NWO Astrochemistry programme

sub-themes selected at kick-off meeting (7 October 2009)

I. Gas-phase Astrochemistry
II. The solid Universe: grans and Ic
III. The aromatic Universe
IV. Pebiotic Universe

Summary of sub-themes

I. Gas-phase Astrochemistry

Topic 1: Photodissociation processes

In any region in which UV photons can penetrate, photodissociation is the major destruction mechanism of (neutral) gas-phase molecules. This includes diffuse and translucent clouds, dense clouds exposed to intense UV from nearby young stars (so-called Photodissociation or Photon-Dominated Regions, PDRs) in our own and other galaxies, the envelopes around evolved stars, and the surface layers of protoplanetary disks. Even deep inside dense clouds, the UV photons produced by the interaction of cosmic rays with H_2 provide a weak field which can dissociate both gas-phase and solid-state molecules (ices). Photodissociation processes may well be responsible for the isotopic anomalies observed in meteorites in our own solar system: how and where did this happen? While data on a number of molecules are available from the older literature (see www.strw.leidenuniv.nl/~ewine/photo for overview), information on several key rates and processes is lacking.

In this topic, unique Dutch theoretical and experimental expertise on photodissociation processes will be brought together to address a number of outstanding questions in (astro)chemistry:

- what are the photodissociation rates and mechanisms of key small molecules in astrochemistry, in particular N_2 and CO? How does this vary among the isotopologues and what is the importance of self- and mutual shielding?

- what are the branching ratios of the photodissociation products, especially for >triatomic molecules? For example, does CH_3OH dissociate primarily to CH_3 + OH, CH_2OH + H, or CH_2 + H_2O, etc.? How does this depend on photon energy?

- do the branching ratios change between molecules in the gas and ices? (link with "Solid-state astrochemistry" theme)

- how stable are complex organic molecules (non-PAHs) against UV radiation? Where does the cross-over from very efficient dissociation as found for small molecules to highly stable because of internal conversion as found for large molecules occur? How does this depend on the structure of the molecule?

Topic 2: Inelastic collisions

Because of their rich energy level structure, molecules are excellent probes of the physical conditions in which they reside. Their abundances can also serve as chemical diagnostics of, for example, the evolutionary state of the sources. In order to extract any information about temperatures, densities and abundances, the state-to-state collisional rate coefficients need to be known over a range of temperatures (typically 10 K - few hundred K). These can only be determined by a combination of experiments and theory. Experimental determinations of individual cross sections at specific energies are crucial to benchmark the theoretical efforts.

Over the last decades, there have been significant efforts in computing and measuring collisional rate coefficients for astrophysically relevant molecules with the main collision partners H_2 and H.

This, in turn, also provides new challenges for basic chemical physics on treating open shell molecules (e.g., OH) and both para- and ortho- H_2 properly. A summary up to a few years ago is given by Schoeier et al. 2005 (Astronomy & Astrophysics) and the relevant data are summarized in an easy-to-access form at www.strw.leidenuniv.nl/~moldata. The Molecular Universe EU network has been very active in the last few years in providing new data on a number of species, most importantly H_2O, crucial for interpretation of upcoming Herschel data.

Most recently, observations using the Spitzer Space Telescope combined with ground-based 8-m class telescopes have detected a wealth of mid-infrared (3-30 micron) lines of small molecules like CO, HCN, C_2H_2, CO_2, H2O and OH. These are mostly the vibration-rotation lines (but in the case of H_2O and OH also very highly excited pure rotational lines). Very little is known about collisional rate coefficients for vibration rotation transitions; even for "simple" systems like CO-H or CO-H_2 uncertainties are still an order of magnitude.

The goal of this topic would be to define a number of astrophysically relevant systems to study in a combined theory-experimental effort.

For both topic 1 and 2, the molecular data would subsequently be incorporated into state-of-the-art molecular excitation and radiative transfer models and applied to interpretations of observations of molecules in star-forming regions, protoplanetary disks and extragalactic regions.

II. The solid universe: Grains and Ice

Large areas of space in our Galaxy are filled by molecular clouds that consist of gas (mainly hydrogen) and (sub)micron sized silicate and carbonaceous dust grains. At some point in time these clouds gravitationally collapse and with decreasing radius, the particle density increases; the cloud becomes optically opaque and turns dark. As light penetration is hindered, the cloud cools down and gas starts accreting onto the dust grain surfaces that act as highly effective cryo-pumps. This results in layered geometries of partially mixed ices on top of the grains. The gravitational collapse goes on until densities get so high that a new star forms and the gas and dust become part of an envelope that feeds the central protostar. Part of the gas and dust ends up in a rotating disc surrounding the young star, the so called proto-planetary disk, and provides the basic material from which icy planetesimals, and ultimately planets are formed. Once the star starts burning these icy dust grains are irradiated by additional UV light and are bombarded by a continuous flux of atoms triggering an exotic solid state chemistry. In a later stage, the grains warm up and thermally induced processes become important. Molecules and reaction products start desorbing from the dust surfaces and start participating in gas phase reactions in which new species are formed.

