We would like to thank the two reviewers for their thorough and constructive reviews of our manuscript. Based on the reviews, we made the following major changes:

- We changed the cited papers referring to methane emissions from sea shelves;
- We split Sect. 4.2 into four sub-parts;
- We acknowledged in Sect. 4.3 the efforts of different teams to measure atmospheric CH$_4$ in Siberia and Canada and the possibility to further improve our results if these measurements are integrated;
- We revised the caption and the label on the y-axis of Fig. 6. We also showed the significance of difference between the simulations with and without lake emissions using a two-sample $t$-test.

This response file includes: (1) the point-to-point response letter to the first reviewer; (2) the point-to-point response letter to the second reviewer; and (3) a marked-up manuscript version.
1. General Comments

The authors took the time to address the issues pointed at by the first round of reviews. The updated manuscript now successfully presents robust and interesting scientific results. It can now be published with only few remaining technical modifications.

The effort to compare the present results to most of the available literature in Sect. 4.2 is really appreciated, as the works on the Arctic are quite scattered and not often compared in comprehensive reviews.

Response: We appreciate the valuable comments from the reviewer. These comments help us improve the manuscript in both readability and scientific values.

2. Technical comments

p.4 l.83: Shakhova’s papers are highly controversial and should not be cited as an absolute reference. More recent works suggest that hydrates emissions to the atmosphere are not that significant in the Arctic. Please prefer some of the following publications rather than Shakhova’s. For Svalbard: Grave et al. (2015; doi: 10.1002/2015JC011084), Lund Myhre et al. (2016; doi: 10.1002/2016GL068999). For Laptev: Berchet et al. (2016; doi: 10.5194/acp-16-4147-2016), Stranne et al. (2016; doi: 10.1002/2015GC006119) or Thornton et al. (2016; 15 doi: 10.1002/2016GL068977).

Response: Thank you for indicating this! We added Berchet et al. (2016) and Myhre et al. (2016) as references here and removed Shakhova’s paper. In addition, for other places referring to methane emissions from East Siberian Shelf, we also added Thornton et al. (2016) as a reference.


p.18 l.388: Sect. 4.1, 4.2, 4.3 are 20, 100 and 30 lines long respectively, which makes the result discussion quite unbalanced. Sect. 4.2 is well structured with high quality content, but please consider splitting it into sub-parts to guide the reader in the discussion.

Response: In the revision, we split Sect. 4.2 into four sub-parts: 4.2.1) Regional CH4 Emissions; 4.2.2) CH4 Emissions from Pan-Arctic Lakes; 4.2.3) CH4 Emissions from Pan-Arctic Wetlands; 4.4.4) Evaluation of Pan-Arctic CH4 Inversions.

p.11 l.221: in "GEOS-5 meteorological (met)", “met” looks a little bit clumsy when reading it the first time. Maybe replace by something like “GEOS-5 meteorological (hereafter GEOS-5 met)”, or more elegant.

Response: We have revised it as suggested.

p.21 l.465: Berchet et al. (2015) applies a regional atmospheric inversion as in this manuscript with surface atmospheric sites and not “flux towers”. Please reformulate this sentence.

Response: We have revised this sentence as “Using the atmospheric CH4 observation data at several sites near Siberian wetlands, Berchet et al. (2015) estimated that CH4 emissions from Siberian wetlands were in the range of 1–13 Tg CH4 yr⁻¹, wider than our estimated range.”.

fig. 1: Siberia looks quite empty here, which is less and less true, fortunately. Somewhere in the discussion should be mentioned the effort by different teams to put instruments in Siberia: JR-STATION by NIES, ZOTTO by MPI, one site near Laptev Sea by FMI, etc. Environment Canada maintains continuous sites in North American Arctic as well. One or two sentences should acknowledge that using all these sites in an inversion system (possible follow-up of the present paper) should improve the inversion results and might reduce (or not?) the gains of using satellite data (though they would be always welcome).

Response: We added the following sentences in our revision: “As shown in Fig. 1, our inverse modeling assimilated few high-precision surface CH4 measurements in Siberia and northern Canada. Since some efforts have already been made by different teams to measure atmospheric CH4 routinely in Siberia (e.g., the JR-STATION network by NIES, the Zotino Tall Tower Observatory by MPI-BGC and the Tiksi site by the Finnish Meteorological Institute) and in North American Arctic (e.g., the Behchoko site by Environment Canada), we would like to take advantage of these measurements to further improve our inversion results and re-evaluate the gains of using satellite data in our future studies.”.
The manuscript has improved significantly, and can be accepted with one final correction: The text describing figure 6 mentions an 'impressive improvement' between the blue end red distributions that are shown. To me, however, they look about the same. The caption doesn't explain the red and blue line extending to the top of the figure, but I guess they represent the means (or medians?). I'm sure that a student t-test of the significance of the difference between these distributions wouldn't justify calling this an 'impressive improvement'. The figure should be explained better in the caption (please also add a label on the y-axis), and the corresponding text should be made compatible with the statistical significance of this result.

Response: We thank the reviewer’s valuable comments for helping improve this manuscript. In the revision, we added the label “Number of SCIAMACHY retrievals” on the y-axis. We also added the following description of the extending red and blue lines “Two extending red and blue lines represent the means of the simulation bias under the “DLEM + Lake” scenario and the “DLEM only” scenario, respectively.”. We tested the significance of the difference between two simulations using a two-sample t-test (MATLAB “ttest2” function). It shows that their means are significantly different: \( p = 0.0032838 < 0.05 \). We revised the text accordingly “A further comparison of model-satellite agreement between the DLEM scenario and this no-lake scenario reveals that the agreement improves when lake emissions are considered (see Fig. 6; \( p = 0.0032838 \) at the two-sample t-test).”.
Inverse modeling of pan-Arctic methane emissions at high spatial resolution: What can we learn from assimilating satellite retrievals and using different process-based wetland and lake biogeochemical models?

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Abstract: Understanding methane emissions from the Arctic, a fast warming carbon reservoir, is important for projecting future changes in the global methane cycle. Here we optimized methane emissions from north of 60°N (pan-Arctic) regions using a nested-grid high-resolution inverse model that assimilates both high-precision surface measurements and column-average SCIAMACHY satellite retrievals of methane mole fraction. For the first time, methane emissions from lakes were integrated into an atmospheric transport and inversion estimate, together with prior wetland emissions estimated with six different biogeochemical models. In our estimates, in 2005, global methane emissions were in the range of 496.4–511.5 Tg yr\(^{-1}\) and pan-Arctic methane emissions were in the range of 11.9–28.5 Tg yr\(^{-1}\). Methane emissions from pan-Arctic wetlands and lakes were 5.5–14.2 Tg yr\(^{-1}\) and 2.4–14.2 Tg yr\(^{-1}\), respectively. Methane emissions from Siberian wetlands and lakes are could be the largest and also have the largest uncertainty. Our results indicate that the uncertainty introduced by different wetland models could be much larger than the uncertainty of each inversion. We also show that assimilating satellite retrievals can reduce the uncertainty of the nested-grid inversions. The significance of lake emissions cannot be identified across the pan-Arctic by high-resolution inversions but it is possible to identify high lake emissions from in some specific regions. In contrast to global inversions, high-resolution nested-grid inversions perform better in estimating near surface CH\(_4\)-methane concentrations.
1. Introduction

Methane (CH$_4$) is the second most powerful carbon-based greenhouse gas in the atmosphere behind carbon dioxide (CO$_2$) and also plays a significant role in the cycles of ozone, hydroxyl radicals (OH) and stratospheric water vapor (Myhre et al., 2013; Shindell et al., 2009). The atmospheric burden of CH$_4$ is now more than factor of 2.5 greater than the pre-industrial value of about 700 ppb (Etheridge et al., 1998), mainly due to anthropogenic emissions. Major sources and sinks of CH$_4$ have been identified (Denman et al., 2007); however their quantification is still of large uncertainties and the annual and inter-annual variability of atmospheric CH$_4$ are not well explained. For instance, scientists have not yet agreed on what caused the leveling off of atmospheric CH$_4$ since the 1980s (Dlugokencky et al., 2003; Bousquet et al., 2006; Aydin et al., 2011; Kai et al., 2011; Levin et al., 2012; Simpson et al., 2012; Kirschke et al., 2013) and the recent rebounding of its growth since 2007 (Rigby et al., 2008; Dlugokencky et al., 2009; Nisbet et al., 2014).

