Global and Regional CH₄ Emissions for 1995–2013 Derived From Atmospheric CH₄, δ¹³C-CH₄, and δD-CH₄ Observations and a Chemical Transport Model

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Abstract To better understand the current global CH₄ budget, biogenic, fossil fuel, and biomass burning CH₄ fluxes for the period 1995–2013 were inversely estimated from the observed mole fraction data of atmospheric CH₄ using a three-dimensional chemical transport model. Then, forward simulations of carbon and hydrogen isotope ratios of atmospheric CH₄ (δ¹³C-CH₄ and δD-CH₄) were conducted using the inversion fluxes to evaluate the source proportion of the global total CH₄ emission. Model-simulated spatiotemporal variations of atmospheric CH₄ reproduce the observational results well; however, the simulated δ¹³C-CH₄ and δD-CH₄ values significantly underestimate their observed values as a whole. This implies that the proportion of biogenic CH₄ sources in the global CH₄ emission, deduced by inverse modeling, is overestimated, although the proportion is fairly comparable with the medians of recent multiple CH₄ inverse modeling. To reduce the disagreement between the observed and calculated isotope ratios, the CH₄ fluxes of individual source categories were adjusted using our atmospheric δ¹³C-CH₄ and δD-CH₄ data observed at Arctic and Antarctic surface stations. The resultant global average biogenic, fossil fuel, and biomass burning CH₄ fluxes over 2003–2012 are 346 ± 11, 162 ± 2, and 50 ± 2 TgCH₄ year⁻¹, respectively. It is also strongly suggested that the leveling-off of atmospheric CH₄ in the early 2000s and the renewed growth after 2006/2007 are, respectively, explainable by the decrease in biogenic and biomass burning CH₄ emissions for 2000–2006 and the increase in biogenic CH₄ emissions after that period. These emission changes mainly originate in the tropics.

1. Introduction

CH₄ plays an important role in global climate change and atmospheric chemistry. The global average atmospheric CH₄ mole fraction in 2018 was approximately 1,858 ppb (https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/#global), which is 2.5 times higher than preindustrial levels (Etheridge et al., 1998). Systematic observations of atmospheric CH₄ also revealed an unexpected trend in the last few decades; the rate of increase in CH₄ slowed down during the 1980s and 1990s, leveled off for 1999–2006, and rose again in 2006/2007 (Dlugokencky et al., 2009; Rigby et al., 2008). However, the cause is still being debated (Nisbet et al., 2016, 2019; Patra et al., 2016; Rigby et al., 2017; Schaefer et al., 2016; Thompson et al., 2018; Turner et al., 2017). Since the atmospheric CH₄ burden is determined by the balance between CH₄ sources (e.g., wetlands, termites, geological processes, agriculture, landfills and waste, fossil fuels, and biomass burning) and sinks (e.g., chemical reactions with OH in the troposphere, consumption by soil, and stratospheric losses), which are unevenly distributed on the Earth’s surface, further studies are required for a better understanding of the global CH₄ budget and its changes. These efforts are indispensable for planning effective mitigation measures in CH₄ emissions in the future.

A great deal of effort has been devoted to quantify global and regional CH₄ budgets using both top-down (i.e., inverse modeling using atmospheric measurements) and bottom-up (i.e., direct flux measurement, statistical database, and process-based modeling) approaches (Kirschke et al., 2013; Saunois et al., 2016, 2017). Despite the considerable progress achieved recently, a large discrepancy remains between the results obtained using the two approaches. Top-down estimation on the global scale can reconcile the entire CH₄ budget by...
constraining with the atmospheric CH₄ mole fraction data, but additional information besides the CH₄ mole fraction, for example, prior distributions, isotopes and tracers, is always required to estimate its sources and sinks. Bottom-up estimation can also be inaccurate due to insufficient spatiotemporal representativeness of CH₄ flux measurements, the uncertainty in activity data and emission factors adopted for statistical databases, and the uncertainty in process-based modeling.

Carbon and hydrogen isotope ratios (δ¹³C-CH₄ and δD-CH₄) of CH₄ are useful for evaluating contributions of individual CH₄ sources or sinks to atmospheric CH₄, since each source or destruction process has its own characteristic isotopic signatures. δ¹³C-CH₄ and δD-CH₄ are commonly defined using a deviation of the sample mole ratio (i.e., ¹³CH₄/¹²CH₄ or CH₃D/CH₄) relative to the given standard ratio:

\[ \delta^{13}C = \frac{\left( \frac{^{13}C}{^{12}C} \right)_{\text{sample}} - 1}{\left( \frac{^{13}C}{^{12}C} \right)_{\text{VPDB}}} \times 1,000 \quad (\%e), \]

\[ \delta D = \frac{\left( \frac{D}{^2H} \right)_{\text{sample}} - 1}{\left( \frac{D}{^2H} \right)_{\text{VSMOW}}} \times 1,000 \quad (\%e), \]

where subscript “sample” denotes the sample and “VPDB” (Vienna Pee Dee belemnite) represents the international standard for δ¹³C-CH₄, with ¹³C/¹²C of 1,123.72 × 10⁻⁶ (Craig, 1957), and “VSMOW” (Vienna standard mean ocean water) for δD-CH₄, having a D/H of 155.76 × 10⁻⁶ (Hagemann et al., 1970).

CH₄ is known to be released through three major processes: biogenic, thermogenic (mainly fossil fuel), and pyrogenic (mainly biomass burning), their isotope ratios being different on average from each other (Sherwood et al., 2017). The δ¹³C-CH₄ values of biogenic and thermogenic CH₄ sources are partly overlapped, but simultaneous analysis of δ¹³C-CH₄ and δD-CH₄ enables us to separate the respective source contributions to atmospheric CH₄ (e.g., Fujita et al., 2018). On the other hand, since lighter isotopes (e.g., ¹³CH₄) are destroyed faster than heavier ones (e.g., ¹²CH₄ and CH₃D), which is called the kinetic isotope effect (KIE) (Saueressig et al., 2001), atmospheric CH₄ becomes isotopically heavier, of which the degree depends on the destruction process. Therefore, for elucidating the global CH₄ cycle, it is useful to perform systematic and high-precision isotope observations at various places around the world and then analyze their results using three-dimensional (3-D) atmospheric chemical transport models. However, the number of the isotope observations is still limited mainly because of technical difficulties in sample analysis, leading to a limited number of modeling studies on δ¹³C-CH₄ (Bousquet et al., 2006; Hein et al., 1997; Monteil et al., 2011; Thompson et al., 2018) and on both δ¹³C-CH₄ and δD-CH₄ (Rice et al., 2016; Warwick et al., 2016).

Recent δ¹³C-CH₄ observations by the Institute of Arctic and Alpine Research, University of Colorado (INSTAAR) and the National Institute of Water and Atmosphere (NIWA) clearly indicated that the global average δ¹³C-CH₄ decreased secularly in step with the regrowth of CH₄ since 2006/2007 (Nisbet et al., 2016; Schaefer et al., 2016; White et al., 2018). This suggests that post-2007 CH₄ growth can be ascribed to enhanced emissions of biogenic CH₄ having lighter isotopes, as was supported by other independent δ¹³C-CH₄ observations (e.g., Morimoto et al., 2017). However, this is inconsistent with an enhancement of fossil fuel emissions that was suggested by the bottom-up emission inventory (EDGAR4.2FT, 2013) and ethane-based approach (Hausmann et al., 2016; Helmig et al., 2016), although more recent ethane-based study pointed out that the results by the previous studies are possibly overestimated (Lan et al., 2019). It was also recently reported that such a conclusion deduced from the atmospheric δ¹³C-CH₄ observations could be inconclusive, since the isotopic signatures of each source are widely ranged (Turner et al., 2017), and the role of OH should be considered (Rigby et al., 2017). Moreover, since previous isotope studies analyzed mainly δ¹³C-CH₄, more detailed analyses using 3-D chemical transport models, including δD-CH₄ data, are required.

In this study, we first perform the inverse modeling of the global atmospheric CH₄ mole fraction observations to estimate global and regional surface CH₄ fluxes for the period 1995–2013 using a 3-D atmospheric chemistry transport model with a priori CH₄ emissions and sink fields. The CH₄ fluxes are estimated...
separately by major three source categories of biogenic, fossil fuel, and biomass burning. After obtaining a posteriori \( \text{CH}_4 \) fluxes, we conduct forward simulations of atmospheric \( \delta^{13}\text{C-CH}_4 \), \( \delta^2\text{H-CH}_4 \), and \( \text{CH}_4 \) mole fraction to evaluate the respective source proportions to the global total \( \text{CH}_4 \) emissions. The calculated \( \delta^{13}\text{C-CH}_4 \) and \( \delta^2\text{H-CH}_4 \) are compared to the data observed by Tohoku University (TU) and the National Institute for Polar Research (NIPR) at two polar surface stations, Ny-Ålesund, Svalbard (NYA: 78°55'N, 11°56'E) and Syowa Station, Antarctica (SYO: 69°00'S, 39°35'E), one of the longest \( \text{CH}_4 \) isotope records obtained by the same group in both polar regions, and in the western Pacific region (NWP: 33°N–40°S, 115°E–180°E) between Japan and Australia/New Zealand. Then, the best-estimates of global and regional \( \text{CH}_4 \) emissions for individual source categories are derived for 1995–2013. By examining the estimates of OH trends by Turner et al. (2017) in our forward simulations, the role of OH in recent \( \text{CH}_4 \) growth is also investigated.

### 2. Method

#### 2.1. Model Setting for Forward Simulation and Inverse Modeling

We use the National Institute for Environmental Studies/3-D chemical Transport Model (NIES-TM) with a horizontal resolution of 2.5° × 2.5° and 32 hybrid sigma-isentropic vertical coordinates (Belikov et al., 2013) to estimate monthly regional \( \text{CH}_4 \) fluxes and to simulate the mole fractions of total \( \text{CH}_4 \) and carbon and hydrogen isotopes (\( ^3\text{C-CH}_4 \) and \( ^2\text{H-CH}_4 \)) in the atmosphere. The model is used to prepare the Level 4 (L4) \( \text{CH}_4 \) data products of the Greenhouse gases Observing SATellite (GOSAT), such as global surface \( \text{CH}_4 \) fluxes and 3-D global \( \text{CH}_4 \) distributions (NIES GOSAT Project, 2016). The model also participated in the recent Global Carbon Project synthesis (Saunois et al., 2016, 2017) by submitting the global inversion \( \text{CH}_4 \) fluxes constrained by surface \( \text{CH}_4 \) observations and GOSAT satellite data for 2009–2013. In this study, we performed the inversion analysis for a longer period of time from 1995 to 2013 using the surface \( \text{CH}_4 \) observations.

