Temporal Variations of the Mole Fraction, Carbon, and Hydrogen Isotope Ratios of Atmospheric Methane in the Hudson Bay Lowlands, Canada

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Abstract We have conducted simultaneous measurements of the mole fraction and carbon and hydrogen isotope ratios \(\delta^{13}C\) and \(\delta D\) of atmospheric methane (CH\(_4\)) at Churchill (58°44'N, 93°49'W) in the northern part of the Hudson Bay Lowlands (HBL), Canada, since 2007. Compared with the measurements at an Arctic baseline monitoring station, Ny-Ålesund, Svalbard (78°55'N, 11°56'E), CH\(_4\) mole fraction is generally higher and \(\delta^{13}C\) and \(\delta D\) are lower at Churchill due to regional biogenic CH\(_4\) emissions. Clear seasonal cycles in the CH\(_4\) mole fraction, \(\delta^{13}C\), and \(\delta D\) are observable at Churchill, and their seasonal phases in summer are earlier by approximately 2 weeks than those at Ny-Ålesund. Using the one-box model analysis, the phase difference is ascribed to the different seasonal influence of CH\(_4\) emissions from boreal wetlands on the two sites. Short-term CH\(_4\) variations are also observed at Churchill throughout the year. The analysis of the observed isotopic signatures of atmospheric CH\(_4\) confirmed that the short-term CH\(_4\) variations are mainly produced by biogenic CH\(_4\) released from the HBL wetlands in summer and by fossil fuel CH\(_4\) transported over the Arctic in winter. Forward simulations of an atmospheric chemistry-transport model, with wetland CH\(_4\) fluxes prescribed by a process-based model, show unrealistically high CH\(_4\) mole fractions at Churchill in summer, suggesting that CH\(_4\) emissions assigned to the HBL wetlands are overestimated. Our best estimate of the HBL CH\(_4\) emissions is 2.7 ± 0.3 Tg CH\(_4\) yr\(^{-1}\) as an average of 2007–2013, consistent with recent estimations by inverse modeling studies.

1. Introduction

Methane (CH\(_4\)) plays an important role in global climate change, as well as in atmospheric chemistry, because CH\(_4\) is the second most important anthropogenic long-lived greenhouse gas after CO\(_2\) and its destruction occurs primarily by chemical reactions in the atmosphere. CH\(_4\) is emitted from natural (wetlands, freshwater, wild animals, wildfires, termites, geological processes, ocean, hydrates, and permafrost) and anthropogenic (rice paddies, ruminants, landfills and waste, fossil fuels, and biomass burning) sources. CH\(_4\) is mainly destroyed by reaction with OH radicals in the troposphere and partly by reactions with OH, Cl, and O\(^{1}D\) in the stratosphere and by bacterial consumption in soils. Since the atmospheric CH\(_4\) mole fraction shows large spatiotemporal variations due to unevenly distributed CH\(_4\) sources/sinks and complicated atmospheric transport, an extensive and dense network of observations is required to depict a global picture of atmospheric CH\(_4\) variations. For this purpose, observations of atmospheric CH\(_4\) with grab sampling and continuous measurement techniques have been conducted mainly at ground-based stations since the 1970s (e.g., Aoki et al., 1992; Blake & Rowland, 1986; Cunnold et al., 2002; Dlugokencky et al., 2011; Rasmussen & Khalil, 1981). In the last few decades, the mole fraction of CH\(_4\) showed unpredictable trends; the rate of increase in atmospheric CH\(_4\) slowed down in the 1980s to 1990s, leveled off from 1999 to 2006, and then rose again in 2006/2007 (Dlugokencky et al., 2009; Morimoto et al., 2017; Rigby et al., 2008). Such a CH\(_4\) trend was examined in terms of ruminants, boreal and/or tropical wetlands, fossil fuels, or change in OH, but the cause is still controversial (e.g., Kirschke et al., 2013; Nisbet et al., 2016; Patra et al., 2016; Rigby et al., 2017; Schaefer et al., 2016; Turner et al., 2017). It is also known that there is a large discrepancy between CH\(_4\) budgets estimated by top-down (inverse modeling using atmospheric measurements) and bottom-up (direct flux measurement,
statistical database, and process-based modeling) approaches, especially for natural sources (Kirschke et al., 2013; Saunois et al., 2016).

Systematic and high-precision observations of carbon and hydrogen isotope ratios (δ13C and δD) of CH4 provide us with additional constraints to understand the contribution of individual CH4 sources to atmospheric CH4 variations because each source has its own characteristic isotope ratio (e.g., Quay et al., 1999; Schwietzke et al., 2016; Sherwood et al., 2017; Whiticar & Schaefer, 2007). Their data would also help to close the gap in the CH4 budget estimation between the top-down and bottom-up approaches through better source apportionment. δ13C and δD are commonly defined by

\[
\delta = \left( \frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \times 1000 \ (\%_\text{o}).
\]  

Here δ represents δ13C or δD and R indicates 13C/12C or D/H. Subscripts “sample” and “standard” denote the sample and the standard, respectively, and as an international standard scale, VPDB is widely used for δ13C and VSMOW for δD. Sherwood et al. (2017) recently reported by compiling a large number of isotope observation data that the biogenic, fossil fuel, and biomass burning CH4 sources have the respective mean isotope ratios of −61.7 ± 6.2 (±1 standard deviation [σ]), −44.8 ± 10.7, and −26.2 ± 4.8‰ for δ13C and −317 ± 33, −197 ± 51, and −211 ± 15‰ for δD. Atmospheric background δ13C and δD were also reported to be approximately −47 and −86‰, respectively (Allan et al., 2001; Whiticar & Schaefer, 2007). However, there have only been a few studies on simultaneous and high-precision measurements of atmospheric δ13C and δD, which aim at examining atmospheric CH4 variations (Rice et al., 2016; Röckmann et al., 2016; Tyler et al., 2007; Umezawa et al., 2012; Warwick et al., 2016).

The Hudson Bay Lowlands (HBL), the second largest continuous wetland in the world, is an important natural CH4 source region in northern latitudes (Glooschenko et al., 1994). Nevertheless, there still remains a large uncertainty in magnitude, seasonality, and spatial distribution of CH4 emissions in the HBL. Previous estimates of CH4 emission rates for the HBL wetlands range from 0.2 to 11.3 Tg CH4 yr−1 (Melton et al., 2013; S. M. Miller et al., 2014, 2016; Pickett-Heaps et al., 2011; Roulet et al., 1994; Thompson et al., 2017; Worthy et al., 2000). In addition to the regional influence, the HBL area is also affected to some extent by anthropogenic CH4 released in Europe and boreal Asia due to long-range air transport, especially in winter (Worthy et al., 1998, 2009). There may also be large anthropogenic CH4 sources in Alberta located to the west of the HBL in association with natural gas production (S. M. Miller et al., 2014; Thompson et al., 2017). It is further pointed out that natural CH4 sources such as ocean, geological seepages, subsea permafrost, and sea ice exist in the Arctic (e.g., Sapart et al., 2017; Walter Anthony et al., 2012). Therefore, to accurately estimate CH4 emissions from the HBL wetlands based on the atmospheric CH4 observations, it is necessary to examine the influence of anthropogenic and other natural CH4 on its atmospheric variations.

