

## High-mass resolution molecular imaging of organic compounds on the Murchison meteorite.

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**Introduction:** Primitive meteorites contain a wide variety of organic molecules. The CHO, CHNO, CHNOS and CHN species have been identified using high-resolution mass spectroscopy for the solvent extracts of carbonaceous chondrites [1-3]. These organic compounds could be synthesized and evolved via interaction of fluid with minerals [3-5]. Therefore, spatial distribution of meteoritic organic compounds is important to reveal the organic-mineral association on the parent bodies. Recently, in-situ analysis of polar organic compounds on the meteorite surface was achieved by desorption electrospray ionization/high-resolution mass spectrometry (DESI/HRMS) [7]. In this study, we further performed the DESI-HRMS imaging on the Murchison meteorite to understand the relationship between spatial distribution of polar organic compounds and minerals.

**Experimental:** The fragment of the Murchison meteorite (CM2) (~ a few mm) obtained by chipping was embedded in indium (In). The DESI-HRMS imaging was performed using a DESI ion source (Omni Spray 2-D, Presolia) equipped with an Orbitrap mass spectrometer (Q-Exactive Plus, Thermo Scientific) with mass resolution of ~140,000 at  $m/z$  200. Methanol (LC-MS grade) was sprayed on the sample surface with a 1 or 2  $\mu\text{l}/\text{min}$  flow rate. The spatial resolution was approximately 100  $\mu\text{m}$ . FESEM-EDS analysis was also carried out without coating and polishing after the DESI-HRMS imaging. The surface of the Murchison meteorite fragment embedded in indium was also extracted with MeOH, and the resulting solution was applied for HPLC-HRMS to obtain chemical structural information on the organic compounds.

**Results and Discussion:** Alkylated homologues of  $\text{C}_n\text{H}_{2n-1}\text{N}_2^+$  ( $n = 7-9$ ) and  $\text{C}_n\text{H}_{2n}\text{NO}^+$  ( $n = 9-14$ ) were detected on the sample surface by the DESI-HRMS imaging. HPLC/HRMS analysis of MeOH extracts from the sample surface after the DESI/HRMS imaging indicated that the  $\text{C}_n\text{H}_{2n-1}\text{N}_2^+$  homologues correspond to alkylimidazole, and that a few structural isomers of the  $\text{C}_n\text{H}_{2n}\text{NO}^+$  homologues were present in the sample. The alkylimidazoles and  $\text{C}_n\text{H}_{2n}\text{NO}^+$  homologues displayed different spatial distributions in the Murchison fragment. Moreover, the distribution pattern of compounds also different among the same homologues. These results may be due to the separation of isomers by similar chromatographic effects, or different synthetic pathways. The alkylimidazoles and  $\text{C}_n\text{H}_{2n}\text{NO}^+$  homologues are mainly distributed in the matrix region of the Murchison by mineralogical observations, which is consistent with previous reports. Altered minerals (e.g., Fe-oxide, Fe-sulfide, and carbonates) occurred in this region. However, no clear relationship was found between these minerals and the organic compounds detected by DESI/HRMS imaging. Although this result might be due to scale differences between the spatial resolution of DESI-HRMS imaging and the grain size in the matrix of the Murchison, our results would indicate that alkylimidazoles and the  $\text{C}_n\text{H}_{2n}\text{NO}^+$  homologues in the Murchison fragment were mainly synthesized by different processes from hydrothermal alteration on the parent body.

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