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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 SCanning Imaging Absorption spectroMeter for Atmospheric CHartogrphY (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 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 and 2.4–14.2 Tg yr^{-1}, respectively. Methane emissions from Siberian wetlands and lakes are 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 some specific regions. In contrast to global inversions, high-resolution nested-grid inversions perform better in estimating near-surface 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 preindustrial 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 interannual 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, b). Spaceborne 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 have been 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 have been 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 colocated (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 $1/2^\circ \times 2/3^\circ$ horizontal resolution Goddard Earth Observing System–Chemistry (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), 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, b). 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, 2007; Bastviken et al., 2011; Tan and Zhuang, 2015a) and the East Siberian Shelf (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 (Sect. 3.1), the pan-Arctic nested-grid CTM (Sect. 3.2), and the adjoint-based inversion method (Sect. 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 (SCIAMACHY) 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 (XCH_4) from the SWIR nadir spectra (channel 6: 1.66–1.67 µm) using the iterative maximum a posteriori differential optical absorption spectroscopy (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 θ is the solar zenith angle and ξ 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 Ω_{CH_4} (molecules cm^{-2}):

$$\Omega_{CH_4} = \Omega_A + \boldsymbol{a}^T(\boldsymbol{\omega} - \boldsymbol{\omega}_A), \quad (1)$$

where $\boldsymbol{\omega}$ is the true 10-layer sub-column densities of CH_4 (molecules cm^{-2}), $\boldsymbol{\omega}_A$ is the 10-layer prior CH_4 sub-column density (molecules cm^{-2}), Ω_A is the corresponding a priori CH_4 total column density, and $\boldsymbol{a}$ is an averaging kernel vector that defines the sensitivity of the retrieved total column to each sub-column in $\boldsymbol{\omega}$. To account for the impact of aerosol scattering and instrument effects on the observed light path, Frankenberg et al. (2006) used the CO_2 column density Ω_{CO_2} as a proxy to normalize and convert Ω_{CH_4} to a column mixing ratio XCH_4 (ppb):

$$XCH_4 = \left(\Omega_{CH_4}/\Omega_{CO_2}\right) XCO_2, \quad (2)$$

where XCO_2 is the column-weighted mixing ratio of CO_2 from NOAA's CarbonTracker CO_2 measurement and modeling system. CO_2 is used as a proxy because it is retrieved in a spectrally neighboring fitting window and, relative to CH_4, 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 scenes that are cloud-free or partially cloudy, 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, XCH_4 retrievals of less than 1500 ppb or larger than 2500 ppb are discarded (Alexe et al., 2015). For the pan-Arctic, most of the qualified XCH_4 retrievals were recorded in the summertime, when local solar zenith angles are higher, surface reflectance is lower, and impact of Arctic vortex is smaller. Figure 1 shows the SCIAMACHY retrievals ($n = 37\,743$) of the weighted column-average CH_4 dry mixing ratio for July–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 in the Supplement. 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). Figure 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 NASA's Arctic Research of the Composition of the Troposphere from Aircraft and Satellite (ARCTAS) mission (Jacob et al., 2010). 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, western 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 Differential Absorption CO Measurement (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.