The horizontal and vertical transport of the model was driven by 6-hr meteorological fields of the Japanese 25-year ReAnalysis (JRA-25) and Japan Meteorological Agency Climate Data Assimilation System for 1995–2013 (Onogi et al., 2007). To separate the well-mixed near-surface layer and the free troposphere, 3-hr planetary boundary-layer height data were taken from the ERA-Interim reanalysis (Dee et al., 2011). To calculate the vertical transport in the stratosphere above the 350-K level in the isentropic coordinate, two-dimensional (2-D) monthly distributions of the climatological heating rates were prepared based on the JRA-25. Atmospheric \( \text{CH}_4 \) was destroyed through chemical reactions with OH, Cl, and O(\( ^3\text{D} \)). Monthly mean global OH fields and the temperature-dependent reaction rates were taken from the TransCom-CH4 intercomparison experiment (Patra et al., 2011). The OH fields were given after tuning those of Spivakovsky et al. (2000) with the observed atmospheric \( ^3\text{C-CH}_3\text{Cl}_3 \) data. The stratospheric reaction rates of \( \text{CH}_4 \) with Cl and O(\( ^3\text{D} \)) were also taken from the TransCom-CH4 data. Note that the monthly mean OH, Cl, and O(\( ^3\text{D} \)) fields used in the model are climatological with no interannual variability, but the sensitivity test was carried out for the interannual OH variability (see section 4). The \( \text{CH}_4 \) consumption by bacteria in soils, given by a process-based terrestrial ecosystem model, VISIT (Ito & Inatomi, 2012), was treated as negative emissions in the model. The \( \text{CH}_4 \) sink processes incorporated into the NIES-TM are summarized in Table 1 (details are presented in Table S1 in the supporting information).

### Table 1

| Sink process               | Data source          | Lifetime (year) | KIE\(^{13}\text{C} \) | KIE\(^{2}\text{H} \) | Time resolution                  |
|----------------------------|----------------------|-----------------|------------------------|------------------------|----------------------------------|
| \( \text{CH}_4 + \text{OH} \) | TransCom-\( \text{CH}_4 \) | 10.0            | 1.0065 \(^a,b \)      | 1.286 \(^b \)         | Monthly, no IAV                   |
| \( \text{CH}_4 + \text{Cl} \) | TransCom-\( \text{CH}_4 \) | 226             | 1.066 \(^b \)         | 1.520 \(^b \)         | Monthly, no IAV                   |
| \( \text{CH}_4 + \text{O} (\^3\text{D}) \) |           | 1.013 \(^b \)  | 1.06 \(^b \)          |                       | Monthly, no IAV                   |
| Soil Oxidation             | VISIT                | 151             | 1.0201 \(^d,e,f \)    | 1.083 \(^d \)         | Monthly, IAV                      |
| Total                      |                      | 9.0             | 1.0065                 | 1.276                  |                                  |

Note: The values of KIEs for the respective sink processes are given at \( T = 296 \text{ K} \). Temperature dependencies of the KIEs are accounted for in the model simulations (see Table S1 for details). The averaged KIE values for \( \text{CH}_4 + \text{Cl} \) and \( \text{CH}_4 + \text{O} (\^3\text{D}) \) are used in the simulations, because only their combined reaction rates were obtained from the TransCom-\( \text{CH}_4 \) (Patra et al., 2011). The “IAV” represents interannual variability.

\(^a\)Cantrell et al. (1990). \(^b\)Saueressig et al. (2001). \(^c\)Saueressig et al. (1995). \(^d\)Snover & Quay (2000). \(^e\)Tyler et al. (1994). \(^f\)Reeburgh et al. (1997). \(^g\)Gierczak et al. (1997). \(^h\)Saueressig et al. (1996).
A priori CH₄ emissions incorporated into the NIES-TM for individual sources are summarized in Table 2. Annual CH₄ emissions from fossil fuel (e.g., coal, natural gas, and residential) sources were obtained from EDGARv4.2 (http://edgar.jrc.ec.europa.eu/overview.php?v=42) for 1995–2008 and EDGARv4.2 FT (http://edgar.jrc.ec.europa.eu/overview.php?v=42FT2010) for 2009–2010, and the values of EDGARv4.2 FT in 2010 were used repeatedly after 2011. Biomass-burning CH₄ emissions for 1996–2011 were taken from GFEDv3.1 (van der Werf et al., 2010), and those for 2012–2013 were estimated using burned area by GFEDv4.0 (Giglio et al., 2013) and the relationship between the burned area and CH₄ emission in GFEDv3.1. Since no data are available from GFEDv3.1 for January–May 1995, the emission data for January–May 1996 were used as a substitute for that period. Biogenic CH₄ emissions from wetlands and rice paddies were calculated using the VISIT (Ito & Inatomi, 2012). The CH₄ emissions from biomass burning, wetlands, and rice paddies are seasonally and interannually variable. Annual biogenic CH₄ emissions from other anthropogenic activities (e.g., ruminants and landfills) were also obtained from EDGARv4.2 and EDGARv4.2 FT for 1995–2013. CH₄ emissions from termites were taken from GISS inventory (Fung et al., 1991).

In the inverse modeling, monthly surface CH₄ fluxes (s) are estimated by optimizing the correspondence between the observed and calculated CH₄ mole fractions, based on the prior flux estimates (sₚrior) and the Bayesian inference (Kim et al., 2011; Saeki et al., 2013). A cost function, J(s), is expressed as

\[ J(s) = \frac{1}{2}(z-Hs)^T R^{-1} (z-Hs) + \frac{1}{2}(s-s_p)Q^{-1}(s-s_p), \]  

(3)

where z is the vector of atmospheric CH₄ observation, H is the matrix of the responses at each observation to pulse emissions of the prior monthly surface CH₄ fluxes, R is the error covariance matrix of the observations, and Q is the error covariance matrix associated with the prior flux estimates. The optimized (a posteriori) CH₄ fluxes, s’, are obtained by minimizing J(s) as

\[ s' = s_p + (H^T R^{-1} H + Q^{-1})^{-1} H^T R^{-1} (z - Hs_p). \]  

(4)

The error covariance matrix associated with s’ is given by

\[ Q' = (H^T R^{-1} H + Q^{-1})^{-1}. \]  

(5)

The size of the vector, s, for each month is given by the product of three CH₄ source categories (i.e., fossil fuel, biomass burning, and biogenic; see Table 2) and 43 source regions (i.e., 42 land regions and the global ocean; see Figure 1). Uncertainties of a priori CH₄ fluxes were set to 20% for fossil-fuel sources, 50% for biomass-burning and biogenic sources, and 20% for the global ocean, following the respective percentage values adopted for anthropogenic sources, biomass-burning and natural sources, and global ocean in GOSAT L4 data processing (NIES GOSAT Project, 2016). Initial 3-D atmospheric distribution of total
CH4 for the inverse modeling was taken from NIES GOSAT Project (2016) after adjusting it to coincide with the observed CH4 value at SYO in January 1995. To reduce computational burden of flux optimization using a large number of observations and the responses to pulse emissions, a fixed-lag Kalman smoother scheme was employed (Bruhwiler et al., 2005). In this study, a 4-month lag window was applied to the inverse modeling, assuming that the signals of pulse emissions from each source region to atmospheric observations decay within 4–6 months and are then mixed into the background. Here we call the a priori and a posteriori CH4 fluxes of the present inverse modeling “Prior” and “PosCH4.”

The mole fractions of atmospheric 13CH4 and CH3D were calculated based on the respective CH4 sinks, KIEs, CH4 emissions, and isotopic signatures (Tables 1 and 2) and were then converted to δ13C-CH4 and δD-CH4 using Equations 1 and 2, respectively. In this calculation, 12CH4 and CH4 were assumed to be equal to the respective residuals obtained by subtracting 13CH4 and CH3D from total CH4 (Tans, 1997). The initial 3-D 13CH4 and CH3D distributions were first roughly prepared using the δ13C-CH4 and δD-CH4 observations at SYO and high-altitude aircraft observations (Rice et al., 2003), and then 36-year spin-up run was performed for total CH4, 13CH4, and CH3D, by which year-to-year changes of δ13C-CH4 and δD-CH4 became negligibly small. The atmospheric CH4, 13CH4 and CH3D distributions at the last time step of the spin-up run were adjusted to correspond to January 1995 by multiplying the respective ratios of the observed CH4, 13CH4, and CH3D at SYO in January 1995 to the calculated values at the last time step. A detailed description of the spin-up method is given in Text S1 in the supporting information.

2.2. Adjustment of Inversion CH4 Fluxes Based on Isotope Observations

Surface CH4 fluxes estimated inversely using the atmospheric CH4 mole fraction data for biogenic, fossil fuel, and biomass burning sources are dependent on the prior source information. Therefore, to find the plausible source proportions, we performed forward simulations of atmospheric δ13C-CH4 and δD-CH4 using the PosCH4 scenario and then minimized their model-observation mismatch by modifying the fractions of the three source categories in the PosCH4 without changing the global total CH4 emission, using a simple mass-balance equation:

\[
E_{\text{PosCH4(tot)}} = E_{\text{PosCH4(ff)}} + E_{\text{PosCH4(bb)}} + E_{\text{PosCH4(bio)}} = f_ff E_{\text{PosCH4(ff)}} + f_bb E_{\text{PosCH4(bb)}} + f_bio E_{\text{PosCH4(bio)}}. \tag{6}
\]

where \(E_{\text{PosCH4}}\) represents the CH4 emissions for fossil fuel (ff), biomass burning (bb), biogenic (bio), and total (tot), and \(f\) means the scaling factors which adjust the CH4 emissions of the respective source.
categories. It can be seen from Equation 6 that if two scaling factors are determined, the remaining one factor can be calculated. Considering that $f_{gg}$ and $f_{bb}$ can be assumed to be larger than 1 and $f_{bb}$ to be smaller than 1 (see section 3.2), we adjusted $f_{gg}$ and $f_{bb}$ independently and also determined $f_{bb}$ by Equation 6. Since atmospheric $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$ observations are spatiotemporally sparse for our analysis period of 1995–2013, it is difficult to constrain surface CH$_4$ fluxes for every 43 regions (Figure 1). Thus, the scaling factors were calculated based on the global budget equation (Equation 6). In this study, the scaling factors were determined so that the simulated $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$ matched our long-term observations at NYA and SYO. The influence caused by using only the two stations will be examined in section 3.3 based on other independent $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$ data.

