Reply to referee report #1

We would like to thank the reviewer for his or her feedback on the manuscript. These comments have been very helpful for improving the manuscript. Below, we have listed the reviewer's comments (in bold) and the corresponding edits to the manuscript.

Page 2, lines 8-9: 'we then use real data to analyze the magnitude, seasonality, and spatial distribution of each model estimate', change 'real data' to 'real data and inversion model results' as some comparison used inversion results (seasonal cycle derived from real data through inversion analysis) rather than directly to 'real data'. Inversion model results with potentially large uncertainties should not be confused with 'real data' or 'atmospheric data'. The distinction between 'real data' and 'inversion model results' has to be clearly noted throughout the manuscript. In fact, the usage of inversion model results in addition to real data should be stated in the title of the manuscript.

We have updated lines 8-9 according to the reviewer's suggestion.

Page 2, lines 10-11: 'Many models predict a seasonality that is narrower than implied by atmospheric CH4 data', change to 'Many models predict a seasonality that is narrower than implied by inversion model results'

We have updated lines 10-11 accordingly.

Page 3, lines 16-17: 'The present study compares the WETCHIMP CH4 flux estimates against atmospheric CH4 data from 2007–2008 through two sets of analyses.' Change 'against atmospheric CH4 data it o 'against atmospheric CH4 data and inversion model results'.

We have updated lines 16-17 accordingly.

Page 4, lines 24-25: 'Based on these synthetic experiments, we conduct a second set of analyses using real atmospheric data.' Change to 'Based on these synthetic experiments, we conduct a second set of analyses using real atmospheric data and inversion model results.'

We have updated lines 24-25 accordingly.

Page 6, lines 11-12: 'H(nxm)', define m.

We have defined this variable in the revised manuscript (The variable "m" refers to the number of emissions or flux grid boxes in both space and time.).

Page 7, lines 18-19: 'As a result, the model selection framework cannot scale other variables in X to reproduce the atmospheric CH4 signal from wetlands' what are the 'other variables' referring to?

We have clarified this sentence in the revised manuscript. These other variables are anthropogenic emissions sources and wetland fluxes in distant geographic regions. To rephrase this sentence, the model selection framework cannot reproduce wetland fluxes by simply upscaling anthropogenic emissions sources that might have a similar spatial distribution to wetlands.

Page 7, lines 22-24: 'For consistency among the synthetic datasets, we scale the annual HBL CH4 budget in each WETCHIMP model to match the overall magnitude estimated by several topdown studies'. Are the fluxes outside the HBL region scaled the same way? If the fluxes elsewhere are scaled differently than the HBL region, then each WETCHIMP model has been altered and the experiments are not evaluating the 'original' WETCHIMP models. How do the results vary with and without scaling HBL as mentioned?

We downscale the WETCHIMP models to match the estimated magnitude in three existing top-down studies. If we did not scale the WETCHIMP models to match observational studies, then we could easily overstate the results and understate the role of model/measurement errors in the analysis. The synthetic data would identify wetlands too easily, and the results would not be representative of the real methane observations.

If we do not scale the magnitude of the WETCHIMP models, then wetland fluxes are a much larger source relative to anthropogenic emissions and modeling errors. As the magnitude of the wetland fluxes increases, the observations appear increasingly powerful and omniscient; the observations have no difficulty seeing the "signal through the noise." The results presented in this analysis are therefore conservative in nature. We have added a condensed version of this discussion to the manuscript.

When we run model selection using real data (as opposed to synthetic data), the observation network only selects flux models in Eastern and Western Canada. This result is broadly consistent with the synthetic data experiments which also show the highest sensitivity to wetland fluxes in Eastern and Western Canada. By contrast, this result is not consistent with synthetic data experiments that use unscaled versions of models with high magnitude (e.g., LPJ-WHyMe and Orchidee). If we ran the synthetic experiment with using these unscaled models, the results would indicate much higher sensitivity to wetland fluxes than the real data experiments actually support.

Page 8, lines 3-5: 'the intercept for each month is represented by a vector of ones in the matrix X, and this intercept is always included within X.' The word 'always' is confusing. Is the 'intercept' included in 'section 2.3 Real data experiments' only or in 'section 2.2 Synthetic data experiment' also (then it should be mentioned in 2.2)?

This sentence applies to Sect. 2.3, not to Sect. 2.2 We have updated the wording accordingly.

Page 10, lines 4-5: 'We run several additional test scenarios to explore why the synthetic observations may not always be able to detect wetland CH4 fluxes.' How many 'repeats' were done in these test scenarios? How do the BIC test results vary with the number of repeats?

These scenarios are displayed in Figs. 3b-3e. We have clarified the wording in this sentence. We have updated lines 4-5 accordingly.

Page 12, line 22: '4 Results and discussion: comparisons with atmospheric data' change to '4 Results and discussion: comparisons with atmospheric data and inversion model results'

We have updated line 22 accordingly.

Page 13, lines 19-20: 'LPJ-Bern and LPJ-WHyMe also use land cover maps and/or land surveys to estimate wetland' This is suggested as a reason why LPJ-Bern was 'selected' by the BIC experiment. Explain why LPJ-WhyMe was not selected and what other factors could be active.

We have added an explanation of this point to the manuscript. Despite similarities in the origin of these two models, LPJ-WHyMe has a relatively different spatial distribution from LPJ-Bern (Fig. S1). Large fluxes in LPJ-WhyMe extend much further south in Ontario and much further east in to Quebec and Labrador relative to LPJ-Bern. Models that use land cover maps or dynamic modeling (e.g., LPJ-Bern and SDGVM) often have a distribution that appears more consistent with atmospheric observations, but this quality does not guarantee consistency.

LPJ-Bern and LPJ-WHyMe use slightly different methane flux schemes, producing different spatial distributions. According to Wania et al. (2013), WhyMe only simulates fluxes from high latitude peatlands and uses an estimated peatland distribution from NCSCD (Tarnocai et al. 2009) to prescribe the distribution of fluxes. LPJ-Bern includes several additional components in its methane flux scheme, including GIEMS soil inundation, water/ice coverage from GICEW, and simulations of soil wetness (refer to Wania et al. 2013). Both models further apply a scaling factor to peatland emissions to account for micro-topography. LPJ-WHyMe uses a scaling factor of 0.75 and LPJ-Bern a scaling factors of 0.26. NCSCD distributes peatlands throughout eastern Canada in Ontario, Quebec, and Labrador. Fluxes in LPJ-WHyMe mirror this distribution. LPJ-Bern, by contrast, uses a lower scaling factor for peatlands and includes fluxes from other soil types. Therefore, LPJ-Bern estimates a distribution of fluxes that is less similar to NCSCD and also less similar to LPJ-WHyMe.

Page 13, line 14: 'all six WETCHIMP models' change six to seven.

We have updated line 14 accordingly.

Reply to referee report #2

We would like to thank the reviewer for his or her feedback on the manuscript. These comments have been very helpful for improving the manuscript. Below, we have listed the reviewer's comments (in bold) and the corresponding edits to the manuscript.

BIC penalty term: Limited information is provided about the use of this term. To avoid confusion about it, some further sentences are needed explaining its role in the experiments that are presented. If I'm right it actually doesn't play a role at all. The number of measurements is the same for every inversion and so is the number of explanatory variables (p=16 in all experiments if I am correct, it certainly doesn't hurt to make this explicit in the text). Because of this the BIC penalty term is the same in every experiment. If this is correct, then please state this explicitly.

We have added more explanation to the manuscript. The penalty term does play a role in the results. The number of measurements (n) is the same for every candidate model, but the number of explanatory variables (p) is not the same for all candidate models.

In each experiment, we divide North America into 4 geographic regions and 4 seasons (for a total of 16 regions and seasons). The model selection framework will consider all possible combinations of these 16 variables. The combination with the lowest BIC score is the best or optimal model. For each combination, the penalty term depends upon the number of variables (p) in that combination. In this case, p can vary from 0 to 16. A variable (i.e., an emissions estimate for one region and one season) must improve the log-likelihood (Eq. 1) by at least ln(n) to be included in the best or optimal model. A variable that does not improve the log-likelihood by at least that amount will not be included in the best or optimal model.

A definition is missing of the word 'selection experiment'. It could either mean the entire procedure of selecting the optimal model or one single inversion. Page 7, line 6 suggests it is the latter (1000 times). Page 8, line 21 suggests it is the first. This should be clarified.

We have clarified this point in the manuscript.

An explanation is needed of why the state vector is different for the synthetic and real data model selection experiments. Strictly speaking, by introducing the monthly offset term we do not know anymore if results that were robust in the synthetic experiment are still robust in the real data experiment.

