

*Supplement of*

**Analysis of atmospheric CH<sub>4</sub> in Canadian Arctic and estimation of the regional CH<sub>4</sub> fluxes**

**Misa Ishizawa et al.**

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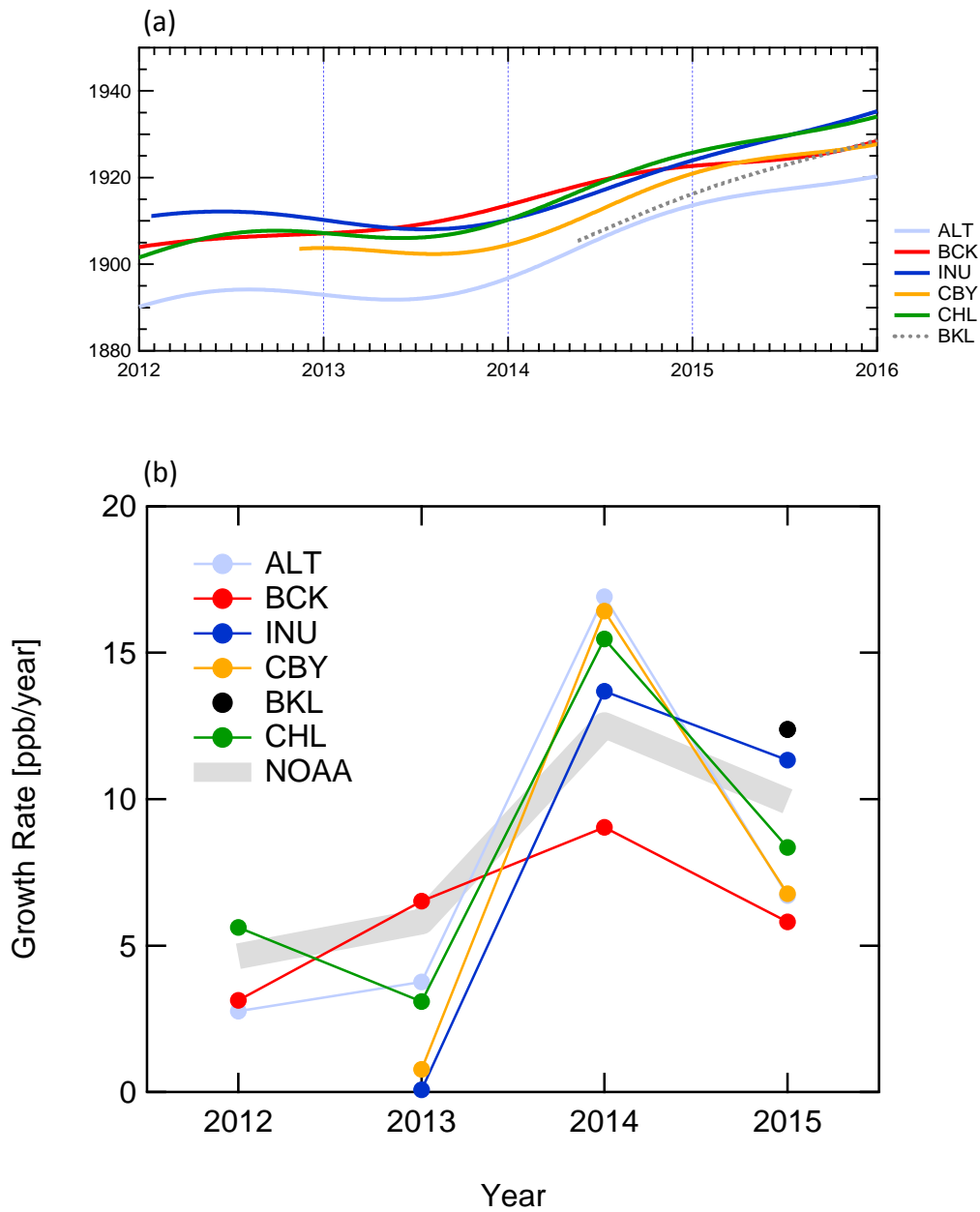
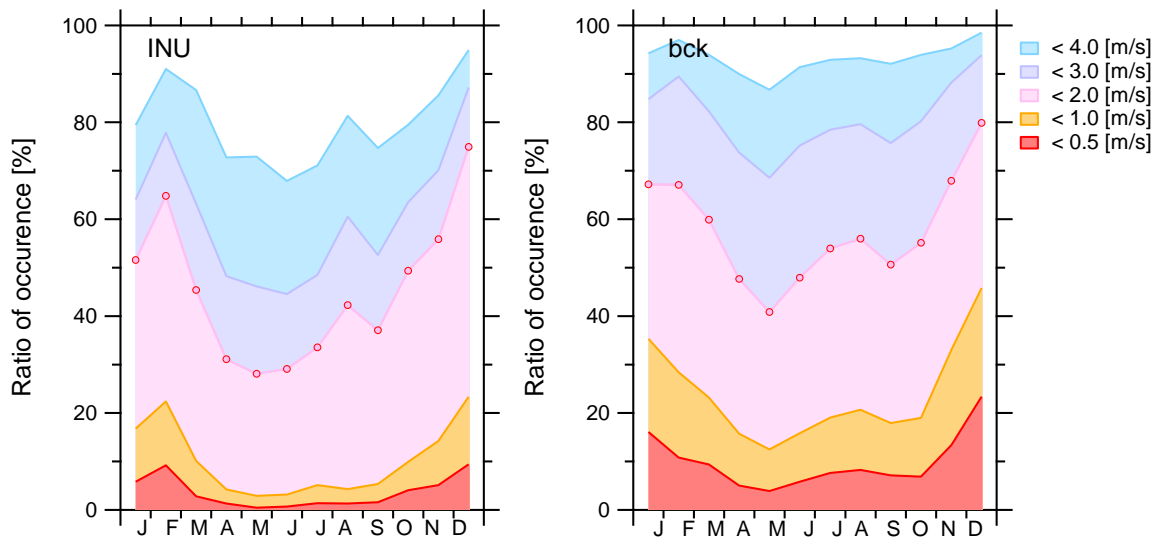
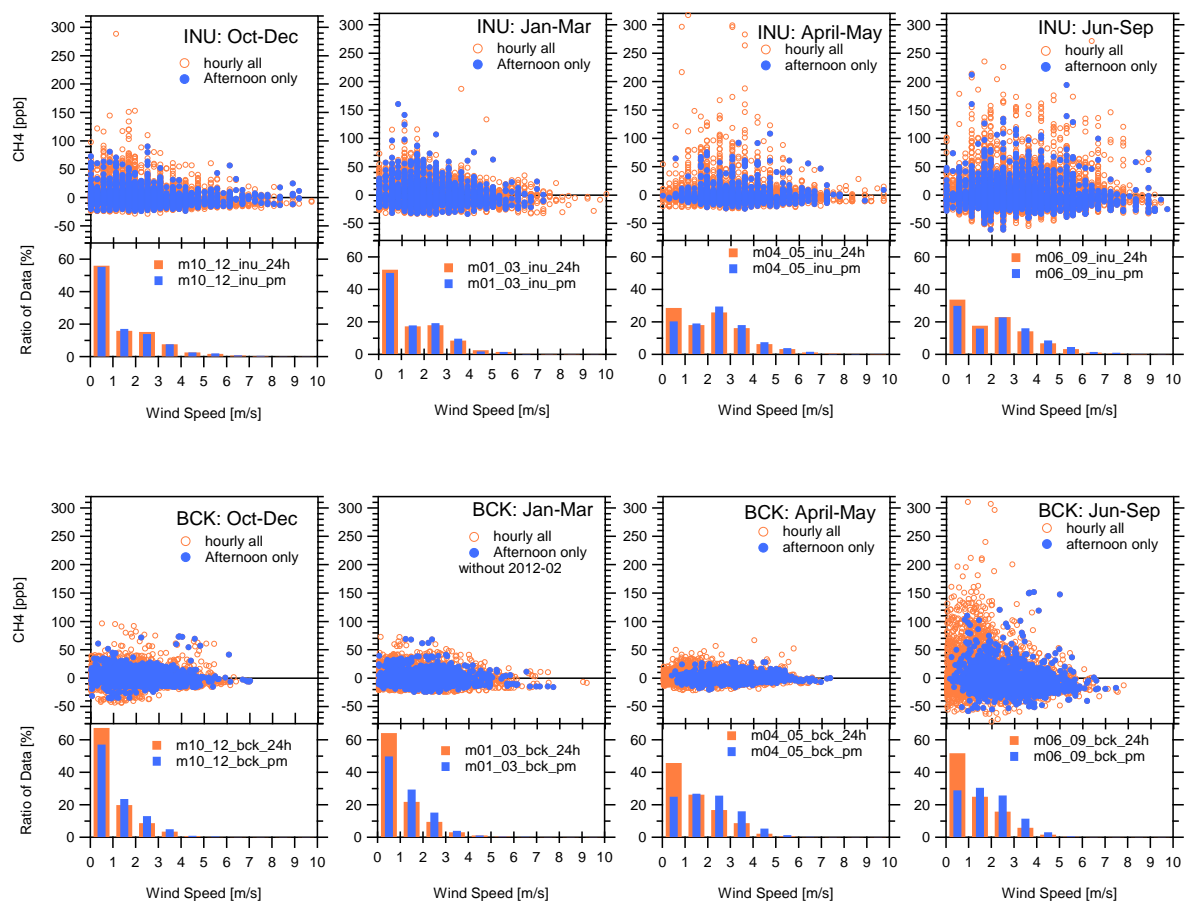


