to setting design requirements at this foundational stage of the project. This announcement outlines how you can get involved and the dates of our first community telecons.

The 2020 Decadal Survey has strongly endorsed the construction and operation of the Next-Generation Very Large Array (ngVLA), for which astrochemistry is a key science driver.

In preparation for the Decadal Survey, the ngVLA project put together a Science Book (<https://ngvla.nrao.edu/page/scibook>) of Scientific Use Cases submitted by the community and organized through a number of Science Working Groups (SWGs). As the ngVLA enters the next major phase of development, the ngVLA Science Advisory Council is reinvigorating these SWGs with the aim of revisiting, updating, and expanding the Scientific Use Cases in the context of new discoveries over the last 5 years. Recognizing the critical role that astrochemistry will play in setting technical requirements and key science drivers for the ngVLA, the project has re-baselined the SWGs to include an SWG specifically focusing on astrochemistry and the molecular emergence of life.

In the coming months, the ngVLA project will undergo a requirements baseline as part of a technical conceptual design review. This will be a major step forward toward entering the formal Design phase as an MREFC (Major Research Equipment and Facilities Construction) candidate for the NSF. At this juncture, ensuring that the ngVLA's science strengths are identified and promoted is of critical importance. It is also important for the project to be able to align its strengths with the science ambitions of Astro2020.

SWG2 (Astrochemistry) will hold two telecons in the month of July to discuss how best to proceed to meet the goals of the project, both in setting critical technical specifications and in messaging to the scientific community, general public, and government agencies and congress. This includes identifying current gaps in science use cases (and the associated design requirement drivers), as well as "Big Questions" we seek to answer of the kind that are appealing to the public and lawmakers.

Tuesday 19 July: 9:00 a.m. ET

Thursday 21 July: 1:00 p.m. ET

If you are interested in being a part of these discussions, we urge you to join our mailing list at the link below. The content of the telecons will be identical - we are just trying to accommodate for different schedules and timezones. Connection details will be distributed via the mailing list shortly in advance of the meetings.

<https://listmgr.nrao.edu/mailman/listinfo/ngvla-swg2>

Thank you in advance for your support and advocacy for this transformational observational facility.
– Brett McGuire & Jenny Bergner (SWG2 Co-Chairs)