To better understand the CH4 cycle around the HBL, we started systematic air sampling at Churchill in 2007, situated in the northern part of the HBL, and analyzed those samples for the CH4 mole fraction, δ13C, and δD. We present long-term, seasonal, and short-term variations of these three variables observed at the site and compare them with those at an Arctic baseline station, Ny-Ålesund, Svalbard (78°55′N, 11°56′E; Morimoto et al., 2006, 2017). We then discuss the potential causes of temporal variations. By comparing the observed atmospheric CH4 mole fractions with those simulated using an atmospheric chemistry transport model, we further examine CH4 emissions in the HBL.

2. Method

2.1. Air Sampling and Analysis of CH4 Mole Fraction, δ13C, and δD

Systematic observations of the CH4 mole fraction and isotope ratios have been conducted at Churchill, Manitoba, Canada (58°44′N, 93°49′W) since April 2007, by a collaborative effort of the National Institute of Polar Research (NIPR), Tohoku University (TU), and Environment and Climate Change Canada (ECCC). The location of Churchill is shown in Figure 1, together with the land cover map of the HBL and its surrounding areas. Details of air sampling procedures and site description are found at the World Data Centre for Greenhouse Gases (WDCGG) website (https://ds.data.jma.go.jp/gmd/wdcg/cgi-bin/wdcg/accessdata.cgi?index=CHL458N00-EC&param=201208150002&select=parameter&parac=observation); thus, a brief explanation is presented here.
Churchill is a small port city on the western shore of Hudson Bay with a population of about 900. The land cover around Churchill is mainly characterized by the Arctic tundra and the boreal forest. Air samples were taken from an intake mounted at the top of a 60-m high tower in the Churchill Northern Studies Centre (https://www.churchillscience.ca/), located 23 km east of the town of Churchill. Each air sample was automatically collected twice a week into a 2-L Pyrex glass flask at a pressure of 0.21 MPa, using a dedicated sampling system consisted of a separated line, a diaphragm pump, and a glass trap submerged in a −80 °C methanol bath. The collected samples with a dew point of around −60 °C were first analyzed at ECCC for mole fractions of various trace gases such as CO2, CH4, CO, N2O, and SF6 and then transported to NIPR, Japan, at approximately 0.16–0.17 MPa for isotope analyses of atmospheric CH4. At NIPR, each sample was divided into four 100-mL Pyrex glass flasks, two for the analysis of δ13C at NIPR and two for δD at TU.

Observations at Ny-Ålesund, Svalbard (78°55′N, 11°56′E), to be compared with those at Churchill have been described by Morimoto et al. (2006) in detail. Air samples were collected once a week into 800-mL stainless steel flasks at 0.8 MPa and then sent to NIPR and TU for the mole fraction and isotope analyses.

Air samples collected at Churchill were analyzed for the CH4 mole fraction at ECCC by using a gas chromatograph (Agilent 6890) equipped with a flame ionization detector (GC-FID) (Worthy et al., 1998) against the WMO-X2004A scale based on a gravimetric method (Dlugokencky et al., 2005, https://www.esrl.noaa.gov/gmd/ccgg/ch4_scale.html). The repeatability of the CH4 mole fraction analysis was estimated to be better than 2 ppb by analyzing the same sample repeatedly. CH4 mole fractions of the air samples collected at Ny-Ålesund were determined using the GC-FID (Shimadzu, GC-8A) at NIPR relative to the TU1987 scale (Aoki et al., 1992; Morimoto et al., 2006). The results of the fifth and sixth WMO (World Meteorological Organization) Round-Robin intercomparison programs (https://www.esrl.noaa.gov/gmd/ccgg/wmorr/wmorr_results.php) showed that the TU2008 scale is higher than the WMO-X2004A scale by 2.5 ± 0.5 ppb on average. The TU2008 scale was also gravimetrically established by the same procedure as the TU1987, but a recently conducted close comparison of the two scales shows that the former provides lower CH4 mole fractions by about 3.0 ppb than the latter at atmospheric CH4 levels. Therefore, the difference between the TU1987 and WMO-X2004A scales is about 0.5 ppb. In this study, we compare the data at Churchill and Ny-Ålesund without any scale correction.

δ13C of CH4 was determined by using a gas chromatography-combustion isotope ratio mass spectrometer (GC-C-IRMS) based on MAT-252 (Thermo Fischer) with repeatability of 0.07‰ (Morimoto et al., 2006, 2009).
2017). The standard used in this analysis was pure CO₂ calibrated using a dual-inlet mass spectrometer against the TU δ¹³C scale prepared from NBS-19 with δ¹³CVPDB of +1.95‰ (Nakazawa et al., 1993). In the daily δ¹³C analysis, we analyzed a CH₄-in-air “test gas” with the known value of δ¹³C, stored in a 47-L aluminum cylinder, at least once a day to confirm the long-term stability of our δ¹³C measurements. δD of CH₄ was obtained by using a gas chromatography-pyrolysis isotope ratio mass spectrometer (GC-P-IRMS) based on Delta Plus XP (Thermo Fischer) with repeatability of 2.2‰ (Umezawa et al., 2009). Our δD scale was established based on VSMOW (δDᵥsmow = 0‰) and SLAP (−428‰) using a dual-inlet mass spectrometer with a chromium reduction system at NIPR. To confirm the internal consistency of our δD analyses over a long period of time, we also analyzed a test gas at least twice on a measurement day and then corrected for potential day-to-day fluctuations of the measured δD arisen from changable conditions of the GC-P-IRMS, assuming that the δD value of the test gas is stable with respect to time (Umezawa et al., 2009). The δD value of the test gas was determined using GC-P-IRMS against a reference gas (purified H₂) calibrated by VSMOW and SLAP.

The comparison of our δ¹³C scale with that of the National Institute of Water and Atmospheric Research (NIWA) was carried out in 2004, and the result showed that our scale is 0.33 ± 0.04‰ higher than the NIWA scale (Morimoto et al., 2006, 2017). The comparison of our δD scale with that of the Institute for Marine and Atmospheric Research Utrecht carried out in 2013–2015 showed that our scale is lower by 13.1 ± 0.6‰ than theirs at ambient air levels (Umezawa et al., 2018). More information on the intercomparison of standard scales used in the CH₄ isotope community, including TU and NIPR, has been given in Umezawa et al. (2018).