Figure S1: Growth rates of atmospheric CH<sub>4</sub> observed at Canadian Arctic sites. For comparison, NOAA annual increase from globally-averaged atmospheric CH<sub>4</sub> is plotted. The growth rates of Canadian Arctic sites are obtained in the similar manner with NOAA annual increase. The growth rate is defined by the difference in CH<sub>4</sub> concentration on January 1st from January 1st of the previous year in the long-term trend curves (shown in Fig. 2, seasonal components are removed).

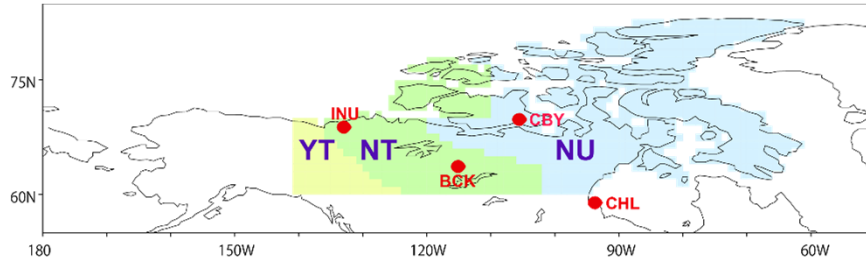


**Figure S2: Mean seasonal cycles of wind speed at Inuvik (INU) and Behchoko (BCK).** At INU, the hourly wind data are obtained at the Inuvik ECCC upper air weather station where the GHG measurement system is situated. At BCK, the wind speed has been measured independently of the ECCC weather network. The wind speed data for 2012-2015 are grouped by wind speed and normalized per month.

