

Analysis of Polycyclic Aromatic Hydrocarbons in Seventeen Ordinary and Carbonaceous Chondrites

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The trace organic analysis of particulates and samples that are inhomogeneous on a microscopic scale presents a major challenge to the fields of geochemistry and cosmochemistry. In order to apply the ultra-sensitive detection method of mass spectrometry, the sample must be both introduced into the gas phase and ionized. The involatility of large organic species combined with their thermal fragility and tendency for fragmentation upon ionization has, however, severely limited this approach. We have developed a microprobe two-step laser mass spectrometer (μ -L²MS) [1] presently capable of detecting sub-femtomole amounts of organic material with a spatial resolution of 40 μ m. We will present the results of the analysis of polycyclic aromatic hydrocarbons (PAHs) in seventeen meteoritic acid residues (HF-HCl) obtained from the interiors of both ordinary and carbonaceous chondrites. In each case, only 2 μ g of material is required for a complete analysis. The instrument appears to show great promise for the analysis of samples of limited size, such as interplanetary dust, and fine spatial heterogeneity, such as meteorite surfaces.

The technique combines the advantages of laser desorption, to volatilize intact neutral molecules from a solid sample, with the molecular selectivity and sensitivity of resonance-enhanced multiphoton ionization (REMPI), and the high ion transmission, unlimited mass range, and multichannel detection of a reflectron time-of-flight (TOF) mass spectrometer. In our μ -L²MS, the output of a pulsed CO₂ laser (10.6 μ m) is focussed through a microscope objective onto a 40 μ m spot of the sample from which efficient desorption of neutral species occurs. After a 20 μ s time delay, the frequency-quadrupled output of a pulsed Nd:YAG laser (266 nm) selectively ionizes any PAHs in the mixture of desorbing species by 1+1 REMPI. The ions thus formed are mass analyzed in a TOF mass spectrometer, equipped with a two-stage reflectron to improve the mass resolution to above 1000. The ability to completely resolve the individual isotopic components of a molecular species offers the potential to determine the carbon isotope ratio for that molecule.

Quantitative and qualitative trends in the PAH composition of the acid residues studied, found listed in Figure 1, have been identified for the five different chondrite classes. Although over 50 different molecular species have been detected, we have no evidence for molecules with masses significantly larger than 350 amu. This supports the suggestion [2] that the building blocks of the macromolecular material may be relatively small molecules. Notable differences are seen between the carbonaceous and ordinary chondrites, reflecting both the degree of alkylation and the extent of aromatization. For example, a comparison between the Bishunpur acid residue, Figure 2a, and the Murchison acid residue, Figure 2b, shows larger concentrations of PAHs in Bishunpur, as well as larger relative abundances of naphthalene (128 amu), methyl-phenanthrene (192 amu), and coronene (300 amu). Moreover, correlations are observed between the degree of alkylation and petrographic type within a single chondrite class. Such observations suggest that the effects of secondary processing, thermal metamorphism [1] and aqueous alteration [3,4], have played a significant role in shaping the organic content of these materials. Perhaps some of these trends might also be explained by chromatographic separation during the aqueous alteration phase of the meteorite parent bodies, as has been recently proposed [5].

References: [1] L.J. Kovalenko, C.R. Maechling, S.J. Clemett, J.-M. Philippoz, R.N. Zare, and C.M.O'D. Alexander (1992) *Analytical Chemistry* (accepted). [2] R. Hayatsu, R.G. Scott, R.E. Winans (1983) *Meteoritics* **18**, 310. [3] E.L. Shock and M.D. Schulte (1990) *Nature* **343**, 728-731. [4] M. Siskin and A.R. Katritzky (1991) *Science* **254**, 231-237. [5] M.R. Wing and J.L. Bada (1991) *GCA* **55**, 2937-2942.

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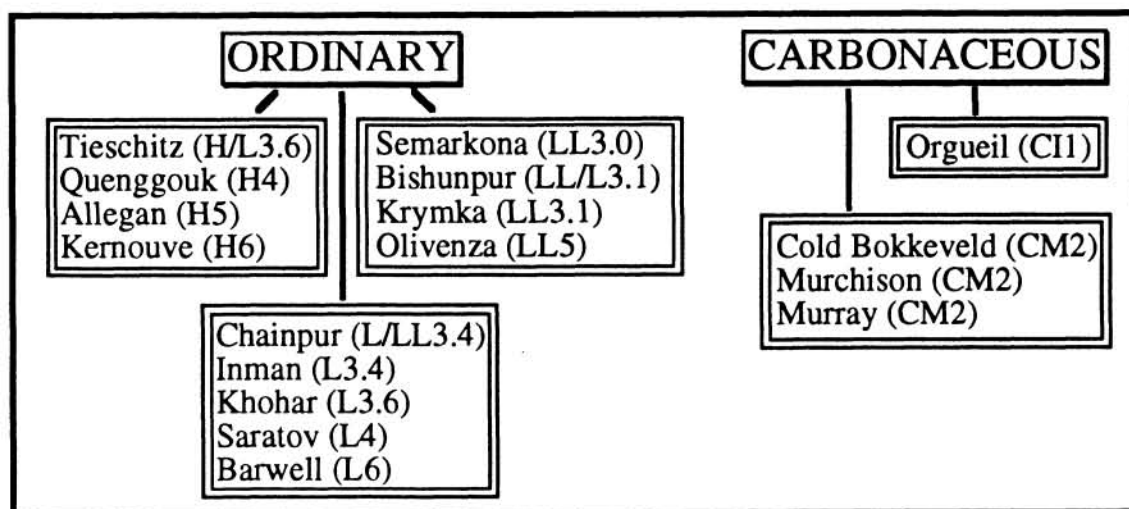