### 2.2. Model Simulation of CH₄ Mole Fraction

To interpret temporal variations of CH₄ in the atmosphere at Churchill and to estimate CH₄ emissions from the HBL, forward simulations of atmospheric CH₄ mole fraction were conducted for 2007–2013 using the CCSR/NIES/FRCGC (Center for Climate System Research/National Institute for Environmental Studies/Frontier Research Center for Global Change) AGCM-based Chemistry Transport Model (ACTM) developed at JAMSTEC (Japan Agency for Marine-Earth Science and Technology), with the setup described in Patra et al. (2016). ACTM uses a horizontal resolution of approximately 2.8° × 2.8° (T42 spectral truncation), with 67 pressure-sigma vertical layers. The atmospheric transport and tropospheric OH radical fields used in the model were validated by Patra et al. (2011, 2014).

Two CH₄ emission scenarios, “P16pri” and “P16pos,” were used in this study, which are a priori and a posteriori CH₄ emissions of the global inverse modeling (Patra et al., 2016, corresponding to their “Case 2. CH₄ags”). In the P16pri scenario, anthropogenic CH₄ emissions are adopted from EDGAR42FT (2013) and kept constant at the value of the year 2000, except for agricultural soils for which annual emissions are given until 2010 and then the value in 2010 is used repeatedly for 2011–2013. CH₄ emissions from biomass burning are taken from the combination of GISS (Goddard Institute for Space Studies) inventory (Fung et al., 1991) and GFED (Global Fire Emission Database) version 3.1 (van der Werf et al., 2010) after multiplying the GISS inventory by an optimal scaling factor (Patra et al., 2011). Biogenic (wetlands and rice paddies) CH₄ emissions are obtained from a process-based terrestrial ecosystem model, VISIT (Ito & Inatomi, 2012). The P16pos scenario is derived by optimizing the P16pri scenario using ACTM and CH₄ mole fraction observations (Patra et al., 2016). In forward simulations with ACTM and the above mentioned two scenarios, atmospheric CH₄ is destroyed through reactions with OH, Cl, and O(1D) and through bacterial consumption in soils. Global OH field obtained by Spivakovskiy et al. (2000) is scaled so that ACTM reproduces the observed decay rate of CH₃CCl₃ in the atmosphere (Patra et al., 2011, 2014). The soil sink is prepared by VISIT (Ito & Inatomi, 2012), and stratospheric loss by OH, Cl, and O(1D) is calculated using their concentration fields obtained by ACTM’s stratospheric model run (Takigawa et al., 1999).

To investigate CH₄ source regions contributing to atmospheric CH₄ variations at Churchill, tagged tracer experiments were also performed using ACTM (Umezawa et al., 2014). In the experiments, the surface CH₄ emission field from the P16pos scenario was used. The global surface was first divided into 17 regions (Figure 2), and the forward simulation was performed for CH₄ released from each region. The region division is slightly different from that in Umezawa et al. (2014). In particular, we divided Boreal North America into four regions to better understand the regional contribution of CH₄ sources around Churchill. We defined the HBL
area as 50°–60°N and 75°–96°W after Pickett-Heaps et al. (2011) (Region 14), the Province of Alberta as 50°–60°N and 110°–120°W after Thompson et al. (2017) (Region 12), and the border of western and eastern Canada as 96°W (Region 11 and Region 13).

3. Results and Discussion

3.1. Variations of CH$_4$ Mole Fraction, $\delta^{13}C$, and $\delta^D$ at Churchill and Ny-Ålesund

Figures 3a–3c show temporal variations of the CH$_4$ mole fraction, $\delta^{13}C$, and $\delta^D$ observed at Churchill and Ny-Ålesund for 2007–2014, together with best fit curves to the data and long-term trends obtained using a digital-filtering technique (Nakazawa et al., 1997). In the filtering, an average seasonal cycle of each variable was approximated by fundamental and its first harmonics, and low-pass filters with cutoff periods of 4 and 24 months were adopted to obtain the best fit curve and the long-term trend, respectively. As seen in Figure 3a, the CH$_4$ mole fraction at Churchill shows a clear seasonal cycle with a prominent minimum in June–July and a broad maximum in late winter, superimposed on an increasing trend. Similar characteristics are also observed at Ny-Ålesund. However, there are noticeable differences between the CH$_4$ variations at Churchill and Ny-Ålesund: (1) the annual mean CH$_4$ mole fraction is higher by 3–16 ppb at Churchill than at Ny-Ålesund for 2007–2013, (2) the timing of the seasonal CH$_4$ minimum is earlier by about 1 week, on average, at Churchill than at Ny-Ålesund (Figure 4a), and (3) episodic high CH$_4$ mole fractions, sometimes over 2000 ppb, are frequently observed at Churchill throughout the year. Similar characteristics are also observed at Ny-Ålesund. However, there are noticeable differences between the CH$_4$ variations at Churchill and Ny-Ålesund: (1) the annual mean CH$_4$ mole fraction is higher by 3–16 ppb at Churchill than at Ny-Ålesund for 2007–2013, (2) the timing of the seasonal CH$_4$ minimum is earlier by about 1 week, on average, at Churchill than at Ny-Ålesund (Figure 4a), and (3) episodic high CH$_4$ mole fractions, sometimes over 2000 ppb, are frequently observed at Churchill throughout the year. A clear seasonal cycle is also observed in $\delta^{13}C$ and $\delta^D$ at Churchill and Ny-Ålesund, showing the maximum in early summer and the minimum in autumn. From inspection of the observation data at the two sites, it is obvious that (1) the annual means are lower by 0.1–0.2‰ for $\delta^{13}C$ and 1–4‰ for $\delta^D$ at Churchill than at Ny-Ålesund, (2) the average seasonal maxima of $\delta^{13}C$ and $\delta^D$ at Churchill precede those at Ny-Ålesund by about 2–3 weeks (Figures 4b and 4c), and (3) anomalously low $\delta^{13}C$ and $\delta^D$ values, below −48.5‰ for $\delta^{13}C$ and −115‰ for $\delta^D$, are often observable at Churchill in the summertime. The differences in annual mean CH$_4$, $\delta^{13}C$, and $\delta^D$ between the two sites suggest that Churchill is more strongly affected by biogenic CH$_4$ sources with low $\delta^{13}C$ and $\delta^D$ than Ny-Ålesund. The seasonal phases of CH$_4$, $\delta^{13}C$, and $\delta^D$ at the two sites and the events with high CH$_4$ and low $\delta^{13}C$ and $\delta^D$ at Churchill are discussed in sections 3.2 and 3.3 in detail, respectively.
The seasonal cycles of CH4 and δ13C at Churchill and Ny-Ålesund are similar to those observed previously at other northern mid-to-high latitude sites (Dlugokencky et al., 2011; J. B. Miller et al., 2002; Nisbet et al., 2016; Tyler et al., 2007; Warwick et al., 2016). There are a few δD observations for the background atmosphere in northern mid-to-high latitudes (Tyler et al., 2007; Warwick et al., 2016). The seasonal cycles of δD observed by Warwick et al. (2016) at three northern high-latitude sites of Alert (82°N, 63°W), Barrow (71°N, 157°W), and Cold Bay (55°N, 163°W) are generally similar to those at Churchill and Ny-Ålesund.

