様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

令和　５ 年　 ６ 月 １７ 日現在

機関番号：１２６０８
研究種目：挑戦的研究（萌芽）
研究期間：2021～2022
課題番号：２１Ｋ１８６４６

研究課題名（和文）Isotopologues as universal tracers of abiotic processes

研究課題名（英文）Isotopologues as universal tracers of abiotic processes

研究代表者
ジルベルト　アレキシー（Gilbert, Alexis）

東京工業大学・理学院・准教授

研究者番号：２０７２６９５５

交付決定額（研究期間全体）：（直接経費）　　4,600,000円

研究成果の概要（和文）：この方法を校正し、その精度を評価することができました (Taguchi et al. 2021)。
私たちは、生物的プロセスと非生物的プロセスの 13C-13C 特徴の体系的な調査を行うことができました。
データには、シミュレーション実験のほか、国際的な同僚から提供され、さまざまな場所からの天然ガスサンプルが含まれています。
そのデータは、(i) 13C13C シグネチャーが非生物的プロセスと生物的プロセスの強力なトレーサーとなり得ること、(ii) 生物的分子と非生物的分子の形成メカニズムや条件を区別するのに役立つ可能性があることを示しています (Taguchi et al. 2022)。

研究成果の学術的意義や社会的意義
There is currently no clear tracer for biological processes. Our method shows a clear distinction between molecules produced by biological and non-bioloigcal processes, which will be of tremendous help in the future for life exploration in the solar system and beyond.

研究成果の概要（英文）：We were able to calibrate the method and evaluate its accuracy (Taguchi et al. 2021). We were able to do a systematic survey of 13C-13C signatures of biotic and abiotic processes. The data include simulation experiments (hydrocarbons cracking, methane polymerization with different energy sources) as well as natural gas samples from different locations provided by international colleagues and sampled by ourselves.
The data shows that (i) 13C13C signature can be a robust tracer of abiotic vs biotic processes (ii) it may help distinguishing mechanisms and/conditions of formation biotic and abiotic molecules. The data has been presented in various conferences and in an article in Nature Communications (Taguchi et al. 2022).

研究分野：Biogeochemistry

キーワード：Biotic　Abiotic　Isotope　Isotopologue　Hydrocarbons

１．研究開始当初の背景

The research project's motivation is built on the lack of clear biological indicators for organic molecules. Stable isotopes have long been used to distinguish biologically-made molecules from non-biological ones, but the current methods show overlap and there is a need to develop new methods for that purpose.
The method developed at Tokyo Tech (Taguchi et al. 2020; Figure 1) allows the measurement of the doubly-substituted 13C-13C isotopologue of C2 molecules such as ethane and ethanol. The method has potential to trace the process by which a molecule is formed, since the 13C-13C isotopologue may record the conditions of C-C bond formation.

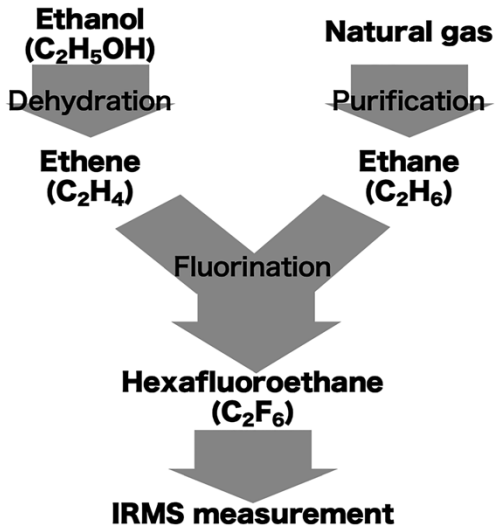

Figure 1. Overall strategy for the measurement of 13C-13C isotopologues of ethane, ethene and ethanol. Fluorination is necessary in order to avoid any interferences due to the presence of ^{2}H atoms in the original molecule. After fluorination, only the 13C atoms contribute to the higher masses of the molecules in the mass spectrometer (from Taguchi et al. 2020).

２．研究の目的

The purpose of the project is to determine whether the doubly-substituted isotopologue 13C-13C of ethane (C2H6) can be used as a tracer for abiotic process, namely, if the 13C13C relative abundance can be used to distinguish molecules made by biological processes from those made by purely geochemical processes not involving biology (abiotic).

３．研究の方法

We used the recently developed method and measured ethane samples from different simulation processes: cracking of biological organic matter (petroleum, natural gas, lignin) and polymerization of methane to higher hydrocarbons using different energy sources (UV, spark discharge, gamma irradiation).

We then collected natural gas samples from different origins, including thermogenic (cracking of biological organic matter) and putatively abiotic samples. The samples were collected by ourselves and also with the help of national and international collaborators.

