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Study on relationship between CH_4 flux and wetting at taiga-tundra boundary in northeastern Siberia using stable isotope ratios of CH_4

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Methane is one of the important greenhouse gases. Methane emission from natural wetlands is the largest individual source of the atmospheric CH_4. The size of this source depends on meteorological conditions such as precipitation, and a significant driver of the interannual variations in the global CH_4 source. Higher water level in wetlands and terrestrial ecosystems generally leads to higher CH_4 emission in spatial variation. On the contrary, the temporal variation in the CH_4 emission tends to be more complicated. Flooding can increase CH_4 emission after a time lag. Higher water level can also occur with lower CH_4 emission. To understand the relationship between wetting and CH_4 emission, it is necessary to assess the underlying processes, CH_4 production, oxidation and transport, which are reflected in stable isotope ratios of CH_4.

Taiga-tundra boundary ecosystem on the Indigirka River lowlands in northeastern Siberia has a gradient in soil wetness conditions, corresponding to the microtopography. Higher micro-relief in the ground level is drier and covered by green mosses, larch trees and shrubs (termed “tree mound”). Lower micro-relief is wetter and covered by wetland vegetation, such as sphagnum mosses and sedges (“wet area”). Thus, this

ecosystem was assumed to have micro-reliefs in the gradient with large interannual variations in soil wetness conditions. Actually, the ecosystem experienced a wetting event with extreme precipitation in 2011.

This study observed interannual variations in CH_4 flux and dissolved CH_4 concentration during the summer from 2009 to 2013 at the taiga-tundra boundary in the vicinity of Chokurdakh (70° 37′ N, 147° 55′ E) to investigate the relationship between CH_4 flux and wetting. The interannual flux variations were observed with portable manual chambers. As a complement, an automatic chamber system was modified to make continuous observations of CH_4 flux. In order to assess variations in CH_4 production and oxidation processes after the wetting event, $\delta^{13}C$ and δD of dissolved CH_4 were analyzed from 2011 to 2013. In addition, soil incubation experiments were conducted under anaerobic and aerobic conditions to investigate the behaviors of $\delta^{13}C$ and δD of CH_4 with regards to CH_4 production and oxidation processes in this ecosystem.

Methane flux showed large interannual variations in wet areas (36–140 mg CH_4 m^{-2} day^{-1} emitted). Increased CH_4 emission was recorded in the summer of 2011 when a wetting event with extreme precipitation occurred. Methane emission remained relatively large in 2012, and increased further in 2013, although water level declined from 2011 to 2013. Thaw depth became deeper from 2011 to 2013, which may partly explain the increase in CH_4 emission. Moreover, dissolved CH_4 concentration showed a sharp increase of one order of magnitude from 2011 to 2012 and increased further from 2012 to 2013.

Variations in individual $\delta^{13}C$ and δD values of dissolved CH_4 (10 cm depth) became smaller from 2011 to 2012 and 2013. These δ valuese clustered around the higher $\delta^{13}C$ and lower δD values, which were seen in the anaerobic incubation experiment along with high CH_4 production rates, suggesting that CH_4

production was enhanced in 2012 and 2013. In addition, the lower δD values in 2012 and 2013 suggests that CH_4 oxidation became less significant after the wetting event, based on the results of aerobic incubation experiment. These enhanced CH_4 production and less significant CH_4 oxidation, which could led to the buildup of dissolved CH_4 and the increase in CH_4 emission, may have been caused by continued soil reduction across multiple years following the wetting event. Additionally, the clustered and higher $\delta^{13}C$ values of dissolved CH_4 in 2012 and 2013 suggests that delayed activation of acetoclastic methanogenesis following the soil reduction could also have contributed to the enhancement of CH_4 production.

This study shows a multi-year effect of wetting on CH_4 emission, which suggests that duration of water saturation in the active layer can be important for predicting CH_4 emission following a wetting event in permafrost ecosystem.

