



Supplement of

Estimating 2010–2015 anthropogenic and natural methane emissions in Canada using ECCC surface and GOSAT satellite observations

Sabour Baray et al.

Correspondence to: Sabour Baray (sabour@yorku.ca)

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32 S1 Supplement

33 S1.1 Monthly GOSAT Data in the Canadian Domain

34 Figure S1 shows the GOSAT data available per month using 2013 as an example year, this corresponds to the data coverage shown in Fig. 1 of the main text but highlights the variability in satellite observational coverage over a single year. GOSAT 35 36 data shown passes all quality assurance flags and includes our domain filter to land data that is within 50°W to 150°W longitude 37 and 45°N to 60°N latitude. The minimum in December observations (n=112) and neighbouring months is due to less solar 38 radiation in the winter resulting in less retrievals. Fewer observations cause the inversion to favour the prior state of emissions. 39 There are less methane emissions from Canadian wetlands in the coldest months of the winter, and the comparison between the prior, the posterior using GOSAT data, and the posterior using ECCC data shows very small differences in emissions 40 41 estimates for these coldest months.





44 Figure S1: GOSAT observations per month in the year 2013 corresponding to Fig. 1 in the main text (n=7656 observations

45 for the entire year). Observations are filtered to land data that is within 50° W to 150° W longitude and 45° N to 60° N latitude.

46 S1.2 Sensitivity of Seasonal Emissions to Climatological Data

We select four climatological stations shown in Table S1 to sample temperature and precipitation data from 2010–2015 in the four provinces where wetland emissions are concentrated (Alberta, Saskatchewan, Manitoba, and Ontario). These stations are not exhaustive and are chosen for their proximity to the stations shown in Table 1. Station measurements are quality-controlled from the National Climate Data Archive from Environment and Climate Change Canada (Hutchinson et al., 2009).

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52 **Table S1:** Climatological sites used for air temperature and total precipitation measurements for the seasonality comparison.

Site Name, Province	Latitude	Longitude
Lac La Biche Climate, Alberta	54.8° N	112.0° W
La Ronge, Saskatchewan	55.1° N	105.3° W
Churchill Climate, Manitoba	58.7° N	94.1° W
Moosonee, Ontario	51.3° N	80.6° W

53

54 Figure S2 shows the mean 2010–2015 seasonal pattern of natural methane emissions constrained by ECCC and GOSAT data 55 corresponding to Fig. 6 in the main text. These emissions are compared to monthly mean air temperature and precipitation 56 averaged over the four climatological stations in Table S1. We consider air temperature a reasonable proxy for the surface skin 57 temperature that is used in WetCHARTS. Surface skin temperature is itself a proxy for soil temperatures deeper beneath the 58 surface where methane is produced (Miller et al., 2016). Hence both metrics may be lagging indicators for the peak of methane 59 emissions. Both air temperature and precipitation show peaks in July which correspond well with the maxima of methane 60 emissions in the prior from WetCHARTS. Methane emissions in the prior begin to accelerate from March to April, however 61 for both months air temperature is below freezing. It is not likely that soil temperatures and subsurface soil temperatures would be above freezing in these months. Air temperature crosses from below 0° to above freezing one month later from April to 62 May, which corresponds to where the posterior ECCC and GOSAT emissions begin to accelerate. Total precipitation shows 63 64 the highest acceleration one month later from May to June. As the peak in July is passed, late-summer and autumn air 65 temperatures are higher than the months opposite of the peak (August is warmer than June, September is warmer than May, October is warmer than April). This pattern is corroborated by the precipitation measurements. Air temperatures go below 66 67 freezing from October to November. As shown by Zona et al. (2016), "zero-curtain" emissions may continue even when the soil is at freezing temperatures. This mechanism may be more likely to occur in the months after the peak if subsurface soils 68 69 are slower to thaw in the spring and slower to freeze in the autumn. These simple climatological measurements and the 70 described mechanisms suggested in other studies corroborate our posterior results of lower spring methane emissions and 71 lower peak methane emissions in the summer. Our results suggest process models may benefit from better parameterization of

- 72 possible lagging effects from air temperature and precipitation for Boreal Canada methane emissions.
- 73



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Figure S2: Mean seasonal pattern of 2010-2015 methane emissions from the prior (gray), posterior constrained with ECCC data (blue), posterior constrained with GOSAT data (green). This is compared to the seasonal pattern of monthly mean air temperature (orange, right axis) and precipitation (pink, left axis) from station measurements listed in Table S1. Both air temperature and precipitation show an asymmetry about the July peak, with higher temperature and precipitation in the fall months than the spring.

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81 S1.3 Evaluation of Bias in the Global Model

In this section we test the GEOS-Chem representation of background methane for both surface ECCC data and column GOSAT data using global and/or boundary condition observations. We show the model representation of methane can be improved using surface and column bias corrections which are presented as the base case in the main text. We test the sensitivity of the posterior emissions to the use of these bias corrections and show the inversions produce consistent results.