４．研究成果

The data shows that (i) 13C13C signature can be a robust tracer of abiotic vs biotic processes (ii) it may help distinguishing mechanisms and/conditions of formation biotic and abiotic molecules. Work is still on progress to determine the factors leading to different signatures, but it appears that the formation of a C-C bond in abiotic (polymerization) processes leads to low 13C-13C abundance in the product, the C-C formation being slower to be produced when 2 13C-atoms are involved.

On the whole, our method shows a clear distinction between molecules produced by biological and non-biological processes, which will be of tremendous help in the future for life exploration in the solar system and beyond. The data has been presented in various conferences and in an article in Nature Communications (Taguchi et al. 2022; Figure 2).

The data has also been covered as a press release (See Fig. 2).

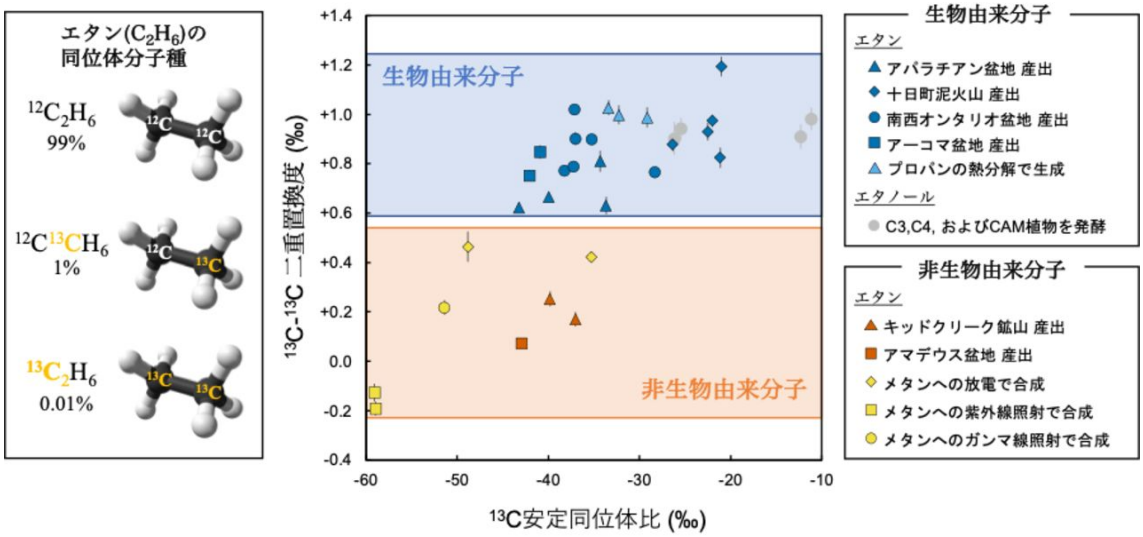

Figure 2. Result of 13C-13C double substitution degree of ethane in natural gas. The vertical axis indicates the degree of 13C-13C double substitution of ethane, and the horizontal axis indicates the carbon isotope ratio of ethane. Ethane abiotically synthesized in the laboratory and ethane in natural gas, which is said to be of non-biogenic origin, tend to have a clearly lower abundance of 13C2H6 than ethanol of biological origin and ethane in natural gas. The 13C-13C double substitution degree is defined as the difference between the 13C-13C double substitution degree probabilistically predicted from the 13C/12C ratio (the square of the 13C/12C ratio) and the actual 13C-13C double substitution degree. be. The measured values shown on the vertical axis are expressed as differences from the standard gas.

５．主な発表論文等

〔雑誌論文〕　計1件（うち査読付論文　1件／うち国際共著　1件／うちオープンアクセス　1件）

１．著者名 Taguchi, Gilbert, Ueno	４．巻 13
２．論文標題 Low 13C-13C abundances in abiotic ethane	５．発行年 2022年
３．雑誌名 Nature Communications	６．最初と最後の頁 5790
掲載論文のDOI（デジタルオブジェクト識別子） 10.1038/s41467-022-33538-9	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

〔学会発表〕　計4件（うち招待講演　2件／うち国際学会　3件）

１．発表者名 Yuichiro Ueno, Koudai Taguchi, Toshiki Katsuta, Mayuko Nakagawa, Naohiro Yoshida, Alexis Gilbert
２．発表標題 Application of fluorination method to isotopologue biogeochemistry
３．学会等名 Goldschmidt 2021（国際学会）
４．発表年 2021年

１．発表者名 Koudai Taguchi, Alexis Gilbert, Yuichiro Ueno
２．発表標題 Low 13C-13C clumping on abiotic hydrocarbons
３．学会等名 Goldschmidt 2021（国際学会）
４．発表年 2021年

１．発表者名 Alexis Gilbert
２．発表標題 Deciphering Hydrocarbons’ Sources and Sinks through Isotopologue measurements
３．学会等名 GRC 2022（招待講演）（国際学会）
４．発表年 2022年

１．発表者名 Alexis Gilbert
２．発表標題 Isotopologues of organic molecules: method developments and applications
３．学会等名 ISI/Isotopes 2022（招待講演）
４．発表年 2022年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

―

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関