We first examined how the simulated $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$ at NYA and SYO change with different combinations of $f_{gg}$ ($f_{gg} = 1.0, 1.5, 2.0$) and $f_{bb}$ ($f_{bb} = 1.0, 2.0, 3.0$) during 1995–2012 (Figure S2). The simulation and comparison were made every 3 years interval at the first stage to reduce computational costs. We looked for the best combination of $f_{gg}$ and $f_{bb}$ that reproduce the observed $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$ fairly well for each interval at NYA and SYO (Text S2 and Figures S3 and S4 for more details) and regarded the combinations as a “base_iso” scenario. The combinations of $f_{gg}$ and $f_{bb}$ derived from the NYA and SYO data were generally similar to each other, but a difference between the results for the two sites was seen in some periods. In such case, the combination derived using SYO data were selected. It is because that the influence of regional/local CH$_4$ sources is less at SYO than at NYA.

By replacing the PosCH4 scenario by the base_iso scenario, we found that the disagreement between the simulated and observed $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$ is greatly improved over the whole period, keeping a good agreement for CH$_4$, but interannual variations of atmospheric $\delta^{13}$C-CH$_4$ were still insufficiently reproduced at both NYA and SYO. To evaluate the year-to-year change of $f_{gg}$ and $f_{bb}$ in the base_iso scenario, as a second stage, we further simulated $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$ at NYA and SYO using a number of combinations for $f_{gg}$ and $f_{bb}$ with small changes from the base_iso scenario and chose the combination which well reproduce the observations. In this adjustment, to avoid unrealistic flux adjustments, the maximum values of $f_{gg}$ and $f_{bb}$ were set to 1.5 and 3.5, respectively, which were roughly equivalent to the upper bounds of CH$_4$ emission estimates for the respective sources in previous top-down studies (Saunois et al., 2016; Schwietzke et al., 2016) when the scaling factors were applied to the PosCH4 scenario. Under these assumptions, we finally derived a plausible set of $f_{gg}$, $f_{bb}$, and then $f_{bio}$ for each year to improve the model-observation mismatch of $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$. The result will be presented and used in section 3.2.

### 2.3. Atmospheric CH$_4$ Mole Fraction, $\delta^{13}$C-CH$_4$, and $\delta$D-CH$_4$ Data Used in Model Studies

#### 2.3.1. CH$_4$ Mole Fraction Data

For the inverse modeling, the CH$_4$ mole fraction data obtained from discrete air sampling and continuous measurements for the period 1995–2013 were taken from the World Data Center for Greenhouse Gases (WDCGG) (https://gaw.kishou.go.jp/) and the NIES/Center for Global Environmental Research (CGER) Joint Japan-Russia project for greenhouse-gas monitoring in Siberia (Sasakawa et al., 2010, 2017) (see Figure 1 and Table S2). The WDCGG data used in this study were collected mostly by the NOAA/ESRL/GMD (Dlugokencky et al., 2019) and partly by other institutes (see Acknowledgments and Data Availability Statement and Table S2), their CH$_4$ mole fraction values being expressed in the WMO-X2004A scale (https://www.esrl.noaa.gov/gmd/ccgg/globalview/ch4_intro.html). The NIES/CGER CH$_4$ data are expressed by the NIES-94 scale (Sasakawa et al., 2010, 2017). The fifth and sixth WMO Round-Robin intercomparison programs showed that the NIES-94 scale is higher by $3.9 \pm 1.0$ ppb on average than the WMO-X2004A scale. Therefore, the NIES data were shifted down by 3.9 ppb to convert to the WMO-X2004A scale. The observation sites used for the inverse modeling were the same as those used for processing the GOSAT L4 products and the Global Carbon Project synthesis results (NIES GOSAT Project, 2016; Saunois et al., 2016, 2017). For the surface continuous observations, we selected daytime data (15:00 local standard time) at inland sites, nighttime data (03:00 local standard time) at mountain sites, and daytime and nighttime data at remote island/coastal boundary layer sites when the atmosphere was well mixed. The data uncertainty for each observation site was given by the mean residual standard deviation from GLOBALVIEW-CH4 (GV-CH4) (https://www.esrl.noaa.gov/gmd/ccgg/globalview/ch4/ch4_intro.html) (see Table S2). To avoid overconstraint on particular observation sites with small uncertainty, the minimum data uncertainties were enforced to be larger than 6 and 20 ppb for discrete and continuous
measurements, respectively (Kim et al., 2011). For the observation data having large differences of over 60 ppb from the model, a large uncertainty (i.e., $10^5$ ppb) was assigned to them to avoid excess adjustment in the inverse modeling.

### 2.3.2. $\delta^{13}$C-CH$_4$ and $\delta^D$-CH$_4$ Data

In addition to the abovementioned CH$_4$ data, we also used our CH$_4$, $\delta^{13}$C-CH$_4$, and $\delta^D$-CH$_4$ measurements at two northern and southern surface baseline stations, NYA and SYO and aboard commercial container ships in the western Pacific, NWP, between Japan and Australia/New Zealand to verify individual source proportions to obtained inversion fluxes. The data at NYA were taken from Morimoto et al. (2017) and Fujita et al. (2018), and the data at NWP were obtained by updating those given in Umezawa et al. (2012), whereas the $\delta^{13}$C-CH$_4$ and $\delta^D$-CH$_4$ data at SYO were used for the first time in this paper. Since our measurements at these sites have already been described in detail elsewhere, a brief explanation is presented here.

Systematic observations of the CH$_4$ mole fraction, $\delta^{13}$C-CH$_4$, and $\delta^D$-CH$_4$ at NYA have been conducted once a week by a grab-sampling method since 1991, 1996, and 2005, respectively (Fujita et al., 2018; Morimoto et al., 2017). The CH$_4$ mole fraction was determined at NIPR using a gas chromatograph with flame ionization detector (GC-FID; Shimadzu GC-8A) relative to the TU1987 scale (Aoki et al., 1992), the difference between the WMO-X2004A scale being 0.5 ppb (Fujita et al., 2018). $\delta^{13}$C-CH$_4$ was also measured at NIPR using a gas chromatograph-combustion isotope ratio mass spectrometer (GC-C-IRMS) based on MAT-252 (Thermo Fischer) with a repeatability of 0.07‰ (Morimoto et al., 2017). $\delta^D$-CH$_4$ was obtained at TU by using a gas chromatograph-pyrolysis isotope ratio mass spectrometer based on Delta Plus XP (Thermo Fischer) with a repeatability of 2.2‰ (Umezawa et al., 2009).

Continuous measurements of atmospheric CH$_4$ have been carried out at SYO since 1988 using the GC-FID (Aoki et al., 1992). Discrete air sampling has also been performed at SYO using a 550-ml Pyrex glass flask once per week since 1983 (Tanaka et al., 1987) while using a 9 L aluminum cylinder for archiving the air once a month or every 2 months since 1995 (hereafter called archive air). $\delta^{13}$C-CH$_4$ has been measured at NIPR for the flask samples and archive air since 2000 and 1995, respectively. $\delta^D$-CH$_4$ has been analyzed only for archive air at TU since 1995.

The samples in the western Pacific region (NWP) were collected into 550 ml Pyrex glass flasks at every 5° latitude interval during a round trip of approximately 30–40 days (Nakazawa, Morimoto, et al., 1997; Umezawa et al., 2012). CH$_4$ mole fractions have been determined using the GC-FID (Shimadzu GC9A and Agilent 6890) at TU relative to the TU1987 scale since 1987. $\delta^{13}$C-CH$_4$ has been measured at TU using the GC-C-IRMS with repeatability of 0.07‰ since 2006. $\delta^D$-CH$_4$ has also been determined at TU since 2006. Because we were required to perform time-consuming isotope analyses in a limited time, only 12 air samples were cautiously selected out of all samples collected on each round-trip cruise so that they would be evenly distributed across the entire sampling latitude band. Note that our $\delta^{13}$C-CH$_4$ data were updated from the earlier publications (Umezawa et al., 2009, 2012) by introducing our new data correction method at TU, that the CH$_4$ observations at NWP were not used for the CH$_4$ inversion, and that the NOAA/ESRL/GMD CH$_4$ data at NYA and SYO were used in the inversion (see Table S2).

Intercomparison of $\delta^{13}$C-CH$_4$ measurements between TU/NIPR and the Institute for Marine and Atmospheric research Utrecht (IMAU) was conducted during 2013–2015, from which the difference from IMAU is found to be $+0.40 \pm 0.04$‰ for TU and $+0.31 \pm 0.03$‰ for NIPR (Umezawa et al., 2018). The difference between the results of TU and NIPR is within their analytical uncertainties. The intercomparison between NIPR and NIWA was also conducted in 2004. The result showed that the difference of NIPR from NIWA is $+0.33 \pm 0.04$‰, which is consistent with the combined results between NIPR and IMAU and between IMAU and NIWA in the intercomparison afterward (Morimoto et al., 2017; Umezawa et al., 2018). In this connection, the comparison of $\delta^D$-CH$_4$ measurements between TU and IMAU was also carried out during 2013–2015, showing that our scale is lower by $13.1 \pm 0.6$‰ than theirs at ambient air levels (Umezawa et al., 2018).