To reduce the quantification uncertainty of CH$_4$ sources and sinks, much effort has been made using Bayesian inference (Bergamaschi et al., 2007, 2009, 2013; Meirink et al., 2008; Cressot et al., 2014; Houweling et al., 2014; Alexe et al., 2015). In these studies, in-situ and/or satellite observations of CH$_4$ that are representative of large spatial scales were assimilated into a chemical transport model (CTM) to constrain the initial estimates of CH$_4$ sources and sinks that are inventoried from field studies, industrial investigations and biogeochemical models (Fung et al., 1991; Zhuang et al., 2004; Walter et al., 2006; Zhu et al., 2013; Tan and Zhuang, 2015a and 2015b). Space-borne observations of atmospheric CH$_4$ are especially useful in inverse modeling because they can deliver dense and continuous coverage unachievable by surface networks or aircraft campaigns (Bergamaschi et al., 2007). There are two types of nadir satellite CH$_4$
retrievals: one from solar backscatter in the shortwave infrared (SWIR) and the other from thermal infrared radiation (TIR). Between them, SWIR retrievals were more widely used in atmospheric inversion of CH$_4$ emissions (Bergamaschi et al., 2007, 2009, 2013; Fraser et al., 2013; Cressot et al., 2014; Houweling et al., 2014; Monteil et al., 2014; Wecht et al., 2014; Alexe et al., 2015; Turner et al., 2015) because they can provide column concentrations with near-uniform vertical sensitivity down to the surface. To date, most of the inversions were operated at coarse spatial resolutions over 300 km. However, partly owing to their coarse resolutions, it is impossible for these inversions to constrain different CH$_4$ sources that are spatially co-located (Fung et al., 1991; Wecht et al., 2014). To address this issue, regional inverse models at fine spatial resolutions were developed (Miller et al., 2013; Wecht et al., 2014; Thompson et al., 2015). For example, Wecht et al. (2014) and Turner et al. (2015) have used the 1/2° × 2/3° horizontal resolution GEOS-Chem adjoint model to constrain CH$_4$ emissions over North America.

Estimating CH$_4$ emissions from the Arctic is important for understanding the global carbon cycle because the fast warming of Arctic permafrost, one of the largest organic carbon reservoirs (Tarnocai et al., 2009), could lead to a rapid rise of CH$_4$ emissions (Zhuang et al., 2006; Walter et al., 2007; Koven et al., 2011). Natural sources dominate the Arctic CH$_4$ inventory (Fisher et al., 2011), e.g. wetlands (McGuire et al., 2012), lakes (Walter et al., 2006; Bastviken et al., 2011), sea shelves (Berchet et al., 2016; Myhre et al., 2016; Shakhova et al., 2013) and oceans (Kort et al., 2012). As the factors governing natural CH$_4$ production (methanogenesis) and oxidation (methanotrophy) are notoriously heterogeneous, estimates of Arctic CH$_4$ emissions are still poorly constrained, even with decades of site-level and modeling studies (Zhuang et al., 2004; Bastviken et al., 2011; Schuur et al., 2015; Tan and Zhuang, 2015a; Tan and Zhuang,
Previous CH$_4$ inversions over the Arctic only assimilated surface measurements that were too sparse to constrain fine-scale CH$_4$ fluxes. Also, possibly important CH$_4$ sources that were newly identified, e.g. CH$_4$ emissions from Arctic lakes (Walter et al., 2006 and 2007; Bastviken et al., 2011; Tan and Zhuang, 2015a) and the East Siberian Shelf (Shakhova et al., 2013; Berchet et al., 2016; Thornton et al., 2016) have not been included in these studies. Given the ill-posed nature of trace-gas inversions, realistic prior fluxes could be important for successful inverse modeling of CH$_4$ emissions from the Arctic (Kaminski and Heimann, 2001).

To address these issues, we used the adjoint of a 3-D CTM at a high spatial resolution (less than 60 km) to improve the quantification of pan-Arctic CH$_4$ emissions in 2005. We explored the feasibility of using satellite CH$_4$ retrievals overpassing the pan-Arctic to further constrain regional CH$_4$ emissions. For the first time, CH$_4$ emissions from pan-Arctic lakes were included in high-resolution inverse modeling of CH$_4$ emissions. As wetland emissions are likely the largest pan-Arctic CH$_4$ source, we also investigated the sensitivity of our estimates to the use of different wetland emission scenarios. Section 2 describes the observation data of atmospheric CH$_4$ that were used to infer CH$_4$ emissions and evaluate posterior estimates. Section 3 details the wetland and lake biogeochemical models that were used in this study (Section 3.1), the pan-Arctic nested-grid CTM (Section 3.2), and the adjoint-based inversion method (Section 3.3). Section 4 presents the posterior CH$_4$ emissions, their evaluation and further discussion.

2. Observations

2.1. Satellite Retrievals

SWIR CH$_4$ retrievals are available from SCanning Imaging Absorption spectroMeter for Atmospheric CHartogrpHy (SCAMACHY) for 2003–2012 (Frankenberg et al., 2006, 2008, 2011)
and Greenhouse Gases Observing SATellite (GOSAT) for 2009 to present (Parker et al., 2011). SCIAMACHY, aboard the European Space Agency’s environmental research satellite ENVISAT, retrieves column-averaged CH$_4$ mixing ratios ($X_{CH_4}$) from the SWIR nadir spectra (channel 6: 1.66–1.67 µm) using the IMAP-DOAS algorithm (Frankenberg et al., 2006, 2008, 2011). The satellite operates in a near polar, sun-synchronous orbit at an altitude of 800 km. At channel 6, the ground pixel size of the retrievals is about 30 km (along-track) × 60 km (across-track). We use version 6.0 proxy CH$_4$ retrievals from Frankenberg et al. (2011) that provide a weighted column average dry-mole fraction of CH$_4$ with 10-layer averaging kernels and prior CH$_4$ profiles. The averaging kernels show near-uniform vertical sensitivity in the troposphere and declining sensitivity above the tropopause (Butz et al., 2010). Some auxiliary data, e.g. the air mass factor $A_F$ ($A_F = 1/\cos \theta + 1/\cos \xi$, where $\theta$ is the solar zenith angle and $\xi$ is the viewing angle of the satellite), water column density and dry air column density, are also published with the IMAP-DOAS v6.0 XCH$_4$ product.

The estimated single-retrieval precision is scene-dependent and averages roughly 1.5% or 25 ppb (Frankenberg et al., 2011). With this order of instrument precision, SCIAMACHY cannot resolve day-to-day variability of emissions but can strongly constrain a multi-year average (Turner et al., 2015). The retrieving algorithm firstly calculates CH$_4$ total column density $\Omega_{CH_4}$ (molecules cm$^{-2}$):

$$\Omega_{CH_4} = \Omega_A + a^T (\omega - \omega_A)$$ (1)

where $\omega$ is the true 10-layer sub-column densities of CH$_4$ (molecules cm$^{-2}$), $\omega_A$ is the 10-layer prior CH$_4$ sub-column density (molecules cm$^{-2}$), $\Omega_A$ is the corresponding a priori CH$_4$ total column density, and $a$ is an averaging kernel vector that defines the sensitivity of the retrieved total column to each sub-column in $\omega$. To account for the impact of aerosol scattering and
instrument effects on the observed light path, Frankenberg et al. (2006) used the CO2 column density $\Omega_{CO_2}$ as a proxy to normalize and convert $\Omega_{CH_4}$ to a column mixing ratio $X_{CH_4}$ (ppb):

$$X_{CH_4} = \left(\frac{\Omega_{CH_4}}{\Omega_{CO_2}}\right)X_{CO_2}$$  \hspace{1cm} (2)

where $X_{CO_2}$ is the column-weighted mixing ratio of CO2 from NOAA’s CarbonTracker CO2 measurement and modeling system. CO2 is used as a proxy because it is retrieved in a spectrally neighboring fitting window and, relative to CH4, its mixing ratio is known with much higher precision.

The quality of SCIAMACHY observations is controlled by a filtering scheme that selects only daytime, over land and with cloud free or partially cloud scenes and good fitting accuracy (http://www.temis.nl/climate/docs/TEMIS_SCIA_CH4_IMAPv60_PSD_v2_6.pdf). Further, a surface elevation filter is applied to filter out observations that are different from the model grids at surface altitude by more than 250 m (Bergamaschi et al., 2009; Alexe et al., 2015). This filtering process ensures that the atmospheric columns seen by SCIAMACHY are well represented by the model columns. To avoid spurious outliers that may have a large impact on the inversion, $X_{CH_4}$ retrievals of less than 1500 ppb or larger than 2500 ppb are discarded (Alexe et al., 2015). For the pan-Arctic, most of qualified $X_{CH_4}$ retrievals were recorded in the summer time when local solar zenith angles are higher, surface reflectance is lower and impact of Arctic vortex is smaller. Fig. 1 shows the SCIAMACHY retrievals ($n = 37743$) of the weighted column-average CH4 dry mixing ratio for July 2005–September 2005 in the pan-Arctic that have passed all quality control tests.