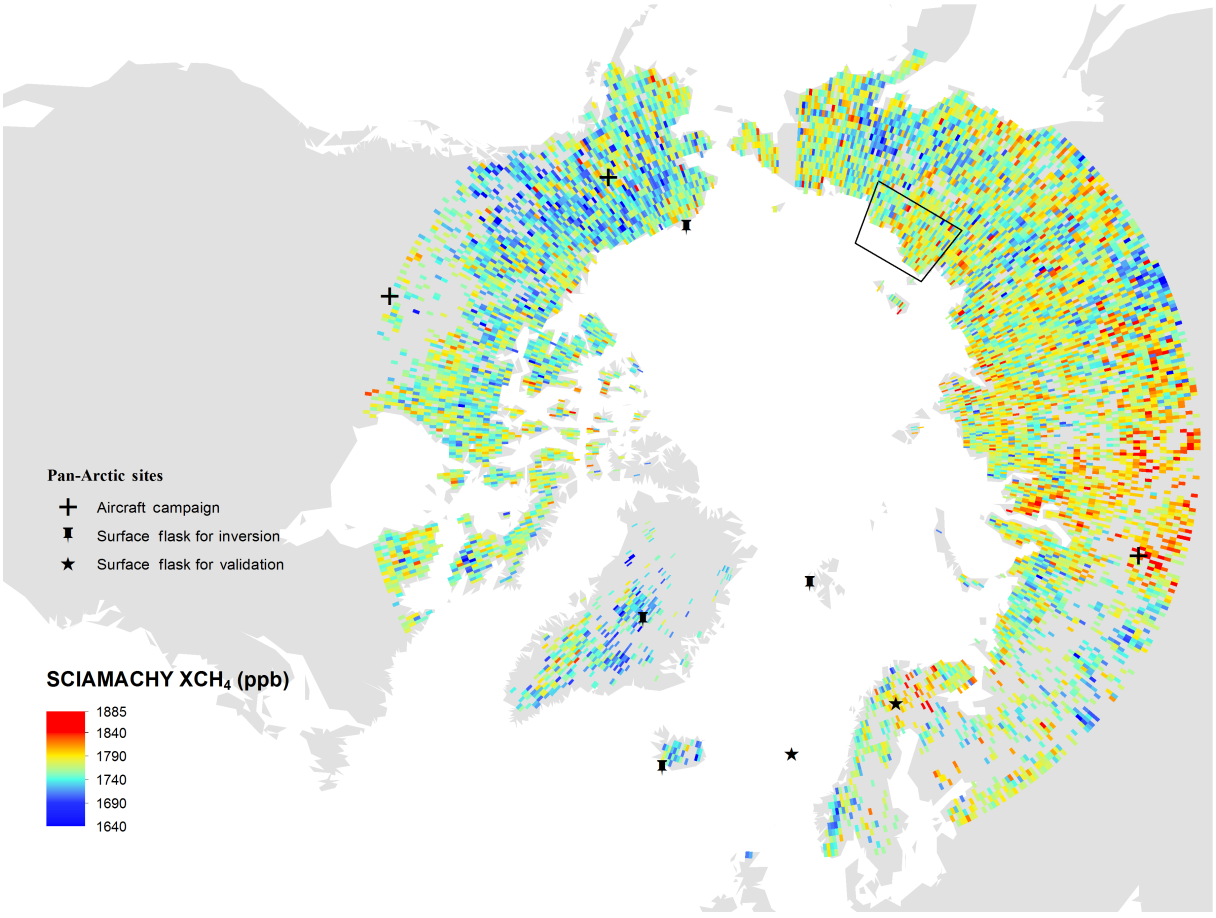

Figure 1. SCIAMACHY retrievals ($n = 37\,743$) of the weighted column-average CH_4 dry-mole fractions for July–September 2005 in the pan-Arctic that have passed all quality control tests described in Sect. 2.1, and the locations of surface flask stations and aircraft missions used for data assimilation or inversion 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: the Community Land Model 4 (CLM4) CH_4 biogeochemistry model; DLEM: the Dynamic Land Ecosystem Model; BERN: the Lund-Potsdam-Jena dynamic global vegetation model – the University of Bern version; WSL: the Lund–Potsdam–Jena dynamic global vegetation model – the Swiss Federal Research Institute version; ORCHIDEE: the Organising Carbon and Hydrology in Dynamic Ecosystems model; SDGVM: the Sheffield Dynamic Global Vegetation Model) to set up six different inverse modeling experiments. All wetland CH_4 simulations follow the same protocol of the 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 in Supplement), and wetland CH_4 emissions are the highest in tropical regions (e.g., the 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