This comment is true. In a previous version of the manuscript, we had included a synthetic data experiment with a monthly offset term. The results were nearly identical to those in the current manuscript (without a monthly offset term). A previous reviewer asked us to omit the synthetic experiment with the monthly offset term; he/she felt that the results of this experiment were redundant, so we changed the manuscript accordingly.

Page 11, line 6: "The atmospheric network can better detect ..." This statement suggests that flux detection using atmospheric data benefits from good priors. I would rather argue that the poorer our priors, the more we can learn from atmospheric data. If the wetland models were an order of magnitude further apart, wouldn't it have been easier to choose the right one?

The reviewer makes a great point here. Let's say that we have a hypothetical trace gas "Y" and have

almost no bottom-up knowledge of where, why, or how that gas is emitted. In that case, we could potentially gain a lot of information about Y from an atmospheric inverse model; any knowledge about the magnitude, spatial distribution, and seasonality of the fluxes would be an improvement over what we knew before. This scenario parallels the reviewer's point above. On the other hand, let's say that we want to differentiate emissions of gas Y from industry A versus industry B. If we know where industries A and B operate and how the spatial locations of those industries differ, then we would stand a better chance of differentiating between those two different emissions sources. In summary, the answer to this question may depend on what type of information that one would like to learn about gas Y (or any other gas): whether one wants to learn about the general magnitude and distribution of emissions or whether one wants to glean more detailed information about who/what is responsible for those emissions.

Page 14, line 24: "Hence, the disagreement ... global wetland budget" What is suggested here is that overestimated emissions over the North America, also mean that global emissions are overestimated. However, this cannot be concluded from that fact that models that show high emissions over North America have large global emissions, in combination with evidence the North American emissions are overestimated. For that, additional evidence is needed that the global emissions are indeed overestimated. Otherwise global emissions could still be correct despite the fact that North American estimates were overestimated.

The reviewer makes a great point here. We have revised this paragraph to clarify its meaning. We intended to say the following: our analysis for North America does not necessarily imply anything about the magnitude of fluxes in other regions of the globe. The models appear to overestimate the magnitude of fluxes across boreal North America, but that result does not necessarily imply that these models have underestimated fluxes elsewhere. For example, models that estimate the largest fluxes for boreal North America often also estimate large fluxes in the tropics relative to other models. I.e., the fluxes in these models should not simply be re-distributed from boreal to tropical regions. Rather, one would want to analyze fluxes from each region separately.

Figure S4: I'm surprised to see that the model is doing a rather poor job in reproducing the measurements at sites that are not much influenced by wetland emissions. To authors point to papers suggesting that EDGAR underestimates anthropogenic emissions over the USA. However, these differences point to an underestimate by about a factor 2, which seems too large. How about sites along the coastline, which directly sample the marine background? Do those measurements confirm that the initial concentrations are correctly dealt with? Some discussion is needed of whether the uncertainty of the anthropogenic emissions is well enough represented in the synthetic experiments. If the real data suggest that uncertainties are larger, then the derived detectability of wetland emissions may have been too optimistic.

We have added additional explanation on this point to the Supplement.

The synthetic experiments do not use EDGAR because that estimate is likely too low for the US and Canada. Instead, we use an anthropogenic emissions estimate from our previous study (Miller et al. 2013) in the synthetic data experiments. That anthropogenic emissions estimate is in good agreement with atmospheric measurements from both tall towers (Fig. S6 in Miller et al. 2013, reprinted below) and from aircraft (Fig. 4 in Miller et al. 2013, reprinted below). Fig. 4 and S6 in Miller et al. 2013 also show that modeled concentrations with the EDGAR inventory underestimate measured concentrations (see below) – a result that is consistent with Fig. S4 in the current paper.

In Miller et al. (2013), we also optimized the estimated "initial" or background concentration values to match observations collected in the free troposphere by regular NOAA aircraft flights. Sections "Methane Boundary Condition" and "Validation of the Boundary Condition" in the Supplement to Miller et al. (2013) describe this boundary concentration validation in greater detail. The same boundary condition estimate is used in the current paper.

Lastly, the uncertainties in the anthropogenic emissions are taken from Miller et al. (2013). In that study, we used an approach called restricted maximum likelihood (RML) to estimate those uncertainties (i.e., variances and covariances) (e.g., Kitanidis and Lane 1985, Michalak et al. 2004). RML guarantees that the estimated uncertainties match against the actual model-data residuals.

A note of caution that is missing in the discussion section is that the WRF-STILT approach relies on an accurate representation of PBL mixing. Care has been taken only too select afternoon values. Nevertheless, it would be useful to have vertical profiles (e.g. from the aircraft measurements) supporting the point that is made in Figure 4.

We have added a discussion of this issue to the Supplement. Our previous paper, Miller et al. (2013), examined this issue in depth, and we have re-printed a number of relevant figures at the end of this document. Fig. 4 in Miller et al. (2013), for example, shows the ability of WRF-STILT to reproduce the vertical structure of the atmosphere at various aircraft observation sites. The present study uses the same WRF-STILT runs, boundary condition estimate, and anthropogenic emissions estimate as in that paper.

Suppl, page 1, line 36: 'At these' io 'At the these'

Thank you for pointing this out. We have updated the text accordingly.

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[Note to the editor: The figures below have already been published elsewhere. It may not be appropriate to post these figures to the online discussion for copyright reasons.]



Relevant figures from Miller et al. (2013)

Fig. 4. A model-measurement comparison at several regular NOAA/DOE aircraft monitoring sites (averaged over 2007–2008). Plots include the measurements; the modeled boundary condition; the summed boundary condition and wetland contribution (from the Kaplan model); and the summed boundary, wetland, and anthropogenic contributions (from EDGAR v4.2 and the posterior emissions estimate).



Fig. S6. A model-measurement comparison at several tower sites. The EDGAR v4.2 and posterior model plots include the boundary condition, wetland contribution modeled from the Kaplan inventory, and the anthropogenic contribution modeled from EDGAR and the posterior emissions, respectively.



Fig. S7. A model-data comparison scatter plot for the posterior emissions estimate and EDGAR 4.2.



Relevant figures from Miller et al. (2014)

Figure 4. A comparison of modeled mixing ratios against measurements at the observation sites. This figure is similar to Figure 2 but compares the deterministic model and posterior emissions estimate instead of existing flux models.



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The ability Evaluation of atmospheric data to resolve differences in wetland methane estimates over emissions across North America using atmospheric data and inverse modeling

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Abstract

Existing estimates of methane (CH₄) fluxes from North American wetlands vary widely in both magnitude and distribution. In light of these disagreements differences, this study uses atmospheric CH₄ observations from the US and Canada to analyze seven different bottom-up, wetland CH₄ estimates reported in a recent model comparison project. We first use synthetic data to explore whether wetland CH₄ fluxes are detectable at atmospheric observation sites. We find that the observation network can detect aggregate wetland fluxes from both eastern and western Canada but generally not from the US. Based upon these results, we then use real data and inverse modeling results to analyze the magnitude, seasonality, and spatial distribution of each model estimate. The magnitude of Canadian fluxes in many models is larger than indicated by atmospheric observations. Many models predict a seasonality that is narrower than implied by atmospheric datainverse modeling results, possibly indicating an over-sensitivity to air or soil temperatures. The LPJ-Bern and SDGVM models have a geographic distribution that is most consistent with atmospheric observations, depending upon the region and season. These models utilize land cover maps or dynamic modeling to estimate wetland coverage while most

¹⁵ models utilize land cover maps or dynamic modeling to estimate wetland coverage while mo other models rely primarily on remote sensing inundation data.

1 Introduction

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 CH_4 fluxes from wetlands play a critical role in global climate change. CH_4 is the second-most important long-lived greenhouse gas, and the radiative forcing of the current atmospheric burden is approximately 26 % of carbon dioxide (Butler, 2014). Wetlands are possibly the largest single source of this gas to the atmosphere and account for roughly 30 % of global emissions

(?) (Kirschke et al., 2013).

Despite the important role of wetland CH_4 fluxes in climate change, existing estimates of this source disagree differ on the magnitude, seasonality, and spatial distribution of fluxes, from regional to global scales. In fact, a recent global model comparison project named WETCHIMP

- ¹⁰ (Wetland and Wetland CH_4 Inter-comparison of Models Project) found large differences among existing CH_4 wetland models (Fig. 1, Melton et al., 2013; Wania et al., 2013). For example, existing estimates of maximum global wetland coverage differ by <u>over about</u> a factor of 6 – from 4.1×10^6 to 26.9×10^6 km². Furthermore, estimates of global natural wetland fluxes range from 92-264 Tg CH_4 yr⁻¹. The relative magnitude of these uncertainties increases at
- ¹⁵ sub-global spatial scales. As a case in point, CH_4 estimates for Canada's Hudson Bay Lowlands (HBL) range from 0.2 to 11.3 Tg CH_4 yr⁻¹. These disagreements in current CH_4 estimates do not bode well for scientists' abilities to accurately predict future changes in wetland fluxes due to climate change (Melton et al., 2013).