To validate the global and regional inversion CH$_4$ fluxes, we also used NOAA/INSTAAR $\delta^{13}$C-CH$_4$ and $\delta^D$-CH$_4$ data (White et al., 2016, 2018) and NIWA $\delta^{13}$C-CH$_4$ data (Lowe et al., 1991, 1994), in addition to the TU/NIPR $\delta^{13}$C-CH$_4$ and $\delta^D$-CH$_4$ data (Fujita et al., 2018). The differences between $\delta^{13}$C-CH$_4$ measurements at NOAA/INSTAAR and TU/NIPR were estimated to be $-0.20 \pm 0.04$‰ in Umezawa et al. (2018), so that the
NOAA/INSTAAR $\delta^{13}$C-CH$_4$ data were shifted by this amount to express in the TU/NIPR scale. Since the differences of $\delta^D$-CH$_4$ measurements between the two groups were reported to be 0.1‰ (Umezawa et al., 2018), no correction was applied to the $\delta^D$-CH$_4$ data. The NIWA $\delta^{13}$C-CH$_4$ data were adjusted by 0.33‰ in accordance with Morimoto et al. (2017) to convert to the TU/NIPR scale values.

3. Results

3.1. Global CH$_4$ Emissions Optimized by Inverse Modeling With the Observed CH$_4$ Mole Fractions

Figure 2 shows the atmospheric CH$_4$ mole fractions at Ny-Ålesund (NYA), Syowa Station (SYO), and selected latitudes in the western Pacific between Japan and Australia/New Zealand (NWP) for the period 1995–2013. Gray and red lines represent the CH$_4$ mole fractions calculated using the a priori (Prior) and posteriori (PosCH4) CH$_4$ emissions, respectively. Thick solid black, gray, and red lines represent the respective long-term trends of the observed and simulated data, obtained using the digital-filtering technique with cutoff periods of 60 months (Nakazawa, Ishizawa, et al., 1997). Also shown are the growth rates (dotted lines against the right axis). For reference, globally averaged marine surface annual mean growth rate obtained by NOAA/ESRL (https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/) are also presented by dotted light blue lines.

Figure 2. Observed (open circles) and NIES-TM simulated CH$_4$ mole fractions (thin solid gray and red lines) at Ny-Ålesund (NYA), Syowa Station (SYO), and selected latitudes in the western Pacific between Japan and Australia/New Zealand (NWP) for the period 1995–2013. Gray and red lines represent the CH$_4$ mole fractions calculated using the a priori (Prior) and posteriori (PosCH4) CH$_4$ emissions, respectively. Thick solid black, gray, and red lines represent the respective long-term trends of the observed and simulated data, obtained using the digital-filtering technique with cutoff periods of 60 months (Nakazawa, Ishizawa, et al., 1997). Also shown are the growth rates (dotted lines against the right axis). For reference, globally averaged marine surface annual mean growth rate obtained by NOAA/ESRL (https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/) are also presented by dotted light blue lines.

NOAA/INSTAAR $\delta^{13}$C-CH$_4$ data were shifted by this amount to express in the TU/NIPR scale. Since the differences of 5D-CH$_4$ measurements between the two groups were reported to be 0.1‰ (Umezawa et al., 2018), no correction was applied to the 5D-CH$_4$ data. The NIWA $\delta^{13}$C-CH$_4$ data were adjusted by 0.33‰ in accordance with Morimoto et al. (2017) to convert to the TU/NIPR scale values.

3. Results

3.1. Global CH$_4$ Emissions Optimized by Inverse Modeling With the Observed CH$_4$ Mole Fractions

Figure 2 shows the atmospheric CH$_4$ mole fractions at NYA, NWP, and SYO obtained from forward simulations by the NIES-TM with the Prior and PosCH4 scenarios with the observed values for 1995–2013. The results calculated for the WDCGG and NIES Siberian sites, of which observed data were used for the inverse modeling, are also presented in Figure S1. As seen from these figures, general features of the observed CH$_4$ variations are reproduced very well by the model simulations with PosCH4 but poorly by simulations with Prior. The observational fact that the global atmospheric CH$_4$ increase slowed down during 1995–2006 and rose again after 2006/2007 is clearly reproduced by PosCH4 even in the western Pacific (NWP) where the CH$_4$ data were not used for the inverse modeling. This suggests that interannually variable CH$_4$ fluxes are properly deduced by inverse modeling. However, high CH$_4$ mole fractions, observed frequently at several continental sites (e.g., IGR, DEM, and KRS in Siberia; see Figure S1), cannot be simulated well, even with Prior or PosCH4. The cause could be partly ascribed to imperfect incorporation of winter-time boundary-layer mixing into the NIES-TM, visible over high latitude continents during winter (Saeki et al., 2013).
The annual mean global CH$_4$ fluxes derived for 1996–2012 are shown in Figure 3a. To avoid the spin-up and spin-down influences, the results for the first and last 4 months of 1995–2013 have been removed. As seen from the figure, the global total flux (TOT) is lower in Prior than in PosCH4 for the entire period, especially until the early 2000s. The global average posterior emissions over the period of 2003–2012 are 413 ± 9, 127 ± 9, 19 ± 2, and 558 ± 14 TgCH$_4$ year$^{-1}$ (average ± standard deviation) for biogenic (BIO), fossil fuel (FF), biomass burning (BB), and TOT, respectively (Figure 3c). The results are generally comparable with the mean values estimated by multiple top-down studies for the same period (Saunois et al., 2016). Anomalies of global TOT CH$_4$ fluxes, derived by subtracting the average CH$_4$ emission over 1996–2012 from deseasonalized monthly emission values, are presented in Figure 3b. The TOT emissions of PosCH4 show large interannual variations of up to ±20 TgCH$_4$ year$^{-1}$ with positive anomalies in 1998 and after 2006/2007 and negative anomalies in 1999–2002 and 2004–2005, superimposed on a positive linear trend of +1.6 ± 0.3 TgCH$_4$ year$^{-2}$ (±95% confidence interval, CI) over the entire period. No significant trend of the TOT emission is found for 2000–2005 and 2008–2012, but a sudden positive emission shift of 22 TgCH$_4$ year$^{-1}$ is observed between 2006 and 2007. Such a behavior is similar to the results reported by Saunois et al. (2017), which show that the global CH$_4$ emissions increase by +2.2 ± 0.2 TgCH$_4$ year$^{-1}$ for 2000–2012 and 22 [16–32] TgCH$_4$ year$^{-1}$ between 2002–2006 and 2008–2012.

3.2. Comparison of Observed and Simulated $\delta^{13}$C-CH$_4$ and $\delta^{13}$D-CH$_4$ in the Atmosphere

Figure 4 shows temporal variations of the atmospheric CH$_4$ mole fraction, $\delta^{13}$C-CH$_4$, and $\delta^{13}$D-CH$_4$ observed at NYA and SYO. The secular trends of $\delta^{13}$C-CH$_4$ and $\delta^{13}$D-CH$_4$ are clearly seen at NYA and SYO. $\delta^{13}$C-CH$_4$ at the two sites shows a slight but monotonous increase from the mid-1990s to 2006/2007 and a distinct decrease after 2006/2007. 5D-CH$_4$ at SYO also shows a secular increase from 1995 to around 2006, followed by a plateau between 2006 and 2013. Such temporal variations of $\delta^{13}$C-CH$_4$ and 5D-CH$_4$ are closely related to the observed atmospheric CH$_4$ trends, which globally leveled off during 2000–2006 and rose again after 2006/2007. The data record of 5D-CH$_4$ at NYA is shorter compared to other records, showing a slight decrease until around 2009, followed by a plateau until 2013. The average rate of increase in $\delta^{13}$C-CH$_4$ at SYO is calculated to be 0.014 ± 0.07 ‰ year$^{-1}$ (±95% CI) for 2000.6–2006.7 (expressed in decimal year) and −0.021 ± 0.03 ‰ year$^{-1}$ for 2006.7–2014.0 (see Table S3 for details). The $\delta^{13}$C-CH$_4$ trend is generally consistent with those deduced from the data at NYA (Morimoto et al., 2017) and globally averaged surface data by NOAA/INSTAAR (Nisbet et al., 2019) but slightly different from that by NIWA, which shows a stabilization rather than an increase for 2000–2006 (Schaefer et al., 2016). The increase rates of 5D-CH$_4$ at SYO are 0.39 ± 0.07 ‰ year$^{-1}$ for 1995.0–2000.6, 0.82 ± 0.06 ‰ year$^{-1}$ for 2000.6–2006.7, and 0.19 ± 0.05 ‰ year$^{-1}$ for 2006.7–2014.0, whereas the rate at NYA is calculated to be −0.14 ± 0.08 ‰ year$^{-1}$ for 2005.8–2014.0. The 5D-CH$_4$ trend at NYA is slightly different from that at SYO, especially before 2009 when NYA shows a clear decrease (see Figures 4c and 4f). Rice et al. (2016) also reported, by analyzing their composite northern midlatitude 5D-CH$_4$ data set, that 5D-CH$_4$ increased clearly
from the 1980s to the early 2000s and then decreased slightly until 2006 (the end of their data period). Therefore, the difference in the $\delta^{13}C$-CH$_4$ trends at SYO and NYA before 2009 could be observed as a result of the hemispherically different behavior of $\delta^{13}C$-CH$_4$.

The results of forward simulations with the PosCH4 scenario for atmospheric CH$_4$, $\delta^{13}C$-CH$_4$, and $\delta D$-CH$_4$ at NYA and SYO are shown in Figure 5, together with their observed values. The observed CH$_4$ mole fractions are reproduced fairly well by the simulation at both sites, but the simulated $\delta^{13}C$-CH$_4$ and $\delta D$-CH$_4$ largely underestimate the observed values, showing a monotonous decrease from 1995 to the early 2000s, followed by a gradual decrease of $\delta^{13}C$-CH$_4$ and a stabilization of $\delta D$-CH$_4$ until 2013. The decrease of $\delta^{13}C$-CH$_4$ and $\delta D$-CH$_4$ since 1995 could be produced by BIO rather than FF and BB, since its mean isotopic signatures ($\delta^{13}C$-CH$_4 = \sim -62\%$ and $\delta D$-CH$_4 = \sim -330\%$) are lower than the source-weighted averages of TOT in equilibrium ($\delta^{13}C$-CH$_4 = \sim -54\%$ and $\delta D$-CH$_4 = \sim -290\%$) which are roughly estimated from the observed atmospheric isotope ratios and KIEs (Sherwood et al., 2017). Since BIO accounts for a major part of the Prior emissions (Figure 3a), the PosCH4 emissions obtained by the inverse modeling could also overestimate the fraction of BIO and underestimate those of FF and BB.