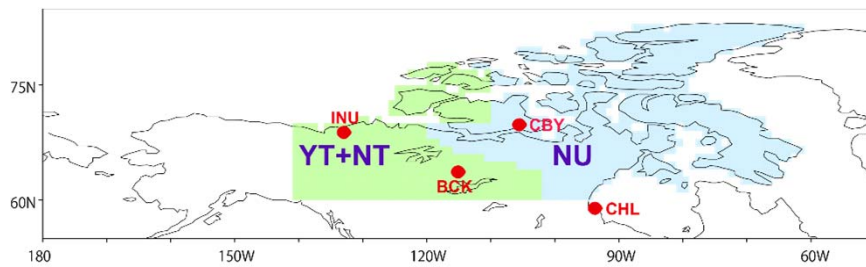


**Figure S3: Variability of observed hourly CH<sub>4</sub> concentrations at Inuvik (INU) and Behchoko (BCK). The variability is defined as the deviations of observed CH<sub>4</sub> from the fitted curves (shown in Fig. 2).**

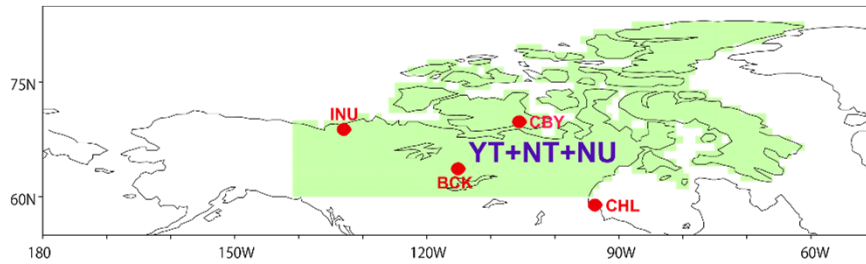
Mask A (3 sub-regions)



Mask B (2 sub-region)



Mask C



**Figure S4: Sub-regions used for the regional inversion. Initially, the Canadian Arctic is sub-divided based on territory boundaries. Mask A: three regions of YT (Yukon), NT (Northwest Territories) and NU (Nunavut), Mask B: Two regions by combining NT and YT, and Mask C: one region as combining all territories YT, NT and NU. The four continuous measurement sites used for the inversion are plotted in the red closed circles.**