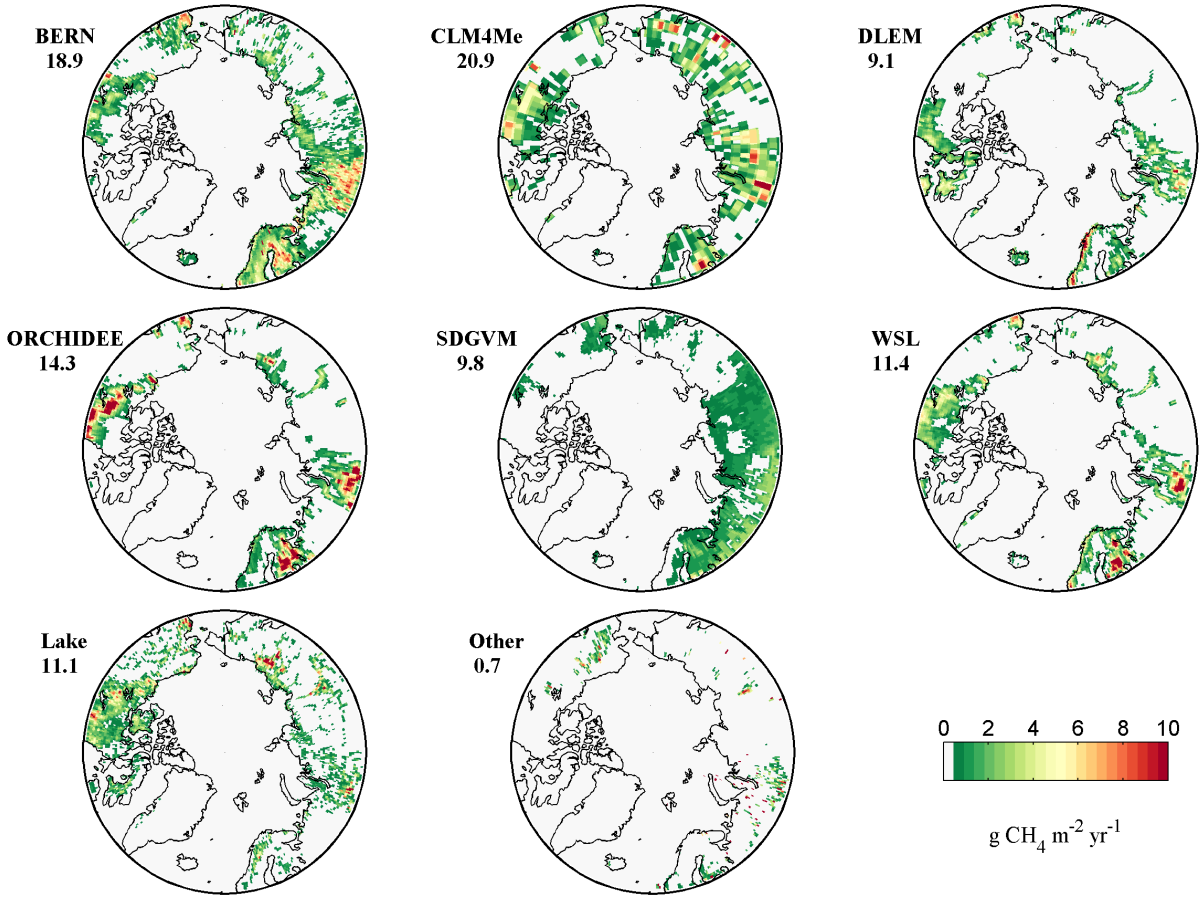

Figure 2. Prior average CH_4 fluxes from wetlands, lakes, and other sources (i.e., anthropogenic and biomass burning) in 2005 used for the pan-Arctic nested-grid inversions at 1/2° × 2/3° resolution. Annual total emission for each pan-Arctic source is presented in units of Tg CH_4 yr^{-1}.

fluxes in the west Siberian lowlands, Finland, and the 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 that 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 deep water 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° N was described by Tan and Zhuang (2015a, b). 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 h, 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 Pole (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 80–56° N) for the pan-Arctic nested grid. To make it consistent with the bLake4Me model, only CH_4 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 have a 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 $4° \times 5°$ resolution global-scale forward runs at 3 h intervals.

The GEOS-Chem CH_4 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 the Emission Database for Global Atmospheric Research v4.2 (EDGAR4.2) with $0.1° \times 0.1°$ resolution and no seasonality (European Commission, Joint Research Centre/Netherlands Environmental Assessment Agency, 2009). CH_4 emissions from termites and biomass burning were obtained from the study of Fung et al. (1991) and the daily Global Fire Emissions Database Version 3 (GFED3) of van der Werf et al. (2010), respectively. CH_4 emissions from wetlands and lakes were simulated by biogeochemical models described in Sect. 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 10.8×10^5 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 regridded 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 were 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 the solution of

$$\boldsymbol{y} = F(\boldsymbol{x}) + \boldsymbol{\varepsilon}. \tag{3}$$

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):