2.2. Surface Observations
The NOAA/ESRL Carbon Cycle Cooperative Global Air Sampling Network provides high-precision weekly flask measurements of surface atmospheric CH$_4$ dry-air mole fraction (Dlugokencky et al., 2014) that were calibrated against the WMO X2004 CH$_4$ standard scale maintained at NOAA (Dlugokencky et al., 2005). Due to the coarse resolution of the GEOS-Chem model, we include only marine and continental background sites and exclude sites that are strongly influenced by sub-grid local sources (Alexe et al., 2015), as listed in Table S1. The flask-air samples in the NOAA/ESRL network that were taken from regular ship cruises in the Pacific Ocean serve to evaluate simulated surface mixing ratios of global inversions over the remote ocean and downwind the continental sources (Alexe et al., 2015). Fig. 1 shows the Arctic sites that were used for data assimilation and nested-grid inversion evaluation.

2.3. Aircraft Campaign Observations

To derive the bias of SCIAMACHY CH$_4$ retrievals overpassing the pan-Arctic and evaluate the modeled CH$_4$ vertical profiles in the troposphere, we used CH$_4$ measurements that were collected by three aircraft campaigns: the NOAA/ESRL Carbon Cycle Cooperative Global Air Sampling Network’s aircraft program (http://www.esrl.noaa.gov/gmd/ccgg/aircraft/data.html; Sweeney et al., 2015), the National Institute for Environmental Studies (NIES) aircraft program (Machida et al., 2001; Sasakawa et al., 2013), and the NASA’s Arctic Research of the Composition of the Troposphere from Aircraft and Satellite (ARCTAS) mission. For the NOAA/ESRL aircraft mission, CH$_4$ was routinely collected using 0.7 L silicate glass flasks on planned flights with maximum altitude limits of 300–350 hPa. The sampling vertical resolution is up to 400 m in the boundary layer and all samples were analyzed by NOAA/ESRL in Boulder, Colorado. For the NIES aircraft mission, air samples were collected in 550 mL glass flasks over Surgut, West Siberia (61.5°N, 73.0°E) at altitude ranging from 0.5 to 7 km with 0.5–1.5 km
intervals. The precision of gas chromatograph analysis for CH$_4$ measurement was estimated to be 1.7 ppb and the NIES-94 scale used in analysis was higher than the NOAA/GMD scale by 3.5–4.6 ppb in a range of 1750–1840 ppb. In ARCTAS, CH$_4$ was measured over northern Canada by the DACOM tunable diode laser instrument with an estimated accuracy/precision of 1%/0.1%. Central locations of their flights in the pan-Arctic are shown in Fig. 1. Table S2 lists the locations and profiles of the NOAA/ESRL aircraft mission flights used in evaluation.

3. Modeling

Here we describe the prior emissions, the forward model, and the inversion method used to optimize CH$_4$ emissions in the pan-Arctic on the basis of SCIAMACHY and NOAA/ESRL observations.

3.1. Wetland and Lake CH$_4$ Emissions

CH$_4$ emissions estimated by the inverse modeling method can be sensitive to the choice of prior wetland CH$_4$ fluxes (Bergamaschi, 2007). To assess this sensitivity, we used wetland CH$_4$ emissions simulated by six well-known wetland biogeochemical models (CLM4Me, DLEM, LPJ-Bern, LPJ-WSL, ORCHIDEE and SDGVM) to setup six different inverse modeling experiments. All wetland CH$_4$ simulations follow the same protocol of WETland and Wetland CH$_4$ Inter-comparison of Models Project (WETCHIMP) as described in Melton et al. (2013) and Wania et al. (2013). Melton et al. (2013) demonstrated that the difference of these estimates primarily arises from the model distinction in CH$_4$ biogeochemistry and wetland hydrology. These models estimated that the annual global CH$_4$ emissions from wetlands during 2004–2005 were in the range of 121.7–278.1 Tg yr$^{-1}$ (Fig. S1) and wetland CH$_4$ emissions are the highest in tropical regions (e.g., Amazon, Southeast Asia and Tropical Africa) where extensive floodplains
and warm environment coexist. In the pan-Arctic, the modeled annual wetland CH$_4$ emissions in 2005 were in the range of 9.1–20.9 Tg yr$^{-1}$ (Fig. 2), and their spatial distribution was mainly controlled by the modeled or mapped wetland coverage (Melton et al., 2013). As shown in Fig. 2, because of some consistency in simulating wetland hydrology, nearly all models suggest that there are high CH$_4$ fluxes in West Siberia Lowlands, Finland and Canadian Shield.

Lakes, permanent still-water bodies without direct connection to the sea, are abundant in the pan-Arctic (Lehner and Döll, 2004). Recent studies indicated that pan-Arctic lakes could contribute a significant amount of CH$_4$ to the atmosphere (Walter et al., 2006; Tan and Zhuang, 2015a) and the emissions could be driven by factors different from wetland emissions, e.g. the supply of labile yedoma permafrost carbon (Walter et al., 2006) and water deep mixing (Schubert et al., 2012). Because the WETCHIMP models cannot account for this source, we used a one-dimension process-based lake biogeochemical model, bLake4Me, to simulate CH$_4$ emissions from pan-Arctic lakes (Tan et al., 2015; Tan and Zhuang, 2015a). The bLake4Me model explicitly parameterizes the control of temperature and carbon substrate availability on methanogenesis, the control of temperature and oxygen level on methanotrophy and the transport of gaseous CH$_4$ by diffusion and ebullition. A detailed model description and evaluation can be found in Tan et al. (2015). Model quantification of CH$_4$ emissions from all lakes north of 60$^\circ$N was described by Tan and Zhuang (2015a and 2015b). On average, the estimated CH$_4$ emissions from pan-Arctic lakes during the studied period are approximately 11 Tg CH$_4$ yr$^{-1}$, see Fig. 2.

3.2. GEOS-Chem Model

Atmospheric CH$_4$ mole fractions are simulated by GEOS-Chem v9-01-03 (http://acmg.seas.harvard.edu/geos/index.html), a global 3-D CTM model (Bey et al., 2001). For
the period of 2004–2005, GEOS-Chem is driven by GEOS-5 meteorological (hereafter GEOS-5 met) data from NASA’s Global Modeling Assimilation Office (GMAO). The GEOS-5 met data have horizontal resolution of 1/2° latitude × 2/3° longitude, temporal resolution of 6 hours and 72 hybrid sigma-pressure levels extending from Earth’s surface to 0.01 hPa. In contrast to the global GEOS-Chem model, the nested-grid version does not include algorithms for handling advection near the North and South Poles (Lin and Rood, 1996). To avoid polar grid boxes, we crop the native 1/2° × 2/3° resolution GEOS-5 met data to a window region (180°W–180°E and 80°N–56°N) for the pan-Arctic nested grid. To make it consistent with the bLake4Me model, only CH₄ emissions north of 60°N are analyzed. We expect that the avoidance of the North Pole only has a minor impact on our inversions because according to Miyazaki et al. (2008) the Northern Hemisphere (NH) extratropics during summer has slow mean-meridional circulation and inactive wave activity but strong vertical transport. Boundary conditions for nested grid simulations are produced using the same period GEOS-Chem 4° × 5° resolution global scale forward runs at 3-hour intervals.