The average growth rate of the CH4 mole fraction at Churchill over 2007–2013 is 3.7 ± 0.5 ppb/yr (±95% confidence interval [C.I.] derived using a residual bootstrap method; Davison & Hinkley, 1997), which is slightly smaller than 4.9 ± 0.5 ppb/yr at Ny-Ålesund. The globally averaged CH4 growth rate derived from the National Oceanic and Atmospheric Administration/Earth System Research Laboratory (NOAA/ESRL) sites (http://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/#global) for the same period is 5.3 ± 0.3 ppb/yr (1σ), which is comparable to the value at Ny-Ålesund. Dlugokencky et al. (2009) reported that the largest CH4 increase of 13.7 ± 1.3 ppb/yr was observed at northern polar latitudes in 2007. A similar rapid CH4 increase of 8.8 ± 3.0 ppb/yr was observed at Ny-Ålesund from 2007 to 2008, while no significant increase was detected at Churchill (1.1 ± 4.5 ppb/yr) for the same period (±95% C.I.). As seen from Figure 3a, high CH4 values were often observed in the warm season of 2007 at Churchill. If data collected in 2007 are excluded from the

![Figure 3](http://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/#global)
Figure 4. Average seasonal cycles of (a) the CH4 mole fraction, (b) δ13C, and (c) δD observed at Churchill (red lines) and Ny-Ålesund (blue lines) for 2007–2013. Dotted lines represent the 95 percentile bootstrap confidence intervals (see text). Each average seasonal cycle is plotted after adding its average value for 2007–2013.

records of the two sites, the growth rate is 3.9 ± 0.7 ppb/yr for Churchill and 4.3 ± 0.6 ppb/yr for Ny-Ålesund, the values being close to each other.

No significant increasing or decreasing trend is found in δ13C at Churchill for 2007–2013, with the rate of change of 0.005 ± 0.005‰/yr (±95% C.I.). In contrast to Churchill, a significant decrease of −0.007 ± 0.004‰/yr was observed at Ny-Ålesund for the same period. The δ13C trend at Churchill is probably due to the same reason as the low CH4 growth rate, but with very low δ13C values in the summer of 2007. By excluding the data for 2007, the rate of change in δ13C is found to be −0.002 ± 0.006‰/yr, which still shows no significant trend. Schaefer et al. (2016) and Nisbet et al. (2016) reported the secular decrease in δ13C after 2006/2007, suggesting that biogenic CH4 sources are predominantly responsible for the CH4 increase after 2006. Long-term variations in CH4 mole fraction and δ13C at Ny-Ålesund in 1996–2013 have been discussed in Morimoto et al. (2017) in detail.

The average rates of increase in δD at Churchill and Ny-Ålesund for 2007–2013 are 0.43 ± 0.13 and 0.12 ± 0.10‰/yr, respectively (±95% C.I.). Since the δD data show relatively large interannual variability and its measurement uncertainty is larger than that of δ13C, it is difficult to robustly determine the trend; thus, we do not discuss the long-term variations in δD at this stage. However, considering that δD is more sensitive to the chemical reaction of CH4 with OH than δ13C because of the larger kinetic isotope effect associated to the destruction of CH3D in comparison with 12CH4 and 13CH4, further studies on atmospheric δD are required to improve our understanding of long-term changes and interannual variability in CH4 sinks (e.g., McNorton et al., 2016; Montzka et al., 2011; Rigby et al., 2017; Turner et al., 2017).

3.2. Seasonal Variations in CH4 Emissions

As mentioned above, the CH4 mole fraction, δ13C, and δD vary seasonally at Churchill and Ny-Ålesund, and the seasonal minimum of CH4 mole fraction and the seasonal maxima of δ13C and δD at Churchill appear about 2 weeks earlier than those at Ny-Ålesund. To examine the contributions of biogenic, fossil fuel, and biomass burning CH4 sources to the observed seasonal CH4 cycle at Churchill and Ny-Ålesund, we employed a simple one-box model expressed by the following equations (Tyler et al., 2007; Umezawa, 2009):

\[
\frac{dC_{ATM}}{dt} = S_{BI} + S_{FF} + S_{BB} - kC_{ATM}.
\]

\[
\frac{d(CATM R_{C,ATM})}{dt} = S_{BI} R_{C,BI} + S_{FF} R_{C,FF} + S_{BB} R_{C,BB} - \frac{1}{KIE} kC_{ATM} R_{C,ATM}.
\]

\[
\frac{d(CATM R_{D,ATM})}{dt} = S_{BI} R_{D,BI} + S_{FF} R_{D,FF} + S_{BB} R_{D,BB} - \frac{1}{KIE} kC_{ATM} R_{D,ATM}.
\]

where \( C_{ATM} \) is the observed value of the CH4 mole fraction in the atmosphere; \( R_C \) and \( R_D \) denote the carbon and hydrogen isotope ratios (i.e., \( ^{13}C/^{12}C \) and D/H) of atmospheric (ATM), biogenic (BI), fossil fuel (FF), and biomass burning (BB) CH4, respectively; \( S \) indicates the seasonally variable contributions of the three CH4 sources; and \( k \) is the pseudo-first-order rate coefficient for OH + CH4. In this analysis, the observed atmospheric monthly values of \( C_{ATM} \) and \( R_{C,ATM} \) and \( R_{D,ATM} \) are derived by adding the average seasonal cycle to the average annual value for 2007–2013 (Figure 4). The respective isotopic signatures of BI, FF, and BB sources \( (R_C \text{ and } R_D) \) were assumed to be −61.7 ± 6.2 \((±1σ)\), −44.8 ± 10.7, and −26.2 ± 4.8‰ for δ13C and −317 ± 33, −197 ± 51, and −211 ± 15‰ for δD (Sherwood et al., 2017). \( k \) was calculated based on the
The present uncertainty range of ε is almost consistent with the estimate by Schaefer et al. (2016). The parameters used in this box model analysis are summarized in Table S1 in the supporting information. For uncertainty estimation of this model analysis, we assumed that the respective source isotopic signatures and KIEs distribute normally around their mean values with 1σ and then ran the Monte Carlo simulation 5000 times by randomly sampling the normally distributed isotopic signatures, KIEs, and average seasonal cycles of CH₄, δ¹³C, and δD (Figure 4). By using the 5000 pseudo-data sets thus generated, we calculated the median and 68 percentile confidence intervals of the monthly contributions of the respective CH₄ sources (Ss).