$$J(\boldsymbol{x}) = (F(\boldsymbol{x}) - \boldsymbol{y})^{\mathrm{T}} \mathbf{C}_{\mathrm{d}}^{-1} (F(\boldsymbol{x}) - \boldsymbol{y}) + \gamma (\boldsymbol{x} - \boldsymbol{x}_0)^{\mathrm{T}} \mathbf{C}_{x_0}^{-1} (\boldsymbol{x} - \boldsymbol{x}_0), \tag{4}$$

where $\boldsymbol{y}$ is a vector of observations from SCIAMACHY and NOAA/ESRL, F is a model operator that maps emissions to observations, $\boldsymbol{x}$ represents CH_4 emissions to be constrained, $\boldsymbol{x}_0$ is the a priori estimate of $\boldsymbol{x}$, $\mathbf{C}_{\mathrm{d}}$ 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 $\mathbf{C}_{x_0}$ is the parameter error covariance matrix (containing the uncertainties of the parameters and their correlations). The regularization parameter γ controls the relative constraints applied by the observational and a priori parts of $J(x)$ (Kopacz et al., 2009). In the adjoint method, γ 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 $\hat{\boldsymbol{x}}$ and its associated error covariance $\hat{\mathbf{C}}_x$ (Rodgers, 2000):

$$\hat{\boldsymbol{x}} = \boldsymbol{x}_0 + \left((\nabla_x F)^{\mathrm{T}} \mathbf{C}_{\mathrm{d}}^{-1} \nabla_x F + \gamma \mathbf{C}_{x_0}^{-1} \right)^{-1} (\nabla_x F)^{\mathrm{T}} \mathbf{C}_{\mathrm{d}}^{-1} (\boldsymbol{y} - F(\boldsymbol{x}_0)), \tag{5}$$

$$\hat{\mathbf{C}}_x^{-1} = (\nabla_x F)^{\mathrm{T}} \mathbf{C}_{\mathrm{d}}^{-1} \nabla_x F + \gamma \mathbf{C}_{x_0}^{-1}, \tag{6}$$

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 4-dimensional variational data assimilation (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 ($e_x = \ln(\boldsymbol{x}/\boldsymbol{x}_0)$) at each grid cell to avoid negative emissions. The posterior error covariance $\hat{\mathbf{C}}_x$ 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 the initial Hessian, so-called preconditioning (Bousserez et al., 2015). In contrast, approximating $\hat{\mathbf{C}}_x$ by stochastic methods, i.e., Monte Carlo sampling and gradient-based randomization, could help avoid the impact of setting the initial Hessian

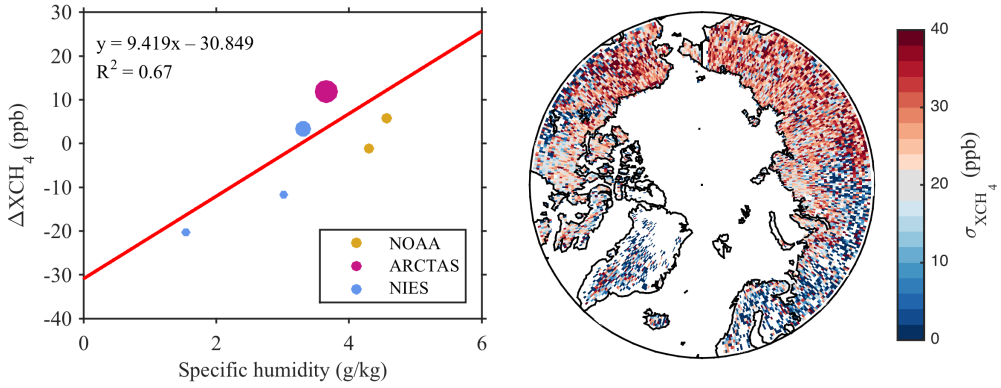

Figure 3. Bias correction function (left) and standard deviation (right) for SCIAMACHY retrievals overpassing the pan-Arctic. ΔXCH_4 is the difference between SCIAMACHY and column-average mixing ratios mapped from aircraft vertical profiles. The red line on the left shows a linear regression weighted by the number (represented by circle size) of SCIAMACHY retrievals.