86 S1.3.1 Evaluation of the ECCC Surface Data Background and Bias Corrections

The left panel of Figure S3 shows the comparison of monthly mean GEOS-Chem surface methane concentrations and methane measured at the ECCC station ESP from 2009 to 2015. ESP is located at the west coast of Vancouver Island (Fig. 1); this site

is used as an evaluation of background methane and tests the bias in the global model as it is the least sensitive to Canadian 89 90 emissions due to westerly prevailing winds. The model reliably reproduces surface observations at this station and the growth rate in background methane due to the source-sink imbalance of +13 Tg a⁻¹ in the model global budget (Maasakkers et al., 91 2019) with a small mean model-observation bias of +5.3 ppb. The right panel of Figure S3 shows the comparison of modelled 92 93 methane to NOAA aircraft profiles at the same site. Aircraft profiles occur approximately once a month continuously over the 94 study period. The data is not averaged here and is directly compared to GEOS-Chem simulated grid boxes at the pressure level 95 of the measurement. The reduced mean axis (RMA) regression shows a slope of 0.86 and a coefficient of regression $r^2 = 0.67$ 96 which shows a reasonable model representation of the measurements. These statistics are consistent with previous inversions 97 using GEOS-Chem that showed relatively unbiased conditions against NOAA surface stations globally (Turner et al., 2015; 98 Maasakkers et al., 2019). A high resolution inversion over North America over the same 2010–2015 time-period using the 99 same prior have shown adjustments to US emissions near the Canadian border are relatively minimal (Maasakkers et al., 2021), 100 so we treat US emissions as constant in the inversion. The acceptable reproducibility of background methane at this site allows 101 us to attribute much larger differences observed at other sites, up to a maximum of ~ 1000 ppb in the summer (Figure 4), to 102 Canadian emissions which are optimized using Canadian observations while holding other global emissions constant.

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- 106

Figure S3: Time-series comparison (left) from 2009–2015 of surface GEOS-Chem simulated methane (red) and measured in situ methane (black) at site ESP off the west coast of British Columbia. Comparison to NOAA aircraft profiles (right) from 2009–2015 at the same site using a reduced major axis (RMA) regression along with the 1:1 line (black).

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While the mean model bias of +5.3 ppb in Figure S3 shows a net over-estimation in the model, the later years 2014 and 2015 show a model underestimation primarily due to underestimated tropical emissions (Maasakkers et al., 2019). This positive-to-

113 negative difference in the model background can project errors onto the trend of ECCC-constrained emissions. This is

addressed by removing the annual-mean background bias at the Canadian boundary conditions from the observation vector.
We use the westmost boundary condition site ESP and a second northernmost background site at Alert, Nunavut (ALT) to diagnose errors in the methane background and show the annual mean model-observation differences in Table S2. The average of these two sites is used to adjust the model for the base-case ECCC inversion in the main text. In Section 1.3.2 of the Supplement, we test the sensitivity of the posterior emissions to the use of these various background corrections and show the consistent results, with the background-adjusted inversion showing slightly more agreement with the GOSAT inversion.

- 120
- 121 **Table S2:** Mean annual model-measurement differences at background sites ESP and ALT.

		Mean Model-Measurement Difference (ppb)					
Year	ESP ^a	ALT ^b	Average ^c				
2010	+5.0	+8.8	+6.9				
2011	+5.8	+8.5	+7.2				
2012	+3.6	+5.9	+4.8				
2013	+2.6	+10.5	+6.6				
2014	+2.1	+11.3	+6.7				
2015	-6.9	-4.7	-5.8				

^aSite ESP is located at 49.38°N, 126.54°W, and is the westernmost boundary condition for Canada.

¹²³ ^bSite ALT is located at 82.45°N, 62.51°W, and is the northernmost boundary condition for Canada.

124 °The average is used in the base-case ECCC inversions shown in the main text. The three alternatives: adjustments using ESP,