The best combination of $f_{\text{pos}}$, $f_{\text{pos}}$, and $f_{\text{pos}}$ derived by the procedures mentioned in section 2.2 for each year is shown in Figure 6. Based on this result, we calculated posteriori CH$_4$ fluxes constrained by isotopes (hereafter called “Pos_iso” scenario). Figure 7 shows temporal variations of the CH$_4$ mole fraction, $\delta^{13}C$-CH$_4$, and $\delta D$-CH$_4$ simulated for NYA and SYO using the Pos_iso scenario. For reference, the simulation results using the base_iso scenario are also presented. As seen in the figure, $\delta^{13}C$-CH$_4$ and $\delta D$-CH$_4$ simulated using the Pos_iso scenario are well consistent with the observed values, keeping good agreement with the simulated and observed CH$_4$ mole fractions. We also compare the simulated $\delta^{13}C$-CH$_4$ and $\delta D$-CH$_4$ using the Pos_iso scenario with the independent observational data in the western Pacific in Figure 8 (also see Figure S5 for the CH$_4$ mole fraction) and with NOAA/INSTAAR, NIWA, and other TU/NIPR data in
The simulations were conducted using the PosCH4 scenario. Figures S6 and S7. The CH4 mole fraction, δ^{13}C-CH4, and δD-CH4, simulated using the Pos_iso scenario, also well reproduce the observations from 33°N to 40°S in the western Pacific, as well as those obtained by other institutes globally. However, it is also seen that the simulated δ^{13}C-CH4 slightly underestimates the values observed at 33°N–25°N and 20°N–10°N in the western Pacific (Figure 8) and at other northern middle/lower latitude sites (e.g., MHD and NWR in Figure S6). This underestimation is probably caused by the fact that the CH4–Cl reaction in the marine boundary layer is not included in our simulations, as discussed in section 4.1.

3.3. Global and Regional CH4 Emissions Constrained by Observed δ^{13}C-CH4 and δD-CH4

Figure 9a shows the annual mean global CH4 emissions given by the Pos_iso scenario for BIO, FF, BB, and TOT, with those of PosCH4. Compared to the result of PosCH4, Pos_iso provides smaller values for BIO and larger values for FF and BB for 1996–2012. The global average CH4 emissions of BIO, FF, and BB over 2003–2012 are calculated to be 346 ± 11, 162 ± 2, and 50 ± 2 TgCH4 year\(^{-1}\), respectively, their fractions of the TOT emission being 62 (±2):29 (±1):9 (±1) (Figure 9c).

Our results are slightly different from the median values of top-down estimations in Saunois et al. (2016), which are rather close to those obtained using the PosCH4 scenario, and are more consistent with the values derived by Schwietzke et al. (2016). Schwietzke et al. (2016) estimated global CH4 emissions for BIO, FF, and BB based on global atmospheric δ^{13}C-CH4 observations and their updated δ^{13}C-CH4 signatures of CH4 sources. The δ^{13}C-CH4 source signatures reported by Schwietzke et al. (2016) are further updated by Sherwood et al. (2017), although the mean difference between the two studies is very small. Therefore, the agreement between the results obtained by us and Schwietzke et al. could be partly attributed to the fact that we also employed the isotopic signatures of Sherwood et al. (2017) for our CH4 forward simulations. However, as seen in Figures 7c and 8b, forward simulations of δD-CH4 with the Pos_iso scenario also well reproduce the observational results at NYA, SYO, and NWP. This suggests that the source partitioning of global CH4 emissions reported by Schwietzke et al. (2016) is also consistent with the present result derived from our δD-CH4 observations and the δD-CH4 source signatures by Sherwood et al. (2017). A slight difference of the TOT emissions between this study and Schwietzke et al. (2016) could be due to the different total atmospheric CH4 lifetimes used in the two studies.

From the trend and interannual variations of each source category in the Pos_iso scenario (Figure 9b), we find that (1) the decrease in the BIO and BB emissions mainly contributes to the reduction in TOT emissions from the late 1990s to the mid-2000s, (2) the reduction is partly offset by an increase of the FF emission during the same period, and (3) the increase in the BIO emissions mainly contributes to the increase in TOT emissions after 2006/2007. The interannual variations are clearly seen in the BIO and BB emissions, especially during the 1997–1998 El Niño, which show a positive anomaly, as well as in the BIO emission in 2004 with the smallest wetland inundation extent during 1993–2004 (Zhuang et al., 2015), which show a negative anomaly. The BIO and BB emissions decrease at the respective rates of −1.1 ± 0.3 TgCH4 year\(^{-2}\) (+95% CI) and −0.7 ± 0.3 TgCH4 year\(^{-2}\) from 1995–2007. Then, the BIO emissions clearly increase by 1.7 ± 0.5 TgCH4 year\(^{-2}\) from 2007–2013, whereas the BB emissions continue to slightly decrease by −0.4 ± 0.4 TgCH4 year\(^{-2}\). On the other hand, the FF emissions of Pos_iso show a clear increase trend of 1.6 ± 0.1 TgCH4 year\(^{-2}\) for 1995–2007, followed by a plateau with 0.2 ± 0.2 TgCH4 year\(^{-2}\) for 2007–2013.

Rice et al. (2016) suggested that FF emissions increased significantly after 2000, that the CH4 emissions from wetlands (i.e., BIO) showed a large decrease after 2000, and that the BB emissions declined from the 1980s to 2009 except for the 1997–1998 El Niño. Bousquet et al. (2006) also pointed out that wetland emissions
Figure 6. Scaling factors for fossil fuel ($f_f$), biomass burning ($f_b$), and biogenic ($f_{bi}$) CH4 sources derived by comparing the observed $\delta^{13}$C-CH4 and $\delta$D-CH4 with the results of model simulations. Color boxes for $f_f$, $f_{bi}$, and $f_b$ represent the scaling factors estimated by the 3-year sensitivity tests with multiple combinations of $f_f$ and $f_b$ (see text). The upper and lower limits of each box represent the uncertainty range derived from the 95-percentile bootstrap confidence intervals of the observed $\delta^{13}$C-CH4 and $\delta$D-CH4 growth rates for the relevant period (see Supporting Information in more detail). Color circles represent the best estimates of the $f_f$, $f_{bi}$, and $f_b$ obtained after $f_f$ and $f_b$ were further adjusted to better reproduce the year-by-year observations at NYA and SYO.

For the emission differences between 1996–2000 and 2002–2006, Temperate S. America and Europe show large decreases of 11 and 6 TgCH4 year$^{-1}$, which are partially offset by a large emission increase in India (+11 TgCH4 year$^{-1}$). The large decrease and increase in Temperate S. America and India are mainly due to decreased for 1999–2003, accompanied by an increase of anthropogenic (mainly FF) CH4 emissions. Their emission scenarios before the first half of the 2000s are generally consistent with our Pos_iso scenario, except that our FF emissions start to increase earlier than 1999/2000. However, our Pos_iso scenario and their results are not consistent with each other after 2006, because the enhanced BIO emissions are primarily responsible for the CH4 regrowth in our scenario. Our result is also inconsistent with EDGAR42FT (2013) and those from some ethane-based approaches (Hausmann et al., 2016; Helmig et al., 2016). On the other hand, $\delta^{13}$C-CH4 observation studies suggested a large contribution of BIO emissions after 2006/2007, which may have originated in the tropics (Nisbet et al., 2016; Schaefer et al., 2016). Additionally, Thompson et al. (2018) recently showed, using their 2-D inversion modeling based on CH4, $\delta^{13}$C-CH4, and C2H6 observations, that both BIO and FF emissions occurred mainly in the tropics and sub-tropics and contributed to the atmospheric CH4 increase after 2007. By inspecting our CH4 emission estimates in terms of latitudinal distribution, we find that the abovementioned temporal changes of FF, BB, BIO, and TOT mainly originate in the tropics/sub-tropics (Figure 10 and Table 3). This is consistent with the result of the mean emission anomaly of the multiple top-down ensembles reported by Saunois et al. (2017) during 2000–2012. We also find that the increase in CH4 emissions in 2006/2007 are mainly caused by BIO sources in the tropics, especially in 30°N–EQ (Figure 10). Similar results were reported in previous studies (Nisbet et al., 2016; Thompson et al., 2018), as mentioned above, but we used 5D observations for the first time as constraints with our 3-D model. From further inspection, the global TOT emission increase of +23 TgCH4 year$^{-1}$ between the period 2002–2006 and 2008–2012 is found to be attributed to 30°N–EQ (+11 TgCH4 year$^{-1}$), EQ–30°S (+8 TgCH4 year$^{-1}$) and 90°N–30°N (+5 TgCH4 year$^{-1}$), respectively (see Table 3). On the other hand, from 1996–2000 to 2002–2006, the 30°N–EQ and EQ–30°S show positive and negative shifts of +9 and −11 TgCH4 year$^{-1}$, which are mainly ascribed by FF and BIO increases for the former and BIO decreases for the latter, respectively. As for the BB source, both 30°N–EQ and EQ–30°S show slight decreases (−2 and −1 TgCH4 year$^{-1}$) between 1996–2000 and 2002–2006. No significant increase or decrease of CH4 emissions are found in 90°N–60°N and 30°S–90°S.