The GEOS-Chem CH₄ simulation was originally introduced by Wang et al. (2004) and updated by Pickett-Heaps et al. (2011). As described by Wecht et al. (2014), the prior anthropogenic sources, including oil/gas production, coal mining, livestock, waste treatment, rice paddies, biofuel burning and other processes, were extracted from Emission Database for Global Atmospheric Research v4.2 (EDGAR4.2) with 0.1° × 0.1° resolution and no seasonality (European Commission, Joint Research Centre/Netherlands Environmental Assessment Agency, 2009). CH₄ emissions from termites and biomass burning were obtained from the study of Fung et al. (1991) and daily Global Fire Emissions Database Version 3 (GFED3) of van der Werf et al. (2010), respectively. CH₄ emissions from wetlands and lakes were simulated by biogeochemical
models described in Section 3.1. Atmospheric CH$_4$ is mainly removed by tropospheric oxidation initiated by reaction with tropospheric OH, which was computed using a 3-D OH climatology of monthly average concentrations from a previous simulation of tropospheric chemistry (Park et al., 2004). The global mean pressure-weighted tropospheric OH concentration is $10.8 \times 10^5$ molecules cm$^{-3}$. For minor sinks, CH$_4$ uptake by upland soils was derived from Fung et al. (1991) and CH$_4$ oxidation in the stratosphere was calculated from the archived CH$_4$ loss frequency described by Murray et al. (2012). The resulting atmospheric lifetime of CH$_4$ is about 8.9 years, consistent with the observational constraint of 9.1±0.9 years (Prather et al., 2012). We re-gridded and cropped the anthropogenic and natural CH$_4$ emissions in EDGAR4.2, GFED3 and Fung et al. (1991) for our nested pan-Arctic domain using the Harvard-NASA Emissions Component (HEMCO) software (Keller et al., 2014), marked as “other” in Fig. 2. Compared to CH$_4$ emissions from natural sources, these emissions are relatively small in 2005 (~2.1 Tg yr$^{-1}$).

3.3. Inversion Method

Atmospheric inversion is a procedure for using observations of atmospheric gases as constraints to estimate surface gas fluxes. The inverse problem can be characterized by solution of

$$ y = F(x) + \varepsilon $$

(3)

By applying Bayesian theorem and assuming Gaussian errors, the inverse problem can be solved by minimizing the cost function, $J(x)$, that measures the model deviations from both prior assumptions and observations (Enting et al., 2002; Kopacz et al., 2009):

$$ J(x) = (F(x) - y)^\top C_d^{-1}(F(x) - y) + \gamma (x - x_0)^\top C_{x_0}^{-1}(x - x_0) $$

(4)
where $y$ is a vector of observations from SCIAMACHY and NOAA/ESRL, $F$ is a model operator that maps emissions to observations, $x$ represents CH$_4$ emissions to be constrained, $x_0$ is the a priori estimate of $x$, $C_d$ is the observational error covariance matrix that includes contributions from model error, representation error (sampling mismatch between observations and the model) and measurement error, and $C_{x_0}$ is the parameter error covariance matrix (containing the uncertainties of the parameters and their correlations). The regularization parameter $\gamma$ controls the relative constraints applied by the observational and a priori parts of $J(x)$ (Kopacz et al., 2009). In the adjoint method, $\gamma$ is not fixed at unity but determined by analyzing its influence on the minimum of $J(x)$ (Henze et al., 2007; Kopacz et al., 2009).

Minimization of $J(x)$ yields the following expression for the maximum a posteriori solution for the state vector $\hat{x}$ and its associated error covariance $\hat{C}_x$ (Rodgers, 2000):

$$\hat{x} = x_0 + \left( (\nabla_x F)^T C_d^{-1} \nabla_x F + \gamma C_{x_0}^{-1} \right)^{-1} \left( (\nabla_x F)^T C_d^{-1} (y - F(x_0)) \right)$$  \hspace{1cm} (5)$$

$$\hat{C}_x^{-1} = (\nabla_x F)^T C_d^{-1} \nabla_x F + \gamma C_{x_0}^{-1}$$  \hspace{1cm} (6)$$

where $\nabla_x F$ is the Jacobian matrix of the forward model. $J(x)$ is minimized iteratively through successive forward and backward simulations with the GEOS-Chem model and its adjoint, developed by Henze et al. (2007) and previously applied to CO, CO$_2$ and CH$_4$ source inversions (Jiang et al., 2011; Deng et al., 2014; Wecht et al., 2014). The GEOS-Chem adjoint model is a 4DVAR inverse modeling system that allows optimization of a very large number of parameters using at the same time very large sets of observational data, such as satellite data. Rather than optimizing CH$_4$ emissions directly, it optimizes an exponential scale factor $e_x$ ($e_x = \ln(x/x_0)$) at each grid cell to avoid negative emissions. The posterior error covariance $\hat{C}_x$ could be
approximated by the Davidon-Fletcher-Powell (DFP) or the Limited-memory Broyden–Fletcher–
Goldfarb–Shanno (L-BFGS) optimization algorithm (Singh et al., 2011; Deng et al., 2014). But
the performances of these deterministic methods are usually not promising, subjecting to the
choice of initial Hessian, so-called preconditioning (Bousserez et al., 2015). In contrast,
approximating $\hat{C}_x$ by stochastic methods, i.e. Monte-Carlo sampling and gradient-based
randomization, could help avoid the impact of setting initial Hessian (Bousserez et al., 2015). For
example, Bousserez et al. (2015) demonstrated that for high-dimensional inverse problems using
a Monte Carlo stochastic approach that samples ensemble members by perturbing $x_0$ and $y$ in
line with $C_{x_0}$ and $C_d$ respectively, could guarantee a low relative error (10%) in the variance with
as few as 50 members. In this study, the posterior uncertainty of nested-grid inversions was
estimated using this method.

For prior emissions, their uncertainties were set as 100% in each grid box and spatial
correlation was set as an e-folding function with spatial correlation lengths of 500 km at the
global $4^\circ \times 5^\circ$ resolution and of 300 km at the nested grid $1/2^\circ \times 2/3^\circ$ resolution (Bergamaschi et
al., 2009). Six global coarse-resolution inversions using different wetland emission scenarios and
assimilating both surface CH$_4$ measurements and satellite CH$_4$ retrievals were performed during
the period of 2005/01–2005/12. These inversions provided boundary conditions for the following
nested-grid inversions. For $1/2^\circ \times 2/3^\circ$ nested-grid inversions, we ran the adjoint model for 50
times over the period of 2005/07–2005/09 for each of twelve scenarios: six wetland scenarios by
two data assimilation scenarios. The two data assimilation scenarios include one scenario
assimilating only NOAA/ESRL measurements and another scenario assimilating both
NOAA/ESRL measurements and SCIAMACHY retrievals. As described above, the 50-member
ensemble run is for the calculation of posterior estimate uncertainty. The steps to construct
optimal initial conditions for global and nested inversions are described in the supplementary materials. As in Wecht et al. (2014), observations in the first week were not assimilated and each optimization was run iteratively at least 40 times until the reduction of its cost function became less than 0.5% with each successive iteration. In the GEOS-Chem adjoint model, optimization changes its course automatically if local minimum reaches.

3.4. Satellite Retrieval Bias Correction

The importance of bias correction for the assimilation of satellite retrievals has been discussed in many earlier studies (Bergamaschi et al., 2007, 2009, 2013; Fraser et al., 2013; Cressot et al., 2014; Houweling et al., 2014; Wecht et al., 2014; Alexe et al., 2015; Turner et al., 2015). Usually, these studies represented satellite retrieval bias as a regression function of one proxy parameter, i.e. latitude, air mass factor or specific humidity. Air mass factor was used as a proxy parameter by some studies due to its correlation to spectroscopic errors and residual aerosol errors (Cressot et al., 2014; Houweling et al., 2014) and specific humidity was used because water vapor is the main cause of SCIAMACHY seasonal bias that lags the variations of solar zenith angle (Houweling et al., 2014). Relative to air mass factor and humidity, latitude can represent the changes in both solar zenith angle and climate variables (Bergamaschi et al., 2007, 2009 and 2013) and thus was used by more studies. Considering that different proxies can account for different errors, the system bias of satellites may be better represented by multiple proxy parameters.

To test this hypothesis, we compared the performance of three traditional one-proxy methods (latitude $\phi$, air mass factor $A_F$, specific humidity $H_S$) and two new two-proxy methods (latitude + humidity, air mass factor + humidity), listed in Table 1. These methods were
evaluated using two reference values: the difference between the satellite-retrieved and the GEOS-Chem modeled CH$_4$ column mixing ratios and the Bayesian Information Criterion (BIC) score. The BIC criterion is widely used for regression model selection and aims to award a model that fit measurements with the least model parameters. In the study, we would select the bias correction method that gives the smallest difference and the lowest BIC score. In our experiments, all bias correction functions were updated monthly. As listed in Table 1, the “latitude only” correction performs the best among the three single-proxy correction methods and is only slightly worse than the “latitude + humidity” correction method. The “air mass factor only” method does not work as well in our experiment. Turner et al. (2015) suggested that it could be attributed to a potential bias in the GEOS-Chem simulation of CH$_4$ in the polar stratosphere. As the “latitude + humidity” method has the smallest model-data difference and the lowest BIC score, we applied it for satellite bias correction in all global inversions.