125 ALT and no background adjustments are shown as sensitivity tests in the Supplement.

126 S1.3.2 Sensitivity Tests of ECCC-Constrained Emissions

127 Figure S4 shows the sensitivity tests comparing the ECCC inversions with an unadjusted model to the two background-adjusted 128 ECCC inversions using either the mean yearly bias from ESP or ALT. The three inversions are consistent with each other 129 within their error intervals, but the adjusted ECCC inversions show improved agreement with the GOSAT results. For anthropogenic sources, the mean yearly emissions are 6.0 \pm 0.4 Tg a⁻¹ in the unadjusted ECCC inversion, 6.1 \pm 0.4 Tg a⁻¹ with 130 131 the ESP-adjusted ECCC inversion, and 6.0 ± 0.4 Tg a⁻¹ with the ALT-adjusted inversion. For natural sources, the mean yearly 132 emissions are 10.5 ± 1.9 Tg a⁻¹ in the unadjusted ECCC inversion, 12.0 ± 1.4 Tg a⁻¹ in the ESP-adjusted ECCC inversion, and 133 11.0 ± 1.2 Tg a⁻¹ in the ALT-adjusted ECCC inversion. The background-adjusted inversions show higher natural emissions in 134 the years 2010–2014 compared to the unadjusted case, and lower natural emissions in 2015 due to the negative background 135 bias that is removed. The background-adjusted inversions show better agreement with the GOSAT mean yearly natural 136 emissions of 11.7 ± 1.2 Tg a⁻¹. In addition, the trend in natural emissions over this time period is reduced by 40-45% from 1.0 137 Tg a^{-1} in the unadjusted inversion to 0.55–0.60 Tg a^{-1} in the adjusted inversions. These results show that the background error 138 does not largely affect the average 2010–2015 results regarding the overall increase in anthropogenic emissions and decrease 139 in natural emissions. Correcting for the model background minimizes the projection of under-estimated tropical emissions onto

the Canadian fluxes in the later years, which improves the consistency with the GOSAT inversion and significantly reducesthe presence of a large trend that was not corroborated by GOSAT.

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Figure S4: Sensitivity analysis of inversion results depending on the use of model background correction for surface pixels. Referred to as the monthly inversion, this approach optimizes annual total Canadian anthropogenic emissions (top) and monthly total natural emissions (bottom) in an n = 78 state-vector element setup. The prior emissions (gray) are compared to the posterior results using GOSAT (green), and the posterior using ECCC data with an unadjusted background (blue), ECCC data using a background adjusted according to the yearly difference at ESP (teal) and ALT (purple) from Table S2.

- 149
- 150 To address the possibility of US emissions influencing the posterior results near the Canadian border, we show a sensitivity
- 151 test where the two stations most influenced by cross-border transport, Egbert (EGB) and Sable Island (SBL) are removed from
- 152 the ECCC inversion. Figure S5 shows posterior-ECCC emissions where EGB and SBL (at latitudes of 44.2° N and 43.9° N,
- 153 respectively) are removed (note in this case, the background is left un-adjusted to avoid overlap in the issues). The mean of
- anthropogenic emissions in the inversion without these stations is 6.4 ± 0.6 Tg a⁻¹, and the mean of natural emissions is 10.9
- 155 \pm 1.5 Tg a⁻¹. These results are similar to the posterior from the unadjusted ECCC inversion (6.0 \pm 0.4 Tg a⁻¹ anthropogenic,
- 156 10.5 ± 1.9 Tg a⁻¹ natural) and the GOSAT inversion (6.5 ± 0.7 Tg a⁻¹ anthropogenic, 11.7 ± 1.2 Tg a⁻¹ natural). This sensitivity
- 157 test shows that the US signal does not substantially affect the results from the optimization of large biases observed by
- 158 Canadian observations due to Canadian emissions.



160

Figure S5: Sensitivity analysis of inversion results depending on the inclusion of sites EGB and SBL which are sensitive to cross-border transport from the United States. Similar to Fig. S4, the monthly inversion optimizes annual total Canadian anthropogenic emissions (top) and monthly total natural emissions (bottom) in an n = 78 state-vector element setup. The prior emissions (gray) are compared to the posterior results using GOSAT (green), and the posterior using ECCC data including all sites (blue) and ECCC data excluding EGB and SBL (yellow).

166 S1.3.3 Evaluation of Global GOSAT Data and Bias Corrections

167 The GEOS-Chem simulation of column averaged methane shows three global biases previously discussed in the literature: (1)

- a latitude-dependent bias, (2) a seasonal bias and (3) a background change for 2014 and 2015 due to differences in the global
- 169 source-sink imbalance in these two years (Turner et al., 2015; Saad et al., 2018; Maasakkers et al., 2019; Stanevich et al.,

170 2020). We apply these corrections to the simulated column of methane on a global basis to produce an unbiased background

171 for our target Canadian domain (45° N to 60°N, 50° W to 150° W). The latitude-dependent bias (1) is likely due to excessive 172 polar stratospheric transport (Stanevich et al., 2020). We correct for this bias by fitting the model-GOSAT difference for global 173 $2^{\circ} \times 2.5^{\circ}$ grid cells according to a second-order polynomial as shown in Figure S6:

174

$$175 \quad \xi = (2.2\theta^2 - 34\theta) \times 10^{-3} - 2.7 \tag{1s}$$

176

177 where ξ is the resulting bias correction in ppb and θ is latitude in degrees. The correction in this work for the latitude bins of 178 our target domain $(45^{\circ} \text{ N to } 60^{\circ} \text{ N})$ is between 0.3 to 2.9 ppb. This correction is lower than what has been shown previously 179 (Turner et al., 2015; Maasakkers et al., 2019) and we attribute this improvement to our use of a $2^{\circ}x2.5^{\circ}$ gridded simulation instead of a $4^{\circ}x4.5^{\circ}$ as recommended by Stanevich et al. (2020) to reduce transport errors. A seasonally oscillating bias (2) 180 181 remains after this correction. The seasonal bias has an amplitude of ± 4 ppb with repeating maxima in June and minima in 182 December. It is not clear whether this seasonal bias is due to emissions and/or transport errors. In our base case we remove the 183 seasonal bias on a monthly basis following Maasakkers et al. (2019) and show a sensitivity test without the correction for our 184 inversion of monthly natural emissions in Canada (Supplement 1.3.4). Inversion results using GOSAT data with and without 185 bias corrections in the model simulation of total column methane do not show major differences (Fig. S7). These scenarios all 186 show agreement with the posterior emissions adjustments determined using unadjusted ECCC in situ data – which is a useful 187 benchmark since modelled methane at the surface is not subject to any bias corrections. The background change (3) that appears 188 in the simulated methane column from 2014 onwards is corrected for in Maasakkers et al. (2019) by optimizing emissions, 189 emissions trends and trends in OH using a global inversion. In that work correction factors do not appear over Canada and the 190 United States that would significantly influence the global change in atmospheric methane, and the main adjustment in 2014 191 and 2015 were to tropical wetland emissions and OH. Here we treat this as a background change and apply a uniform correction 192 to the simulated column since emissions outside of Canada and changes in OH are treated as fixed in our Canada-focused 193 inversion. The background change (3) is 5 ppb in 2014 and 10 ppb in 2015. The right panel of Figure S6 shows the latitude 194 dependent bias correction and the left panel shows the resulting global time-series of GEOS-Chem total column methane from 195 2010–2015 after corrections are applied. The global GEOS-Chem – GOSAT differences in the methane column can be limited 196 globally to within 10 ppb without including the seasonal bias correction, and within 5 ppb with its inclusion. This shows a 197 steady background in methane for the entire time period from 2010–2015 so global emissions do not affect the optimization 198 of Canadian emissions. While biases within 10 ppb have been treated as acceptable for methane inversions (Buchwitz et al., 199 2015), we evaluate our GOSAT inversion results against inversions with independent ECCC in situ measurements that do not 200 require any bias corrections in the model to produce more robust emissions estimates.





204 Figure S6: Time series (left) from 2010–2015 of the difference between GEOS-Chem simulated total column methane and 205 GOSAT observations after applying bias corrections, showing a consistent global background for methane. Data used in the 206 inversion for Canada is from 45° N to 60° N (purple line) and shows acceptable differences within 5 ppb over the entire global latitude band. To produce the left figure, the latitude-dependent bias (right) is shown with the polynomial correction 207 208 that is applied (gray dash) that is within a magnitude of 0.3 to 2.9 ppb for the same latitude.

210 S1.3.4 Sensitivity Tests of GOSAT-Constrained Emissions

211 We test the sensitivity of the posterior GOSAT-constrained methane emissions in our analysis to the use of latitude-dependent 212 and seasonal bias corrections in the GEOS-Chem simulated total column of methane. The latitude-dependent bias correction 213 has a magnitude less than 3.5 ppb for our domain of interest (45 to 60°N). On a global basis the seasonal bias correction has 214 an amplitude of ± 4 ppb with a maximum in June and a minimum in December. Figure S7 shows the sensitivity of posterior monthly emissions to these bias corrections using 2013 as an example. We show four versions of the posterior methane 215 216 emissions using GOSAT data: GOSAT11 (green) is the base case which applies the latitude-dependent bias correction and the 217 seasonal bias correction, GOSAT10 (purple) applies the latitude-dependent bias correction and does not apply the seasonal 218 correction, GOSAT01 (orange) does not apply the latitude-dependent bias correction and applies the seasonal correction, and 219 GOSAT00 (light blue) uses neither bias correction. The range of emissions from all four examples is 9.7 - 10.7 Tg a⁻¹, which are all consistent with the unadjusted ECCC emissions of 10.0 Tg a⁻¹ and lower than the prior emissions of 14.3 Tg a⁻¹. Not 220 applying the latitude-dependent bias correction results in a decrease in the resulting emissions and maintains the same seasonal 221 222 pattern. Not applying the seasonal bias correction results in a change in the temporal distribution of emissions that better 223 matches the August peak in the posterior with ECCC data. Emissions are lower than the base case in the spring and higher 224 than the base case in autumn. This change enhances the autumn-shift in emissions that has been described in Section 3.2 of the

main text. While this may be more consistent with our interpretations, it is not clear whether the difference is due to emissions 225 226 or transport biases. Stanevich et al. (2020) showed that the latitude dependent bias is most likely due to excessive polar 227 stratospheric transport at high latitudes. If the seasonal bias is indeed due to mischaracterized natural emissions, it is not clear 228 why the bias would be equally large in December (-4 ppb) as June (+4 ppb) on a global basis. The magnitude of natural 229 emissions in December is much lower than June and emissions mischaracterization would not itself produce an equally large 230 bias as the largely overestimated summertime emissions. Our analysis with ECCC data shows most of the adjustments to 231 wetlands are in the peak of summer with some extension into the autumn. These results show that the bias corrections produce 232 minor differences in the magnitude and seasonal pattern of emissions.