Figure 11 shows the annual CH4 emissions of BIO, FF, BB, and TOT in individual regions for 1996–2012, obtained from the PosCh4 and Pos_iso scenarios after aggregating the land regions from 42 to 14 (also see Table 3). For comparison, the regional CH4 emissions for Prior and PosCh4 are shown in Figure S8. It is seen from Figure 11 that the TOT emissions and their interannual variations of PosCh4 and Pos_iso in each region are similar to each other, primarily because the BIO emissions decreased by changing the scenario from PosCh4 to Pos_iso are offset by the corresponding increase in the FF and BB emissions. As a result, the atmospheric CH4 variations simulated using Pos_iso are nearly consistent with those using PosCh4 at any observation sites (see Figures S5 and S9). Also seen from Figure 11 is that larger CH4 emissions exist in tropical regions (e.g., India, Tropical S. America, Temperate S. America, and Southeast Asia) and populous temperate regions (Europe, Temperate N. America, and East Asia). In the Prior scenario, East Asia (mainly China; regions 31 and 32) is especially a quite strong CH4 source region, where a significant FF CH4 emissions is seen after 2000 (EDGAR42FT, 2013) (Figure S8). By replacing PosCh4 with Pos_iso, the increase in FF emissions exceeds the decrease in BIO emissions in East Asia, resulting in slight increase of TOT emissions there, but the Pos_iso TOT emissions are still lower than the Prior emissions (also see Figure S8). This is consistent with other inversion studies, suggesting that the EDGAR v4.2 CH4 emissions are overestimated for East Asia after the 2000s (Saunois et al., 2017, and references therein).
to BIO sources, which mainly explains the negative and positive emission shifts in EQ–30°S and 30°N–EQ, respectively, as mentioned above. The emission decrease in Europe is caused by both BIO and FF, which is offset by slight FF increase of East Asia and West Asia (see Figure 11), resulting in the stabilization of TOT emissions above 30°N (Figure 10). Between 2002–2006 and 2008–2012, the large emission increase occurs in East Asia, Southeast N. Asia, and Tropical S. America of 9, 7, and 5 TgCH4 year\(^{-1}\), respectively, with a continued decrease in Europe (−6 TgCH4 year\(^{-1}\)). Most of the increase in South-East N. Asia and Tropical S. America are caused by BIO sources, resulting in a large TOT emission increase in the tropics (+18 TgCH4 year\(^{-1}\); see Table 3), whereas the changes in East Asia and Europe are caused by both BIO and FF. Because the emission increase in East Asia exceeds the decrease in Europe, and slight increases in West Asia and Boreal Asia are accompanied, the TOT CH4 emission in 90°N–30°N shows a positive shift (+5 TgCH4 year\(^{-1}\); see Table 3).

4. Discussion

As mentioned above, by comparing \(\delta^{13}C\)-CH4 and \(\delta^D\)-CH4 simulated using the PosCH4 and Pos_iso scenarios with the observed values, it is suggested that the source partitioning in the global CH4 emissions based on our \(\delta^{13}C\)-CH4 and \(\delta^D\)-CH4 data is similar to that of Schwietzke et al. (2016), which shows that FF and BB sources are much stronger and BIO sources are weaker, compared with the PosCH4 and medians of multiple top-down estimates summarized by Saunois et al. (2016). The results obtained in this study also indicate that the plateau of atmospheric CH4 in the early 2000s, and its renewed growth after 2006/2007 can be explained by the reduced BIO and BB emissions for 2000–2006 and the enhanced BIO emissions after 2006/2007 mainly in the tropics, respectively. These findings are obtained under the assumptions that (1) our forward and inverse model settings (i.e., prior CH4 emissions and sinks, including their isotopic signatures) are appropriate, (2) our \(\delta^{13}C\)-CH4 and \(\delta^D\)-CH4 observations at NYA and SYO reflect their hemispheric scale changes, and (3) the atmospheric OH concentration shows no significant trend for the whole simulation period of 1995–2013. Here we discuss our findings by inspecting these assumptions and future issues required to better comprehend the global CH4 budget using isotope observations.

4.1. Prior CH4 Emissions and Sinks

Our prior CH4 emissions were obtained from the bottom-up inventory for FF (coal, natural gas, and residential [EDGAR v4.2]), a part of BIO (ruminants, landfills [EDGAR v4.2], and termites [GISS inventory]), and BB (GFED v4.0) and from the process-based model for the other part of BIO (wetlands and rice paddies [VISIT]) (see Table 2). The Prior scenario, thus obtained, is not special but rather common, and the a posteriori CH4 fluxes (i.e., PosCH4) of the three source categories in 43 regions were derived reasonably by the CH4 inversion within their given uncertainties. The obtained global fluxes are close to the median values reported by Saunois et al. (2016). These results suggest that our inverse modeling is comparable to other recent top-down modeling studies. However, it should be noted that the Prior scenario does not include CH4 emissions from natural geological seepage in FF. Etiope et al. (2008) reported that the emissions from this source amount is approximately 51 ± 20 TgCH4 year\(^{-1}\), which is equivalent to as much as 43% of our prior FF emissions for 1995–2013. As mentioned, Schwietzke et al. (2016) also obtained higher FF emissions than traditional top-down estimates as the sum of anthropogenic FF and natural geological seepage. In this regard, it may be worthy to note that the Prior scenario would underestimate the total FF CH4 emissions and that the FF emissions given by the Pos_iso scenario would be regarded as the sum of CH4 emissions from anthropogenic FF and natural geological seepage. Since Etiope et al. (2019) recently reported for the first time
1° × 1° global gridded maps of geological CH4 sources, more reliable inverse CH4 modeling can be performed by including their emission data.

For the δ\(^{13}\)C–CH4 and δD–CH4 source isotopic signatures, we adopted the mean values summarized by Sherwood et al. (2017), in which δ\(^{13}\)C–CH4 of FF is 3–4‰ lighter than the typical values employed in previous studies (e.g., Whiticar & Schaefer, 2007). From their δ\(^{13}\)C–CH4 database, Schwietzke et al. (2016) proposed that the CH4 emissions from FF, including natural geological seepage, are larger by 60%–110% than the previous estimates by the CH4 inversion. In this regard, if we employ the flux-weighted mean δ\(^{13}\)C–CH4 values of Whiticar and Schaefer (2007) for FF (δ\(^{13}\)C–CH4 = −40.9‰), our finally obtained CH4 source partitioning would decrease the fractions of FF and/or BB with heavier isotopes and increase that of BIO with lighter isotopes to balance the atmospheric 13CH4 budget. As for the δD–CH4 source signatures, the data listed in Sherwood et al. (2017) are much less than those for the δ\(^{13}\)C–CH4 signatures. For example, the δD–CH4 source signatures for BB were sampled only in Brazil, so the value would have sampling bias. Further δ\(^{13}\)C–CH4 and δD–CH4 measurements for each CH4 source are needed to account for their spatial distributions and global representative values for more proper model analysis.

The KIE values of CH4 sink processes are also crucial for simulating atmospheric δ\(^{13}\)C–CH4 and δD–CH4, especially the value for CH4–OH reaction, which is most dominant in CH4 destruction. In this study, we set the value to 1.00475 for KIEC, which is the average value of Cantrell et al. (1990) and Saueressig et al. (2001). The KIE\(^{C}\) value of Cantrell et al. (1990) (KIE\(^{C}\) = 1.0054) was used by Hein et al. (1997), Bousquet et al. (2006), and Rice et al. (2016) for their model analyses, whereas other studies, such as Nisbet et al. (2016), Schaefer et al. (2016), and Warwick et al. (2016), used the value of Saueressig et al. (2001) (KIE\(^{C}\) = 1.0039). The difference between the two KIE\(^{C}\) values appears to be small, but we confirmed by performing forward simulations with invariant CH4 fluxes over 36 years that such a small difference produces a change of approximately 1.3‰ in atmospheric δ\(^{13}\)C–CH4 at steady state. Similar discussion can be also made.

Figure 8. Comparison of the observed (closed circles) and model-simulated (red and green lines) δ\(^{13}\)C–CH4 and δD–CH4 at the respective latitude bands between Japan and Australia/New Zealand in the western Pacific (NWP) for the period 2006–2013. The simulations were conducted using the PosCH4 (red) and Pos_iso (green) scenarios. Correlation coefficient (R) and root-mean-square error (RMSE) between the observed and simulated δ\(^{13}\)C–CH4 and δD–CH4 for the Pos_iso scenario are also shown in the figure. The results are presented after aggregating into 5 latitudinal bands.
on the KIE\textsuperscript{D} value. There are four eminent experimental studies on estimating the KIE\textsuperscript{D} value (DeMore, 1993; Gierczak et al., 1997; Joelsson et al., 2016; Saueressig et al., 2001). In this study, we used the value of Gierczak et al. (1997), which covers a wide temperature range, including typical atmospheric temperatures. The value of Gierczak et al. (1997) (KIE\textsuperscript{D} = 1.286 at 296 K) is slightly lower than the value of Saueressig et al. (2001) (KIE\textsuperscript{D} = 1.294 reported only at 296 K) and Joelsson et al. (2016) (KIE\textsuperscript{D} = 1.320 at 296 K) but much higher than the value of DeMore (1993) (KIE\textsuperscript{D} = 1.172 at 296 K). Because the value of Gierczak et al. (1997) is comparable to the median among these previous studies, we assumed their value to be most promising at this stage. It was confirmed that the respective differences of KIE\textsuperscript{D} between Gierczak et al. (1997) and the other three studies affect the atmospheric $\delta$D-CH\textsubscript{4} value by approximately 5‰, 16‰, and −68‰ at a steady state.

