Organic constitution of the CO3 chondrites and implications for asteroidal processes

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Introduction: By comparison with other carbonaceous chondrite groups, the CO3s have received little attention regarding their organic constitution. This has largely been due to the low carbon abundance of these samples (typically less than 1 wt% for falls [1]) inhibiting the use of many conventional analytical techniques, and the metamorphic history of the group [2, 3, 4], which implies any indigenous organic species will have undergone significant secondary alteration. Previous work [e.g. 5, 6, 7] has focused largely on the free organic components, which have been characterized by isotopic analysis to be of terrestrial origin. However, [8] utilized pyrolysis techniques to degrade the CO3 macromolecular material into its constituent fragments, elucidating up to 2-ring aromatic and alkyl aromatic species and O-heteroatomic structures.

The fall and rapid collection of the Moss CO3 meteorite in July 2006 presents a unique opportunity to study the indigenous organic species present in CO3 chondrites and investigate the metamorphic sequence of the CO3s defined by [3] in terms of organic species. We have performed a Py-GCxGC-TOFMS study of six CO3 chondrites, which, as demonstrated in [9], provides improved resolution for these low C samples. [10] details advantages of this experimental technique in greater detail.

Samples: The samples chosen for analysis were limited to CO3 falls (Kainsaz, Lancé , Ormans, Felix, Warrenton and Moss) in order to minimise terrestrial contamination and investigate the pre-defined metamorphic scale [3]. Two sub-samples of Moss were analysed. Crushing of whole rock meteorite samples was carried out using an agate pestle and mortar within a Class 100 clean room.

Method: 1-2 mg of each sample was subjected to thermal desorption at 300°C to remove labile organic species, then flash pyrolysis at 610°C. Exact temperatures were recorded for each step using filament resistance (<600°C) and a photodiode (>600°C). Sample heating was undertaken using a Pyrolyt 2000 (Pyrolab, Sweden) for 2s in a total flow of He at 1ml/min. The interface was held at 200°C and coupled to the injector of a Pegasus 4D (GCxGC) TOFMS (LECO Corporation) maintained at 250°C. The Pegasus 4D system comprises an Agilent 6890 Gas Chromatograph, a LECO GCxGC module and a Pegasus IV time-of-flight mass spectrometer.

The primary (first dimension) column was a 30mx250µmx0.25µm BPX5 column and was press-fit connected to the secondary GC column (1.5m×100µm×0.1µm BPX50). The secondary column was installed in its own oven inside the main GC oven. The carrier gas was helium (99.9999%) supplied at a column flow rate of 1ml/min. Samples were analysed under the following GC oven program: The primary GC oven was held at 35°C for 1 min and then raised to 300 °C at a rate of 5°C/min and held for 5 mins. The secondary oven was offset +15°C from the primary oven temperature. A liquid N cooled modulator was used with a temperature offset of +30°C above the primary oven temperature for modulation on the second column. Modulation frequency was 4s. Acquisition rate was 133 spectra/sec for a mass range of 33-400amu. Data processing was performed on LECO ChromaTOF software.

Results: Moss: Moss has been classified as a CO3.5/6 according to the classification of [4] which equates to a CO3.4/5 on the metamorphic scale defined by [3]. In a companion report [12] it is shown that Moss contains 0.25 wt% C, with some present as organic material. Figure 1A displays the total ion chromatogram (TIC) as a contour map for Moss. Each point on the map represents an organic component within the pyrolysate, with abundant components displaying larger contour profiles. Only one pyrolysate of Moss yielded any organic species, suggesting the organic components are heterogeneously distributed throughout the sample. Benzene, toluene and up to C2-alkyl benzenes are evident along with traces of biphenyl, benzonitrile and some aliphatic hydrocarbons. Naphthalene is the highest molecular weight aromatic species evident but only C1-alkyl species are identifiable.