233

Figure S7: Sensitivity of 2013 posterior GOSAT constrained methane emissions to bias corrections used in the GEOS-Chem simulated total column of methane. For comparison, the prior in 2013 (gray) and the posterior in 2013 constrained by ECCC data (unadjusted, blue) are shown. The digits in the GOSAT label represent the binary use of bias corrections (1 = applied, 0 = not applied). The first digit corresponds to the use of the latitude bias correction, the second digit corresponds to the use of the monthly bias correction, hence GOSAT11 is the base case that applies both bias corrections and GOSAT00 is the case with no bias corrections applied.

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241 S1.3.5 Evaluation of the Prior and Posterior Fluxes Using Global Observations Outside of the Canadian Domain

The inverse model design in this study uses a simplified approach, where Canadian emissions are optimized using only observations in Canada. The results from this approach may be sensitive to errors in the global model projected onto the Canadian domain if errors in the global model are sufficiently large relative to the local biases in Canada (Figure 4 in the main

245	text) and the observational error used in the inversion procedure (16 ppb for GOSAT, 65 ppb for ECCC). Figure S8 shows an
246	independent evaluation of the prior global model and the posterior in this study to 2010-2015 background observations from
247	the NOAA cooperative flask sampling network (https://gml.noaa.gov/ccgg/flask.html) outside of the Canadian domain. We
248	use a simple version of the posterior where Canadian anthropogenic emissions are scaled up by 37% to 6.0 Tg a ⁻¹ and natural
249	emissions are scaled down by 24% to 11.2 Tg a ⁻¹ . This captures the central results of the monthly, sectoral, and provincial
250	inversions in the main text and avoids a large number of model comparisons. The analysis shows that the prior model
251	reasonably reproduces the methane background, and the posterior from adjusted Canadian emissions does not degrade this
252	result. In the reduced-major axis regression, the prior r^2 coefficients are in the range of 0.77–0.92 and the prior slopes are in
253	the range of $0.94-0.97$ across the three surface, ship, and aircraft datasets. In the posterior, the r ² is in the range of $0.76-0.91$
254	and the slope is in the range of 0.93–0.96. The posterior reflects a decrease of 2.0 Tg a ⁻¹ in the global budget due to a net
255	decrease in Canadian emissions, which is shown in the improvements to the mean bias comparisons. This decrease in emissions
256	slightly improves the global model agreement with independent data in the years 2010-2013 (since the model overestimates
257	emissions) and slightly degrades the agreement in 2014-2015 (since the model underestimates tropical emissions), which is
258	understandable considering only Canadian emissions are adjusted and the global model is not optimized. A net decrease in
259	Canadian emissions is consistent with previous global inversion studies using GEOS-Chem (Turner et al., 2015; Maasakkers
260	et al., 2019). The results from the Canada-focused inversion with subnational details in this study show that the net-decrease
261	in Canadian natural emissions masks an increase in anthropogenic emissions in Western Canada which should be considered
262	in global inverse studies.
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Figure S8: Model comparison to independent NOAA observations globally from 2010–2015. The top panel shows data used in the global model comparison. Red diamonds indicate NOAA surface flasks, purple circles indicate NOAA ship data, and blue lines indicate HIPPO III, IV and V aircraft data. Comparison of the prior and posterior emissions in GEOS-Chem is shown using a reduced-major axis regression against NOAA Surface flasks (bottom-left), HIPPO III, IV and V aircraft data (bottommiddle), and NOAA Ship data (bottom-right).

278 S1.4 Diagnostics of Sectoral and Provincial Inversions

In this analysis we first evaluate the correlations and/or independence of the state vector elements from the posterior error covariance matrix $\hat{\mathbf{S}}$ as follows (Heald et al., 2004):

282
$$r_{ij} = \frac{\hat{s}_{ij}}{\sqrt{\hat{s}_{ii}}\sqrt{\hat{s}_{jj}}}$$
(2s)

The error-normalized posterior correlation matrix r provides information on the independence of the state vector elements. This is corroborated by the averaging kernel matrix \mathbf{A} which shows which state vector elements contain independent pieces of information, with the trace of \mathbf{A} providing the total degrees of freedom for signal for the inversion. To further evaluate the signal-to-noise ratio of the observation-constrained state vector elements and their independence from each other we use an eigenanalysis. The Jacobian matrix \mathbf{K} is normalized about the observational and prior error covariance matrices as follows (Rodgers, 2000):

290

$$\mathbf{\check{K}} = \mathbf{S}_{o}^{-1/2} \mathbf{K} \mathbf{S}_{a}^{1/2}$$
(3s)

292

The singular value decomposition of $\mathbf{\check{K}}$ gives its rank which is the number of singular values greater than one. The singular values also correspond to the signal-to-noise ratio of state vector elements and hence quantify the strength of the observational constraints on individual emissions categories.