Entire Canadian Arctic

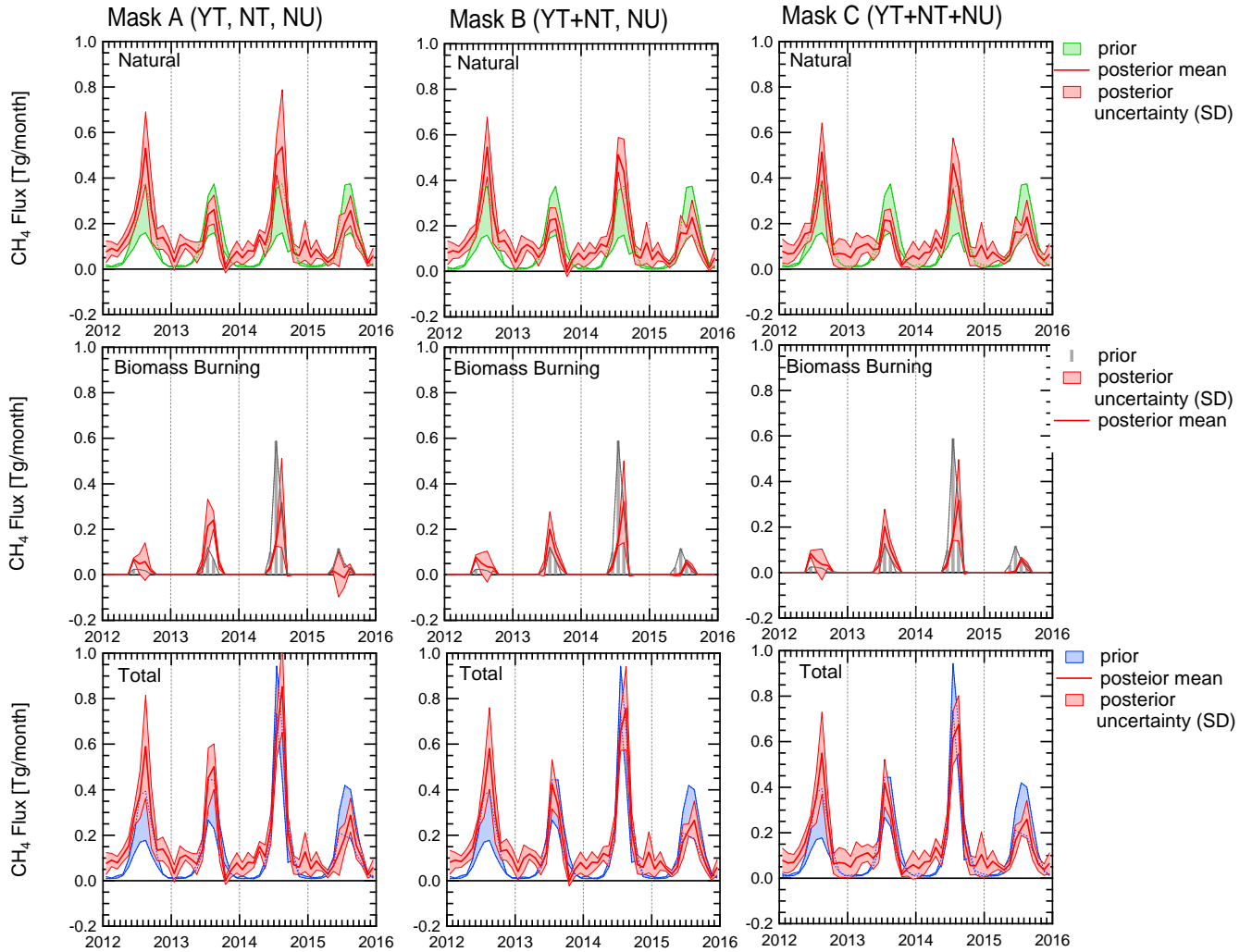
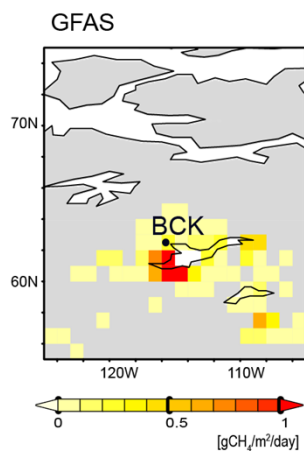
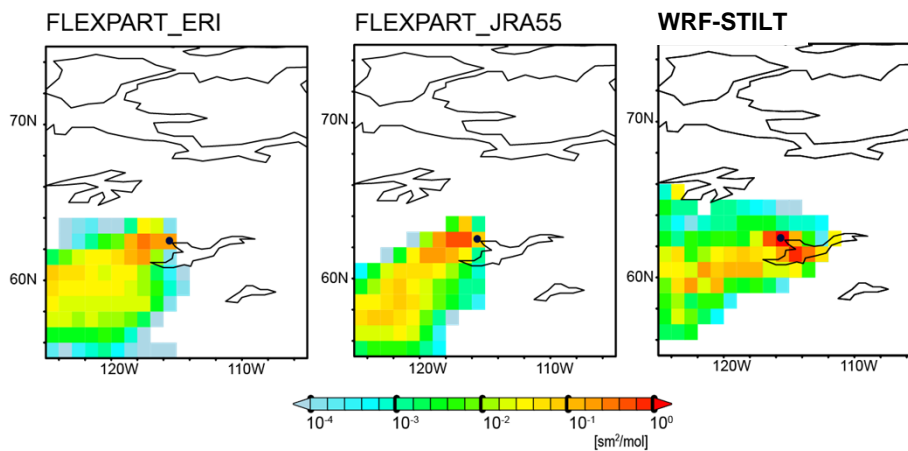


Figure S5: Monthly posterior fluxes, same with Fig. 8, but for the entire Canadian Arctic with three different sub-region masks. The sub-regional fluxes with Mask A and Mask B are aggregated.