A number of studies have used chamber measurements of CH₄ to parameterize or evaluate
²⁰ biogeochemical CH₄ models (e.g., Livingston and Hutchinson, 2009). These measurements usually encompass fluxes from a relatively small area, and fluxes can often vary greatly with landscape heterogeneity at these spatial scales (Waddington and Roulet, 1996; Hendriks et al., 2010). CH₄ data collected in the atmosphere sees the cumulative effect of CH₄ fluxes across a broader region (e.g., Winderlich et al., 2010; Pickett-Heaps et al., 2011; Miller et al., 2014).
²⁵ Hence, atmospheric data can provide a unique tool for evaluating existing CH₄ flux estimates across different countries or continents.

The present study compares the WETCHIMP CH_4 flux estimates against atmospheric CH_4 data and inverse modeling results from 2007–2008 through two sets of analyses. First, we con-

struct a set of synthetic data experiments to understand whether the atmospheric CH_4 observation network can detect CH_4 fluxes from wetlands. We also explore the factors that might prevent the network from detecting wetland fluxes. To answer these questions, we utilize a model selection procedure based upon the Bayesian Information Criterion (BIC) (Sect. 2.2 Shiga et al.,

- $_{5}$ 2014; Fang et al., 2014; Fang and Michalak, 2015). This procedure determines whether wetland fluxes from different regions and seasons are necessary to describe variability in synthetic atmospheric CH₄ observations. Based on these synthetic experiments, we conduct a second set of analyses using real atmospheric data and inverse modeling results. We use this data to analyze the magnitude, seasonal cycle, and spatial distribution of each WETCHIMP CH₄ estimate.
- ¹⁰ We investigate these questions over the US and Canada using CH_4 data collected from towers and regular aircraft flights operated by NOAA and its partners and from towers operated by Environment Canada.

2 Methods

This section first describes the atmospheric CH₄ data and the atmospheric model that allows direct comparison between the data and various flux estimates. Subsequent sections describe both the synthetic and real data experiments outlined in the introduction (Sect. 1).

2.1 Data and atmospheric model

The present study utilizes atmospheric CH₄ observations from aircraft and tower platforms across the US and Canada, a total of 14 703 observations from 2007–2008. These observation sites include four towers operated by Environment Canada , and 10 towers in the US operated by NOAA and its partners. Observations at the NOAA towers consist of daily flasks (occasionally weekly), and observations at the Environment Canada sites are continuous measurements. As in Miller et al. (2014), we use afternoon averages of this continuous data. In addition to these towers, we utilize observations from 17 regular NOAA aircraft monitoring locations in the US and Canada (Fig. 2). We incorporate aircraft data up to 2500m altitude as was done in Miller et al. (2013). Observations above that height are usually representative of the free troposphere with limited sensitivity to surface fluxes. The tower and aircraft observations used here These observations and the associated model runs (described below) are the same as those in Miller et al. (2013) and Miller et al. (2014).

- We then employ an atmospheric transport model to relate CH₄ fluxes at the Earth's surface to atmospheric concentrations at the observation sites. The modeling approach here combines the Weather Research and Forecasting (WRF) meteorological model and a particle-following model known as STILT, the Stochastic Time-Inverted Lagrangian Transport model (e.g., Lin et al., 2003; Nehrkorn et al., 2010; Hegarty et al., 2013). WRF-STILT generates a set of footprints; these footprints quantitatively estimate the sensitivity of each observation to fluxes at each surface location (with units of ppb per unit surface flux). We multiply the footprints by a flux model and add this product to an estimate of the "background" concentration – the CH₄ concentration of air entering the North American regional domain. We estimate this background concentration using CH₄ observations collected near or over the Pacific Ocean and in
- the high Arctic, a setup described in detail by Miller et al. (2013) and Miller et al. (2014). The resulting modeled concentrations can be compared directly against atmospheric CH₄ observations. This modeling setup is the same as in Miller et al. (2013) and Miller et al. (2014). The observations, the WRF-STILT modelruns, background concentrations, and uncertainties in the modeling framework are described in greater detail in the Supplementand in those papers, Miller et al. (2013) , and Miller et al. (2014).

Using this setup, we can evaluate predicted CH₄ concentrations using the WETCHIMP flux estimates (Fig. 1) against observed atmospheric concentrations. The WETCHIMP project was designed to compare simulated wetland distributions and modeled CH₄ fluxes at multi-year, continental scales (Melton et al., 2013; Wania et al., 2013). The project entailed several sets of model runs, but Melton et al. (2013) primarily reported on one set of runs – runs for 1901– 2009 that used the same observed climate and CO₂ concentration datasets to force all models. Each CH₄ model utilized its own parameterization for wetland area and distribution. We use the outputs from this set of model runs in the present study. Of the WETCHIMP models, seven provide a flux estimate on a suitable time step for boreal North America and six provide an estimate for temperate North America. These models include CLM4Me (Riley et al., 2011), DLEM (Tian et al., 2010), LPJ-Bern (Spahni et al., 2011), LPJ-WHyMe (Wania et al., 2010), LPJ-WSL (Hodson et al., 2011), ORCHIDEE (Ringeval et al., 2010), and SDGVM (Singarayer et al., 2011). All flux model outputs used from the WETCHIMP study have a temporal resolution of one month. These models are described in Melton et al. (2013); Wania et al. (2013), and the Supplement.

2.2 Synthetic data experiments

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We assess the ability of the CH₄ observation network to detect wetland fluxes and use a model selection framework adapted from the BIC. A model selection framework can sort through a
large number of potential explanatory variables and will choose the smallest set of variables that best describe the dataset of interest (e.g., Ramsey and Schafer, 2012). In the current setup, we generate synthetic atmospheric CH₄ observations. The model selection framework then indicates whether a wetland model and/or an anthropogenic emissions inventory are necessary to describe variability in these observations. In this way, model selection can indicate the sensitivity of the observation network to wetland CH₄ fluxes.

We use a form of the BIC that has been adapted for use within a geostatistical inverse modeling framework. This setup has previously been used to select either bottom-up models or environmental drivers of CO_2 and CH_4 fluxes (e.g., Mueller et al., 2010; Yadav et al., 2010; Gourdji et al., 2012; Miller et al., 2013, 2014; Shiga et al., 2014; Fang et al., 2014; Fang and Michalak, 2015). The implementation here mirrors that of Fang et al. (2014), Shiga et al. (2014), and Fang and Michalak (2015):

$$BIC = \underbrace{\ln |\Psi| + (z - HX\beta)^{\mathrm{T}} \Psi^{-1}(z - HX\beta)}_{\text{negative log-likelihood}} + \underbrace{p \ln(n)}_{\text{penalty term}}$$
(1)

The first two terms in Eq. (1) are the negative log-likelihood, a measure of how well the model fits the data. The last term penalizes a particular model based upon the number of explanatory variables (*p*). The best combination or candidate model has the lowest BIC score.

The variable z ($n \times 1$) represents the observations minus background concentrations, H $(n \times m)$ the footprints, and Ψ $(n \times n)$ a covariance matrix derived from an atmospheric inversion framework. The variable m refers to the total number of flux or emissions grid boxes in both space and time. These variables are based upon two existing inverse modeling studies by Miller et al. (2013) and Miller et al. (2014) (refer to the Supplement). The matrix \mathbf{X} ($m \times p$) contains p explanatory variables. In the current setup, X can include a wetland flux estimate and/or individual emissions sources from an anthropogenic inventory. β ($p \times 1$) is a set of coefficients that scale the variables in X. We set these coefficients to one in the the synthetic data experiments. As a result, the model selection framework cannot scale other variables in X to reproduce the atmospheric signal from wetlands. reproduce wetland fluxes by simply upscaling anthropogenic emissions sources that might have a similar distribution to wetlands.

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The first experiments described here use synthetic atmospheric CH_4 data. We generate the synthetic data using one of the WETCHIMP models and the anthropogenic emissions estimates from Miller et al. (2013) and Miller et al. (2014). For consistency among the synthetic datasets, we scale the annual HBL CH₄ budget in each WETCHIMP model to match the overall magnitude estimated by several top-down studies (Pickett-Heaps et al., 2011; Miller et al., 2014;

15 Wecht et al., 2014). We then multiply these fluxes by the footprints (H) and add error that is randomly generated from the covariance matrix (Ψ). This covariance matrix represents errors in atmospheric transport and in the measurements – collectively referred to as model-data mismatch. This matrix also represents uncertainties in the prior flux estimate. In a geostatistical inverse model, this prior flux model is given by $X\beta$ (refer to the supplement for more detail). 20

We divide the WETCHIMP wetland fluxes into four regions (Fig. 2) and four seasons (DJF, MAM, JJA, and SON). The model selection framework then chooses variables that are necessary to reproduce the synthetic data, variables that include EDGAR and the 16 wetland flux maps. We run this model selection experiment. The penalty term in Eq. 1 increases as we add

wetland maps or add EDGAR to the X matrix. Each variable added to X will increase the 25 penalty term by $\ln(n)$; an additional variable must improve the log-likelihood by more than this penalty term to be chosen by model selection.