296

297 Figure S9 shows this series of diagnostics for the sectoral (5 state vector element) inversion and Figure S10 shows the same 298 analysis for the provincial (16 state vector element) inversion. Figure S9 (top left) shows the error-normalized correlation 299 matrix for the sectoral inversion. The most important result is that the primary source of natural emissions, wetlands, is not 300 correlated with the primary source of anthropogenic emissions, energy. Within the anthropogenic category however, we see 301 that energy is strongly correlated with agriculture, showing that these two elements cannot be distinguished by the observation 302 system. For natural emissions, other natural sources are weakly correlated with wetlands and are not completely independent. 303 Emissions from waste are shown to be slightly more independent and can be distinguished from the other sources. The 304 averaging kernel matrix corroborates this result, and shows the three independent pieces of information are energy, wetlands 305 and waste, with partial information content from other natural sources and a lack of information on agriculture. The singular 306 values show strong constraints on wetlands with a signal-to-noise ratio of 37.3, and strong constraints on energy with a signal-307 to-noise ratio of 5.2. Waste sources are 2.2, other natural are 1.2 and agriculture is below the noise at 0.4. These diagnostics 308 demonstrate that a joint ECCC in situ and GOSAT satellite inversion system can successfully provide constraints on and 309 distinguish the three major categories of methane emissions in Canada: wetlands, energy and waste. Emissions from agricutlure 310 cannot be distinghised in this system and should be aggregated with energy, this is likely because of the strong spatial overlap 311 between these emissions in Western Canada and the lower signal from lower magnitude agriculture emissions. Emissions from other natural sources (biomass burning, seeps, and termites) also are at the noise and should be aggregated with wetlands. This 312 is because minor natural sources are much lower in magnitude (0.8 Tg a^{-1} out of 14.8 Tg a^{-1}) and also show spatial overlap 313 314 with wetlands.

316 Figure S10 shows the diagnostics on the provincial (16 state vector element) inversion. This choice of state vector elements 317 challenges the observing system and results in a largely underdetermined solution. These diagnostics allow us to identify where 318 the limitations of the ECCC + GOSAT observing system are. The posterior error correlation matrix r shows the provincial 319 emissions are somewhat correlated a) between anthropogenic/natural emissions of the same province and b) with neighboring 320 provinces in the same category of emissions. For example, AB anthropogenic emissions show a small inverse correlation with 321 AB natural emissions. AB anthropogenic emissions also show a small correlation with the anthropogenic emissions of nearby 322 provinces BC and SK. For the natural emissions, natural emissions within a province in most cases extend correlations into 323 the provinces to the east and west. These correlations are not as large as the case of Energy and Agriculture emissions in Fig. 324 S9, and show a more moderate influence of nearby provinces on the optimized emissions. The primary limitation of the 325 provincial inversion is the inability to distinguish provinces with a very small magnitude of emissions. This is shown in the 326 averaging kernel matrix, which has a degrees of freedom for signal of 7.9 out of 16 elements. The 6 regions that are best 327 constrained are AB anthropogenic, ON anthropogenic, AB natural, SK natural, MB natural, and ON natural, with partial 328 constraints on BC anthropogenic, SK anthropogenic, QC anthropogenic, BC natural, QC natural and NOR natural. The singular 329 vectors corroborate this result and show that there are 8 regions that are above the noise and 8 that are at or below the noise. 330 The best constraints on anthropogenic emissions are in Alberta, with a signal to noise ratio as good as 15.1 (solid blue line), 331 followed by Ontario (2.5-2.8).

332

333 These diagnostics show that the ECCC+GOSAT observing system for Canada is limited in its ability to characterize 334 agricultural emissions, and somewhat limited in its ability to characterize non-wetlands natural emissions. Hence we present 335 Energy+Agriculture and Wetlands+Other Natural together for our conclusions. More precise and more dense measurements 336 at a finer scale would better disaggregate these sources, although the use of the precise in situ data is primarly limited by the 337 model error (Section 2.3 of the main text). In the provincial inversion, the observing system provides good constraints on 338 anthropogenic emissions from AB and ON and is capable of distinguishing these emissions from natural sources in the same 339 province. However, anthropogenic sources from other provinces with much lower emissions cannot be distinguished. Natural 340 emissions can be characterized from the provinces that are most responsible for wetland emissions (AB, SK, MB, ON), 341 however the observing system struggles in Atlantic and Northern Canada where the surface and satellite observations we use 342 are limited. The emissions adjustments to state vector elements beneath the noise are due to aliasing with other sources and 343 compensation effects due to interprovincial transport. We limit our conclusions to simple interpretations, we use the limited 344 provincial inversion for spatial attribution to show higher posterior anthropogenic emissions are primarily from the total in 345 Western Canada (BC+AB+SK+MB), and not emissions in Central Canada (ON+QC).



