

COMPOSITIONAL TRENDS IN CHONDRITIC ORGANIC SOLIDS WITHIN AND BETWEEN

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Introduction: The organic matter in carbonaceous chondritic meteorites has the potential of adding to our understanding of the formation of the solar system. The challenge is that the organic matter constitutes, generally, a small percentage of the total mass of a given chondritic meteorite. Additionally, the physical nature of this carbon provides particular difficulties for analysis. Specifically, the vast majority of organic carbon in chondritic meteorites exists as an insoluble (possibly macromolecular) solid; consequently the most effective means of chemically analyzing this material is the use of solid state nuclear magnetic resonance NMR spectroscopy.

Experimental : In order to obtain self consistent chemical structural data on meteoritic organic matter we employ a series (seven) of independent solid state NMR experiments on organic residues obtained from demineralized meteorite samples. Details on the specific experiments and the demineralization protocol can be found in [1]. The essential information obtained includes the fraction of aromatic carbon, the fraction of aromatic carbon that is bonded to hydrogen, the fraction of aliphatic and aromatic hydrogen, and a measure of the content of nano-diamond in each sample.

Using this protocol we have discovered that in every meteorite analysed so far, the fraction of aromatic carbon linked to hydrogen is low (on the order off 30-35%). Thus, in each case the aromatic moieties are highly substituted. We have found no evidence for significant abundances of large polycyclic aromatic molecules (PAHS), i.e. larger than coronene. In each case, the aliphatic carbon is highly branched rather than linear. The chemical structural data obtained from these experiments allows us to construct self-consistent molecular structural representations providing an averaged picture of the molecular structure of each meteorites organic solids.

In addition to solid state NMR we are also applying pyrolysis GC/MS. We find that the combination of these to techniques provides a very powerful means of elucidating structural relationships as well as discontinuities among the meteorites analyzed thus far.

Discussion: Cross the groups CR2, CM2, CI1, and Tagish lake (group uncertain) there exists enormous differences in the organic carbon. This is evident in the spectra presented in Figures 1 and 2.

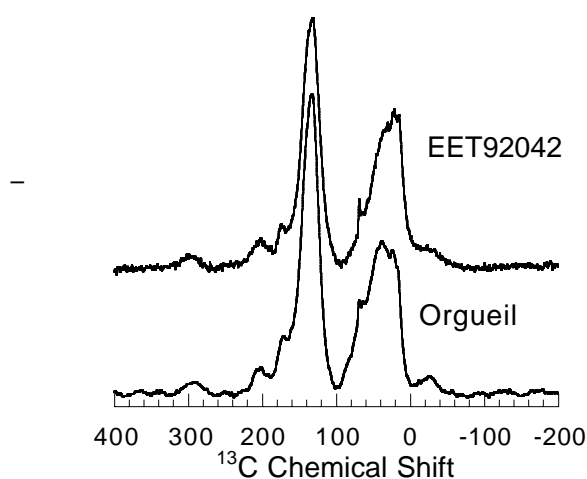


Figure 1: ¹³C solid state NMR spectra of CR2 (EET92042) and CI1 (Orgueil) organic solids. The spectra are normalized to equal total integrated intensity. The fraction of aromatic carbon in the CR2 and the CI1 are 0.44 and 0.49 respectively.

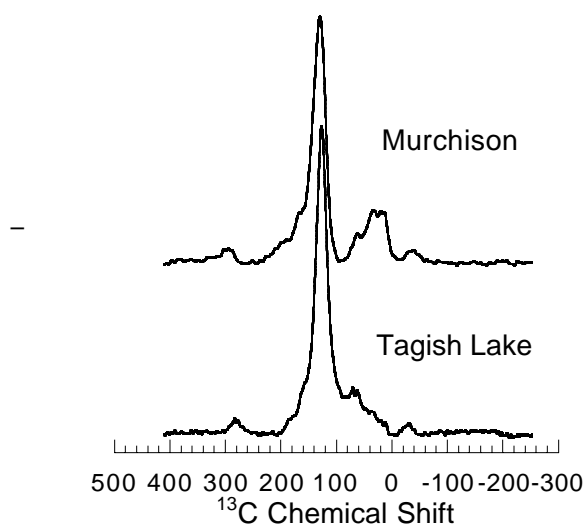


Figure 2: ¹³C solid state NMR spectra of CM2 (Murchison) and Tagish Lake organic solids. The spectra are normalized to equal total integrated intensity. The fraction of aromatic carbon in the CM2 and Tagish Lake are 0.63 and 0.79 respectively.

Each of the organic solids exhibit a broad range of organic functional groups including methyl, methylene, and methine carbon (CH_x); oxygen substitute methylene and methine (CH_xO) [there is no methoxyl carbon], carboxyl and carbonyl (ketone) (CO), and multiply substituted aromatic carbon.