Today, the origin of planetary systems around young stars, including the enticing implications for the distribution of complex molecules in the universe, are key avenues pursued in modern astrochemical studies by observational astronomers, astrochemists and experimentalists. Given the unresolvable scale size at interstellar distances the embedded environment, and the uncertainty of the dominating solid state processes, it is a challenging task to quantitatively characterize processes that are astrophysically and astrochemically relevant. This is the goal of the solid state theme within the NWO-astrochemistry proposal that addresses key questions related to the structure and aggregation of dust grains, grain mediated chemistry and chemical reactions in ices on dust grains.

The Netherlands has a long standing tradition in the study of astronomical dust. This proposal takes this expertise as a starting point, extending it to new ideas with techniques that come from observational astronomy, laboratory astrophysics, and theoretical chemistry.

Structural studies of grains, comparing synchrotron laboratory data of home made samples with observational data (XMM-Newton / Chandra X-ray) are possible at SRON Utrecht. Aggregation of dust grains can be studied with facilities available at the van ‘t Hoff laboratory in Utrecht, using colloids of granular systems with long-range (electrostatic) repulsion, and short-range attraction as model systems. Grain mediated chemistry is likely responsible for the so far mysterious conversion mechanism of gas-phase sulphur to FeS, as found in meteorites. It is also unclear whether metal atoms can be liberated upon X-ray and/or cosmic ray impact of dust grains. Interest for these projects exists at the astronomical Institutes in Amsterdam and Utrecht. Laboratory ice studies are possible at the Sackler Laboratory for Astrophysics. Several solid state setups are dedicated to the study of chemical processes in ices upon thermal and UV-photoprocessing, and atom bombardment, both for simple and complex (PAH-containing) ices. The laboratory data are used to guide astrochemical models and to interpret gas-grain interactions (UL, RUG). Theoretical chemical support is particularly available for structural calculations (VU) and dynamical simulations (UL) of photo-induced processes in the ice. Further characterization is possible through collaborations with surface (RUG) and plasma (TUE) groups to understand ice geometry and surface plasma interaction, respectively.

III. The Aromatic Universe

Broad emission features dominate the mid-infrared spectra of most astronomical objects, including regions of star and planet formation, photodissociation regions where massive stars interact with their surrounding material, planetary nebula, and the interstellar medium of galaxies. These emission features are due to IR fluorescence of UV pumped, Polycyclic Aromatic Hydrocarbon molecules. These large molecules are a ubiquitous and abundant component of the interstellar medium of galaxies. PAHs are thought to form in the ejecta of stars at the end of their evolution in chemical processes akin to those in sooting flames. In the interstellar medium, PAHs evolve through photoprocessing, weeding out less stable forms, and through energetic ion interactions in shocks. In dense clouds, PAHs may cluster, become incorporated into ices, or complex with gas phase atoms or molecules.

The aromatic theme within the NWO astrochemistry initiative will focus on PAH species but include a wider set of species with a possible connection to the interstellar PAH family. These include carbon chains, large carbon clusters including fullerenes, nitrogenated PAHs, clusters of PAH molecules, complexes of PAHs with astrophysically relevant atoms or molecules, deuterated PAHs, …

Key question in the aromatic universe are the spectroscopic signatures of PAHs from the UV through the visible into the mid- and far-IR and the fragmentation and isomerization of PAHs by UV and X-ray photons and energetic ions. These studies are uniquely enabled in Holland through the infrared laser facility, FELIX in Nieuwegein, and the spectroscopic facilities at the UvA and the VU Laser Center. The THz free-electron laser facility in development in Nijmegen will provide another unique tool. The KVI in Groningen may be able to study energetic ion interaction at astrophysically relevant energies.

The aim is to systematic study a series of related gas phase species and in that way develop an understanding of PAH spectra and PAH stability in the ISM. The IR is key to analyzing observed interstellar spectra in terms of the properties of the carriers. UV and visible spectroscopy are key to understanding the energy balance of PAHs and related species as well as their putative role as DIB carriers. The processing aspects take aim at the formation and evolution of PAHs in the ISM and their role in molecule formation. There is wide chemical interest in these aspects as well. These include understanding molecular properties in a systematic way. Of particular interest are the electronic structure of PAHs and their link to graphene and in a more general sense the properties of (semi)conductors. Some of the techniques that will be developed under this initiative (eg., helium droplet techniques) are aimed to become facility-like instrumentation at Dutch laboratories.