Figure S9: Diagnostics of the sectoral inversion used to evaluate the independence and information content of the 5 state vector elements. The error-normalized posterior correlation matrix (top left) shows the correlations between elements. The averaging kernel matrix (top right) shows where the independent pieces of information are (DOFS = 3.3). The singular vector decomposition of the pre-whitened jacobian (bottom) quantifies the signal-to-noise ratio of the significant elements – these are the singular values listed above one (4 in total). The singular vector below noise (agriculture) is shown as a dashed line.



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Figure S10: Similar to Fig. S9 for the 16 state vector provincial inversion. The DOFS from the averaging kernel matrix are 7.9, which are consistent with the number of singular values greater than unity in the pre-whitened jacobian matrix (8 in total). In the bottom panel, the singular vectors below the noise (corresponding to singular values less than one) are shown as light-dashed lines, these show which emissions are not constrained by observations.

A possible solution to improving the resolution of the solution is to combine all six years of data to constrain finer scale emissions for the sectoral and provincial inversions. In the presented approach inversions were completed on a yearly basis for six years to produce an average result for 2010–2015. We used the year to year variance as a representation of noise in

364 the system and real yearly variability in the state (due to emissions and/or transport). In principle using more years of data

365 provides a better signal to noise ratio. However, due to the way our state vector elements are defined in the sectoral and 366 provincial inversions, the inverse approach is sensitive to aggregation error and overfitting the fewer number of well-defined 367 state vector elements. Overfitting can be diagnosed using the reduced chi-squared metric:

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369
$$\chi_{\nu}^2 = \frac{\chi^2}{\nu} \cong \frac{\Sigma \frac{(y - K_X)^2}{So}}{m}$$
 (4s)

370

Where χ_{ν}^2 is the chi-square per degree of freedom ν . Here, the χ^2 is equal to the ratio of the square of the innovation, **S**₀ is the diagonal element of the observational error covariance matrix corresponding to the same observation, *m* is the number of rows of the observation vector and *n* is the number of state vector elements. A value of χ_{ν}^2 less than one indicates overfitting. We calculate a value of 0.65 for the total vector containing ECCC and GOSAT data which shows evidence of overfitting. Hence using a larger amount of data for the same number of state vector elements would exasperate the issue.

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377 We further test the improvement from combining 6 years of data against independent measurements. To evaluate the 378 differences between using a repeated 1-year approach and a 6-year approach we use independent observations from NOAA 379 ETL aircraft measurements and ECCC CHA in situ surface measurements. Table S3 lists the metrics of agreement that were 380 in Figure 8 and compares them to the results using all 6 years of data simultaneously, using inversions with no model background corrections for the ECCC observation vector. For the sectoral inversion, using 6 years of data provides a small 381 382 improvement in the slope (0.96 vs. 0.91), no improvement in the R^2 (0.20) and degrades the mean bias (+4.3 ppb vs. +0.4 383 ppb) when comparing to NOAA ETL. Similarly with ECCC CHA data, using 6 years of data for the sectoral inversion 384 provides an improvement in the slope (1.01 vs. 0.98), a slightly worse R^2 (0.43 vs. 0.44) and largely degrades the mean bias 385 comparison (+10.6 ppb vs. +5.9 ppb). For the provincial inversion evaluation at NOAA ETL, using 6 years of data slightly degrades the slope (0.83 vs. 0.86), gives an improvement in the R^2 (0.27 vs. 0.22), and degrades the mean bias (+3.2 ppb vs. 386 387 +0.5 ppb). The same comparison at ECCC CHA degrades agreement in the slope (0.87 vs. 0.91), improves the R^2 (0.51 vs. 388 (0.47), and improves the mean bias (+4.1 ppb vs. +4.9 ppb). These results show that using 6 years of data for the subnational 389 inversions does not improve agreement against independent data and in many cases degrades the mean bias. The inversion 390 converges on a solution within our defined prior error matrix S_0 with only one year of data. These tests show that using one 391 year of data at a time and calculating the average and variance of the repeated results is reasonable considering the limits of 392 the observation system towards resolve low magnitude emissions.

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		NOAA Aircraft Observations ETL			ECCC Surface Observations CHA		
		Slope	\mathbb{R}^2	Mean Bias (ppb)	Slope	\mathbb{R}^2	Mean Bias (ppb)
	Prior	1.15	0.14	+6.8	1.17	0.36	+16.4
Sactoral	Posterior (1 yr)	0.91	0.20	+0.4	0.98	0.44	+5.9
Sectoral	Posterior (6 yr)	0.96	0.20	+4.3	1.01	0.43	+10.6
Drovinsial	Posterior (1 yr)	0.86	0.22	+0.5	0.91	0.47	+4.9
FIOVINCIAI	Posterior (6 yr)	0.83	0.27	+3.2	0.87	0.51	+4.1