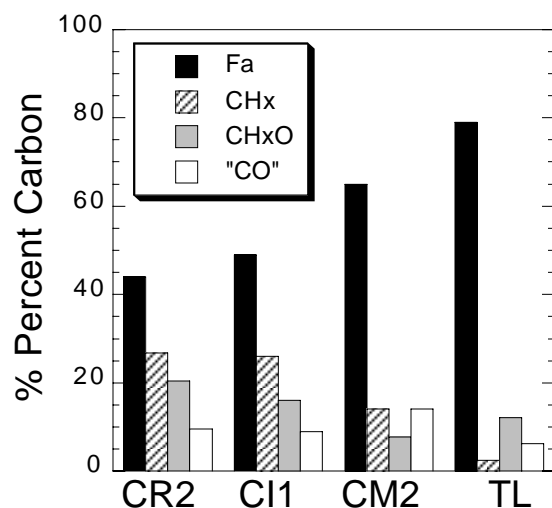


Figure 3: The molar percentage of carbon contained within aromatic (black), aliphatic (cross-hatched), oxygen substituted aliphatic (dots), and carbonyl (carboxyl + ketone) (white) functionality. Data obtained from curve fitting of ^{13}C VACP NMR data of organic residues from meteorites.

Each meteorite may be compared by the relative concentration of each class organic carbon as is shown in Figure 3. It is seen that the fraction of aromatic carbon varies widely from the CR2 (EET92042) to Tagish Lake. Whereas this increase in aromatic carbon is superficially reflected in a reduction in aliphatic carbon (CH_x) (Figure 3), detailed inspection of Figures 1 and 2 reveal that the distribution of aliphatic carbon within the class CH_x varies significantly from meteorite to meteorite. There exists a much more complex relationship between aromaticity and CH_xO or CO ; e.g. Murchison has proportionally the most CO functional groups.

The degree to which a given organic residue is more oxidized by virtue a greater proportion of oxygen-bearing functional groups is shown in Figure 4. In this figure, Tagish lake would appear to be the least oxidized. However, our analysis indicate that the aromatic fraction of each meteorite is very similar in degree of substitution and perhaps mean size, therefore, a more appropriate assessment of mean state of organic

oxidation would compare only the CH_x vs $\text{CH}_x\text{O} + \text{CO}$ as is shown in Figure 5. This essentially assumes that aromatic carbon is less susceptible to oxidation. It is now seen that the organic carbon in the meteoritic residues is progressively more oxidized moving from the CR2 through to Tagish lake.

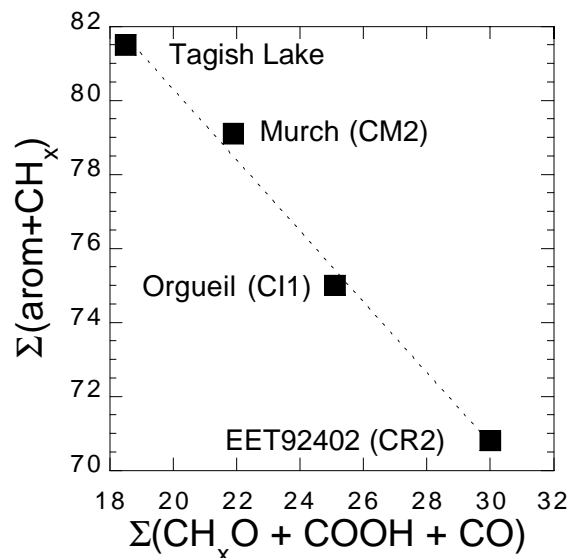


Figure 4: Chondritic organic carbon chemistry grouped as percentages of oxygen and non-oxygen containing organic carbon functionality.

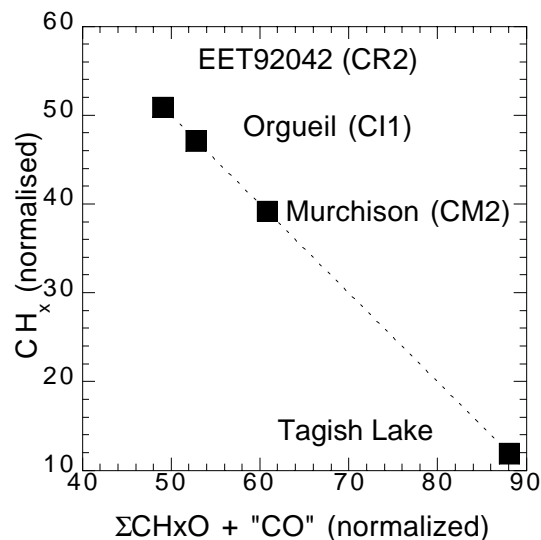


Figure 5: Chondritic organic carbon chemistry grouped as percentages of oxygen and non-oxygen containing organic carbon functionality. Normalized to exclude aromatic carbon.

Additional samples are currently being analyzed.

References: [1] Cody, G. D., C. M. O D. Alexander, F. Tera (2002) GCA, 66,1851-1865