Areas which are presently not covered are quantum chemical theory of PAH vibrational and electronic spectra as well as their chemical kinetics. Perhaps, a link to the soot community can be made as well through the role of PAHs in sooting flames.

IV. Prebiotic Universe – an overview

Carbon is a key element in the evolution of prebiotic material. Our understanding of the evolution of organic molecules and their voyage from molecular clouds to the early solar system and Earth provides important constraints on the emergence of life on Earth and possibly elsewhere. Astronomical observations have shown that carbonaceous compounds in the gas or solid state, refractory or icy are ubiquitous in our and distant galaxies (van Dishoeck & Blake 1998; Ehrenfreund & Charnley 2000; Pendleton & Allamandola 2002, van Dishoeck 2004, Tielens 2008). Interstellar molecular clouds and circumstellar envelopes are factories of complex molecular synthesis (Kwok 2004). During the formation of the solar system, this interstellar organic material (gas and dust) was chemically processed and integrated in the solar nebula from which planets and small solar system bodies formed. Small bodies, such as comets and asteroids and their fragments impacted the young planets in the early history of the solar system (Gomes et al. 2005). A surprisingly high number of biomolecules and molecules that are used in contemporary biochemistry on Earth are found in the interstellar medium, planetary atmospheres and surfaces, comets, asteroids and meteorites as well as interplanetary dust particles. Planet Earth was formed through a hot accretion process that allowed only the rocky material from the inner solar system to survive. Ices were sublimed and existing carbon material (volatile and refractory) combusted. Therefore, organic molecules found on terrestrial planets must have formed after the planetary surface cooled, or were delivered via impacts by small bodies in the young solar system. All terrestrial planets have been seeded with organic compounds through the impact of small bodies during solar system formation. Part of this material may have been important starting material for life (Chyba et al. 1990; Ehrenfreund et al. 2002).

Volatile and robust carbon compounds residing in interstellar clouds were recycled during solar system formation (Ciesla 2008). The dynamic environment of the solar nebula with the simultaneous presence of gas, particles, and energetic processes, including shock waves, lightning, and radiation (e.g. Gorti et al. 2009) can trigger a rich organic chemistry leading to organic molecules. Turbulent motion led to radial mixing of the products within the disk (Visser et al. 2007; Dullemond et al. 2008). The carbonaceous inventory of our solar system therefore contains highly processed material that was exposed to high temperatures and radiation, newly formed compounds, and some relatively pristine material with significant interstellar heritage. Organic compounds observed or sampled from our solar system, such as planetary surfaces/atmospheres, comets, and interplanetary dust, thus hold clues to processes that occurred during the origin of our solar system (Ehrenfreund & Charnley 2000; Cruikshank et al. 2005; Septhon & Botta 2005; Raulin 2008).

More than 50 molecules have been identified in cometary comae (Crovisier et al.2009). Many small organic molecules observed in cometary comae probably originate wholly or partially from the decomposition of larger molecules or particles, indicating that large polymers such as polyoxymethylene and HCN-polymers are present in comets (Ehrenfreund et al. 2004; Cottin & Fray 2008). Carbonaceous chondrites (meteorites) and micrometeorites do contain a variety of organics (e.g. see Alexander 2005; Sephton 2002; Septhon & Botta 2005 for reviews). They are fragments of cometary and asteroidal bodies. Investigating their organic composition often indicates the nature of the parental body (Hiroi et al.1993; Ehrenfreund et al.2001; Nesvorny et al.2009).

Summary of the organic inventory of comets and meteorites

The large quantities of extraterrestrial material delivered to young terrestrial planetary surfaces in the early history of our solar system may have provided the material necessary for the emergence of life (Chyba et al. 1992; Ehrenfreund et al. 2002). Comets are icy bodies containing silicates and refractory organic material formed in the region beyond Jupiter (Greenberg 1998; Di Santi & Mumma 2009). The NASA Stardust comet sample-return mission captured recently cometary dust intact at a velocity of 6.1 km/s in 300 km vicinity of comet Wild-2. Most of the > 5 µm solid particles collected by the mission are mineral grains or assemblages of high-temperature minerals that condense at 1400 K or above. The data provided evidence for radial transport of large solid grains from the center of the solar nebula to the Kuiper belt (Brownlee et al. 2006). Comets probably contributed most of the carbonaceous compounds during the heavy bombardment phase 4.5–4 billion years ago (Ehrenfreund et al. 2002). Fragments of asteroids and comets such as interplanetary dust particles (IDPs) and carbonaceous meteorites were probably among the other major extraterrestrial contributors of carbon (Chyba et al. 1990).