For SCIAMACHY retrievals overpassing the pan-Arctic, because the modeled atmospheric CH$_4$ could be less reliable, we used another bias correction method. According to a comparison between SCIAMACHY and the high-precision Total Carbon Column Observing Network (TCCON) measurements, the system bias of SCIAMACHY retrievals could be closely correlated with specific humidity averaged over the lowest 3 km of the atmosphere (Houweling et al., 2014). And Wecht et al. (2014) has demonstrated that this humidity-proxy method shows promising performance in debiasing SCIAMACHY retrievals overpassing North America. In this study, we sought a similar linear regression relationship between SCIAMACHY bias and specific humidity. First, we detected the SCIAMACHY bias by comparing SCIAMACHY retrievals with CH$_4$ vertical profiles measured by the NOAA/ESRL aircraft mission over Alaska, USA, the NIES aircraft mission over Siberia, Russia and the NASA/ARCTAS aircraft mission
over Alberta, Canada. Before comparison, these CH$_4$ vertical profiles had been mapped to the SCIAMACHY retrieval pressure grid using Eq. (1) and (2). Fig. 3 (left) shows that the retrieved system bias ($\Delta$XCH$_4$) has a negative relationship with air humidity. Because the pan-Arctic is normally dry, SCIAMACHY retrievals could be lower than atmospheric CH$_4$ column average mixing ratios in most of days.

After bias correction, the error variances of SCIAMACHY retrievals were estimated using the relative residual error (RRE) method described by Heald et al. (2004). Fig. S2 shows the error variances of SCIAMACHY retrievals in the global scale and Fig. 3 (right) shows the error variances in the nested grid. In both global and nested grid inversions, the total error of individual SCIAMACHY retrievals is assumed to be at least 1.5% (Bergamaschi et al., 2007; Frankenberg et al., 2011). The observational error of the NOAA/ESRL CH$_4$ mixing ratios is estimated as the sum of measurement error (~0.2%) and representation error. Similar to satellite retrievals, the representation error of surface measurements is defined as the standard deviation of surface CH$_4$ concentration differences between NOAA/ESRL measurements and GEOS-Chem.

4. Results and Discussion

4.1. Optimized Global CH$_4$ Emissions

As listed in Table 2, when both NOAA/ESRL measurements and SCIAMACHY retrievals were assimilated, the posterior estimates of total emissions in 2005 show good convergence at a narrow range of 496.4–511.5 Tg CH$_4$ yr$^{-1}$, albeit our six prior scenarios span in a wide range (471.5–627.8 Tg CH$_4$ yr$^{-1}$). Because the total of global emissions is constrained by the atmospheric CH$_4$ burden and lifetime, this convergence probably suggests that surface
measurements from the NOAA/ESRL network are of sufficient density and accuracy to represent
the global CH$_4$ burden if the CH$_4$ lifetime is correct. In contrast, the posterior CH$_4$ emissions
differ largely between different wetland emission scenarios in the TransCom3 land regions. For
example, in the DLEM inversion, the estimated CH$_4$ emissions from the Eurasian temperate
region are as large as 146.1 Tg CH$_4$ yr$^{-1}$. But in the CLM inversion, the total of these emissions is
only 84.9 Tg CH$_4$ yr$^{-1}$. Also, for CH$_4$ emissions from the South American tropical region, the
estimate is 31.4 Tg CH$_4$ yr$^{-1}$ in the DLEM inversion but nearly two times larger (62.3 Tg CH$_4$ yr$^{-1}$)
in the SDGVM inversion. There are several possible explanations for the large differences
between the scenarios: high-precision surface measurements could be not of sufficient density in
regional scales, satellite retrievals could be not of sufficient accuracy, and the GEOS-Chem
model and its priors could be not of high temporal and spatial resolutions to resolve satellite
retrievals. A detailed comparison between our estimates and previous inversion studies at the
global scale is presented in the supplementary materials.

4.2. Optimized Pan-Arctic CH$_4$ Emissions

4.2.1. Regional CH$_4$ Emissions

When using both surface measurements and satellite retrievals, our estimated CH$_4$
emissions over the pan-Arctic are in the range of 11.9–28.5 Tg CH$_4$ yr$^{-1}$. The simulation is the
largest in the ORCHIDEE scenario and the smallest in the SDGVM scenario: 24.9±3.6 Tg CH$_4$
yr$^{-1}$ and 16.1±4.2 Tg CH$_4$ yr$^{-1}$, respectively. Regionally, posterior CH$_4$ emissions from Alaska,
northern Canada, northern Europe and Siberia are 0.3–3.4 Tg CH$_4$ yr$^{-1}$, 1.3–7.9 Tg CH$_4$ yr$^{-1}$, 0.8–
8.1 Tg CH$_4$ yr$^{-1}$ and 4.4–14.9 Tg CH$_4$ yr$^{-1}$, respectively. Same as the global inversions, the
difference of the nested-grid inversions between different scenarios is much larger than the total
uncertainty of priors and observations of each scenario: 16.6 Tg CH$_4$ yr$^{-1}$ vs. 5.5 Tg CH$_4$ yr$^{-1}$. In
these regions, CH₄ emissions from Siberia are more uncertain (Fig. 5), a possible indication of
the lack of high-quality measurements in Siberia for assimilation. Our results also indicate that
the assimilation of SCIAMACHY retrievals overpassing the pan-Arctic can reduce the estimate
uncertainty. For example, for the BERN scenario, the posterior uncertainty is about 18%, much
smaller than the inversion that only assimilates NOAA/ESRL measurements (27%). And for the
CLM scenario, the posterior uncertainty increases from 16% to 23% when only surface
measurements were assimilated. Our estimates are consistent with other inverse modeling
estimates. For example, Kirschke et al. (2013) reviewed a series of top-down estimation of CH₄
emissions and suggested that CH₄ emissions north of 60°N could be in the range of 12–28 Tg
CH₄ yr⁻¹, very close to our estimate. This consistency could reflect the robustness of our nested-
grid GEOS-Chem adjoint model and the good constraint of the NOAA/ESRL sites over the pan-
Arctic on the atmospheric CH₄ field. Our estimates also imply that CH₄ emission from the pan-
Arctic could constitute a large fraction of CH₄ emissions in the northern high latitudes (> 50°N).
Based on the estimate (50 Tg CH₄ yr⁻¹) of Monteil et al. (2013), we calculated that 29.2–60.8% of
CH₄ emissions in the northern high latitudes could be emitted from the pan-Arctic (> 60°N). For
all scenarios, the inverse modeling adjusts total CH₄ emissions downward compared to prior
emissions. It is possible that CH₄ emissions are overestimated by the biogeochemical models or
double counted between the wetland and lake models or both. This adjustment could also be
explained by the underestimate of CH₄ absorption by soils in biogeochemical models due to the
missing of high-affinity methanotrophy (Oh et al., 2016).