(a) Biomass Burning  
CH<sub>4</sub> fluxes



(b) Footprints



(c) prior and posterior concentrations

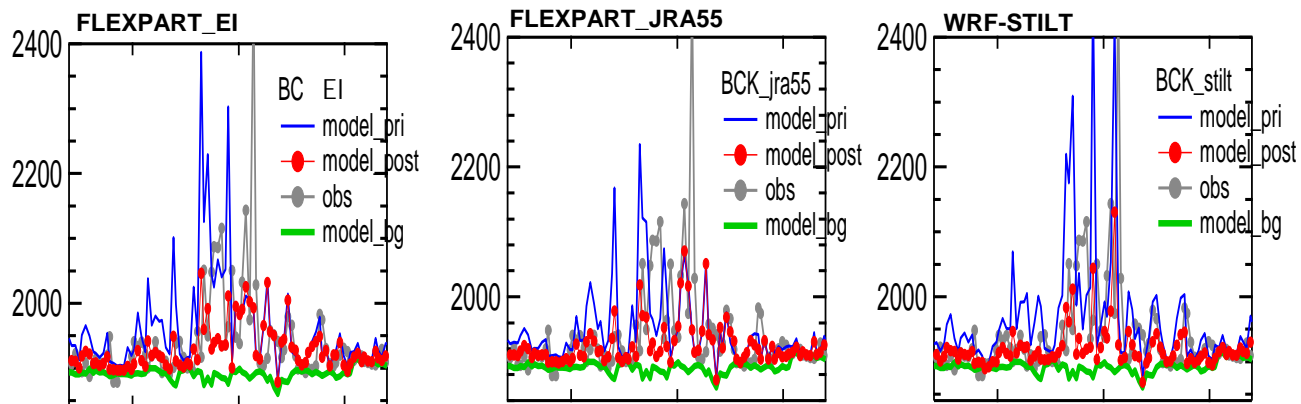
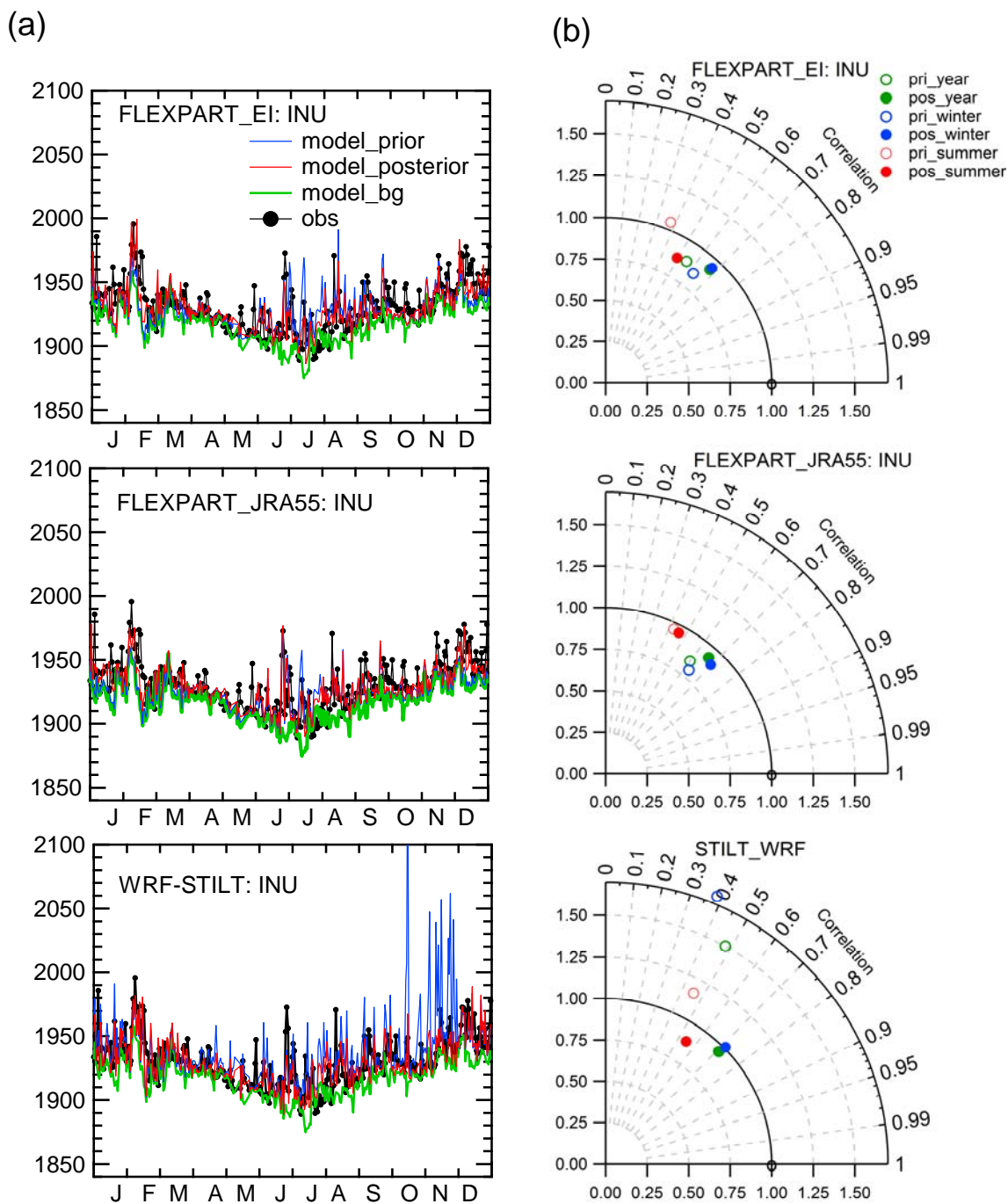
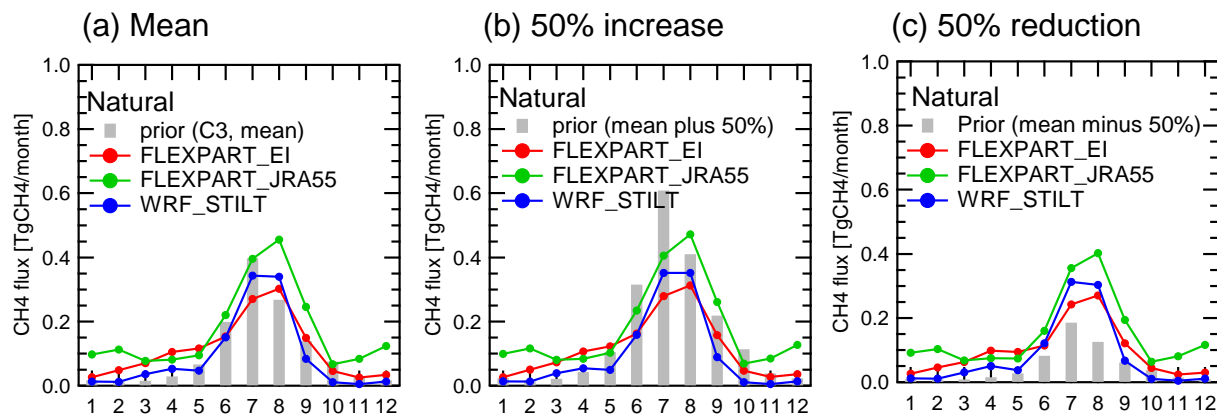