We then run this framework 1000 times, generating new synthetic data each time, and calculate the percentage of all trials in which the model selection chooses a wetland model. The 1000 repeats are needed due to the random or stochastic nature of the synthetic data experiment; the results of the model selection can vary slightly, depending on the particular random errors that we generate based upon the covariance matrix (Ψ). This procedure ensures that the model selection results are not the output of a single realization. We then report on how frequently each

- ⁵ of the 16 wetland flux maps is chosen. If a wetland flux map is chosen with high frequency, then a wetland flux map is necessary to describe variability in the synthetic CH_4 observations, and the synthetic observation network can detect aggregate wetland CH_4 fluxes from the given region and season. This setup mirrors that of Shiga et al. (2014), who employed a model selection framework to explore the detectability of anthropogenic CO_2 emissions.
- We also explore why the synthetic CH₄ observations may not be able to detect wetland fluxes.
 We run a series of case studies and in each case remove a different confounding factor that might prevent the network from detecting wetland CH₄ fluxes. In one case, we remove anthropogenic emissions. In subsequent cases, we remove model-data mismatch errors and/or prior flux errors. In each case, we re-run the model selection experiment and examine whether the results improve
 when each of these confounding factors is removed.

2.3 Real data experiments

This paper subsequently compares the spatial distribution, magnitude, and seasonality of each WETCHIMP estimate against real atmospheric CH_4 observations, using the synthetic experiments to guide the analysis.

- ²⁰ We first explore the spatial distribution of the WETCHIMP flux estimates. We modify the model selection setup in Sect. 2.2 to focus on the spatial distribution of each estimate using a procedure developed by Fang et al. (2014) and Fang and Michalak (2015). Instead of fixing the coefficients (β) to one, we instead estimate the coefficients using real atmospheric CH₄ observations. We also include an intercept term that can vary by month; the intercept for each
- ²⁵ month is represented by a vector of ones in the matrix **X**, and this intercept is always included within included as part of each candidate model for **X**. We then run model selection using real

observations. As a result of this setup, a wetland model is not necessary to reproduce either the magnitude or seasonality of the atmospheric CH_4 data; the model selection framework can simply scale the intercept term or scale EDGAR to reproduce the magnitude or seasonality of the observations. The spatial distribution of wetland fluxes, however, can only come from a wetland model. The model selection procedure will only choose a wetland model if the spatial distribution of that model describes sufficient, additional variability in the observations (e.g., Fang et al., 2014).

Model selection can therefore indicate which WETCHIMP models have the best spatial distribution relative to the atmospheric observations; any WETCHIMP model chosen by model selection must have has a spatial distribution that improves model-data fit, and the model must improve improves that fit more than the penalty term in Eq. 1. A negative result does not necessarily indicate that a WETCHIMP model has a poor spatial distribution. In that case, the observations may not be very sensitive to the spatial distribution of fluxes for the given region or given season. Similarly, the spatial distribution in a WETCHIMP model may improve modeldata fit but not by more than the penalty term in Eq. 1. By contrast, a positive result indicates that a WETCHIMP model likely has a particularly good spatial distribution. As in Sect. 2.2, we divide the wetland fluxes into four sub-continental regions and four seasons. The Supplement

describes this setup in greater detail.

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We then analyze the magnitude and seasonality of the WETCHIMP fluxes using a number of model-data time series. We model CH_4 concentrations at a number of US and Canadian observation sites using the WRF-STILT model, the WETCHIMP estimates, and the EDGAR v4.2FT2010 emissions inventory (Olivier and Janssens-Maenhout, 2012; European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL), 2013). We average the observations and model output at the monthly scale and then compare the magnitude of these model estimates for each month against the averaged observations.

We further compare the seasonality of existing bottom-up models against the seasonality of a recent inverse modeling estimate by Miller et al. (2014). We plot the monthly budgets for both the bottom-up models and the inversion estimate, and we plot the monthly CH_4 budget as a fraction of the annual total.

Note that inter-annual variability in existing CH_4 flux models is small relative to the differences among these models; as a result, conclusions from the 2 year study period (2007–2008) likely hold for other years. For example, the inter-annual variability in the total US/Canadian budget is $\pm 7.3-9.7\%$ (standard deviation), depending upon the model in question (Note that LPJ-Bern has even larger inter-annual variation due to an issue with model spin-up described in Wania et al. (2013)).

3 **Results and discussion: synthetic experiments** 5

The synthetic experiments presented here explore the limits of existing atmospheric data for constraining wetland fluxes. If atmospheric observations are to constrain wetland CH_4 fluxes, those observations must be able to detect wetland CH_4 fluxes above errors in the transport model and above other emissions sources such as fossil fuels and agriculture.

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much of the west.

The four columns in Fig. 3a display the results from an individual season in each of four geographic regions. In this experiment, the synthetic CH_4 observations can detect aggregate wetland CH_4 fluxes from Eastern Canadian wetlands in greater than 75 % of all trials for the summer and fall seasons. In the eastern US, the model selection framework chooses a wetland model in 25–50 % of all trials in multiple different seasons. By contrast, the synthetic CH₄ data are least sensitive to wetland fluxes in the western US, and the model selection framework 15 chooses wetland fluxes from that region in fewer than 25 % of all trials irrespective of the season. This result may be due, in part, to the scant wetlands and sparse atmospheric observations in

The results also vary by season. Of any region, the atmospheric CH_4 network is best able to constrain fluxes across multiple seasons in eastern Canada. The largest wetland fluxes in the 20 WETCHIMP models are in Ontario and Quebec (Fig. 1). It is therefore unsurprising that the network is best able to detect wetland fluxes in that region, even though there are relatively few observation sites in the area. In other regions, the atmospheric CH_4 network is less sensitive to wetlands during the winter, fall, and spring shoulder seasons.

We run several additional test scenarios model selection experiments to explore why the synthetic observations may not always be able to detect wetland CH_4 fluxes -(Fig. 3b-e). We remove anthropogenic emissions from the synthetic dataset for the experiment in Fig. 3b. We remove all model data mismatch errors in Fig. 3c; model-data mismatch encompasses errors in atmospheric transport and in the measurements. Subsequently, we remove all errors due to the prior flux estimate in Fig. 3d. In Fig. 3e, we remove both types of errors. In each case, we re-run the model selection experiment to see if the sensitivity of the atmospheric CH_4 network to wetland fluxes improves.

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Anthropogenic emissions have only a modest effect on the results in specific regions and seasons. In case (b) without anthropogenic emissions, the results improve by $\sim 25-50\%$ in the fall and spring shoulder seasons for several geographic regions.

By contrast, the model-data mismatch and prior flux errors have a much larger effect on the model selection results. The results improve incrementally across many regions and seasons 10 when we remove model-data mismatch errors in case (c). The results improve across the spring, summer, and fall seasons and improve across all four geographic regions. However, the magnitude of this improvement is never more than 25%. Model-data mismatch errors are likely dominated by errors in modeled atmospheric transport. These results imply that transport errors play an incremental yet pervasive role in the utility of the atmospheric observations. 15

The prior flux errors have the largest effect on the results, particularly during the warmest seasons. In case (d), the results show great improvement during fall, spring, and summer and show little improvement during winter or in the western US. In the setup here, the prior flux uncertainties scale with the seasonal magnitude of the fluxes. When we remove the prior flux errors, the results concomitantly show the greatest improvement in seasons that have larger 20 overall CH_4 fluxes. These results indicate that the prior estimate greatly impacts the utility of the atmospheric CH_4 observations. A geostatistical inverse model can leverage any combination of land surface maps, meteorological maps, and/or anthropogenic inventory estimates in the inversion prior. These maps or estimates are incorporated into the X matrix in Eq. 1. If accurate maps or estimates are not available, then the prior flux errors will be large, and the 25 model selection framework will be less likely to choose any particular variable. If these maps or estimates have high explanatory power, then the prior flux errors will be small, and the model selection framework will be more likely to choose any one variable. As a result, the detectability of wetland CH_4 fluxes partly depends on the availability of land surface or meteorological data that matches those fluxes. The atmospheric network can better detect differentiate wetland CH_4 fluxes from other CH_4 sources when accurate prior information can guide that identification.