4.2.2. CH₄ Emissions from Pan-Arctic Lakes

In contrast to CH₄ emissions from pan-Arctic wetlands, CH₄ emissions from pan-Arctic
lakes at large spatial scales are still largely unknown. Consensus has not been reached yet on
how to apply the knowledge learnt from individual lakes to the pan-Arctic scale, because even
lakes in a small area could have much different transport pathways (ebullition vs. diffusion),
morphology (deep vs. shallow and large vs. small), eutrophication (eutrophic vs. oligotrophic)
and carbon source (thermokarst vs. non-thermokarst and yedoma vs. non-yedoma). Because
wetlands and lakes, both inundation landscapes, are usually neighbored, it is difficult to use
inverse modeling at coarse spatial scales to detect strong CH$_4$ emissions that are emitted solely
by lakes. To test whether high-resolution inversions can better represent CH$_4$ emissions from
lakes, we conducted a comparison test (“DLEM only”) over the East Siberia Coastal Lowlands
(Fig. 1) using the DLEM model and excluding CH$_4$ emissions from lakes. We chose the East
Siberia Lowlands to test our hypothesis as lakes there occupy 56% of the water-inundated
landscapes, i.e. lakes, wetlands and rivers (Lehner and Döll, 2004) and a large fraction of lakes
in the region are high-flux yedoma lakes (Walter et al., 2006). We chose the DLEM model
considering that the simulated wetland CH$_4$ emissions in this model are weak for the East Siberia
Lowlands. This design is also aimed to alleviate the impact of one major shortcoming: because
there are not sufficient high-quality observations, we optimized CH$_4$ emission in each grid cell
separately for wetlands and lakes and in this manner a fraction of lake emissions could be
attributed incorrectly to wetlands or vice versa. The inversion of the “DLEM only” scenario is
shown in Fig. S5. In comparison to Fig. 4c, CH$_4$ emissions from the East Siberia Coastal
Lowlands are low in Fig. S5. A further comparison of model-satellite agreement between the
DLEM scenario and this no-lake scenario reveals that the agreement improves impressively
when lake emissions are considered (see Fig. 6; $p = 0.0032838$ at the two-sample t-test). It
implies that CH$_4$ emissions from regional lakes could be significant. As illustrated above,
however, the spatial neighborhood of wetlands and lakes makes it difficult to conduct similar
experiments in other areas. Thus we are cautious to claim that CH$_4$ emissions from lakes are
ubiquitously strong across the pan-Arctic. Rather, since we used six wetland models that can
simulate very different wetland emission distributions at spatial and temporal scales, our
estimates of 2.4–14.2 Tg CH$_4$ yr$^{-1}$ for lake emissions could be more useful in explaining the
range of this source. The lower bound of our estimate is much smaller than the estimate of 7.1–
17.3 Tg CH$_4$ yr$^{-1}$ by Bastviken et al. (2011) in the use of extensive site-level observations. In
contrast, the upper bound of our estimate is within the range. Given the wide span of this
estimate, it is difficult to say whether CH$_4$ emissions from pan-Arctic lakes can be significant
across the region.

4.2.3. CH$_4$ Emissions from Pan-Arctic Wetlands

Arctic tundra is regarded as an important source of CH$_4$ in the northern high latitudes. By
using process-based models and atmospheric CH$_4$ observations, McGuire et al. (2012) estimated
that Arctic tundra was a source of 25 Tg CH$_4$ yr$^{-1}$ to the atmosphere during 1990–2006. By using
the TM5-4DVAR inverse model and assimilating SCIAMACHY and NOAA/ESRL observations,
Alexe et al. (2015) estimated that CH$_4$ emissions from Arctic wetlands were 18.2 Tg CH$_4$ yr$^{-1}$ for
2010–2011. A similar estimate of 16±5 Tg CH$_4$ yr$^{-1}$ was also made by Bruhwiler et al. (2014)
using the CarbonTracker-CH$_4$ assimilation system. Our estimate of 5.5–14.2 Tg CH$_4$ yr$^{-1}$
overlaps with the estimate of Bruhwiler et al. (2014) but is much lower than the estimates of
Alexe et al. (2015) and McGuire et al. (2012). However, McGuire et al. (2012) did not use
complex inverse models and Alexe et al. (2015) used the coarse-resolution TM5-4DVAR inverse
model. As our global inversions (Table 2) are consistent with the estimate of Alexe et al. (2015),
this difference is likely introduced by the use of the nested-grid inverse model. In other words,
the nested-grid inverse model reveals some information that could be missed in global coarse-
resolution inversions. For Siberian wetlands, they could emit much more CH$_4$ (1.6–7.6 Tg yr$^{-1}$) than any other areas. But the uncertainty of this source is also the largest. Using the atmospheric CH$_4$ observation data at several sites near to Siberian wetlands, Berchet et al. (2015) estimated that CH$_4$ emissions from Siberian wetlands were in the range of 1–13 Tg CH$_4$ yr$^{-1}$, wider than our estimated range. In addition, our estimate is also much smaller than the estimate of 21.63 ± 5.25 Tg CH$_4$ yr$^{-1}$ by Kim et al. (2012) for annual mean CH$_4$ emissions from Siberian wetlands during 2005–2010. According to our inversions, CH$_4$ emissions from wetlands in Alaska, northern Canada, northern Europe are 0–1.2 Tg CH$_4$ yr$^{-1}$, 0.4–4.8 Tg CH$_4$ yr$^{-1}$ and 0.7–3.6 Tg CH$_4$ yr$^{-1}$, respectively. For Alaskan wetlands, the total of posterior CH$_4$ emissions is much lower than the inferred value of 4.1 Tg CH$_4$ yr$^{-1}$ for the Alaskan Yukon River basin during 1986–2005 using the modeling of process-based CH$_4$ biogeochemistry and large-scale hydrology (Lu and Zhuang, 2012) and also much lower than the inferred value of 3 Tg CH$_4$ yr$^{-1}$ for the whole Alaska (Zhuang et al., 2007). Our estimate of wetland emissions from northern Europe compasses a European-scale estimate of 3.6 Tg CH$_4$ yr$^{-1}$ by Saarnio et al. (2009), agreeing with the investigation that wetlands in Europe are predominantly located north of 60°N.

### 4.2.4. Evaluation of Pan-Arctic CH$_4$ Inversions

As shown in Fig. 7, in most of scenarios, the nested grid inversions perform much better than both the forward simulations and the global inversions at NOAA/ESRL pan-Arctic flask sites (Fig. 1). For example, for the ORCHIDEE scenario, the nested grid inversion reduces the model bias by 44 ppb relative to the forward run and by 20 ppb relative to the global inversion, respectively. Also, for the SDGVM scenario, it reduces the model bias by 22 ppb relative to the forward run and by 13 ppb relative to the global inversion, respectively. But for aircraft CH$_4$ measurements, it is more complex. The nested grid inversions can reduce the model bias in some
scenarios greatly, i.e. the CLM4Me scenario and the SDGVM scenario. But in many cases, they
do not perform visibly better than the forward runs and the global inversions. One possible
reason is that the aircraft CH$_4$ RMS has already been low and thus the remaining errors,
including the representation error of model diurnal variability, cannot be resolved by our current
inversion system. For example, CH$_4$ emissions from Alaska can be well constrained by three
NOAA/ESRL surface sites in Alaska (BRW, CBA and SHM) and the CH$_4$ mixing ratios at the
aircraft PFA site are representative of the interior of Alaska as pointed out in Sweeney et al.
(2015). It is also possible that the increase of grid cells in the nested grid inversions introduced
more transport and computation errors.

4.3. Further Discussion

Both the global and nested-grid inversions indicate that the inverse modeling is more
sensitive to different wetland models than prior emission error and data error. Thus, to gain
better understandings of the global and pan-Arctic CH$_4$ cycles, it is important to develop more
realistic biogeochemical models. Especially, from the perspective of inverse modeling, focus
should be on improving the spatial and temporal representation of the models rather than
emission magnitude.

For the high-resolution inverse modeling, transport and computation errors of the nested-
grid CTMs need to be reduced for better performance. These CTMs can also benefit the efforts
to assimilate aircraft CH$_4$ measurements. For the purpose of satellite data bias correction, more
coordination between satellite missions and aircraft missions is demanded. The treatment of the
SCIAMACHY bias could be an important uncertainty source for our estimates, as suggested by
Houweling et al. (2014). Future top-down studies could benefit from a more reasonable bias correction method, even for low bias satellite products, e.g. GOSAT (Alexe et al., 2015).

The attribution of CH$_4$ fluxes to spatially overlapped sources, e.g. wetlands and lakes, could be problematic for even high-resolution inversions. Carbon isotope measurements ($\delta^{13}$CH$_4$) are widely used to separate biogenic and geologic CH$_4$ sources (Langenfelds et al., 2002) but are not useful for two biogenic sources with similar carbon isotope ratios (Walter et al., 2008; Fisher et al., 2011). In our study, lake and wetland emissions were simulated separately by different models. This raised the possibility of double counting emissions of the two sources. A possible solution is to simulate them together in one earth system model and use a consistent method to identify wetland and lake pixels.

Our nested grid adjoint model currently does not cover the regions near the North Pole. While it could be rare in the summer time, if air mass transports across the Arctic Ocean, it may not be represented in the model. In the following studies, we will adapt the advection algorithm for the polar region from the global adjoint model to the nested-grid model and validate the adaptation. These refinements shall reduce the uncertainty of our estimates. It is also valuable to discuss the integration of other natural CH$_4$ sources found in the pan-Arctic, such as CH$_4$ emission from subsea permafrost of East Siberian shelf (Shakhova et al., 2013; Berchet et al., 2016; Thornton et al., 2016). As shown in Fig. 1, our inverse modeling assimilated few high-precision surface CH$_4$ measurements in Siberia and northern Canada. Since some efforts have already been made by different teams to measure atmospheric CH$_4$ routinely in Siberia (e.g., the JR-STATION network by NIES, the Zotino Tall Tower Observatory by MPI-BGC and the Tiksi site by the Finnish Meteorological Institute) and in North American Arctic (e.g., the Behchoko site by Environment Canada), we would like to take advantage of these measurements to further
improve our inversion results and re-evaluate the gains of using satellite data in our future studies.

