超高分解能質量分析による宇宙・地球有機化合物の解析

Organic compounds in cosmogeochemical samples revealed by ultrahigh-resolution mass spectrometry

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Various types of organic matter are distributed widely in terrestrial and extraterrestrial environments. The molecular and isotopic occurrence in various natural samples will provide important clues for the chemical evolution and origins of life in the universe as well as ecological and environmental changes associated with biological activities in a natural history. However, chemical structures of organic compounds consisting of C, H, N and/or O are significantly diverse and complex (e.g. structural and optical isomers), and their concentrations are generally very low with significant mixtures in natural samples. Therefore, high mass resolution (m/Dm > 100,000) and high sensitivity are preferred with high chromatographic separation under a clean analytical condition.

Recent ultrahigh-resolution mass spectral (UHRMS) analysis using Fourier transform-ion cyclotron resonance/mass spectrometry (FT-ICR/MS) detected tens of thousands of different mass peaks consisting of C, H, N, O, and/or S in the solvent extracts of the Murchison meteorite [1]. Considering the structural and optical isomers, the current organic contents identified in the meteorite correspond to only approximately 1% of the total compounds present. We also found > 600 alkylated homologous compounds consisting of C\textsubscript{n}H\textsubscript{2n}N and C\textsubscript{n}H\textsubscript{2n-1}N\textsubscript{2} in elemental composition by UHRMS using HPLC coupled with an Orbitrap MS [2]. An advantage of Orbitrap MS compared to FT-ICR/MS is that the UHRMS analysis can be achieved in a clean room.

Trace organic compounds have been generally analyzed using the solvent extracts of powdered samples. In-situ analysis of organic compounds has been limited for volatile compounds such as PAHs. In order to clarify the distribution for various organic compounds, the molecular imaging on the surface of carbonaceous meteorites has been performed by UHRMS using desorption electrospray ionization (DESI) coupled with an Orbitrap MS in a clean room. Alkylated N-containing cyclic compounds including alkylimidazoles (C\textsubscript{n}H\textsubscript{2n}N\textsuperscript{+}) and alklypyridines (C\textsubscript{n}H\textsubscript{2n-4}N\textsuperscript{+}) were identified on the surfaces of the carbonaceous meteorites [3]. The distribution of alkylimidazoles and alklypyridines appeared different on the meteorite surface, suggesting different their source regions or asteroidal chromatographic effect on the parent body.

High precision measurement of compound- and site-specific isotope composition has been achieved by UHRMS recently using a Orbitrap MS [4]. The UHRMS isotope analysis does not require a combustion or pyrolysis interface to convert organic compounds into CO\textsubscript{2}, N\textsubscript{2} or H\textsubscript{2} gases, and has a great benefit for the simultaneous multi-isotope measurement as well as for the sample preparation.

The technical development of molecular and isotopic analysis by UHRMS will allow for the improved identification of organic compounds and will advance comprehensive studies of the formation pathways and origins of organic compounds in terrestrial and extraterrestrial environments.


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