Case (e) (no model-data mismatch errors and no errors in the prior flux estimate) shows large,
ubiquitous improvements; the model selection chooses a wetland model 100% of the time in almost all regions and seasons. The results for Eastern Canada during winter are the exception. In winter, the wetland model cannot always explain enough variability in the synthetic observations to overcome the BIC penalty term in Eq. 1.

The density of the atmospheric CH_4 network may also play a role in these results. Wetlands in the Eastern US are sparse relative to eastern Canada, but the higher density of observations in the Eastern US may contribute to a moderate success rate (25-50%) for that region. Recent and planned network expansions in the eastern US and in Canada could play a key role in future efforts to constrain wetland fluxes across these regions.

Overall, the synthetic experiment results indicate that the observation network cannot detect wetland fluxes from the US (i.e., model selection has a success rate <50%). Across Canada, the results are more promising (i.e., near 100% success rate in some regions/seasons), despite the relative sparsity of the observation network there.

4 Results and discussion: comparisons with atmospheric data and inverse modeling results

4.1 Spatial distribution of the fluxes

We compare the spatial distribution of the WETCHIMP flux estimates against CH₄ data from the atmospheric observation network. To this end, we use a version of the model selection framework that chooses wetland models based upon their spatial distribution (Fang et al., 2014; Fang and Michalak, 2015). WETCHIMP models that are chosen by the framework have a spatial distribution that is more consistent with atmospheric observations relative to those that are not selected.

The results of this model selection analysis are displayed in Table 1. This table lists the regions and seasons that had a success rate > 50 % in the synthetic data experiment; the atmospheric CH₄ network is most sensitive to wetland CH₄ fluxes in those regions and seasons. Two of the WETCHIMP models were chosen by the model selection framework – LPJ-Bern (in eastern Canada) and SDGVM (in eastern and western Canada). The spatial distribution of these models improve the model-data fit more than the penalty term in Eq. 1.

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The LPJ-Bern and SDGVM models have several unique spatial characteristics that could explain these results. Over eastern Canada, LPJ-Bern and SDGVM concentrate the large fluxes in the HBL. Other models, by contrast, often distribute the fluxes more broadly across Ontario and Quebec or put the largest fluxes in Ontario outside of the HBL. In western Canada, SDGVM distributes fluxes across northern, boreal Saskatchewan and Alberta.

The LPJ-Bern and SDGVM models share another common characteristic: both model wetland area independently instead of relying solely on remote sensing inundation datasets. LPJ -WSL, ORCHIDEE, DLEM, and CLM4Me use remote sensing inundation datasets like GIEMS (Global Inundation Extent from Multi-Satellites, Prigent et al., 2007) to construct a wetland map. Other models, like LPJ-Bern and LPJ-WHyMe also use land cover maps and/or land surveys to estimate wetland (or at least CH₄-producing) area. SDGVM estimates this area dynamically as a function of soil moisture (Melton et al., 2013; Wania et al., 2013). Wetland maps generated using these different approaches show substantial differences. Remote sensing datasets estimate relatively high levels of inundation in regions of Canada that are not forested

- or have many small lakes (see further discussion in Melton et al., 2013; Bohn et al., 2015). By contrast, modeling approaches that dynamically generate wetland area or use land cover maps assign more wetlands over regions with high water tables but little surface water as seen by remote sensing based inundation datasets). As a result of these differences, models like LPJ-Bern assign more wetlands and CH_4 fluxes in the HBL relative to other regions of eastern Canada.
- ²⁵ Of note, LPJ-Bern and LPJ-WhyMe have many structural model similarities but predict relatively different spatial distributions of CH₄ fluxes. The latter estimates fluxes that are more

broadly distributed across Quebec and Labrador. LPJ-WhyMe only simulates fluxes from high latitude peatlands and uses an estimated peatland distribution from Tarnocai et al. (2009); this distribution extends across Quebec and Labrador. LPJ-Bern, by contrast, includes fluxes from non-peatland regions and applies a smaller scaling factor to peatland fluxes relative to LPJ-WHyMe Wania et al. (2013). As a result, the fluxes in LPJ-Bern have a spatial distribution that is different from the peatland map and also different from LPJ-WHyMe.

4.2 Flux magnitude

We next compare the magnitude of predicted concentrations using the WETCHIMP models against atmospheric observations at individual locations. Unlike previous sections that utilized model selection, this section employs several model-data time series, displayed in Fig. 4. This model estimate consists of several components: the background (in green) is the estimated background concentration of CH₄ in clean air before entering the model domain as in Miller et al. (2013, 2014). The estimated contribution of anthropogenic emissions from EDGAR v4.2FT2010 is added to this background (in red). The contribution of wetland fluxes from the WETCHIMP models is then added to the previous inputs, and the sum of all components (blue lines) can be compared directly against measured concentrations.

The various WETCHIMP flux estimates produce very different modeled concentrations at the atmospheric observation sites (Fig. 4). Overall, modeled concentrations with the WETCHIMP fluxes usually exceed the CH₄ measurements during summer. At Chibougamau, Fraserdale, and Park Falls in early summer, all <u>six seven</u> WETCHIMP models predict CH₄ concentrations that equal or exceed the observations. The ORCHIDEE, LPJ-WHyMe, and LPJ-Bern models always exceed the measurements during summer while DLEM and SDGVM better match the observations at these sites. Notably, a number of previous studies report that the EDGAR inventory may underestimate US anthropogenic CH₄ emissions (e.g., Kort et al., 2008; Miller et al., 2013; Wecht et al., 2014; Turner et al., 2015). If EDGAR underestimate emissions, then the WETCHIMP models would be an even larger overestimate relative to the atmospheric data.

Many models appear to overestimate the magnitude of fluxes across boreal North America, but this result does not necessarily imply that these models have underestimated fluxes elsewhere. CH₄ models that overestimate fluxes in estimate the largest fluxes across boreal North America do not always compensate with smaller fluxes elsewherein other regions of the globe. For example, the ORCHIDEE model not only estimates large fluxes over North America but also estimates higher fluxes over the tropics than any other model (Melton et al., 2013). Hence, the disagreement in magnitude over North America not only reflects uncertainty in the global distribution of wetland fluxes but also reflects uncertainty in the global wetland budget.

4.3 Seasonal cycle

- Bottom-up CH_4 flux estimates show variable features when compared to atmospheric observa-5 tions, and the seasonal cycle of these estimates is no exception. Figure 5 compares the seasonal cycle of the existing estimates over Canada's HBL. Eastern Canada is one of the largest wetland regions in North America (Fig. 1), and nearby atmospheric observation sites see a much larger CH_4 enhancement from wetlands relative to other regions (Fig. 4 and S4).
- In this region, the bottom-up estimates diverge on the seasonal cycle of fluxes. Most estimates 10 predict peak fluxes in July or August, though two variations of the LPJ model predict seasonal peaks in September and October - LPJ-WHyMe and LPJ-Bern, respectively. LPJ-WHyMe is a module inside of LPJ-Bern, a possible explanation for the similar seasonal cycle in these two models. Differences among models are also notable during the fall and spring seasons. For example, fluxes in June account for anywhere between 6 and 21 % of the annual CH₄ budget, 15 depending upon the model. Fluxes in October account for between 1 and 23 % of the annual budget (Fig. 5b).

Figure 5 also displays the seasonality of an inverse modeling estimate from Miller et al. (2014) for comparison. That estimate incorporates observations from Chibougamau, Quebec,

(2014) for comparison. That estimate incorporates observations from Chibougamau, Quebec, and Fraserdale, Ontario, atmospheric measurement sites that are strongly influenced by fluxes from the HBL. The discrepancies among Differences between this inverse modeling estimate and the WETCHIMP models often exceed the 95 % confidence interval of the inversion estimate inverse 20 model. The WETCHIMP estimates are often comparable to Miller et al. (2014) in magnitude during fall and spring months but exceed the inversion-inverse modeling estimate in summer

months (Fig. 5a). On whole, the WETCHIMP models have a narrower relative seasonal cycle 25

than the <u>inversion inverse modeling</u> estimate (Fig. 5b). That estimate assigns a greater portion of the annual budget to the fall and spring shoulder seasons.

Additional top-down studies exist for the HBL, but those studies use a seasonal cycle drawn from an existing bottom-up model and do not estimate the seasonal cycle independently from CH_4 observations (Pickett-Heaps et al., 2011; Wecht et al., 2014; Turner et al., 2015). By comparison, a recent inverse modeling study of the Western Siberian Lowlands found parallel results for that region – existing models also predict a seasonal cycle that is narrower than the seasonality implied by atmospheric observations (Winderlich, 2012; Bohn et al., 2015).