Other minor CH\textsubscript{4} sinks, such as uptake by soils, stratospheric destruction, and reaction with Cl in the marine boundary layer (tropospheric Cl), can also strongly influence the sink-weighted mean KIE values. Many model studies, including this study, accounted for soil sinks and stratospheric destruction (Bousquet et al., 2006; Monteil et al., 2011; Rice et al., 2016; Schwietzke et al., 2016; Warwick et al., 2016), whereas others included those and tropospheric Cl (Lassey et al., 2007; Nisbet et al., 2016; Rigby et al., 2017; Schaefer et al., 2016). A few studies considered the OH sink only (Turner et al., 2017). In this study, we used the average KIE values for the soil sinks, which are calculated from four KIE\textsubscript{C} values and two KIE\textsubscript{D} values obtained experimentally (Reeburgh et al., 1997; Snover & Quay, 2000; Tyler et al., 1994) and the KIE values for stratospheric reactions with Cl and O(1D) (Saueressig et al., 1995, 1996) (see Table S1 for details). Because our model simulations do not account for the tropospheric Cl sink, the $\delta^{13}$C values, simulated for 33°N–25°N and 20°N–10°N in the western Pacific and other northern middle/low latitude sites, are possibly underestimated, because the tropospheric Cl would account for >20% of boundary layer CH\textsubscript{4} sink in the polluted northern hemisphere (i.e., downwind of East Asia) (see Figure 12b in Hossaini et al., 2016), which significantly enriches $^{13}$CH\textsubscript{4} because of its large KIE. This being the case, our estimated global BB emissions (~50 TgCH\textsubscript{4} year\textsuperscript{−1}) could be slightly overestimated, because the BB emission increases $^{13}$CH\textsubscript{4} in our model owing to its heavier isotope ratios. However, Gromov et al. (2018) recently suggested that the contribution of Cl to the CH\textsubscript{4} removal in the troposphere is much smaller than that estimated by Hossaini et al. (2016) and references therein.

From the above discussion, further studies are required to comprehend the influence of not only the isotope source signatures but also the KIE values on the global and regional CH\textsubscript{4} budgets when the inverse modeling is performed using the atmospheric isotope observations as constraints. It is also crucial to determine the KIE values of CH\textsubscript{4} sink processes more precisely by conducting more laboratory experiments and compiling the KIE database thus far, as performed by Sherwood et al. (2017) to characterize globally representative isotope source signatures. To improve our model simulations of seasonal and short-term variations in atmospheric $\delta^{13}$C-CH\textsubscript{4} and $\delta$D-CH\textsubscript{4}, noting that the simulated amplitudes were underestimated (Figures 6 and 8), spatially high resolution maps of isotope source signatures (e.g., Ganesan et al., 2018, for wetland $\delta^{13}$C-CH\textsubscript{4}) would be required with increasing direct measurements and updating the database. Such a database is particularly useful for forward simulations with high-resolution models (e.g., Wang et al., 2019), by which short-term variations of not only the atmospheric CH\textsubscript{4} mole fraction, but also $\delta^{13}$C-CH\textsubscript{4} and $\delta$D-CH\textsubscript{4}, would be well represented.

![Figure 9](source_url)

**Figure 9.** (a) Same as Figure 3 except for (a) the PosCH\textsubscript{4} (dash lines) and Pos_iso (solid lines) scenarios, (b) FF, BB, BIO, and TOT of the PosCH\textsubscript{4} (dash lines) and Pos_iso (solid lines) scenarios, and (c) the Pos_iso scenario. The results of Schwietzke et al. (2016) for the period 2003–2013 are also shown in (c).
Figure 10. Same as Figure 9b but for (a) 90°N–60°N, (b) 60°N–30°N, (c) 30°N–EQ, (d) EQ–30°S, and (e) 30°S–90°S.

Table 3
Averaged Global, Latitudinal, and Regional CH₄ Emissions (TgCH₄ year⁻¹)

| Period            | 1996–2000 | 2002–2006 | 2008–2012 | 2002–2006 minus 1996–2000 | 2008–2012 minus 2002–2006 |
|-------------------|-----------|-----------|-----------|---------------------------|---------------------------|
| **Global**        |           |           |           |                           |                           |
| Pos_iso (PosCH₄)  |           |           |           |                           |                           |
| 90°N–60°N         | 19 (19)   | 20 (20)   | 22 (21)   | 1 (1)                     | 2 (1)                     |
| 60°N–30°N         | 197 (186)| 195 (187)| 198 (194)| −1 (1)                   | 2 (7)                     |
| 30°N–EQ           | 181 (188)| 190 (198)| 201 (210)| 9 (10)                   | 11 (11)                   |
| EQ–30°S           | 140 (143)| 129 (128)| 137 (133)| −11 (−15)                | 8 (4)                     |
| 30°S–90°S         | 11 (12)  | 11 (11)  | 9 (10)   | 0 (−1)                   | −1 (−1)                   |
| Tropics (30°N–30°S) | 121 (205)| 215 (207)| 220 (215)| 0 (2)                    | 5 (8)                     |
| NH (90°N–EQ)      | 396 (393)| 405 (405)| 421 (425)| 9 (13)                   | 16 (20)                   |
| SH (EQ–90°S)      | 151 (155)| 140 (139)| 146 (142)| −12 (−15)                | 7 (3)                     |
| **S17**           |           |           |           |                           |                           |
| 90°N–60°N         | 20 [14–24]| 22 [15–31]|          |                           |                           |
| 60°N–30°N         | 175 [158–194]| 184 [164–203]|          |                           |                           |
| 30°N–90°S         | 349 [330–379]| 363 [344–391]|          |                           |                           |
| **Regional**      |           |           |           |                           |                           |
| Pos_iso (PosCH₄)  |           |           |           |                           |                           |
| Boreal N. America | 15 (17)  | 15 (16)  | 14 (15)  | 0 (−1)                   | −1 (−1)                   |
| Temperate N. America | 57 (58)| 56 (59)  | 54 (58)  | −1 (0)                   | −2 (−1)                   |
| Tropical S. America | 50 (43)| 49 (52)  | 54 (57)  | −2 (−2)                  | 5 (5)                     |
| Temperate S. America | 51 (59)| 40 (45)  | 43 (47)  | −11 (−14)                | 3 (1)                     |
| Northern Africa   | 46 (44)  | 45 (43)  | 48 (46)  | −1 (−1)                  | 2 (3)                     |
| Southern Africa   | 38 (35)  | 40 (37)  | 42 (36)  | 1 (2)                    | 2 (0)                     |
| Boreal Asia       | 24 (21)  | 25 (22)  | 28 (25)  | 1 (2)                    | 3 (3)                     |
| West Asia         | 15 (13)  | 19 (17)  | 22 (20)  | 4 (4)                    | 3 (3)                     |
| India             | 44 (50)  | 55 (63)  | 51 (57)  | 11 (13)                  | −3 (−6)                   |
| East Asia         | 42 (37)  | 46 (40)  | 55 (52)  | 3 (2)                    | 9 (12)                    |
| South-East S. Asia| 26 (23)  | 23 (20)  | 23 (20)  | 3 (−3)                   | 0 (1)                     |
| South-East N. Asia| 36 (39)  | 35 (37)  | 43 (45)  | −1 (−1)                  | 7 (8)                     |
| Oceania           | 9 (9)    | 9 (9)    | 10 (10)  | 0 (0)                    | 0 (0)                     |
| Europe            | 64 (60)  | 58 (55)  | 52 (50)  | −6 (−5)                  | −6 (−5)                   |
| Ocean and others  | 29 (28)  | 31 (29)  | 31 (30)  | 2 (1)                    | 0 (1)                     |

Note: Values of the Pos_iso are shown over the periods 1996–2000, 2002–2006, and 2008–2012 and differences between the two adjacent periods. The corresponding results of PosCH₄ are shown with the values of Pos_iso. The results shown in Saunois et al. (2017) for the period of 2002–2006 and 2008–2012 and the differences between the two periods are also presented (represented as S17). Left and right numbers in each bracket in S17 represent the minimum and maximum top-down estimates summarized by Saunois et al. (2017), respectively. The CH₄ emissions differences between the two adjacent periods exceeding ±5 TgCH₄ year⁻¹ are shown in boldface. The individual values include a rounding error of 1 TgCH₄ year⁻¹.
4.2. $\delta^{13}$C-CH$_4$ and $\delta$D-CH$_4$ Observations

Recently, some isotope studies attempted to estimate the global CH$_4$ budget and its temporal variations for the plateau and renewed growth of atmospheric CH$_4$ in the 2000s using independent or combined $\delta^{13}$C-CH$_4$ measurement records (Morimoto et al., 2017; Nisbet et al., 2016; Rice et al., 2016; Rigby et al., 2017; Schaefer et al., 2016; Schwietzke et al., 2016; Thompson et al., 2018; Turner et al., 2017). However, no study has constrained the CH$_4$ budget after the 2000s using the atmospheric $\delta$D-CH$_4$ data. Only Rice et al. (2016) presented a long-term $\delta$D-CH$_4$ record over recent decades, but they did not use their data to constrain the CH$_4$ budget, just used them to validate their estimated budget. Therefore, some differences are found in the CH$_4$ budget and its temporal behavior between present and previous studies. For example, our conclusion on the temporal CH$_4$ stagnation in the early 2000s is inconsistent with that deduced by Schaefer et al. (2016), using globally averaged $\delta^{13}$C-CH$_4$ data collected by NIWA, INSTAAR, and Heidelberg University, Germany (UHEI) from 17 sites. They suggested that the CH$_4$ plateau was caused by diminished CH$_4$ emissions from FF, whereas our study ascribes the diminished emissions to BIO and BB, with an

Figure 11. Annual mean regional CH$_4$ emissions of fossil fuel (FF: red), biomass burning (BB: green), biogenic sources (BIO: blue), and their total (TOT: black) given by the PosCH4 (dash lines) and Pos_iso (solid lines) scenarios for the period 1996–2012. The region number (rgXX) corresponds to that in Figure 1, and the results are represented after aggregating the land regions from 42 into 14.
increase in FF emissions. Our positive δD-CH₄ trend at SYO until 2006 would not favor the diminished FF emissions to balance the atmospheric CH₃D budget. The slight difference of δ¹³C-CH₄ trend in the early 2000s between the two studies, as mentioned in section 3.2, would also be related to the difference.

Regarding the growth of atmospheric CH₄ after 2006/2007, Schaefer et al. (2016) reported that the global average δ¹³C-CH₄ decreased secularly in parallel because of enhanced CH₄ emissions from BIO rather than from FF. The same conclusion was obtained by Nisbet et al. (2016) using the δ¹³C-CH₄ data collected by INSTAAR, UHEI, and Royal Holloway, University of London. Our result also suggests that the increase in the BIO emissions, especially in the tropics, is a major contributor to the post-2006/2007 CH₄ growth and that the FF emissions generally remain constant after 2007. This suggestion is also supported by the plateau of δD-CH₄ at SYO after 2007. In this regard, Rice et al. (2016) reported that the FF emissions increased significantly from 2000 to 2009, based on the δ¹³C-CH₄ data taken by INSTAAR, Portland State University, University of Washington, and University of California, Irvine. However, considering that their analysis was limited until 2009, it would be uncertain whether FF mainly contributed to the CH₄ regrowth after 2006/2007.