(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 $\boldsymbol{x}_0$ and $\boldsymbol{y}$ in line with $\mathbf{C}_{x_0}$ and $\mathbf{C}_d$, 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° × 5° resolution and of 300 km at the nested-grid 1/2° × 2/3° 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 January–December 2005. These inversions provided boundary conditions for the following nested-grid inversions. For 1/2° × 2/3° nested-grid inversions, we ran the adjoint model 50 times over the period of July–September 2005 for each of 12 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 Supplement. 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 is reached.

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, e.g., latitude, air mass factor, or specific humidity. The air mass factor was used as a proxy parameter by some studies due to its correlation with 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 the air mass factor and humidity, latitude can represent the changes in both solar zenith angle and climate variables (Bergamaschi et al., 2007, 2009, 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 φ, 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 fits 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-

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.

	Bias correction function*	Mean absolute difference	ΔBIC	R^2
No correction		9.271		
Latitude only	$p_0 + p_1\varphi + p_2\varphi^2$	6.305		0.62
Air mass factor only	$p_0 + p_1 A_F$	7.071	161	0.52
Humidity only	$p_0 + p_1 H_S$	6.786	73	0.56
Latitude + humidity	$p_0 + p_{11}\varphi + p_{12}\varphi^2 + p_{21} H_S$	6.230	−7	0.62
Air mass factor + humidity	$p_0 + p_{11} A_F + p_{21} H_S$	6.396	12	0.60

* p_0, p_1, p_2, p_{11}, p_{12}, and p_{21} are regression parameters.

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 Eqs. (1) and (2). Figure 3 (left) shows that the retrieved system bias (ΔXCH_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 on most 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). Figure S2 shows the error variances of SCIAMACHY retrievals on a 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 are 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}, although 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 (Atmospheric Tracer Transport Model Intercomparison Project) 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 CLM4Me 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 2 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 enough tem-