5. Conclusion

In this study, we used a high-resolution nested-grid adjoint model in the pan-Arctic domain to constrain CH$_4$ emissions from pan-Arctic wetlands, lakes and anthropogenic sources. The sensitivity of the method to different prior wetland CH$_4$ fluxes was tested. When assimilating both NOAA/ESRL measurements and SCIAMACHY retrievals, we estimated that in 2005, the total of global CH$_4$ emissions was in the range of 496.4–511.5 Tg CH$_4$ yr$^{-1}$, with wetlands contributing 130.0–203.3 Tg CH$_4$ yr$^{-1}$. Both of these estimates are consistent with some widely accepted expert assessments. The estimated CH$_4$ emissions in the pan-Arctic were in the range of 11.9–28.5 Tg yr$^{-1}$, with wetland and lake emissions ranging from 5.5 to 14.2 Tg yr$^{-1}$ and from 2.4 to 14.2 Tg yr$^{-1}$, respectively. The largest CH$_4$ emissions in the pan-Arctic are from Siberian wetlands and lakes. The study demonstrates that the assimilation of satellite retrievals can reduce the uncertainty of the nested grid inversions. Evaluation with independent datasets shows that the nested inversions can better improve the representation of CH$_4$ mixing ratios in lower boundary layer rather than top boundary layer and free troposphere.

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Figure Captions

Figure 1. SCIAMACHY retrievals (n = 37743) of the weighted column-average CH$_4$ dry mole fractions for July 2005–September 2005 in the pan-Arctic that have passed all quality control tests described in Section 2.1 and the locations of surface flask stations and aircraft missions used for data assimilation or inversion evaluation.

Figure 2. Prior average CH$_4$ fluxes from wetlands, lakes and other sources (i.e. anthropogenic and biomass burning) in 2005 used for the pan-Arctic nested grid inversions at 1/2° × 2/3° resolution. Annual total emission for each pan-Arctic source is presented in units of Tg CH$_4$ yr$^{-1}$.

Figure 3. Bias correction function (left) and standard deviation (right) for SCIAMACHY retrievals overpassing the pan-Arctic. ΔXCH$_4$ is the difference between SCIAMACHY and column-average mixing ratios mapped from aircraft vertical profiles. The red line in the left shows a linear regression weighted by the number of SCIAMACHY retrievals.

Figure 4. Optimized pan-Arctic CH$_4$ fluxes in 2005 at 1/2° × 2/3° resolution using both SCIAMACHY and NOAA/ESRL observations. a) BERN; b) CLM4Me; c) DLEM; d) ORCHIDEE; e) SDGVM; f) WSL.

Figure 5. Comparison of prior and posterior pan-Arctic CH$_4$ emissions and their uncertainties. “NOAA only” represents posterior emissions assimilating only surface measurements. “NOAA + SCIA” represents posterior emissions assimilating both surface measurements and satellite
retrievals. The uncertainty of prior emissions is 100%. Scenarios are represented by their name initials: “B” for BERN, “C” for CLM4Me, “D” for DLEM, “O” for ORCHIDEE, “S” for SDGVM and “W” for WSL.

Figure 6. Distribution of the relative difference between the observed and simulated posterior SCIAMACHY column-average mixing ratios. The “DLEM + Lake” scenario includes CH$_4$ emissions from both wetlands and lakes and the “DLEM only” scenario only includes CH$_4$ emissions from wetlands. Relative difference is calculated as a percentage of absolute differences between GEOS-Chem and SCIAMACHY relative to SCIAMACHY retrievals. Two extending red and blue lines represent the means of the simulation bias under the “DLEM + Lake” scenario and the “DLEM only” scenario, respectively.

Figure 7. Evaluation of the posterior GEOS-Chem CH$_4$ mole fractions from the pan-Arctic nested-grid inversions with independent data sets from the NOAA flask stations, the NOAA aircraft PFA profiles and the NIES aircraft Surgut profiles. Black symbols indicate the rms of the forward GEOS-Chem runs and red symbols indicate the rms of the global inversions.
Table 1. Summary of bias correction methods and of mean absolute satellite-model difference (ppb) for 2003-2005 before and after applying bias correction. ΔBIC is the BIC score increase of a bias correction method when referring to the latitude only method.

<table>
<thead>
<tr>
<th>Bias correction function*</th>
<th>Mean absolute difference</th>
<th>ΔBIC</th>
<th>R²</th>
</tr>
</thead>
<tbody>
<tr>
<td>No correction</td>
<td></td>
<td>9.271</td>
<td></td>
</tr>
<tr>
<td>Latitude only</td>
<td>$p_0 + p_1 \varphi + p_2 \varphi^2$</td>
<td>6.305</td>
<td>0.62</td>
</tr>
<tr>
<td>Air mass factor only</td>
<td>$p_0 + p_1 A_F$</td>
<td>7.071</td>
<td>161</td>
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<tr>
<td>Humidity only</td>
<td>$p_0 + p_1 H_S$</td>
<td>6.786</td>
<td>73</td>
</tr>
<tr>
<td>Latitude + Humidity</td>
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<td>6.230</td>
<td>-7</td>
</tr>
<tr>
<td>Air mass factor + Humidity</td>
<td>$p_0 + p_{11} A_F + p_{21} H_S$</td>
<td>6.396</td>
<td>12</td>
</tr>
</tbody>
</table>

* $p_0$, $p_1$, $p_2$, $p_{11}$, $p_{12}$ and $p_{21}$ are regression parameters.
Table 2. Estimated annual CH$_4$ emissions (units: Tg CH$_4$ yr$^{-1}$) for TransCom 3 land regions (NAB: North American Boreal, NAT: North American Temperate, SATr: South American Tropical, SAT: South American Temperate, NAf: Northern Africa, SAf: Southern Africa, ErB: Eurasian Boreal, ErT: Eurasian Temperate, TrA: Tropical Asia, Aus: Australasia, and Eur: Europe). The priors are the range of the initial CH$_4$ emissions given by the six scenarios.

<table>
<thead>
<tr>
<th>Region</th>
<th>Priors</th>
<th>Posterior Bern</th>
<th>CLM4Me</th>
<th>DLEM</th>
<th>ORCHIDEE</th>
<th>SDGVM</th>
<th>WSL</th>
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<th>Alexe et al. (2015)</th>
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Figure 1.
Figure 2.
Figure 3.
Figure 4.
Figure 5.
Figure 6.
Figure 7.
S1. Methods and Results

In the text S1, the steps to construct optimal initial conditions for global and nested grid inversions are described. We also describe the steps to construct an optimal GEOS-Chem CH$_4$ field for SCIAMACHY bias correction purpose and the comparison between our estimates and previous inversion studies in the global scale.

To start global and nested-grid inversions, the initial CH$_4$ field of the GEOS-Chem model needs to be optimized to minimize its error. As our focus is in the period of 2004–2005, to speed up the whole process, we only ran one inversion from 1993 to 2003 using the LPJ-WSL scenario and NOAA/ESRL measurements. The main purpose of this inversion is to construct initial CH$_4$ field in 2004. As presented in Fig. S2, without optimization, the LPJ-WSL scenario gives the best fit of the GEOS-Chem modeled CH$_4$ to the GLOBALVIEW-CH4 data (GLOBALVIEW-CH4, 2009). During the 1993–2003 inversion, GEOS-Chem was driven by GEOS-4 meteorological (met) data from NASA’s Global Modeling Assimilation Office (GMAO). Relative to GEOS-5, the GEOS-4 met data has the same horizontal resolutions but less vertical hybrid sigma-pressure levels (55 vertical levels).