Previous measurements of CH₄ fluxes indicate that the CH₄ emissions from boreal wetlands peak in June–August (e.g., Whalen & Reeburgh, 1992). However, there are large differences in the strength and seasonality of their measured CH₄ fluxes, mainly due to large spatial and temporal variability of CH₄ emissions. Pickett-Heaps et al. (2011) estimated the CH₄ emissions from the HBL using the GEOS-Chem chemical transport model and the atmospheric CH₄ mole fraction data at Fraserdale and Alert, Canada, and found that the seasonal maximum occurs in July. S. M. Miller et al. (2014) also suggested from their regional inversion that CH₄ emissions from the HBL reach a maximum in July. The Bayesian atmospheric inversion model results by Thompson et al. (2017) showed that the CH₄ flux in the HBL increases gradually in spring, reaches a maximum in August–September, and declines rapidly in September–October.

Seasonal variations of the contribution of biogenic CH₄ estimated for Churchill and Ny-Ålesund are slightly different from each other (Figures 5a and 5b). For example, the biogenic CH₄ is discernible at Churchill in May, but there is no appearance of such a contribution at Ny-Ålesund. Moreover, the seasonal maximum of the biogenic CH₄ contribution appears in July at Churchill and in August at Ny-Ålesund. This difference is presumably attributable to the influence of local/regional wetland CH₄ emissions on Churchill and to different latitudes of the two sites. Churchill is located on the northern perimeter of the HBL; thus, CH₄ emitted from HBL wetlands could directly affect the CH₄ mole fraction at Churchill. On the other hand, since
Ny-Ålesund is far from strong CH4 sources, seasonal signals of CH4 emissions from boreal wetlands may reach the site with a time lag. It is also noteworthy that the onset of wetland CH4 emissions is earlier at lower latitudes due to the latitude-dependent seasonal temperature pattern.

It is also found in Figure 5 that fossil fuel and biomass burning are minor contributors to the seasonal CH4 cycle. However, more detailed inspection of the results indicates that fossil fuel sources significantly influence the atmospheric CH4 mole fraction both at Churchill and Ny-Ålesund in early winter. Fossil fuel sources of CH4 emissions are usually regarded as nonseasonal sources (EDGAR42FT, 2013), but the contribution of fossil fuel CH4 emissions could be enhanced in winter, especially in northern high-latitude regions. For example, natural gas is consumed in large quantities during the cold season, during which the transport pipelines are pressurized so that a significant leakage of CH4 may occur (Lowry et al., 2001). In addition to fossil fuel CH4 emissions, slow vertical air mixing due to the strong inversion layer and weak destruction of CH4 with OH may strengthen the influence of the fossil fuel CH4 source on the wintertime increase of atmospheric CH4.

Biomass burning is also known to have seasonality in CH4 emission, mainly due to seasonally varying rainfall and temperature. GFED 3 shows that the maximum CH4 emissions from biomass burning occur in July in northern high latitudes (>50°N) (van der Werf et al., 2010). However, in this study, only small seasonal variations are detected for CH4 emissions from biomass burning. Although the summertime maximum of biomass burning CH4 emissions is detected both at Churchill and Ny-Ålesund, the values are not statistically significant.

We also see at Churchill and Ny-Ålesund that the chemical destruction by OH varies seasonally as large as biogenic CH4. Since CH4 is a long-lived species whose atmospheric lifetime is longer than 1 year even in summer, OH in remote areas would play an important role in the observed CH4 variations both at two sites. Therefore, we used OH concentration and temperature data averaged over 30°–90°N and 700–1000 hPa in the present one-box model analysis. Other CH4 sinks, such as soil oxidation and stratospheric loss, also contribute to the CH4 seasonal cycle to some extent. The results of the one-box model analysis are further affected by values adopted for the isotopic signatures of CH4 sources and KIEs. To inspect the sensitivity of our model analysis results to these variables, we made the one-box model analysis again using the parameters different from the initial set (see Table S1). The results of the sensitivity tests obtained for the two sites, shown in Figures S1–S3, indicate that the seasonal contributions obtained under various conditions are generally consistent to that derived with our initially set parameters.

### 3.3. Short-Term Variations of CH4 Mole Fraction, δ13C, and δD

The CH4 mole fraction sometimes shows extremely high values at Churchill throughout the year. Similar anomalous data are also found in δ13C and δD with extremely low values, although such data are observed only in the summertime. In this study, 596, 605, and 600 data are available for the CH4 mole fraction, δ13C, and δD, respectively. By defining the data deviated from the best fit curve by more than 3σ of the fit as outliers, 50, 41, and 19 data were selected out from the respective records of the CH4 mole fraction, δ13C, and δD.

To investigate the cause of the 50 outliers with extremely high CH4 mole fractions in terms of emission sources, the "Miller/Tans plot" represented by

\[
C_{\text{obs}} \delta_{\text{obs}} - C_{\text{BGD}} \delta_{\text{BGD}} = (C_{\text{obs}} - C_{\text{BGD}}) \delta_S
\]
was applied to the CH₄ mole fraction, δ¹³C, and δD data (J. B. Miller & Tans, 2003; Umezawa et al., 2012). Here C and δ represent the CH₄ mole fraction and corresponding δ¹³C (or δD), respectively, and subscripts obs, BGD, and S denote the observed, background, and source values, respectively. The mean isotope ratio of the source, δS, can be obtained as a slope of the regression line of C₀obsδobs/C₀BGDδBGD and C₀obs/C₀BGD. In this analysis, the background value for each variable is given by the best fit curve of the related observation data.