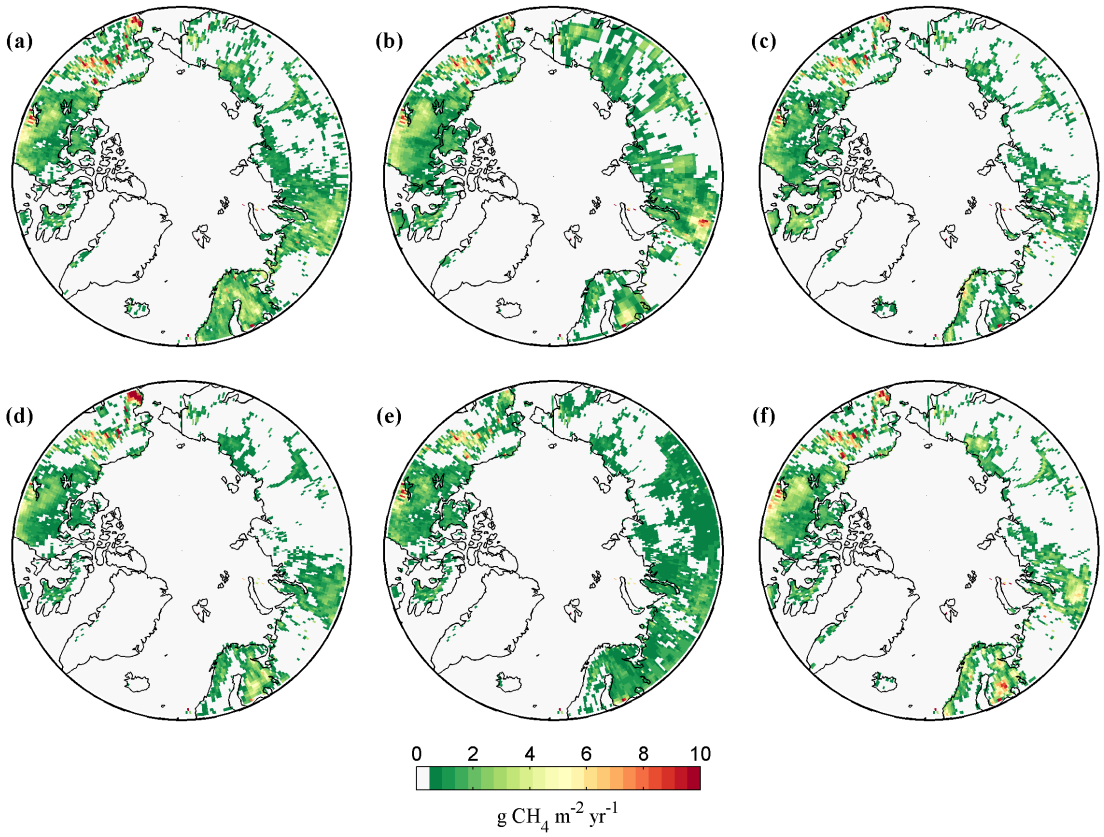

Figure 4. Optimized pan-Arctic CH_4 fluxes in 2005 at 1/2° × 2/3° resolution using both SCIAMACHY and NOAA/ESRL observations. **(a)** BERN; **(b)** CLM4Me; **(c)** DLEM; **(d)** ORCHIDEE; **(e)** SDGVM; **(f)** WSL.

poral 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 Supplement.

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 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, 1.3–7.9, 0.8–8.1 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, CH_4 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 CLM4Me scenario, the posterior uncertainty increases from 16 to 23 % when only surface measurements are assimilated. Our estimates are consistent with other inverse modeling estimates. For example, Kirschke et al. (2013) reviewed a series of top-down estimation of CH_4 emissions and suggested that CH_4 emissions north of 60° N could be in the range of 12–28 Tg CH_4 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 CH_4 field. Our estimates also imply that CH_4 emission from the pan-Arctic could constitute a large fraction of CH_4 emissions in the northern high latitudes (> 50° N). Based on the estimate (50 Tg CH_4 yr^{-1}) of Monteil et al. (2013), we calculated that 29.2–60.8 % of CH_4 emissions in the northern high latitudes could be emitted from the pan-Arctic (> 60° N). For all scenarios, the inverse modeling adjusts total CH_4 emissions downward compared to prior emissions. It is possible that CH_4 emissions are overestimated by the biogeochemi-

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.

Region	Priors	Posterior						Fraser et al. (2013)	Alexe et al. (2015)
		BERN	CLM4Me	DLEM	ORCHIDEE	SDGVM	WSL		
NAB	7.9–26.0	24.3	16.2	16.8	27.4	12.0	20.7	5.1 ± 1.1	10.3
NAT	38.5–59.2	33.2	32.8	42.8	49.2	51.2	39.7	62.5 ± 4.4	45.6
SATr	29.6–100.0	43.0	60.8	31.4	61.0	62.3	42.1	49.6 ± 6.4	71.8
SAT	29.1–55.8	31.2	27.1	35.2	39.1	25.6	30.5	55.8 ± 9.5	40.2
NAf	26.8–31.2	34.0	41.3	27.9	28.0	27.7	32.0	46.9 ± 7.3	50.6
SAf	16.0–27.0	18.4	16.2	19.0	24.2	15.6	18.7	36.6 ± 5.8	42.0
ErB	11.5–32.7	19.2	14.3	16.5	18.7	22.2	14.9	16.5 ± 3.8	15.4
ErT	114.9–133.5	97.0	84.9	146.1	92.7	98.3	99.8	115.9 ± 7.3	109.6
TrA	33.1–45.8	47.3	51.4	35.8	33.1	36.4	45.1	43.5 ± 3.2	76.8
Aus	5.8–8.3	7.3	7.7	6.6	7.9	6.3	7.3	17.6 ± 2.7	4.3
Eur	43.6–53.5	54.9	52.2	46.4	43.5	56.5	54.1	39.6 ± 3.7	28.9
Wetlands	121.7–278.1	166.8	164.6	130.0	203.3	161.8	160.7	192.1 ± 16.1	169
Global	471.5–627.8	501.0	497.7	511.5	511.0	496.4	502.9	510.6 ± 18.4	540.5

cal models or double-counted between the wetland and lake models or both. This adjustment could also be explained by the underestimate of CH_4 absorption by soils in biogeochemical models due to the lack of high-affinity methanotrophy (Oh et al., 2016).

4.2.2 CH_4 emissions from pan-Arctic lakes

In contrast to CH_4 emissions from pan-Arctic wetlands, CH_4 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 neighboring, 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 Siberian coastal lowlands (Fig. 1) using the DLEM model and excluding CH_4 emissions from lakes. We chose the east Siberian 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 Siberian lowlands. This design is also aimed to alleviate the impact of one major shortcoming: because there are not sufficient high-quality observations, we optimized the total CH_4 emission in each grid cell, 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 Siberian 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 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.