Keywords: methane emission, interannual variations, carbon isotopes, hydrogen isotopes, permafrost ecosystem, Arctic

博士論文要旨

北海道大学 大学院環境科学院 地球圏科学専攻（論文博士）

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北東シベリアタイガ - ツンドラ境界域における湿潤化前後のCH_4フラックスとCH_4安定同位体比の利用

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CH_4は重要な温室効果気体の一つである．大気CH_4の代表的な放出源として自然湿地生態系が挙げられる．当放出源の大きさは，気温・降水量等の気象条件に依存して変動することから，全球CH_4放出量の年々変動を駆動する主な因子であるとされている．湿地やその他陸域生態系においては，一般に水位が高い場所で大きなCH_4放出フラックスが観測される．ところが，時間変動はより複雑であり，湿潤化の後タイムラグを経てCH_4フラックスの上昇が起きた例や，水位とフラックスが逆相関した例が報告されている．このように，湿潤化とCH_4放出フラックスの関係は不明瞭であり，その理解のためにはCH_4生成過程・酸化過程・輸送過程からなるメカニズムを調べる必要がある．これら3つの過程によってCH_4の炭素・水素安定同位体比が変動するため，逆に同位体比を利用して3過程の変動に関する情報を得ることができる．

北東シベリア・インディギルカ川下流域に広がるタイガ - ツンドラ境界域では，陸面の微地形に応じて乾湿の勾配が見られる．比高の高い地点は乾燥しており，コケやカラマツ，低木に覆われている．一方，比高の低い地点はより湿潤であり，ミズゴケやワタスゲ・スゲをはじめとする湿地植生に覆われている（wet area）．このような勾配のなかには，乾湿時間変動の大きい地点があると予想される．実際，当生態系では2011年に著しい降雨に伴う湿潤化が見られた．

湿潤化とCH_4放出フラックスの関係を調べるため，本研究はチョクルダ（70° 37′ N, 147° 55′ E）周

辺のタイガ - ツンドラ境界域において，2009～2013年の夏季にCH_4フラックスと溶存CH_4濃度を観測した．CH_4フラックスの年々変動は，簡易型のチャンバー法により観測した．また補足的に，土壌呼吸用に製作された自動開閉チャンバーシステムを改変し，CH_4フラックスの連続観測を行った．さらに，湿潤化後のCH_4生成およびCH_4酸化過程の変動を調べるため，溶存CH_4の$\delta^{13}C$およびδDを分析した．併せて，土壌培養実験を好気および嫌気条件で行い，当生態系のCH_4生成・酸化過程におけるCH_4の$\delta^{13}C$およびδDの挙動を調べた．

得られたCH_4フラックスは，wet areaにおいて大きな年々変動を示した（36–140 mg CH_4 m^{-2} day^{-1} emitted）．著しい降水に伴う湿潤化が起こった2011年夏に，CH_4フラックスは増大した．さらに，その後2011年から2013年にかけてwet areaの水位が低下したにも関わらず，CH_4フラックスは2012年に比較的大きな値を維持し，2013年には更なる上昇を示した．2011–2013年には融解深の増加が見られ，CH_4フラックス上昇の一因となった可能性がある．そのうえ，溶存CH_4濃度値は2011–2012年に1桁分の著しい上昇を示し，2012–2013年にもさらに上昇した．

溶存CH_4の$\delta^{13}C$およびδD（10 cm深）は，2011年に大きく変動し，2012–2013年には変動が小さくなった．$\delta^{13}C$は高い値，δDは低い値に集まり，これらの値はCH_4生成培養実験で生成速度が大きい場合に見られたδ値におよそ一致した．したがって，2012および2013年に，CH_4生成が活発化したことが示唆された．また，2012–2013年に見られた低いδD値と，CH_4酸化培養実験の結果から，湿潤化の翌年にCH_4酸化の寄与が小さくなったことが示唆された．このように，CH_4生成が促進され，CH_4酸化の寄与が小さくなったことで，溶存CH_4濃度が上昇し，CH_4フラックスが増大したことが考えられる．これらは，湿潤化のあと土壌の還元が複数年にわたって進行することで，引き起こされた可能性がある．また，2012–2013年に溶存CH_4の$\delta^{13}C$が高い値に集まったことから，土壌還元に続いて酢酸分解経路によるCH_4生成が遅れて活性化したことが示唆され，このプロセスもCH_4生成の促進に寄与した可能性がある．

本研究は，湿潤化がCH_4放出フラックスに対して2年後まで持続的影響を与えたことを示すものであり，永久凍土生態系において湿潤化後のCH_4フラックスを予測するにあたり，活動層に水の滞留する時間が重要であることを示唆している．

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