COUPLED H-ISOTOPIC FRACTIONATION AND STRUCTURAL EVOLUTION IN EXPERIMENTALLY IRRADIATED ORGANIC MATTER.

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Introduction: Organic matter found in carbonaceous chondrites is highly enriched in deuterium, relative to H_2 in the protosolar nebula (PSN) [1]. In the scenario where the insoluble organic matter (IOM) is a PSN product [2], ion-molecule or gas-grain reactions could lead to a D-enrichment of organics [3]. The ionizing irradiations in the protosolar nebula potentially enhanced such mechanisms [4]. Recent experimental irradiations of natural and synthetic analogues of the IOM under high energy (300 keV) have shown that electron irradiation could act as a driving mechanism for D-enrichment [5,6]. In order to quantify the isotope effects induced by ionizing particles on pre-accretional organic matter, we conducted irradiation experiments on well constrained analogues of this IOM.

Methods: All the samples consist of thin polymer films, from 1 to 10 µm thick. The polymers of interest bear three different C-H groups (benzylic, aliphatic and aromatic) in various proportions. For this purpose, Polyethylene, (PE), Polyethyleneterephtalate (PET) and Polystyrene (PS) have been used. The ionizing excitation was obtained by electron irradiation with a scanning electron microscope (SEM) at moderate energy (30 keV) at room temperature.

Results: The chemical, structural and isotopic properties of all samples exhibit the same evolution as the electron dose increases. The kinetics of the transformations can be described by a first-order rate formalism. The rate-constant is similar for all the properties of a given polymer and marginally depends on the initial nature of the polymer. From an isotopic point of view, a large D-enrichment is observed. After a continuous increase, the D/H levels up. The isotopic signature at this plateau depends on the polymer, from 320±85‰ up to 590±85‰.

Discussion: From these isotopic maxima, we derived the D-enrichment of each C-H group (benzylic, aliphatic and aromatic). Assuming a local equilibrium, these values can in turn lead to the intra-molecule hydrogen isotopes fractionation factors associated to the irradiation process. These fractionation factors are in excellent agreement with those measured in the CI chondrite Orgueil [2], and those experimentally measured after an interaction between a D-rich ionized gas and an organic solid [7]. These correlation may indicate that irradiation promotes such interaction between solids and ionized gas at the molecular scale, shaping the isotopic signature of the Orgueil IOM. Finally, our experimental conditions were extrapolated to the irradiation conditions that prevailed in the early solar system. We show that such a systematic D-enrichment of the organic residue is produced in only a few hundred of years.

References: [1] Robert F. and Epstein S. 1982. *GCA* 46:81–95. [2] Remusat L. et al. 2006. *EPSL* 243:15–25. [3] Sandford S. A. et al. 2001. *MAPS* 36:1117–1133. [4] Glasshold A. et al. 2000. *Protostars and Planets* IV:429–455. [5] De Gregorio B. et al. 2010. *GCA* 74:4454–4470. [6] Le Guillou C. et al. 2013. *Icarus* 226:101–110. [7] Robert F. et al. 2011. *GCA* 75:7522–7532.