End members: In order to investigate whether the organic species reflects the accepted metamorphic sequence [3], it is useful to consider the end members, Warrenton (CO3.6) and Kainsaz (CO3.1). Benzene and toluene dominate the Warrenton pyrolysate; however alkyl-benzenes up to C5 are also evident, which have also been reported in Murchison using the same method as this study [9]. As with Moss, naphthalene is the highest molecular weight polyaromatic species evident but only C1-alkyl species are identifiable.

In contrast, Kainsaz, the low metamorphic grade CO3.1 [3], yields comparatively few organic species when pyrolysed. Only benzene and toluene are identifi-
able in the pyrolysate. The lack of aliphatic species is consistent with the results of [8], and this study also reported 2-ring aromatic species in the Kainsaz pyrolysable fraction. [11] suggested on the basis of isotopic analyses that Kainsaz may exhibit geochemical (C, N and D) characteristics inconsistent with its metamorphic grade, which would support its apparent lack of organic species, however, reanalysis of this meteorite is required to validate these results.

**Ornans, the type specimen:** Ornans (CO3.3), the type specimen for the CO3 group, displays a TIC similar to that of Moss with benzene, toluene, up to C_{2} alkylenes, some low molecular weight aliphatic components and traces of naphthalene. No higher molecular weight species are evident.

**Felix and Lancé:** Fig 1B illustrates that Lancé (CO3.5) displays abundant organic species compared with Moss. The pyrolysate yields benzene, toluene, acetonaphthone and naphthalene and selected ion monitoring elucidates up to C_{9} n-alkyl benzenes, up to C_{4} branched aliphatics, up to C_{3} alkyl naphthalenes, biphenyl and methyl biphenyls. Significantly, Lancé also elucidates 3-ring phenanthrene and methyl phenanthrenes, and 4-ring fluoranthene and pyrene. These polyaromatic rings have not previously been reported in CO3 chondrites. Alkanes and alkenes up to C_{20} are evident, which decrease in abundance with increasing molecular weight. Felix (CO3.2) displays an almost identical pyrolysate as Lancé, with generally lower relative abundances but comparatively higher abundances of high molecular weight hydrocarbons.

**Discussion:** The pyrolysates of Ornans, Moss and possibly Kainsaz are similar in terms of abundance and distribution of organic species when compared to Felix, Lancé and to some extent, Warrenton. Previous studies have identified sub-groupings and anomalies within the CO3 group [e.g. 2, 8] that are inconsistent with a metamorphic continuum. The “Ornans paradox” has been evident in many geochemical studies of CO3 chondrites [2], particularly in reference to Ornans’ C-chemistry [e.g. 13, 14]. Ornans displays a more restricted organic inventory compared to other CO3 chondrites, even those of higher petrographic type e.g. Lancé and Warrenton. This is consistent with its low C abundance relative to the remaining group members [1, 13, 14] and supports proposals that the geochemistry of Ornans is more consistent with a higher metamorphic grade than its current classification [12]; a higher grade sample would be expected to elucidate fewer pyrolysable organic species due to increased condensation of the aromatic network.

Similarly, Moss should display an organic inventory consistent with its metamorphic grade, and approximately similar to that of Lancé (assuming all CO3s accreted a similar organic component). However, like Ornans, it displays an organic inventory consistent with a higher metamorphic grade, restricted to 2-ring aromatics with few alkylated species, and shows a similarly low C abundance for its metamorphic grade [12]. This implies that Moss and Ornans may represent a sub-group within the CO3 chondrites that have undergone unique parent body alteration processes.


Figure 1. Total Ion Chromatograms (TIC) of A. Moss and B. Lancé displayed as contour maps. Moss clearly displays a lower diversity of organic species than Lancé. Key: B = benzene, N = naphthalene, MN = methylphenanthrene isomers, C2-C4-N = C_{2}-C_{4} alkylphenanthrenes, P = phenanthrene, A = anthracene, MP = methylphenanthrene isomers.