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Macromolecular organic acids in the Murchison meteorite

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MACROMOLECULAR ORGANIC ACIDS IN THE MURCHISON METEORITE. J. S. Watson, M A. Sephton, I. Gilmour, ¹Planetary and Space Sciences Research Institute, Open University, Walton Hall, Milton Keynes, MK7 6AA, UK (j.watson@open.ac.uk).

Introduction: There have been relatively few studies of polar moieties in the macromolecular organic matter in carbonaceous chondrites (e.g. [1]). NMR data suggests that there are various oxygen-containing functionalities in Murchison [2]. This study aims to characterize the polar moieties associated with Murchison macromolecular material. Conventional pyrolysis gas chromatography mass spectrometry (Py-GC-MS) is unable to detect a large number of polar O- and N-containing compounds as these species are either not GC-amenable or defunctionalization occurs during pyrolysis.

This study has detected bound organic acids within the Murchison meteorite organic macromolecule. A HF/HCl residue from a sample of Murchison meteorite was subjected to online thermochemolysis followed by analysis by gas chromatography-mass spectrometry (GC-MS). Thermally assisted hydrolysis and methylation (thermochemolysis) using tetramethylammonium hydroxide (TMAH) results in the cleaving of ether and ester bonds and methylation of the released products. Temperatures employed (280 °C) are below those required for pyrolytic cracking of organic material. Thus thermochemolysis is an excellent method for determining oxygen bound moieties within organic macromolecules such as that in the Murchison meteorite.

Results and discussion: The distribution of compounds released by thermochemolysis are displayed in Fig 1. Benzoic acid was the most abundant compound released, other abundant compounds include methyl and dimethyl benzoic acids together with methoxy benzoic acids (Figs. 1 and 2). Short chain α,ω -dicarboxylic acids (C_{4-8}) were also released from the macromolecule similar to those observed in solvent extracts of the same meteorite [3]. Other compounds detected include fluoranone, methoxymethylsulfanylbenzenes and dimethylsulfone as well as aromatic hydrocarbons.

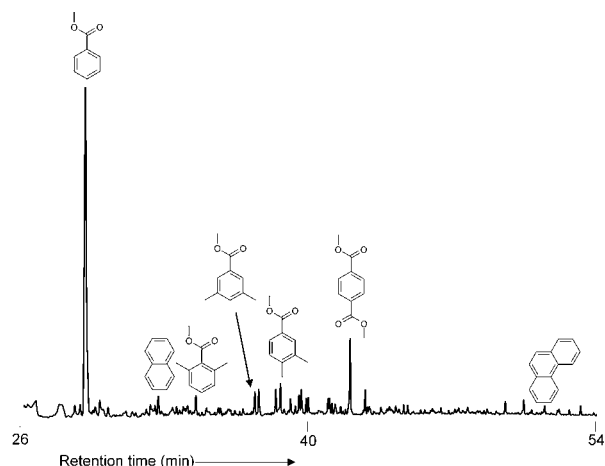


Fig 1: Total ion chromatogram of products released upon thermochemolysis of HF/HCl residue from Murchison.

The distribution of the C_1 and C_2 benzoic acids (BA) contain all possible structural isomers (except the ethyl BA). The most abundant isomers include 3,4-dimethylbenzoic acid (DMBA), 3,5-DMBA, 2,6-DMBA and phenylacetic acid. Unlike terrestrial samples extraterrestrial samples are expected to contain all possible isomers. Naphthoic acids are also present and represent the largest ring size aromatic acids detected.

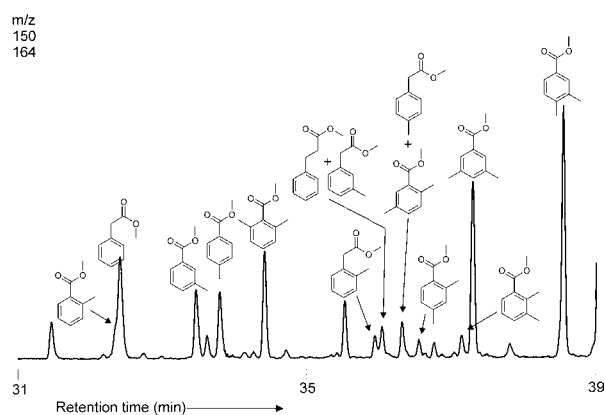


Fig 2: Partially reconstructed mass chromatogram (m/z 150 + 164) displaying the C_1 and C_2 benzoic acids (as methyl esters) released upon thermochemolysis of HF/HCl residue from Murchison.

Hydrocarbons were also detected that were not present in the thermally desorbed (280 °C, no TMAH) sample. Compounds identified include naphthalene,

methylnaphthalenes, biphenyl, methylbiphenyls, acenaphthylene, acenaphthene, phenanthrene, anthracene, fluoranthene and pyrene (see Fig 3). This discrepancy between thermally desorbed and thermochemolysis products suggest that hydrocarbons are released when the HF/HCl residue is treated with TMAH and may be occluded in the organic framework but not covalently bound. Such an interpretation is supported by the predominance of the parent molecule compared to the alkylated homologue. This is the first time that occluded hydrocarbons have been suggested to be present within the macromolecular material.

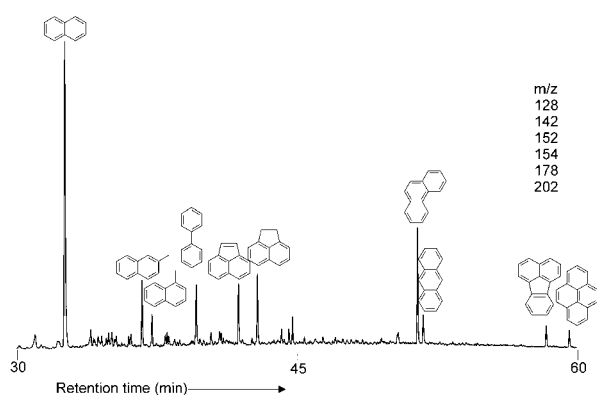


Fig 3: Partially reconstructed mass chromatogram (m/z 128 + 142 + 152 + 154 + 178 + 202) displaying aromatic hydrocarbons released upon thermochemolysis of HF/HCl residue from Murchison

There are two probable mechanisms for the formation of organic acids firstly irradiation of cosmic ices in the interstellar medium. The more likely process involves aqueous alteration upon the parent body, either oxidation of free hydrocarbons or oxidation of macromolecular material. The distribution of the acids present will reflect whether they are derived from the free fraction or the macromolecule. Bound acids will be formed from free acids being transesterified into the macromolecule. The aliphatic acids will probably be destroyed by further oxidation whereas the aromatic acids require ring cleavage for complete oxidation and are therefore more resistant to complete mineralisation.

References:

- [1] Hayatsu, R., et al., (1980) *Science*, 207, 1202-1204. [2] Cody G. D. et al.. *Geochimica et Cosmochimica Acta*, 66, 1851-1865. [3] Lawless, J. G. and Yuen, G. U., (1979). *Nature*, 282, 396-398.