Numerous possible explanations could underly differences in the seasonal cycle of CH_4 fluxes. For example, the temperature threshold for CH_4 production may be too high in some models. Relative to summer months, the bottom-up models predict small fluxes during fall/spring months when air temperatures are near freezing but soils are still unfrozen (Fig. S3). According to estimates from the North American Regional Reanalysis (NARR) (Mesinger et al., 2006), surface soils in the HBL (0 and 10 cm depth) begin to thaw in April and are largely unfrozen in May (Fig. S3). In the fall, surface soils (0 cm depth) begin to freeze in November, but deeper soils (10 and 40 cm) remain largely unfrozen until December. Compared to the bottom-up models, the inversion-inverse modeling estimate predicts a wider seasonal window, a result that

¹⁵ would be consistent with dates of deep soil freeze and thaw.

5 Conclusions

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A recent model comparison study revealed wide differences among several estimates of wetland CH_4 fluxes – differences at global to regional scales. In the first component of this study, we use a synthetic data experiment to understand whether the atmospheric observation network can detect wetland CH_4 fluxes. We find that the network can reliably identify aggregate wetland fluxes from both eastern and western Canada. The network can detect wetland fluxes from the eastern US in a smaller fraction of trials and rarely from the western US. This analysis also accounts for distracting signals in the atmosphere from anthropogenic sources or simulated atmospheric transport errors.

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In a second component of the study, we analyze each bottom-up CH_4 model from the WETCHIMP. 25 study using real atmospheric data. We find that the LPJ-Bern and SDGVM models have spatial distributions that are most consistent with atmospheric observations, depending upon the region and season of interest. In addition, almost all models overestimate the magnitude of wetland CH_4 fluxes when compared against atmospheric data at individual observation sites. The model ensemble may also estimate a seasonal cycle for eastern Canada that is too narrow (i.e., places too much of the total annual flux in the summer relative to the fall and spring shoulder seasons). 5

The results of this paper suggest possible pathways to improve future top-down estimates of wetland CH_4 fluxes. The ability of the atmospheric observation network to detect wetland fluxes depends in large part upon the prior flux model. In a geostatistical inverse model, this model can incorporate land surface maps – wetland maps, estimates of land surface processes, and maps of anthropogenic emissions sources. This information plays a large role in whether 10 atmospheric observations can detect wetland fluxes; the observations can more adeptly identify wetland fluxes when accurate land surface maps are available to guide that identification. By contrast, atmospheric transport and measurement errors (i.e., model-data mismatch errors) have a ubiquitous but smaller effect on the utility of atmospheric CH_4 observations.

- The results presented here also hold a number of suggestions for future bottom-up modeling 15 efforts:
 - 1. Spatial distribution: Bottom-up estimates that use surface water inundation as the only proxy for wetland area do not perform as well relative to atmospheric observations. Bottomup models that use satellite inundation data should incorporate additional tools like wetland mapping or dynamic modeling to capture wetlands covered by vegetation.
 - 2. Magnitude: Existing top-down studies that use a diverse array of in situ and satellite CH_4 observations show good agreement on the magnitude of CH₄ fluxes from the Hudson Bay Lowlands (HBL) region (e.g., Pickett-Heaps et al., 2011; Miller et al., 2014; Wecht et al., 2014; Turner et al., 2015). These studies could be used to calibrate the magnitude of future bottom-up estimates, at least over the HBL where CH₄ observations provide a strong constraint on wetland fluxes.

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3. Seasonal cycle: Bottom-up models do not show consensus on the seasonal cycle of wetland fluxes across Canada. Few top-down studies estimate the seasonal cycle independently using atmospheric observations. Additional top-down studies would indicate the range of seasonal cycle estimates that are consistent with atmospheric observations, particularly studies that use a diverse set of atmospheric models and/or diverse observational datasets. These efforts could help reconcile differences in the seasonal cycle among bottom-up models and between bottom-up models and the few, existing top-down studies.

These steps will hopefully lead to better convergence among wetland CH_4 estimates for North America.

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Table 1. Spatial flux patterns chosen by the model selection framework.

Region	Season	Chosen models
E. Canada	summer	LPJ-Bern, SDGVM
E. Canada	fall	LPJ-Bern
W. Canada	summer	SDGVM

Discussion Paper



Figure 1. Mean of the annual methane fluxes estimated by the WETCHIMP models (**a**) and the range of fluxes estimated by the ensemble (**b**). Note that the range in estimates is larger than the mean. The fluxes shown above are the average flux per m^2 of land area, not per m^2 of wetland area.



Figure 2. The US and Canadian atmospheric methane observation network for 2007–2008 (14 703 total observations). Small yellow dots indicate observations from the START08 measurement campaign (Pan et al., 2010). Larger dots indicate tower and aircraft sites with regular observations over the two year period (Andrews et al., 2014). The grey background delineates the four regions used in the synthetic data experiments (Sect. 2.2).



Figure 3. This figure displays the results of the synthetic data experiments. These experiments examine whether the observation network can detect aggregate wetland CH_4 fluxes. The figure shows the percentage of trials that are successful. Darker shades indicate that the network is more sensitive to wetland fluxes in the given region and season. Panel (a) shows the results for the standard setup while panels (**b-e**) show the results of several test cases in which anthropogenic emissions or different errors are set to zero.



Figure 4. These time series compare atmospheric methane measurements at several observation sites against model estimates using the WETCHIMP ensemble and the EDGAR v4.2FT2010 anthropogenic emissions inventory. Refer to Fig. S4 for model-data time series at additional sites, particularly sites that are distant from large wetlands.



Figure 5. The seasonal cycle in methane fluxes estimated for the HBL ($50-60^{\circ}$ N, $75-96^{\circ}$ W). We include both the WETCHIMP estimates and an inverse modeling estimate from Miller et al. (2014). Panel (a) displays the monthly budget from each estimate while (b) displays each month as a percentage of the annual budget estimated by a given model.

This supplement provides more detail on the atmospheric observations, the wetland methane (CH_4) flux estimates, and the statistical methods used throughout the paper.

S1 Atmospheric observation sites

Here we describe, in greater depth, the atmospheric CH_4 observations collected across the US and Canada in 2007–2008. The observations used here are the same as those in Miller et al. (2013) and Miller et al. (2014b), and the discussion below summarizes the data descriptions in those papers.

The CH₄ analysis in the main article uses either real or synthetic data at US and Canadian observation sites – a total of 14703 observations. Of those measurements, 2009 are from observation towers in Canada. These towers (from east to west) include Chibougamau, Quebec (CHM, 50°N, 74°W, 30m above ground level); Fraserdale, Ontario (FSD, 50°N, 83°W, 40m agl); East Trout Lake, Saskatchewan (ETL, 54°N, 104°W, 105m agl); and Candle Lake, Saskatchewan (CDL, 54°N, 105°W, 30m agl, 2007 only). These sites, operated by Environment Canada, measure CH₄ continuously. In this study, as in Miller et al. (2014b), we use only afternoon averages of the CH₄ data and WRF-STILT model output (1pm - 7pm local time); small scale heterogeneities in the continuous data caused by turbulent eddies and incomplete mixing make it difficult to model finer scale temporal patterns in the data. The 2009 observations at these Canadian sites represent the total after averaging.

An additional 4984 CH₄ observations were collected from US towers operated by the National Oceanic and Atmospheric Administration (NOAA) and its partners. These observations include daily flask samples from a number of tower sites (weekly at Argyle and Ponca City): Argyle, Maine (AMT, 45 °N, 69 °W, 107m above ground level (agl)); Erie, Colorado (BAO, 40 °N, 105°W, 300m agl); Park Falls, Wisconsin (LEF, 46°N, 90°W, 244m agl), Martha's Vineyard, Massachusetts (MVY, 41°N, 71°W, 12m agl); Niwot Ridge and Niwot Forest, Colorado (NWF,

NWR, 40°N, 105°W, 2,3,23m agl); Ponca City, Oklahoma (SGP, 37°N, 97°W, 60m agl); West Branch, Iowa (WBI, 42°N, 93°W, 379m agl); Walnut Grove, California (WGC, 38°N, 121°W, 91m agl), and Moody, Texas (WKT, 31°N, 97°W, 122, 457m agl).

A further 7710 CH_4 measurements were obtained from flask samples on regular NOAA aircraft flights and from the START08 (Stratosphere-Troposphere Analyses of Regional Transport

- ³⁰ 2008) measurement campaign (Pan et al., 2010). As in Miller et al. (2013), we only use aircraft observations up to 2 500m above ground level. Observations at higher altitudes are less sensitive to surface emissions and were instead used by Miller et al. (2013) to optimize the estimated CH₄ boundary condition or background concentrations. In this study, we only use aircraft and tower-based observations collected during daytime hours.
- ³⁵ We further screen the data for biomass burning influence at the Canadian sites and at Park Falls, Wisconsin. At the these sites, we remove all days with CO that peaks above 200 ppb, as was done in Miller et al. (2014b). When these sites see influence from distant anthropogenic emissions, CO is often elevated, but it rarely exceeds 200 ppb except during time periods with known fires (Miller et al., 2008).