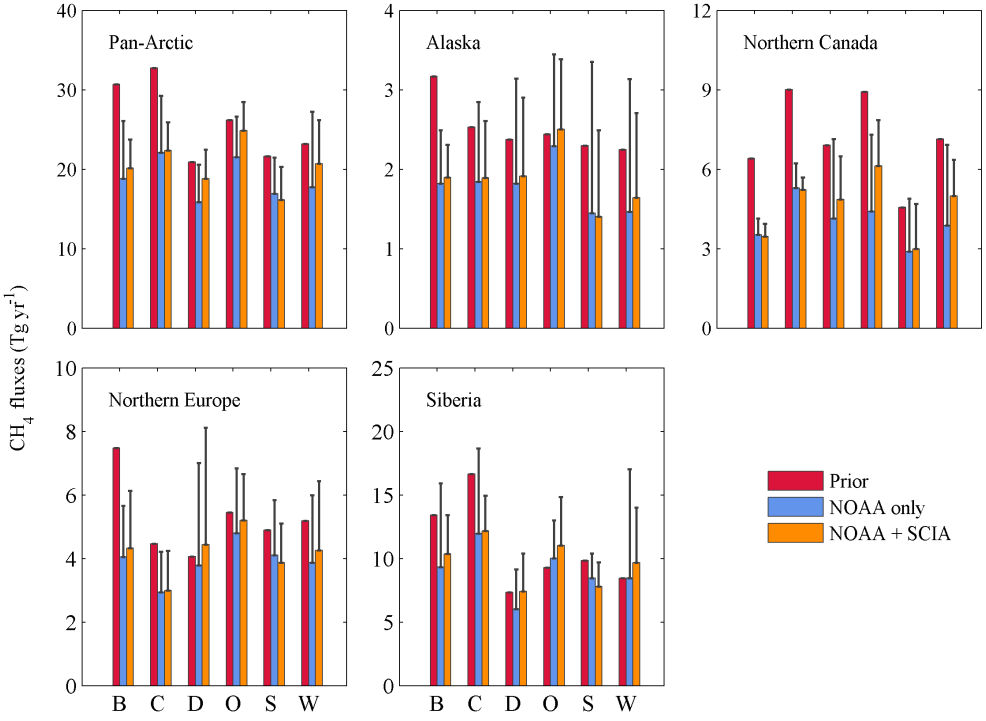

Figure 5. Comparison of prior and posterior pan-Arctic CH_4 emissions and their uncertainties. "NOAA only" represents posterior emissions assimilating only surface measurements. "NOAA + SCIA" represents posterior emissions assimilating both surface measurements and satellite retrievals. The uncertainty 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.

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 Transport Model 5 (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 the Siberian emissions is also the largest. Using the atmospheric CH_4 observation data at several sites near 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, and northern Europe are 0–1.2, 0.4–4.8, 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 of 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 finding 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 simula-