To construct optimal atmospheric CH$_4$ fields for the bias correction of SCIAMACHY retrievals at the global scale, we ran a global inversion during 2004–2005 using the LPJ-WSL wetland emission scenario and NOAA/ESRL measurements. In this inversion, the GEOS-Chem model was driven by the GEOS-5 met data. The global inversions of different scenarios that assimilated both surface measurements and satellite retrievals were then run in two sequential time windows: 2004/01–2004/12 and 2005/01–2005/12. Only the inversions in the second time window are for analysis and the first time window is designed to minimize the impacts of the
transition from GEOS-4 to GEOS-5 and from the LPJ-WSL scenario to other scenarios. In the above inversions, we included surface measurements from pan-Arctic sites but excluded satellite retrievals out of 50°S–50°N. The global inversions during 2005 also provided initial conditions and time-dependent boundary conditions for the nested grid simulations of the adjoint model. Following Turner et al. (2015), we did not optimize boundary conditions in the nested-grid inversions as did in Wecht et al. (2014). The nested grid inversions of the pan-Arctic were run at 1/2° × 2/3° resolution from July 1, 2005 to Oct 1, 2005.

Specific humidity for bias correction was retrieved from the European Centre for Medium-Range Weather Forecasts (ECMWF)’s ERA-20C reanalysis product (http://apps.ecmwf.int/datasets/data/era20c-daily), averaged by the column between the surface and 3 km altitude (Houweling et al., 2014). The air mass factor and coordinates of satellite CH₄ retrievals have been included in the SCIAMACHY IMAP v6.0. For global-scale bias correction, we first optimized the GEOS-Chem 4-D CH₄ mixing ratios using only surface measurements and then sampled the modeled XCH₄ at the coordinates and time of SCIAMACHY retrievals and with local averaging kernels applied. Following Bergamaschi et al. (2009) and Houweling et al. (2014), only satellite retrievals between 50°S and 50°N were utilized. The XCH₄ differences between SCIAMACHY and GEOS-Chem are shown in Fig. S3a. A regression relationship was then built to represent the satellite system bias by proxy factors. Turner et al. (2015) suggested that it is more likely that grid squares residual standard deviation (RSD) in excess of 20 ppb are dominated by model bias in prior emissions. Thus, we excluded such grid squares in regressions. And satellite retrievals with low precisions (the ratio of retrieval precision error to retrieval is larger than 3%) were also removed from analysis. Following Houweling et al. (2014), we did not optimize bias correction functions in the inversion cycle in the concern that this process could
cause bias correction to incorrectly account for the uncertainties caused by unaccounted model errors or even the uncertain sources and sinks. As shown in Fig. S3d, bias correction reduced model-satellite differences greatly in tropical areas of America, Africa and South Asia and also reduced the differences in Australia and some areas of the United States. And the agreement between GEOS-Chem and SCIAMACHY is also improved at the global scale (Fig. S3c). However, the model-data agreement is deteriorated in East Asia. It could be caused by the overestimate of anthropogenic CH₄ emissions from China in the EDGAR dataset (Peng et al., 2016).

The results of the global inversions are presented in Table 2 and Fig. S4. There have been many studies that assimilated surface measurements and/or satellite retrievals into a CTM inverse model to constrain global CH₄ emissions, see Kirschke et al. (2013) for review. For instance, using the same observations suite, Bergamaschi et al. (2009) estimated that in 2004, CH₄ emissions in global, tropical (30°S–30°N), northern extratropical (30°N–90°N) and southern extratropical (90°S–30°S) zonal areas were 506.7 Tg CH₄ yr⁻¹, 323.5 Tg CH₄ yr⁻¹, 172.8 Tg CH₄ yr⁻¹ and 10.4 Tg CH₄ yr⁻¹, respectively. These large-scale estimates are consistent with our calculations: 284.5–319.6 Tg CH₄ yr⁻¹ (tropical), 165.3–206.6 Tg CH₄ yr⁻¹ (northern extratropical) and 10.0–13.9 Tg CH₄ yr⁻¹ (southern extratropical). This agreement could imply that the GEOS-Chem adjoint and TM5-4DVAR are consistent in the atmospheric transport, chemistry and inverse modeling methods. In contrast to Bergamaschi et al. (2009), our inversions allocate more emissions to extratropical regions. As a result, the tropical total (SATr + NAF + SAF + TrA) of the six inversions is in the range of 114.1–169.7 Tg CH₄ yr⁻¹, which is much lower than their estimate of 203.2 Tg CH₄ yr⁻¹. The likely reason for this discrepancy is that we did not optimize bias correction functions in the inversion cycle. Our posterior wetland CH₄ emissions estimated
in the Bern, CLM4Me, SDGVM and WSL scenarios are close to the estimate of 161 Tg CH$_4$ yr$^{-1}$ for 2003–2007 in Bloom et al. (2010). The latter was based on CH$_4$ and gravity spaceborne data to constrain large-scale methanogenesis. Our estimates are also close to the inferred wetland CH$_4$ emissions (175±33 Tg CH$_4$ yr$^{-1}$) by Kirschke et al. (2013). By using artificial neural networks, Zhu et al. (2013) estimated that from 1990 to 2009, annual wetland CH$_4$ emissions from northern high latitudes (> 45°N) were in the range of 44.0–53.7 Tg CH$_4$ yr$^{-1}$, agreeing with the estimates of the Bern, CLM4Me and SDGVM scenarios.

Fig. S4a shows that CH$_4$ fluxes are the highest in the Amazon, China, Southeast Asia, North America and Europe where there are either a large area of wetlands and rice paddies or advanced coal and oil industries or both. Our results indicate that the Eurasian temperate zone, including China, North America and Europe, emitted much more CH$_4$ than any other geographic zones (Table 2), implying the dominance of anthropogenic sources in the global CH$_4$ inventory. As presented in Fig. S4c, our inverse modeling reduced the CH$_4$ emissions from China, the Amazon basin and the Eurasian boreal region (scale factor < 1) but increased the emissions in Europe and Southeast Asia (scale factor > 1) relative to the prior.

Fig. S6 shows the difference between the modeled and observed CH$_4$ mixing ratios at NOAA ship board sampling stations and aircraft vertical profile sites under different wetland scenarios before and after the global scale inversions. For most scenarios, inversion improves the representation of CH$_4$ mixing ratios in GEOS-Chem at both marine and inland boundary layers and free troposphere. For example, the BERN scenario inversion reduced the bias by about 18 ppb for ship stations and about 6 ppb for aircraft sites. Also the DLEM scenario inversion reduced the bias by about 20 ppb for ship stations and about 19 ppb for aircraft sites. For the
CLM4Me and SDGVM scenarios with low prior biases, the inversions did not improve the performance. This could be caused by the errors introduced by the inversion process itself. For example, as the optimization is designed to address total emissions, the representation of diurnal variability in GEOS-Chem could be made worse during inversion.
Table S1. NOAA/ESRL stations used in the inversion.

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Figure S1. Average of prior wetland CH$_4$ annual emissions during 2004–2005 from six different wetland biogeochemical models used for the GEOS-Chem global inversion at 4° × 5° resolution. Annual total emission (orange) is presented in units of Tg CH$_4$ yr$^{-1}$. 
Figure S2. The comparison between the GEOS-Chem simulated and GLOBALVIEW-CH4 atmospheric CH$_4$ (units: ppbv) at five stations (Mace Head, Ireland; Trinidad, California; Ragged Point, Barbados; Cape Matatula, Samoa; Cape Grim, Tasmania). The wetland CH$_4$ emissions used are pre-optimized model simulations provided by the WETCHIMP project.
Figure S3. Comparison of column averaged CH$_4$ mole fractions from SCIAMACHY with those from GEOS-Chem model calculated with prior emissions. (a and b) show the mean bias and residual standard deviation of the satellite-model difference, (c) shows the comparison of the model (x axis) and satellite (y axis) XCH$_4$ after applying the “latitude + humidity” correction from the linear regression (weighted R$^2$ is shown inset and the red 1:1 line is also shown), and (d) shows the satellite-model difference after bias removal.
Figure S4. Optimized global CH₄ emissions and emission scale factors in 2005 at 4° × 5° resolution. Emission scale factor is defined as posterior emissions relative to prior emissions. a) Posterior CH₄ emissions averaged over inversions of six scenarios; b) standard deviation of posterior CH₄ emissions over inversions of six scenarios; c) optimized emission scale factors averaged over inversions of six scenarios.
Figure S5. Posterior CH$_4$ emissions from the pan-Arctic in 2005 estimated by the inversion of the “DLEM wetland only” scenario. The “DLEM wetland only” scenario uses the simulated wetland CH$_4$ emissions from the DLEM model and does not incorporate CH$_4$ emissions from pan-Arctic lakes.
Figure S6. Evaluation of posterior GEOS-Chem CH₄ mole fractions from the global inversions with independent data sets. The plot shows the root mean square (rms) of differences between the modeled and the observed CH₄ mixing ratios. Black symbols indicate the rms of the forward GEOS-Chem runs.