Figures 6a and 6b show the Miller/Tans plots for δ¹³C and δD, respectively. It is found from the figures that the summertime (May–October) and wintertime (November–April) slopes are significantly different from each other. By applying an ordinary least squares regression to each cluster, the summertime data yield the slopes of

\[ \text{sum: } \delta^{13}C = 63.3 \pm 2.8 \text{‰ (±95\% C.I; } R^2 = 0.96) \]

and

\[ \delta D = 327 \pm 26 \text{‰ (} R^2 = 0.92) \]

while the wintertime data provide the corresponding values of

\[ \text{winter: } \delta^{13}C = 47.7 \pm 4.3 \text{‰ (} R^2 = 0.96) \]

and

\[ \delta D = 241 \pm 48 \text{‰ (} R^2 = 0.89) \]

The summertime slopes agree well with those expected from biogenic CH₄ sources (e.g., Sherwood et al., 2017; Whiticar & Schaefer, 2007), suggesting the influence of CH₄ emissions from the HBL wetlands. On the other hand, the wintertime slopes result in much heavier isotope ratios than the summertime slopes, the values being close to the isotopic signatures of fossil fuel CH₄.

Previous studies reported that the δ¹³C and δD values of CH₄ released from wetlands in northern high latitudes range from −60 to −80‰ and from −300 to −420‰, respectively (e.g., Nakagawa et al., 2002; Walter et al., 2008). Our summertime δ¹³C and δD slope values fall in previously reported ranges for the respective variables. Measurements taken by Kuhlmann et al. (1998) for two days at Fraserdale, Ontario, in August 1995 show that the isotopic signature of CH₄ from regional wetlands is

\[ \delta^{13}C = 60.0 \pm 3.2 \text{‰ and } \delta D = 442 \pm 142 \text{‰} \]

These values are consistent with our summertime values within estimated uncertainty limits, although their δD estimate is more negative than ours on average.

Worthy et al. (1998, 2009) show that the air is often transported from Siberia and Europe to the Canadian high Arctic region in winter, by which Canada is widely covered with polluted air masses originated in the Eurasian Continent. To investigate the highly elevated CH₄ mole fractions observed at Churchill in winter, a 7-day backward trajectory analysis was conducted using the HYSPLIT model (Stein et al., 2015). In this analysis, each air parcel was released from 500 m above sea level over Churchill at the time when the high CH₄ mole fraction was observed. The results show that the air parcels wander around Churchill in summer, while the wintertime air parcels go back to more distant areas, mainly northern high latitudes (Figure 7). The backward trajectory analysis also shows that some air parcels assigned to high CH₄ mole fractions observed at Churchill in winter originated from Western Canada (Figure 7b). In this connection, S. M. Miller et al. (2014) and Thompson et al.
(2017) reported recently that a large amount of CH4 is presumably released from Alberta, Western Canada, in association with natural gas production.

There are also other minor natural CH4 sources, such as ocean, geological seepages, subsea permafrost, and sea ice, in the Arctic region, of which isotopic signatures are close to the values of wetlands and/or fossil fuel sources (e.g., Sapart et al., 2017; Walter Anthony et al., 2012). As mentioned above, the backward trajectory analysis indicates that Churchill is strongly influenced by air masses from the HBL and its surroundings in summer. This suggests that the summertime CH4 enhancement at Churchill is mainly due to wetlands rather than these minor sources. On the other hand, it is difficult to distinguish anthropogenic fossil fuel origin from natural geologic origin in winter using the backward trajectory analysis. However, some wintertime high CH4 events at Churchill were found to be coincident with high CO and CO2 mole fractions, suggesting the influence of human activities.

3.4. Model Simulation of Atmospheric CH4 Variations

To investigate CH4 emissions from the HBL in more detail, we simulated the atmospheric CH4 mole fraction at Churchill by using ACTM and two CH4 emission scenarios, P16pri and P16pos. The atmospheric CH4 mole fractions simulated for 2007–2013 are shown in Figure 8a, together with the observed values. For comparison, the results for Ny-Ålesund are also shown in Figure 8b. As seen in the figures, CH4 mole fractions simulated for Ny-Ålesund reproduce general features of the observed CH4 variations, while obvious discrepancies between the simulated and observed mole fractions are seen at Churchill. The CH4 mole fractions simulated using both scenarios for Churchill frequently overestimate and underestimate the summertime and wintertime values, respectively. It is also found at the two sites that the model-simulated CH4 mole fractions based on P16pri are higher than the observations for 2007–2010 as a whole. In this connection, Patra et al. (2016) mentioned that a priori emissions used in their inversion (i.e., P16pri scenario) are too high in the 2000s. On the other hand, P16pos reproduces fairly well the long-term trends of atmospheric CH4 at the two sites.

To see the degree of model-observation agreement at each site, the correlation coefficient ($R$) and the root-mean-square error (RMSE) of the simulated and observed CH4 mole fractions for each scenario are summarized in Table 1. These statistical parameters were calculated from the respective curves fitted to the simulated and observed data (Nakazawa et al., 1997). $R$ generally indicates the degree of agreement between the model calculation and observation for the seasonal phase of atmospheric CH4, since the seasonal CH4 cycle is larger in amplitude than interannual variations. RMSE is a measure of how well the model reproduces the observed CH4 variations.

The respective correlation coefficients obtained for the P16pri and P16pos scenarios are 0.22 and 0.36 for Churchill and 0.57 and 0.95 for Ny-Ålesund (Table 1). The results of Ny-Ålesund show that the observed seasonality of atmospheric CH4 is reproduced fairly well by the model for either scenario and that the agreement between the model and observation is much improved by employing P16pos rather than P16pri. RMSE is also decreased by replacing P16pri with P16pos, suggesting an improvement of the model-observation agreement. On the other hand, the two statistics, $R$ and RMSE, for Churchill indicate that there is no appreciable improvement even if the scenario is altered. It should be noted that the P16pos scenario was derived from the inversion calculation by including the CH4 mole fraction data observed at Zeppelin Station, Ny-Ålesund, but with no observation data around the HBL (Patra et al., 2016). Therefore, the model with P16pos shows a much better agreement with observed CH4 variations at Ny-Ålesund rather than at Churchill.

To improve the agreement between the model-simulated and observed seasonal CH4 cycles at Churchill, we first examined the cause for this discrepancy. The average seasonal CH4 cycle at Churchill and Ny-Ålesund,
derived by applying the digital filtering technique to the observed and model-calculated CH₄ mole fractions, is plotted in Figure 9 after adding the average CH₄ mole fraction over 2007–2013 at the respective sites. Since the OH fields and the atmospheric transport of ACTM are validated (Patra et al., 2011, 2014), the difference between the observed and model-generated seasonal CH₄ cycles could be mainly attributable to CH₄ emissions adopted in model simulations. Figure 9c shows the difference between the average seasonal CH₄ cycles at Churchill and Ny-Ålesund (defined as ΔCH₄) for each scenario or the observation. Since the variations at Ny-Ålesund are representative of northern high latitudes, ΔCH₄ would be closely related to CH₄ emissions around Churchill. The observations yield the maximum ΔCH₄ of approximately 15 ppb in late July and December–January. On the other hand, model simulations show the maximum ΔCH₄ of up to 40–60 ppb in late July, which is 3 to 4 times the observational result. This suggests that both P16pri and P16pos scenarios overestimate the summertime CH₄ emissions around Churchill. As shown in Figure 5, we found at Churchill and Ny-Ålesund that CH₄ emissions from biogenic and fossil fuel sources dominate the CH₄ mole fractions in summer and winter, respectively, and biomass burning is not important for the seasonality of atmospheric CH₄. Therefore, the summertime and wintertime maxima of ΔCH₄ are likely associated with the respective emissions of CH₄ from wetlands and fossil fuels around Churchill.