40 S2 WETCHIMP CH₄ flux models

This section of the supplement details the WETCHIMP CH_4 estimates from Melton et al. (2013) and Wania et al. (2013). The seven CH_4 estimates used in this study are shown in Fig. S1. The wetland CH_4 fluxes estimated by these models varies widely – both in magnitude and in spatial distribution. For example, the SDGVM model places large fluxes over the US Corn Bolt relative to other regions while other models like Orchidee place large fluxes in Northern

Canada that extent far into the Northwest Territories. For a more in-depth inter-comparison of these flux estimates, refer to Melton et al. (2013) and Wania et al. (2013).

S3 Additional information on the model selection setup

In the main article, we use synthetic CH_4 data and a model selection framework to examine whether atmospheric observations can detect aggregate wetland CH_4 fluxes (Sect. 2.2 and 3). This section first describes the synthetic data experiments (Sect. 2.2) followed by additional detail on the model selection runs that use real data (Sect. 2.3). The methods described here are adapted from Fang et al. (2014), Shiga et al. (2014), and Fang and Michalak (2015), and the discussion below parallels the descriptions in those studies.

⁵⁵ The synthetic observations include contributions from anthropogenic sources, from wetlands, and from simulated model and measurement errors:

$$\boldsymbol{z}_{\text{synthetic}} = \mathbf{H}(\boldsymbol{s}_{\text{anthro}} + \boldsymbol{s}_{\text{wetland}}) + \boldsymbol{\epsilon}$$
 (S1)

In this equation, $z_{\text{synthetic}}$ $(n \times 1)$ represents the synthetic observations generated for an observation site. The vector s_{anthro} $(m \times 1)$ represents emissions from anthropogenic sources, and s_{wetland} $(m \times 1)$ represents wetland fluxes. The footprint or sensitivity matrix **H** $(n \times m)$, generated from WRF-STILT, models the impact of these emissions at the observation sites.

In this study, we use the a priori anthropogenic emissions estimates from Miller et al. (2013) and Miller et al. (2014b) for s_{anthro} . Those studies used activity data from the EDGAR inventory and a model selection framework to construct a prior anthropogenic emissions estimate. These EDGAR activity datasets include economic or demographic data that may predict the spatial distribution of CH₄ emissions (e.g., human or ruminant population maps).

The wetland fluxes $(s_{wetland})$ in Eq. S1 are taken from the WETCHIMP CH₄ flux models (experiment two in Melton et al. (2013)). For the synthetic data experiments, we scale these models to match the Hudson Bay Lowlands (HBL) budget estimated by Pickett-Heaps et al. (2011), Miller et al. (2014b), and Wecht et al. (2014). This scaling ensures more consistent or

⁷⁰ representative results. The larger the wetland flux, the more likely that the observation network can detect a CH_4 fluxes from wetlands. Therefore, if we conduct the synthetic data experiment using a flux model that has an anomalously large magnitude, we would concomitantly obtain anomalously optimistic results.

As in Miller et al. (2013) and Miller et al. (2014b), the emissions (s_{anthro} and $s_{wetland}$) are regridded to a spatial resolution of 1° latitude by 1° longitude. The EDGAR activity data do not have any seasonality, so the anthropogenic emissions (s_{anthro}) are seasonally invariant. The WETCHIMP models have a monthly temporal resolution, as in Melton et al. (2013). That study provides flux estimates for the years 1993-2004; we use the mean of these ten years for all analysis in this study.

The final term in equation S1, ϵ ($n \times 1$), represents simulated errors in the measurements, in WRF-STILT, and in the fluxes (s_{anthro} and s_{wetland}). The errors in ϵ are distributed according to the covariance matrix Ψ ($n \times n$) (Eq. 1):

$$\boldsymbol{\epsilon} \sim \mathcal{N}(\mathbf{0}, \boldsymbol{\Psi})$$
 (S2)

$$\Psi = \mathbf{H}\mathbf{Q}\mathbf{H}^T + \mathbf{R}$$
(S3)

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The variances and covariances within Ψ fall into two different categories. The first category are errors due to imperfect emissions, described by covariance matrix \mathbf{Q} ($m \times m$). In atmospheric inversion studies, this matrix is typically termed the a priori covariance matrix. The diagonal elements of \mathbf{Q} describe a set of variances – differences between the prior fluxes and the unknown true emissions over long spatial or temporal scales. The off-diagonal elements of \mathbf{Q} describe any



Figure S1: Annual mean wetland CH_4 fluxes from seven different WETCHIMP estimates (Melton et al., 2013; Wania et al., 2013). The fluxes shown here are averaged over the 1993-2004 study period. Note that the fluxes shown above are averaged over the entire grid cell, not per m^2 of wetlands.

spatial and/or temporal covariances in these differences. In Eq. S3, the footprint or sensitivity matrix (**H**) projects **Q** from units of $(flux)^2$ into units of parts per billion squared, $(ppb)^2$.

We refer to the second type of errors as model-data mismatch errors, denoted by covariance matrix \mathbf{R} $(n \times n)$. These include all errors in the WRF-STILT model or the measurements that are unrelated to an imperfect flux estimate. Examples of model-data mismatch errors include measurement error, atmospheric transport error, or errors due to the spatial or temporal

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resolution of WRF-STILT. The synthetic data simulations in this study use values of **Q** and **R** estimated in Miller et al. (2013) and Miller et al. (2014b). In the synthetic data studies, we construct a statistical model that is representative of a prototypical real data inverse model. Similarly, we want to use values for **Q** and **R** that are representative of what one would likely encounter in a real-data setup. Miller et al. (2013) and Miller et al. (2014b) constructed real data inverse models over the US and Canada, respectively, using the same atmospheric observations and WRF-STILT simulations used in this study. Those studies used a model selection framework to find prior models that show optimal fit against available observations. In each study, the authors then estimated the elements of **Q** and **R** using that prior model. The resulting estimates of **Q** are representative of prior models that shows optimal agreement with atmospheric observations.

- representative of prior models that shows optimal agreement with atmospheric observations. For case study (b) (no anthropogenic emissions), we estimate **Q** using the same approach as in Shiga et al. (2014). In that study, the authors used the estimated variances and covariances of the remaining fluxes (in this case wetland fluxes) to populate **Q**.
- In the real data experiments (Sect. 2.3), we estimate unique values of **Q** and **R** each time we run the model selection framework. We estimate these parameters using Restricted Maximum Likelihood (RML) (Corbeil and Searle, 1976; Kitanidis, 1995; Michalak et al., 2004; Gourdji et al., 2012), the same procedure used in Miller et al. (2013) and Miller et al. (2014b).

We use these covariance matrices to compute ϵ through several steps. First, we compute the Cholesky decomposition of the combined covariance matrix Ψ :

$$\Psi = \mathbf{C}\mathbf{C}^T \tag{S4}$$

The covariance matrix Ψ has units of $(ppb)^2$, but its Cholesky decomposition (**C**) has units of ppb. With this decomposition in hand, we next simulate a set of errors, ϵ (e.g., Fang et al., 2014; Shiga et al., 2014):

$$\boldsymbol{\epsilon} = \mathbf{C}\boldsymbol{u} \tag{S5}$$

$$\boldsymbol{u} \sim \mathcal{N}(\boldsymbol{0}, \boldsymbol{1})$$
 (S6)

where \boldsymbol{u} represents a set of randomly-generated numbers with a mean of zero and variance of one.

We simulate 1000 synthetic datasets for each experiment to adequately sample the random errors in ϵ . We then use the model selection framework to find the optimal candidate model for each of these datasets. The results presented in Fig. 3 are therefore the composite of thousands of model selection runs: one model selection run for each synthetic dataset. We use a branch and bound algorithm from Yadav et al. (2013) to improve the computational efficiency of these model selection runs. Furthermore, we also estimate the coefficients (β) in Eq. 1 using Lagrange multipliers to ensure that none of the estimated coefficients have unrealistic negative values (e.g., Miller et al., 2014a).

In the real data setup (Sect. 2.3), we run the model selection procedure once for each of the seven WETCHIMP flux estimates. We only include one of the seven WETCHIMP flux models in each model selection run. As a result, the WETCHIMP models do not compete against one another for selection. In each run, the model selection framework can select the

¹²⁵ given WETCHIMP model in any of the four geographic regions and any of the four seasons.