Figure 1: Schematic of the five chondrite classes analyzed

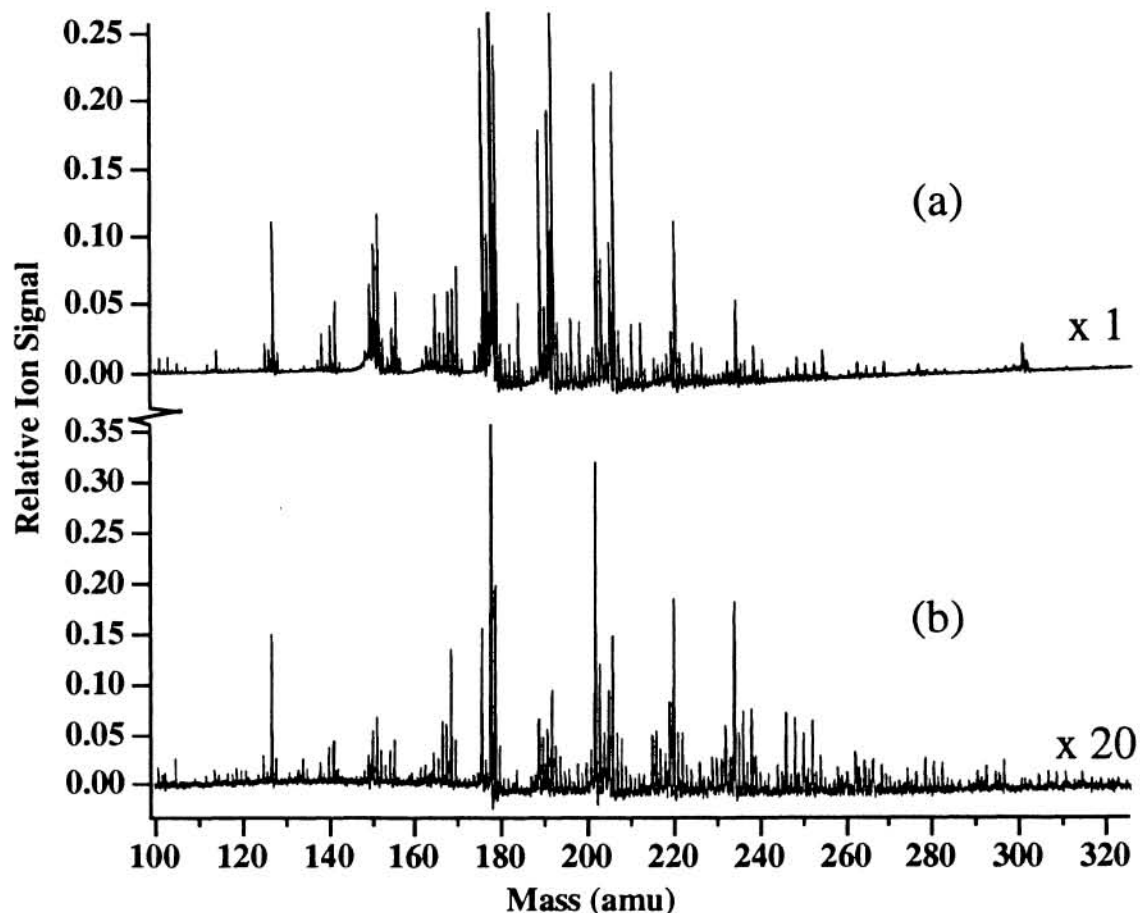


Figure 2a: Bishunpur acid residue mass spectrum. Figure 2b: Murchison acid residue mass spectrum. Both spectra represent 100-shot averages; intensity scaling of the y-axis is relative to phenanthrene (178 amu); relative intensities of the phenanthrene peak are shown by the multiplicative factor in the lower right hand corner of each spectrum.