Since the model-observation disagreement is remarkably larger in summer than in winter, we focus our discussion on the summertime events. To clarify which regions contribute to the summertime overestimated CH₄ mole fractions, we conducted tagged tracer experiments as described in section 2.2. The calculated contributions of the respective regions to the average seasonal CH₄ cycles at Churchill and Ny-Ålesund are shown in Figure 10. Also shown in the figure are the observed and model-simulated (P16pos) average seasonal CH₄ cycles.

It is obvious from Figure 10 that the HBL (rg14), Western Canada/Alaska (rg11), and Europe (rg02) have a large influence on the seasonal CH₄ cycle at Churchill. However, the model-generated seasonal CH₄ cycles for the HBL and Western Canada/Alaska are quite different from the observed result, particularly in seasonal phase. CH₄ originated in these regions are emitted mostly from boreal wetlands. Therefore, the reproduction of the observed seasonal CH₄ cycle by the model can be greatly improved by reducing the summertime CH₄ emissions, especially from the HBL. In this regard, the forward simulations with the P16pos scenario can reproduce relatively well the CH₄ mole fractions at Alert (82°N, 63°W), Barrow (71°N, 157°W), Cold Bay (55°N, 163°W), and Estevan Point (49°N, 127°W) (Patra et al., 2016), which suggests that the CH₄ emissions around the four background sites (Western Canada/Alaska) are constrained fairly well.

### 3.5. CH₄ Emissions From the HBL Wetlands

Assuming that the seasonal CH₄ cycle at Churchill is strongly affected by nearby CH₄ sources in the warm months, we made a rough estimation of CH₄ emissions from the HBL. In this estimation, (1) the seasonality of CH₄ emissions from the HBL for May–October was set so as to follow the seasonal variations in biogenic CH₄ sources derived by the one-box model analysis and (2) the annual CH₄ emission strength of the HBL was adjusted to minimize the RMSE between the modeled and observed seasonal CH₄ cycles at Churchill over 2007–2013, based...
Figure 10. Comparison of the observed average seasonal CH4 cycles (black solid lines) with the contributions of the respective regions (cf. Figure 2) estimated by tagged tracer experiments (color lines) at (a) Churchill and (b) Ny-Ålesund. Purple, yellow, green, and red lines represent the contributions from Region 2 (Europe), Region 5 (Western Siberia), Region 11 (Western Canada/Alaska), and Region 14 (Hudson Bay Lowland), respectively. The other regions with minor contributions are shown in gray. Black dotted line in each panel is the average seasonal CH4 cycle calculated using ACTM with the P16pos scenario, which is equivalent to the sum of contributions from Regions 1–17.

annual CH4 emissions as 0.5 ± 0.3 Tg CH4 yr\(^{-1}\) for the HBL (Roulet et al., 1994). The inverse approach based on the atmospheric CH4 observations at Fraserdale and Alert estimated the annual CH4 emissions as 0.2–0.5 Tg CH4 yr\(^{-1}\) (Worthy et al., 2000), similar to the result obtained by Roulet et al. (1994). On the other hand, Pickett-Heaps et al. (2011) calculated CH4 emissions from the HBL as 2.3 Tg CH4 yr\(^{-1}\) using a chemical transport model and surface observations of atmospheric CH4. A process model intercomparison project (The Wetland and Wetland CH4 Intercomparison of Models Project; WETCHIMP) showed CH4 emissions from the HBL at the range of 2.2–11.3 Tg CH4 yr\(^{-1}\) (Melton et al., 2013). Wetland CH4 emissions calculated by VISIT, used as a priori flux to derive the P16pos scenario, yield 5.7 ± 0.5 Tg CH4 yr\(^{-1}\) for the HBL region, which lies near the middle of the nine results from WETCHIMP. Recently, two inversion studies based on atmospheric CH4 data reported the HBL CH4 emissions as 2.4 ± 0.3 Tg CH4 yr\(^{-1}\) (S. M. Miller et al., 2014) and 2.7–3.4 Tg CH4 yr\(^{-1}\) (Thompson et al., 2017), which are lower than the results of most process model studies but close to the estimate by Pickett-Heaps et al. (2011). Our estimate of 2.7 ± 0.3 Tg CH4 yr\(^{-1}\) is also comparable to the results of these top-down studies and to the lower values of WETCHIMP.

As mentioned above, CH4 emissions reduced in the HBL were transferred to the Province of Alberta. By this additional amount of CH4, the annual emissions of 2.6 ± 0.3 Tg CH4 yr\(^{-1}\) allocated by P16pos to Alberta is now increased to 6.9 ± 0.5 Tg CH4 yr\(^{-1}\) in the P16pos_rev scenario. Thompson et al. (2017) estimated the CH4 flux in Alberta to be 5.0–5.8 Tg CH4 yr\(^{-1}\) based on their Bayesian inversion, which is smaller than our estimate by 1.1–1.9 Tg CH4 yr\(^{-1}\). By adopting P16pos_rev instead of P16pos, we found that the CH4 mole fractions observed at two continental tower sites operated by ECCC, Lac La Biche (55°N, 113°W) and East Trout Lake (54°N, 105°W) (http://ds.data.jma.go.jp/gmd/wdcgg/wdcgg.html; also see Figure 1a), are better reproduced by the ACTM forward simulation; the two towers are located in and near the Alberta region, defined as on the forward simulation of ACTM with the emission scenario modified above (“P16pos_rev” in Table 1 and Figure 9). To keep the global CH4 emissions unchanged, the same amount of CH4 as the reduced summertime emission for the HBL was added to the Province of Alberta (Region 12) as nonseasonal CH4 emissions. This method is based on the results of the previous studies that the anthropogenic CH4 emissions in Alberta could be underestimated in EDGAR4.2FT (S. M. Miller et al., 2014; Thompson et al., 2017). As mentioned above, the model simulations with P16pos made for Churchill underestimate the observed CH4 mole fractions in winter. Additional CH4 emissions in Alberta would contribute to improving this discrepancy.