It should also be noted that the δ¹³C-CH₄ and δD-CH₄ values measured by individual laboratories differ because of the differences in instrument settings, correction methods, traceability to reference materials, and long-term data management (Umezawa et al., 2018). The differences of δ¹³C-CH₄ and δD-CH₄ among the laboratories amount to 0.5‰ and 13‰, respectively, at present atmospheric CH₄ levels. The combined isotope dataset prepared with no careful correction of the interlaboratory offset would yield erroneous result in source contributions to the recent atmospheric CH₄ growth by using it as a constraint (Levin et al., 2012). To avoid the artifact arisen from the interlaboratory offset, we used only our δ¹³C-CH₄ and δD-CH₄ data based on the TU/NIPR scale to determine the global CH₄ source partitioning.

### 4.3. Methane Sinks by OH

In this study, we employed climatological tropospheric OH data from the TransCom-CH₄ (Patra et al., 2011) to account for the CH₄-OH reaction as the major CH₄ sink. These OH data were based on the global OH fields of Spivakovsky et al. (2000), which have been widely used in the CH₄ forward and inverse modeling studies (e.g., Houweling et al., 2014). Recently, several studies have suggested that the year-to-year changes of the OH concentration played a substantial role in the plateau and renewed growth of atmospheric CH₄ mole fraction in the 2000s (McNorton et al., 2016; Rigby et al., 2017; Turner et al., 2017).

McNorton et al. (2016) reported, using their 3-D global chemistry transport model with global OH concentrations estimated from CH₃CCl₃ observations, that the stagnation of atmospheric CH₄ observed during 1999–2006 could be explained by relatively small variations in the OH concentration. Additionally, Rigby et al. (2017) and Turner et al. (2017) showed, using their simple box models constrained by CH₃CCl₃, CH₄, and δ¹³C-CH₄, that the decrease in OH could be primarily responsible for the increase of atmospheric CH₄ after 2006/2007. However, these studies did not use the δD-CH₄ observation as a constraint.

To test the sensitivity of our isotope simulations results on OH changes, we adopted the “most likely solution” of global OH anomalies presented by Turner et al. (2017). The OH anomalies were first calculated for the period of 1996–2013 relative to the average OH concentration over 1996–1999 (Figure 12d). The
period 1996–1999 was selected to keep the consistency of the average total CH$_4$ lifetime between this study (9.0 year) and Turner et al. (2017). Then, interannually variable (IAV) OH data were obtained by multiplying the climatological OH fields by the anomalies after 1996, and forward simulations were performed for 1996–2013 using the IAV OH, initial values of the Pos$_{iso}$ scenario in January 1996, and climatological CH$_4$ fluxes derived by averaging Pos$_{iso}$ CH$_4$ emissions in 1996–1999. For reference, the simulation using the climatological OH given by the Pos$_{iso}$ scenario and the climatological CH$_4$ fluxes was also performed (hereafter called OH1.00 scenario).

Figure 12 shows long-term variations of the simulated CH$_4$, $^{13}$C-CH$_4$, and D-CH$_4$ at NYA and SYO with the two OH scenarios, that is, OH1.00 and Turner et al. (2017), together with those calculated using the Pos$_{iso}$ scenario and the observations. The simulated CH$_4$ mole fractions based on the OH1.00 scenario overestimate the observations after the early 2000s and reach an almost constant value around 2010. The simulated $^{13}$C-CH$_4$ and D-CH$_4$ show long-term positive trends over 1996–2013, which is not consistent with the observational results. On the other hand, the simulations with the Turner et al.’s (2017) OH scenario well reproduce the observed plateau and renewed growth of the atmospheric CH$_4$ in the 2000s, which are achieved by the increase of $\sim$4% in CH$_4$ destruction by OH between 1997 and 2000, followed by the respective decreases of $\sim$4% and $\sim$3% between 2004 and 2007 and between 2010 and 2012 (see Figure 12d). However, the simulated $^{13}$C-CH$_4$ values still overestimate the observation result after 2000, while good reproducibility is obtained for D-CH$_4$.

The sensitivity tests with the Turner et al.’s (2017) OH scenario revealed that temporal changes in the CH$_4$ emissions are also required to simultaneously reconcile the observed and simulated CH$_4$, $^{13}$C-CH$_4$, and D-CH$_4$ over 1995–2013. In this connection, Turner et al. (2017) and Rigby et al. (2017) were cautious about attributing the recent CH$_4$ trend to the emission changes in specific sources because of the uncertainty of not only OH variability but also $^{13}$C-CH$_4$ source signatures. To interpret the CH$_4$ trend using isotope observations, it is also necessary to inspect the uncertainty of KIEs in more detail. In this regard, it is also noted that the good reproducibility of the D-CH$_4$ trend, as shown here, is achieved by employing the CH$_4$-OH KIE$^D$ value from Gierczak et al. (1997). Further advanced 3-D inverse modeling studies, including CH$_4$ isotopes and multiple 3-D OH fields calculated using photochemical models (Zhao et al., 2019), considering the uncertainty of both isotope source signatures and KIEs, will be helpful in deepening our understanding of the recent global CH$_4$ cycle.

5. Summary and Conclusions

To estimate the global and regional CH$_4$ emissions for the period of 1995–2013, we performed the inverse modeling of atmospheric CH$_4$ observations, based on the global 3-D atmospheric chemistry transport model with a priori fluxes and sink fields of CH$_4$. Seasonal and long-term variations of atmospheric CH$_4$ simulated with the a posteriori CH$_4$ fluxes, are generally close to the observational results, not only at the sites where the CH$_4$ data were used for the inverse modeling but also at other independent sites in the western Pacific region. The respective contributions of three major CH$_4$ source categories (i.e., biogenic, fossil fuel, and biomass burning) to the global total a posteriori CH$_4$ emission are fairly comparable with the medians of multiple CH$_4$ inverse modeling reported by Saunois et al. (2016). However, forward simulations of CH$_4$, $^{13}$C-CH$_4$, and D-CH$_4$ performed using the a posteriori CH$_4$ fluxes, significantly underestimate the values of $^{13}$C-CH$_4$ and D-CH$_4$ observed at Ny–Ålesund, Syowa Station, and in the western Pacific for 1995–2013. This result suggests that the conventional global CH$_4$ fluxes obtained by the CH$_4$ inverse modeling could overestimate the proportion of biogenic CH$_4$ with lighter isotopic signatures in the total emissions and underestimate those of fossil fuel and biomass burning CH$_4$ with heavier isotopic signatures, probably because of an over-estimation of prior biogenic CH$_4$ emissions.

The global CH$_4$ emissions derived from the inversion were further modified to reproduce the observed long-term trends of $^{13}$C-CH$_4$ and D-CH$_4$ at Syowa Station and Ny–Ålesund. By this modification, the fractions of fossil fuel and biomass burning sources are increased and that of biogenic sources is decreased. After improving the disagreement in atmospheric $^{13}$C-CH$_4$ and D-CH$_4$, the respective contributions of biogenic, fossil-fuel, and biomass-burning source categories to the global total a posteriori CH$_4$ emission are found to be 62 (±2):29 (±1):9 (±1) over 2003–2012. This result slightly differs from that of Saunois et al. (2016) but is
fairly comparable to that of Schwietzke et al. (2016), constrained by their global atmospheric δ13C-CH4 data and isotopic source signatures. By inspecting the CH4 emission trends of individual source categories, the decrease in biogenic and biomass burning CH4 emissions in the first half of the 2000s is responsible for the total CH4 emissions reduced for this period. On the other hand, the biogenic CH4 emission, mainly in the tropics/subtropics, increases remarkably after 2006/2007, contributing to the renewed growth of atmospheric CH4. It is also found that the increase rate of fossil fuel CH4 emissions after 2006 is much smaller than those derived from both a priori (based on EDGAR4.2 FT, 2013) and conventional inversion fluxes using only CH4 observation. As suggested by Turner et al. (2017) and Rigby et al. (2017), the global OH concentration may have significantly affected the atmospheric CH4 variations in recent decades, but our independently observed CH4, δ13C-CH4, and 5Δ-CH4 data appear to indicate that all their trends cannot be consistently explained only by the temporal changes in OH.

This study indicates that our long-term observations of atmospheric δ13C-CH4 and 5Δ-CH4 are useful for estimating the proportions of individual CH4 sources in the global total CH4 emission using the 3-D atmospheric chemistry and transport model. However, our data analysis in this study remains simple. Further advanced CH4 inversion approaches, including δ13C-CH4 and 5Δ-CH4 as constraints in inversion, are needed to quantify the regional CH4 budgets in more detail. For this approach, further 5Δ-CH4 measurements for the atmosphere and sources/sinks, as well as appropriate correction of large interlaboratory 5Δ-CH4 measurement offsets, are especially important. It will also be important to incorporate interannual variations of OH concentrations into the inversion modeling and to carefully consider the uncertainties of the source and KIE isotopic signatures. In this study, we suggest that the biogenic CH4 emission mainly in the tropics is responsible for the recent CH4 growth. However, we cannot conclude which biogenic source plays a main role (i.e., wetlands [Nisbet et al., 2016; Thompson et al., 2018], ruminants [Patra et al., 2016], and/or others [e.g., rice paddies, landfills]), because of the limitations of the isotope-based top-down approach. To resolve this issue, further studies using top-down and bottom-up approaches are required.

**Data Availability Statement**

Our δ13C-CH4 and 5Δ-CH4 data at NYA, SYO, and NWP are available from the WDCGG website (https://gaw.kishou.go.jp/search/station#NYA, https://gaw.kishou.go.jp/search/station#SYO, https://gaw.kishou.go.jp/search/station#NWP). The CH4 mole fraction data are also available from the WDCGG website, except for Tibetan continuous and aircraft measurement data available from NIES website (http://db.cger.nies.go.jp/portal/geds/atmosphericAndOceanicMonitoring).

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