Carbonaceous meteorites contain a substantial amount of carbon (up to 3 % by weight) and exhibit evidence of thermal and aqueous alteration believed to have occurred on their parent bodies (Sephton 2002; Botta & Sephton 2005; Martins et al. 2007). In the soluble fraction of the Murchison meteorite, more than 70 extraterrestrial amino acids have been identified in addition to many other organic compounds, including N-heterocycles, carboxylic acids, sulfonic and phosphonic acids, and aliphatic and aromatic hydrocarbons (Cronin et al. 1993; Martins et al. 2007, 2008). However, the major carbon component in meteorite samples is composed of a macromolecular organic fraction (Alexander et al. 2007). Although several classes of organic compounds important in contemporary biochemistry are found in the interstellar medium, comets and meteorites, the dominant form of carbonaceous material that will be delivered is likely aromatic in nature.

Prebiotically-significant organic material identified in meteorites is formed within the solar system in radiation shielded environments and in the presence of liquid water. This is strongly supported by recent results that provide evidence that the organic composition in carbonaceous meteorites is dependent on parent body alteration processes, in particular aqueous alteration (Martins et al. 2007; Glavin & Dworkin 2009). Amino acids are likely formed via the process of Strecker-cyanohydrin synthesis. This formation route requires the presence of aldehydes or ketones, liquid water, HCN and NH_3. Additionally to amino acids and sugar-related compounds another important precursor molecule, namely nucleobases, were recently detected in the Murchison meteorite (Martins et al. 2008). Possible formation routes include polymerization of HCN, although many other mechanism have been proposed as well (Martins et al. 2008). Consequently, nucleobases delivered to young planets together with sugar-related species and amino acids might have been beneficial to the origin of life on Earth, Mars, or elsewhere. Life on Earth is based on amino acids that are left-handed. An L-excess of several meteoritic amino acids has been measured indicating that chirality for some biomolecules may have been predetermined (Glavin & Dworkin 2009). The inorganic matrix of meteorite parent bodies has undoubtedly played an important role in the formation and evolution of solar system organics (Hazen et al. 2001). The high abundance of aromatic material in meteorites led to the hypothesis of the "aromatic world" that describes PAH-based transitions from non-living to living matter as an interesting alternative to traditional origin of life models (Ehrenfreund et al. 2006).

In contrast, the endogenous synthesis of prebiotic organic compounds may have been constrained by the conditions on the young Earth. Whatever the inventory of endogenous organic compounds on the ancient Earth, it would have been augmented by extraterrestrial material. It is estimated that these sources delivered ~10^9 kg of carbon per year to the Earth during the heavy bombardment phase 4.5–3.9 billion years ago (Chyba & Sagan 1992). Therefore it is important to understand how biomolecules identified in comets and meteorites are formed, what their interstellar heritage is (gas and dust) and how stable they are during and upon extraterrestrial delivery.

Summary

The investigation of the life cycle of biomolecules in space is highly relevant to astrochemistry. Understanding the implications of extraterrestrial delivery requires substantial knowledge of planetary dynamics, disk and solar nebula chemistry. Recent ground-based observations as well as data from the Spitzer telescope provide important insights into the molecular inventory and water content in planetary formation regions around young circumstellar disks (e.g. Lahuis et al. 2006, Carr & Najita 2008).

Working on a synergy of recent:

- astronomical results regarding inter/circumstellar dust and gas and cometary volatiles
- theoretical models investigating the physical and chemical evolution of protoplanetary disks
- simulation experiments monitoring the formation and evolution of organics in space
- analyses of extraterrestrial matter

will help to reconstruct the processes occurring during the formation of our solar system and provide crucial insights into the "prebiotic universe".

Key questions to address

The knowledge and expertise of surface chemists and organic chemists is crucial to reveal formation pathways and chirality investigations:

- **What are the key chemical routes towards molecular complexity in regions of star and planet formation ?**

Proposal: The building blocks of life in interstellar ice - a combined astrophysical and biochemical laboratory study – (Linnartz et al.)

- **What is the organic inventory of regions of star and planet formation particularly in the habitable zone ?**
- **What does the organic inventory of regions of star and planet formation tell us about the chemical history of solar system objects such as comets, meteorites, and interplanetary dust particles ?**
- **How can we differentiate between interstellar prebiotic chemistry and the molecular "signatures of life" ?**

- **How does the evolution of interstellar gas and dust proceed in the protoplanetary disks?**
- **What are the formation routes of meteoritic and cometary organics ?**
- **How stable are biomolecules in space environments ?**
- **How is chirality of biomolecules determined in space environment ?**
- **…….**

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