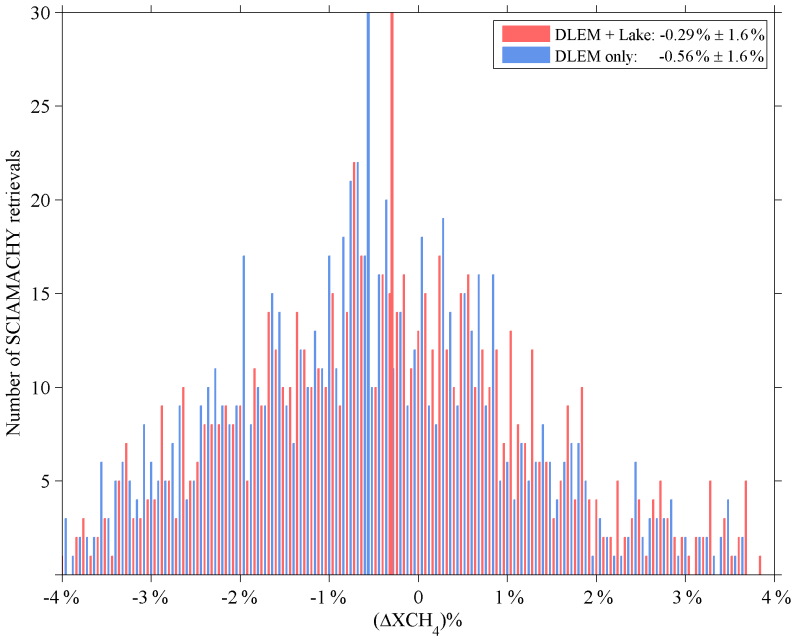

Figure 6. Distribution of the relative difference between the observed and simulated posterior SCIAMACHY column-average mixing ratios. The "DLEM + lake" scenario includes CH_4 emissions from both wetlands and lakes, and the "DLEM-only" scenario only includes CH_4 emissions from wetlands. Relative difference 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.

tions 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. 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. But for aircraft CH_4 measurements, it is more complex. The nested-grid inversions can reduce the model bias in some scenarios greatly, e.g., 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 root mean square error (RMSE) of aircraft CH_4 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 (Poker Flat, Alaska) 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

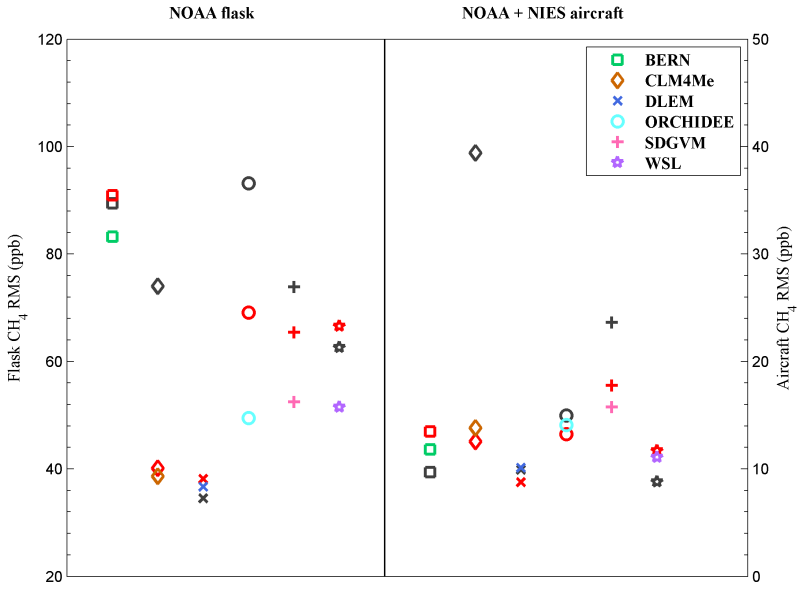

Figure 7. Evaluation of the posterior GEOS-Chem CH_4 mole fractions from the pan-Arctic nested-grid inversions with independent data sets from the NOAA flask stations, the NOAA aircraft PFA profiles, and the NIES aircraft Surgut profiles. Black symbols indicate the RMSE of the forward GEOS-Chem runs, and red symbols indicate the RMSE of the global inversions.

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 summertime, if air mass is transported 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 the East Siberian Shelf (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 Japan–Russia Siberian Tall Tower Inland Observation Network(JR-STATION) by NIES, the Zotino Tall Tower Observatory by the Max Planck Institute for Biogeochemistry (MPI-BGC), and the Tiksi site by the Finnish Meteorological Institute) and in the 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 Conclusions

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 data sets shows that the nested inversions can better improve the representation of CH_4 mixing ratios in the lower boundary layer rather than the top boundary layer and free troposphere.

6 Data availability

SCIAMACHY retrievals are available at http://www.temis.nl/climate/methane/methane_month_scia.php. NOAA/ESRL surface and aircraft observations are available at ftp://aftp.cmdl.noaa.gov/data/trace_gases/ch4/flask/surface/ and http://www.esrl.noaa.gov/gmd/ccgg/aircraft/data.html, respectively. NASA/ARCTAS aircraft observations are available at ftp://ftp-air.larc.nasa.gov/pub/ARCTAS/DC8_AIRCRAFT/DISKIN.GLENN/. NIES aircraft observations, prior lake CH_4 emissions, and all model-optimized CH_4 emissions are available upon request.

The Supplement related to this article is available online at doi:10.5194/acp-16-12649-2016-supplement.

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