

Enrichment of deuterium in insoluble organic matter from primitive meteorites: A solar system origin?

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Abstract

Because of a systematic enrichment in deuterium, the insoluble organic matter (IOM) of the carbonaceous chondrites is considered to have formed in the interstellar medium. However, the D/H ratios in IOM remain much lower than those measured in the organic molecules commonly observed in the dense interstellar medium. In this study, the D/H ratio of different aromatic and aliphatic molecular fragments of IOM from the Orgueil meteorite was measured by GC-irMS (gas chromatography–isotopic ratio mass spectrometry). No correlation was observed between the D/H ratios and structural parameters characterizing the IOM, such as the H/C ratio. However, the δD of the benzylic, aliphatic and aromatic hydrogen into the IOM can be determined to be 1250‰, +550‰ and +150‰, respectively, relative to SMOW. This indicates that D-enrichment in IOM is correlated with the C–H bond dissociation energy. Such a correlation rules out IOM formation from observed interstellar molecules and suggests instead that the different components of IOM have acquired their D/H ratios by an exchange with a deuterium-rich reservoir after its synthesis. The same process can be invoked to account for the D/H composition of meteoritic water. Findings point to a common process for deuterium enrichment in the solar system.

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1. Introduction

The origin of water and organic molecules in the early solar system has been under intense scrutiny since it was realized that these compounds were systematically enriched in deuterium relative to molecular hydrogen in the protosolar nebula (referred to as PSN).

$(D/H)_{\text{PSN}} = 25 \times 10^{-6}$ i.e. $\delta D = -840\%$ [1]. All δD numbers in this contribution are relative to SMOW. For example, in the Orgueil meteorite [2], water has a D/H ratio of 160×10^{-6} or $\delta D = +40\%$ and some components of insoluble organic matter (IOM) are characterized by D/H ratio of 350×10^{-6} or $\delta D = +1250\%$. With regard to water, it has been proposed that the D/H ratio decreased with time and temperature in the PSN [3], implying that water was initially enriched in deuterium by interstellar reactions before its incorporation into the

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PSN (theoretical calculations show that the D/H ratio of interstellar H_2O can reach 10^{-3} at 10 K; [4]) Because of the lack of experimentally measured isotopic exchange rates, similar modeling was not performed for IOM.

An interstellar origin was considered for IOM based on its systematic enrichment in deuterium [5]. However, the D/H ratio in IOM remains much lower than those measured in the organic molecules commonly observed in the dense interstellar medium (ISM; $D/H \approx 10^{-2}$) [6]. Two qualitative interpretations have been proposed to account for the isotopic difference between the chondritic IOM and the organic molecules in the ISM. First, because stepwise pyrolysis of the IOM reveals a large isotopic heterogeneity [2,7], the bulk D/H ratio represents a mixture of various components enriched in deuterium to different degrees [5,7,8]. It is possible that the most deuterium-rich of these components have D/H ratios as high as those observed in the ISM or in some IDPs [9], but that these components sufficiently diluted in IOM to prevent their direct detection. Second, it has been proposed that the IOM is indeed an interstellar product (as also proposed for some soluble organic compounds [10,11]), but that the IOM formed at temperatures much higher (120 K) than canonical ISM temperatures (10–20 K) [6]. Indeed, to first order (and as observed in Hot Cores [12,13]); the enrichment in deuterium is expected to decrease with increasing formation temperature of IOM. Here we show, however, that neither of these interpretations can account for the measured deuterium distribution in the organic moieties of the IOM. Conversely, it seems that the enrichment in deuterium of the IOM (and water) is a secondary process and consequently that these molecules were primary formed in the PSN.

A large body of data on the molecular structure of the IOM in meteorites have been reported recently in the literature [14–19]. The IOM consists of small aromatic units highly substituted and cross-linked by short aliphatic linkages (less than seven carbons), which can link more than two aromatic units. Overall, such chemical structure indicates (1) that signatures of condensation from a gas phase are preserved, (2) the IOM has suffered very limited thermal alteration on the parent body [15] and (3) there is a clear difference between the size distribution of the aromatic moieties constituting the IOM and those observed in the ISM [14].

In the present study, we have determined the D/H ratio of aliphatic and aromatic moieties in Orgueil IOM. Although aromatic moieties in chondritic IOM are known to be easily released by pyrolysis and have been widely studied [19–21], molecular information on the aliphatic linkages was only recently provided with the

development of the ruthenium tetroxide oxidation technique [18]. This chemical degradation results in oxidation of the aromatic units and release of the aliphatic chains, mainly as dicarboxylic acids (Fig. 1) [22,23]. Compound specific D/H isotope measurements can then be performed on both the pyrolysis products and on the acids after derivatisation into their trimethylsilyl (TMS) esters.

2. Methods

2.1. Sample preparation and degradation of the IOM

The IOM was isolated from the Orgueil meteorite by the standard HF/HCl treatment, which was preceded by and followed by thorough extractions to remove soluble compounds and any terrestrial contamination [17,18]. A sample of Orgueil carbonaceous chondrite (CI) was ground in an agate mortar. The powder was then submitted to several extractions: water (stirring for 24 h under reflux), acetone (stirring two times for 2 h at room temperature) and a $CH_2Cl_2/MeOH$ mixture (volume ratio 2/1; stirring overnight at room temperature). After each extraction, the solvent extract was removed by centrifugation. The remaining sample was then stirred into HCl (6 N) for 24 h at room temperature and after neutralization, the insoluble residue was added to a mixture of HF/HCl (volume ratio 2/1) for 24 h at room temperature under nitrogen flow. Finally, the sample was submitted to HCl (6 N) for 4 h at 60 °C and neutralized. The remaining sample, which mainly consisted of organic matter, was submitted again to acetone and $CH_2Cl_2/MeOH$ extractions.

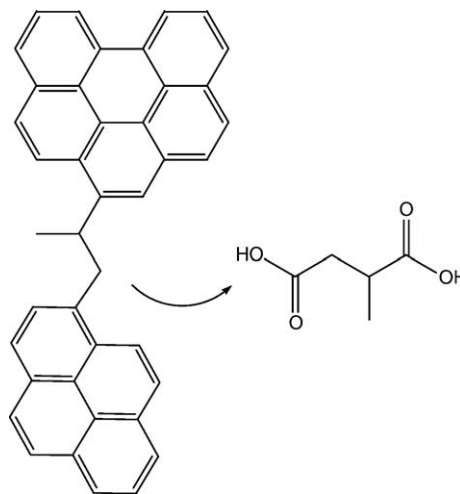


Fig. 1. Schematic pathway of ruthenium oxidation of two polyaromatic units linked by an aliphatic chain (aromatic moieties are not representative of the chemical structure of the chondritic IOM).

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