The best agreement between the observed and model-simulated CH4 variations is obtained by reducing the HBL CH4 emissions for May–October to 30%, as an average for 2007–2013, of their original values given by the P16pos. This reduction corresponds to a fall to 39% of the original annual emission given by the P16pos scenario (6.9 ± 0.4 Tg CH4 yr\(^{-1}\)). The result yields 2.7 ± 0.3 Tg CH4 yr\(^{-1}\) as the average HBL CH4 emission for 2007–2013. Note that this emission value includes CH4 released not only from wetlands but also from other sources such as human activities and biomass burning. However, total CH4 emissions from sources other than wetlands could be very small (~0.2 Tg CH4 yr\(^{-1}\)), since the a priori P16pri indicates that wetland CH4 emissions account for 94% of the total in the HBL. With respect to the reduction in CH4 emissions for May–October in our results, the resultant percent value could be an upper limit in the estimation of CH4 emissions from the HBL, since the summertime CH4 mole fractions observed at Churchill are affected not only by the HBL but also by the other regions to some extent.

There still remain large differences in CH4 emissions estimated for the HBL region. For example, the ABLE-3B/NOWES airborne and ground observation campaign, conducted in the summer of 1990, estimated CH4 emissions unchanged, the same amount of CH4 as the reduced summertime emission for the HBL was added to the Province of Alberta (Region 12) as nonseasonal CH4 emissions. This method is based on the results of the previous studies that the anthropogenic CH4 emissions in Alberta could be underestimated in EDGAR4.2FT (S. M. Miller et al., 2014; Thompson et al., 2017). As mentioned above, the model simulations with P16pos made for Churchill underestimate the observed CH4 mole fractions in winter. Additional CH4 emissions in Alberta would contribute to improving this discrepancy.

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50°–60°N and 110°–120°W in this study. It is also seen in Figure 9a that not only the summertime minimum but also the wintertime maximum of the average seasonal CH4 cycle at Churchill is simulated well by P16pos_rev rather than by P16pos. Consequently, our analyses support the results of S. M. Miller et al. (2014) and Thompson et al. (2017) that EDGAR42FT underestimates the anthropogenic CH4 emissions in the Province of Alberta.

4. Summary and Conclusions

We measured the mole fraction, $\delta^{13}C$, and $\delta D$ of atmospheric CH4 at Churchill (58°44′N, 93°49′W) on the northern perimeter of the Hudson Bay Lowlands (HBL), Canada, from a grab sampling method for 2007–2014. Compared to the measurements at Ny-Ålesund, Svalbard (78°55′N, 11°56′E), which is away from regional CH4 sources, the CH4 mole fraction is generally higher and $\delta^{13}C$ and $\delta D$ are lower at Churchill, suggesting CH4 emissions from regional/local boreal wetlands around the site.

The seasonal cycle of CH4 ($\delta^{13}C$) is clearly observable, with the maximum value in January–February (May) and the minimum in June (October). $\delta D$ also shows high values in June and low values in cold months of September to March. The summer minimum (maximum) of the CH4 mole fraction ($\delta^{13}C$ and $\delta D$) appears approximately 2 weeks earlier at Churchill than at Ny-Ålesund. The simple mass balance analysis with the one-box model indicates that the seasonal maximum of biogenic CH4 influence at Churchill precedes the maximum at Ny-Ålesund, contributing to the phase difference of atmospheric CH4, $\delta^{13}C$, and $\delta D$ between the two sites.

Short-term variations in the CH4 mole fraction are observed throughout the year at Churchill, with higher values especially in the summertime. By inspecting the relationship between the short-term variations of the CH4 mole fraction and isotope ratios, $\delta^{13}C$ and $\delta D$ of related CH4 sources are estimated to be respectively $-63.3 \pm 2.8$ and $-267 \pm 26\%$ for the summertime (May–October) and $-47.7 \pm 4.3$ and $-241 \pm 48\%$ for the wintertime (November–April). These values indicate that short-term CH4 variations observed at Churchill are produced mainly by biogenic CH4 emissions from wetland in summer and fossil fuel sources in winter.

To investigate the seasonal cycle of atmospheric CH4 in terms of CH4 sources, we simulated the atmospheric CH4 mole fractions using ACTM with two CH4 emission scenarios and then compared them with the observed results at Churchill and Ny-Ålesund. ACTM overestimates the CH4 mole fraction at Churchill in summer, although the seasonal CH4 cycle at Ny-Ålesund is reproduced well. Tagged tracer experiments indicate that the summertime high CH4 mole fractions at Churchill are mainly caused by the air transported from the HBL. This implies that the wetland CH4 fluxes prescribed for the region in the ACTM simulations are overestimated. By adjusting the CH4 fluxes prescribed for the HBL in ACTM so that the seasonal CH4 cycle observed at Churchill is reproduced well, average CH4 emission from the HBL for 2007–2013 is estimated to be $2.7 \pm 0.3$ Tg CH4 yr$^{-1}$, which is in good agreement with the results of previous modeling studies based on atmospheric CH4 observations.

This study shows that simultaneous and high-precision measurements of the mole fraction, $\delta^{13}C$, and $\delta D$ provide us with valuable information on CH4 sources. It is also shown from the model-observation comparison that systematic observations of the atmospheric CH4 mole fraction in nearby source regions are important for assessing the local/regional CH4 emissions. Inclusion of $\delta^{13}C$ and $\delta D$ into the model analysis would provide additional strong constraints on a better understanding of CH4 sources and sinks. For this purpose, further efforts are needed not only to increase systematic observations of $\delta^{13}C$ and $\delta D$ but also to undertake an extensive intercomparison program of $\delta^{13}C$ and $\delta D$ scales among related institutes.

Acknowledgments

We express our sincere thanks to Lee Ann Fishback, scientific coordinator at the Churchill Northern Studies Centre, for her efforts to air sample collection and Michele Ernst at ECCC for her logistical coordination of the flask program and air sample analyses. We are also grateful to Hiroko Nagamoto, NIPR, and Morimasa Sato, TU, for their support to isotope analyses. This work was partly supported by NIPR through the Project Research Nos. KP-15 and KP-304, the Arctic Challenge for Sustainability (ArCS) project by the Ministry of Education, Culture, Sports, Science and Technology, Japan, and the JSPS KAKENHI grants 23310012 and 15H03722. The $\delta^{13}C$ and $\delta D$ data are available from the website at TU (https://caos.sakura.ne.jp/tgr/data/en/). The CH4 mole fraction data at Churchill, Lac La Biche, and East Trout Lake are available from the WDCGG website (http://ds.data.jma.go.jp/gmd/wdcgg/wdcdg.html).

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