Table S3: Sensitivity test against independent observations

We show a comparison of emissions estimates and methods to derive errors for the sectoral inversion in Table S4 and for the provincial inversion in Table S5. The tables compare two error estimates to three sensitivity tests. They show the error estimates from the diagonal elements of the posterior error covariance matrix $\hat{\mathbf{S}}$ and compares to the 1 σ variance in the repeated yearly inversions. In both the sectoral and the provincial inversions, the error estimates from the diagonal elements of $\hat{\mathbf{S}}$ often show a more optimistic estimate of the uncertainties. This is likely due to spatial and temporal correlations in the daily-mean ECCC in situ observations and correlations in the GOSAT data that are difficult to quantify in the absence of a full OSSE study. We compare the 1σ variance from repeated yearly inversions from 2010–2015 to the relative change in posterior emissions from using only ECCC data, only GOSAT data, and using 6 years of ECCC+GOSAT data simultaneously. The 1_o yearly variance captures these differences except for state vector elements that were shown to be below the noise and highly correlated with other emissions in Figure S9 and S10. The lack of improvement against the comparison to independent data in Table S3 suggests that this may be suggestive of overfitting. We consider the agreement between the independent use of ECCC and GOSAT data to be a reliable sensitivity test to check the robustness of our results.

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425 Table S4: Sensitivity analysis of the Sectoral (5 state vector) inversion. The error estimates from the posterior error

	Prior	Posterior	Posterior Ŝ	1σ Yearly Variance	ECCC-only	GOS-only	6-year
	(Tg a ⁻¹)	(Tg a ⁻¹)	Relative Error (%)	Relative Error (%)	(% change)	(% change)	(% change)
Energy	2.4	3.6	±11	±25	+9	-6	-24
Agriculture	1.0	1.5	± 28	±25	-1	-19	+64
Waste	0.9	0.8	±25	±25	-8	+50	-29
Wetlands	14.0	9.6	<u>+</u> 4	±11	+3	+3	+2
Other Natural	0.8	1.7	±20	±55	-31	-9	+69

426 covariance matrix are compared to the yearly variance and the change in emissions using alternative observation vectors.

429 Table S5: Sensitivity analysis of the Provincial (16 state vector) inversion. As per S4 error estimates from the post	sterior error
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430	covariance matrix are comp	bared to the yearly van	riance and the o	change in emiss	sions using alternative observation	tion vectors.

	Prior	Posterior	Posterior Ŝ	1σ Yearly Variance	ECCC-only	GOS-only	6-year
	(Tg a ⁻¹)	(Tg a ⁻¹)	Relative Error (%)	Relative Error (%)	(% change)	(% change)	(% change)
BCA	0.5	0.8	±24	±41	-20	-11	+115
ABA	2.3	3.3	± 5	±16	-6	+2	-2
SKA	0.3	0.3	<u>+</u> 44	±37	+18	-1	+6
MBA	0.2	0.2	± 50	±25	+2	+6	+22
ONA	0.5	0.5	±17	±14	-4	+11	+2
QCA	0.4	0.3	±51	±40	-4	+19	+14
ATLA	0.0	0.0	±51	<u>±</u> 4	+1	+3	-8
NORA	0.0	0.0	± 50	±1	0	0	+1
BCN	0.4	0.6	±32	±50	+2	+5	-76
ABN	2.4	1.9	± 14	±34	+67	-29	-25
SKN	1.6	0.7	±28	±37	+7	-7	-4
MBN	1.5	1.4	±22	±32	+27	-6	-11
ONN	3.5	1.0	±32	±37	+12	-3	-13
QCN	1.6	1.2	±40	±41	+9	34	-51
ATLN	0.7	0.8	±39	±26	-29	+21	+48
NORN	0.7	1.9	±15	±35	-41	-2	+72

431 S1.5 Combined ECCC+GOSAT Monthly Inversion

- 432 Figure S11 shows the monthly inversion comparing the results from the ECCC-only inversion, the GOSAT-only inversion and
- 433 the combined ECCC+GOSAT inversion. The mean 2010–2015 anthropogenic emissions in the combined inversion is $6.0 \pm$
- 434 0.4 Tg a^{-1} . The mean 2010–2015 total natural emissions in the combined inversion is 12.0 ± 0.9 Tg a^{-1} . The combined inversion
- 435 agrees with the ECCC and GOSAT results and appears to follow the seasonality of natural emissions in the GOSAT-only
- 436 inversion more closely. Combining the two datasets does not appear to improve the results of the individual inversions, hence
- 437 the intercomparison between the ECCC-only and GOSAT-only inversions adds more value as a consistency test of the
- 438 posterior results.
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Figure S11: Sensitivity analysis of the results from the monthly inversion including a comparison to the combined ECCC+GOSAT inversion. Following Fig. 4 in the main text, the monthly inversion optimizes annual total Canadian anthropogenic emissions (top) and monthly total natural emissions (bottom) in an n = 78 state-vector element setup. The prior emissions (gray) are compared to the posterior results using GOSAT (green), and the posterior combining both ECCC and GOSAT data (purple).

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