Figure S2: Total, summed footprint from the (a) Canadian and (b) US observation networks. The observation sites incorporated into this figure are shown in Fig. 2. Each individual footprint (associated with an individual atmospheric observation) has units of concentration per unit flux (ppb per μ mol m⁻² s⁻¹). In this figure, we sum all footprints for 2007–2008.

S4 Overall sensitivity of the observation network to CH_4 fluxes

In this section, we describe the overall footprint or sensitivity of the observation network to CH_4 fluxes. This sensitivity will play at least some role in network's ability to detect wetland CH_4 fluxes. The WRF-STILT model quantifies this sensitivity in terms of a footprint. Each row the matrix **H** is the footprint associated with a different atmospheric CH_4 observation. In Fig. S2, we plot these footprints, summed over all of 2007–2008.

This figure show several distinctive patterns. First, the US network has a higher sensitivity than the Canadian network. This pattern is due to the larger number of observation sites over the US. Second, the highest sensitivities are clustered in distinctive regions with multiple observation sites – Wisconsin, Texas/Oklahoma, and California, among other regions.

S5 Soil freeze/thaw estimates from NARR

Figure S3 shows the soil freeze/thaw cycle at different depths averaged across the HBL. These estimates are taken from North American Regional Reanalysis (NARR) (Mesinger et al., 2006), and the values shown in Fig. S3 are average values for each month. The main article references this figure in a discussion of the CH_4 flux seasonal cycle (Sect. 4.3).

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S6 Additional model-data time series

This section includes additional model-data time series analogous to those in Fig. 4. That figure compares averaged concentrations modeled by WRF-STILT against monthly-averaged observations at four different observation sites. The sites displayed in that figure are located



Figure S3: This figure displays the fraction of soil water that is unfrozen for the HBL in different seasons and at different soil depths. Estimates are taken from NARR (Mesinger et al., 2006).

near large wetlands and in regions where the synthetic data experiments had a high success rate (Fig. 3). The sites displayed in Fig. S4 in this section are located further from wetlands and in regions that had a low success rate in the BIC experiments. At many of the sites in Fig. S4, the modeled wetland signal is difficult to distinguish. These sites contrast with those in Fig. 4 which "see" a relatively large CH₄ increment from wetlands.

¹⁵⁰ S7 Validation of the WRF-STILT model

This section describes work that validates the atmospheric transport estimated by WRF-STILT. The supplements to Miller et al. (2013) and Miller et al. (2014b) provide detailed validation of the atmospheric transport and boundary condition estimate; refer to those papers for additional information. Those papers use the same WRF-STILT simulations and boundary condition estimate as in the present paper. This section of the Supplement discusses a number of key points or highlights.

A number of figures in Miller et al. (2013) and Miller et al. (2014b) illustrate the ability of the WRF-STILT model to reproduce daily and seasonal patterns in the observations at different sites across the US and Canada. Those studies used an geostatistical inverse model to estimate

- ¹⁶⁰ CH₄ fluxes for the US and Canada, respectively. Figures S6 and S7 in Miller et al. (2013) compare modeled concentrations using this estimate against observed concentrations. The figures also display the estimated boundary condition and modeled concentrations with the EDGAR inventory for comparison. Modeled concentrations using the flux estimate in that paper can reproduce day-to-day variations in CH₄ concentrations at tall tower sites in Wisconsin, California, and
- ¹⁶⁵ Texas (Fig. S6 in Miller et al. (2013)), among other tall tower locations. Figure 4 in Miller et al. (2014b) further compares modeled concentrations against observed concentrations at sites in Canada. WRF-STILT is able to reproduce seasonal variability in CH₄ concentrations at tower sites across Canada



Figure S4: This figure is analogous to Fig. 4 and displays monthly-averaged model-data time series for additional atmospheric observation sites.

using the flux estimate from that study.

Miller et al. (2013) validate WRF-STILT's ability to reproduce the vertical structure of the atmosphere, Figure 4 in Miller et al. (2013) shows modeled and observed concentrations at 170 regular NOAA aircraft sites, averaged across the 2007–2008 study period. The first panel of that figure displays observed and modeled concentrations averaged across all aircraft sites and the remaining panels display individual aircraft sites in Oklahoma, New Jersey, and Iowa. At all of these sites, WRF-STILT is able to reproduce CH_4 enhancements near the surface and can reproduce vertical patterns in the aircraft observations. 175

The next section provides additional discussion of uncertainties and errors in the WRF-STILT model.

S8 Uncertainty in the model-data framework

A number of modeling and measurement uncertainties influence the results presented in this paper. These uncertainties are discussed in detail by Miller et al. (2013), Miller et al. (2014b), 180 and Miller et al. (2014a). This section provides a summary of those discussions.

The model selection framework in this study accounts for modeling or measurement errors in the **R** covariance matrix (Eq. S3). This covariance matrix is typically included in a Bayesian synthesis or geostatistical inverse model (e.g., Michalak et al., 2004). The errors described by R are collectively referred to as model-data mismatch – any errors in the model-data framework

185 that are unrelated to an imperfect flux estimate. This mismatch includes errors in the modeled winds, errors in the CH_4 boundary condition, and any errors due to the finite spatial or temporal resolution of the model, among other possible error sources. This section of the supplement first discusses the overall magnitude of these model-data mismatch errors and then discusses individual components of the model-data mismatch, including potential errors in the estimated

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winds and in the boundary condition.

Both Miller et al. (2013) and Miller et al. (2014b) estimate the magnitude of model-data mismatch errors for observation sites in the US and Canada, respectively. These studies used a procedure known as Restricted Maximum Likelihood (RML) to estimate the parameters that

define both the **R** and **Q** covariance matrices (e.g., Corbeil and Searle, 1976; Kitanidis, 1995; 195 Michalak et al., 2004; Gourdji et al., 2012). The estimated mismatch errors range in magnitude from 12-13 ppb (standard deviations) at Canadian tower sites to 20-30 ppb at tower sites near oil and gas operations in the southern US (refer to Fig. S2 in Miller et al. (2013) and Fig. S6 in Miller et al. (2014b)). This magnitude (12-30 ppb) is equivalent to 25-70% of the average CH₄ signal from North American emissions as seen at the various observation sites. 200

These model-data mismatch errors encompass numerous sources of error, but these errors are likely dominated by uncertainties in atmospheric transport. Nehrkorn et al. (2010) generated WRF meteorology for use in STILT and compared the estimated winds against US and Canadian radiosondes. They computed a root mean squared error (RMSE) of $2.5-4 \text{ m s}^{-1}$ in

the horizontal winds and found no change in error statistics at the top of the boundary layer. 205 Hegarty et al. (2013) further coupled the STILT model with several weather models and found that simulations with WRF produced lower error statistics relative to other weather models. Several existing studies have shown consistent results between WRF-STILT and other atmo-

spheric models; this consistency may indicate a lack of large-scale bias in atmospheric transport estimated by WRF-STILT. For example, constraints on summertime US carbon monoxide emissions estimated with STILT and the GEOS-Chem model match to within 10% (Miller et al.,

2008; Hudman et al., 2008). CH₄ budgets estimated for the HBL in Canada using WRF-STILT and GEOS-Chem are similar to within 10% (Pickett-Heaps et al., 2011; Miller et al., 2014b; Wecht et al., 2014). Furthermore, CH_4 budgets estimated for the US with WRF-STILT and GEOS-Chem match to within $\sim 10\%$ (Miller et al., 2013; Turner et al., 2015). 215

The CH₄ boundary condition is or background concentrations are another, potentially large source of uncertainty in the CH₄ modeling framework. To create this boundary condition, we interpolate atmospheric CH₄ observations near or over the Pacific and Arctic Oceans to create a boundary "curtain." This curtain estimates CH₄ concentrations over the Pacific and Arctic; it varies by latitude, altitude, and time (see Fig. S4 in Miller et al. (2014b)). We then sample concentrations along this curtain depending upon the ending latitude, altitude, and time of each WRF-STILT trajectory. These sampled concentrations become the boundary condition – an estimate of the CH₄ concentration in air before that air reaches North America. Miller et al. (2013) and Miller et al. (2014b) describe this setup in greater detail along with the

- associated uncertainties. For example, Miller et al. (2013) compared the boundary condition estimate against aircraft data collected above 3000m over the United States. They found an average difference of 2.7 ppb between the aircraft observations and boundary condition estimate. Miller et al. (2013) then adjusted the boundary condition based upon this aircraft data. They subsequently estimated a total US CH_4 budget using boundary conditions with and without
- the aircraft adjustment. The total CH_4 budget using the aircraft-corrected boundary condition was approximately 5% higher than the unadjusted boundary condition estimate. This result indicates the possible effects of boundary condition uncertainties on a national-scale CH_4 budget estimate.

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