Figure S6: (a) Prior biomass burning CH<sub>4</sub> emissions from GFAS for August 5, 2014. (b) 5-day back trajectory footprints of BCK for August 5, 2014, by FLEXPART\_ERI, FLEXPART\_JRA55 and WRF-STILT. (c) prior (blue) and posterior (red) concentrations at BCK for the summer in 2014 with Mask B and prior flux case C3. Observations (gray) and modelled background concentrations (green) are also plotted.



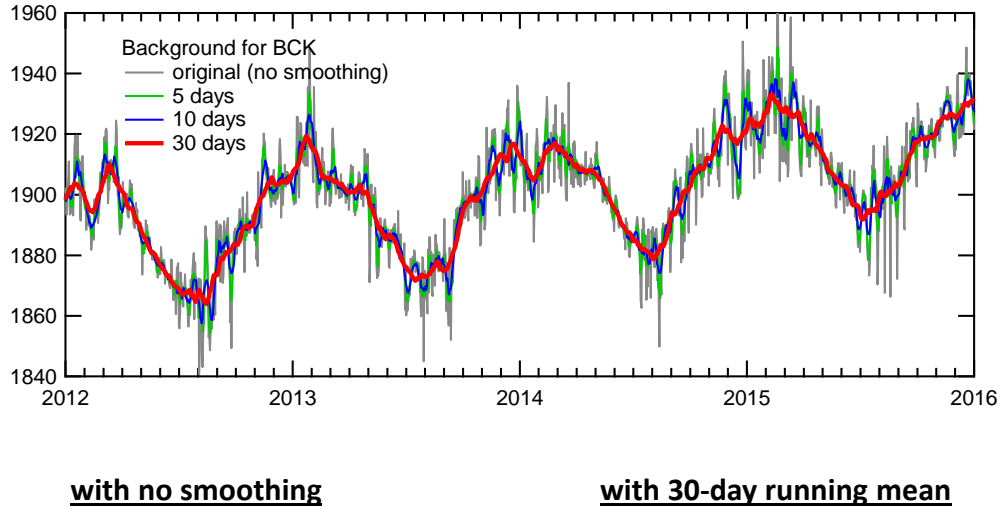
**Figure S7:** (a) Simulated concentrations for Inuvik with prior and posterior fluxes by three different transport models for 2015, with inversion set up of Mask B and prior flux case C3. The modelled background concentrations and observations are also plotted. (b) Taylor diagrams for comparison between prior and posterior concentrations. The correlation coefficients and normalised standard deviations (NSD) are calculated for summer (June–September), winter (October–May) and throughout years for 2012–2015.



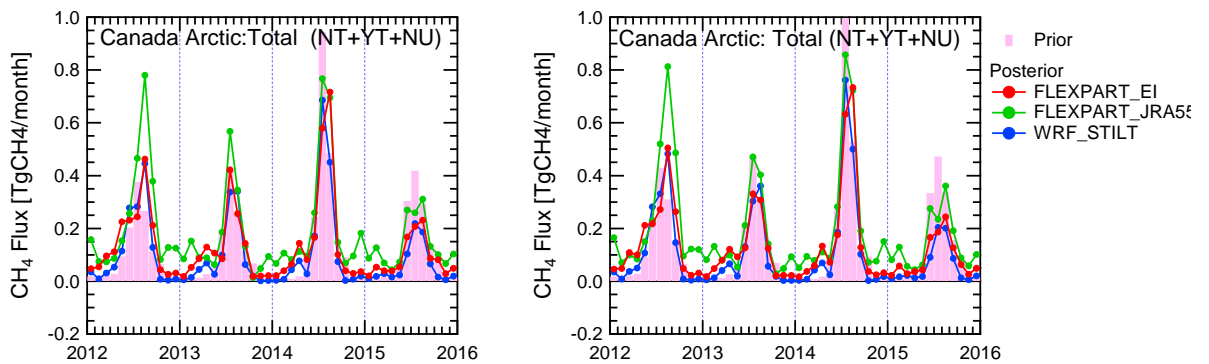


**Figure S8: Four-year (2012–2015) mean optimised natural CH<sub>4</sub> fluxes for the entire Canadian Arctic, with prior flux case C3, but three different prior wetland fluxes, (a) WetCHARTs mean (same as C3), (b) WetCHARTs mean with 50% increase, and (c) WetCHARTs mean with 50 % reduction.**

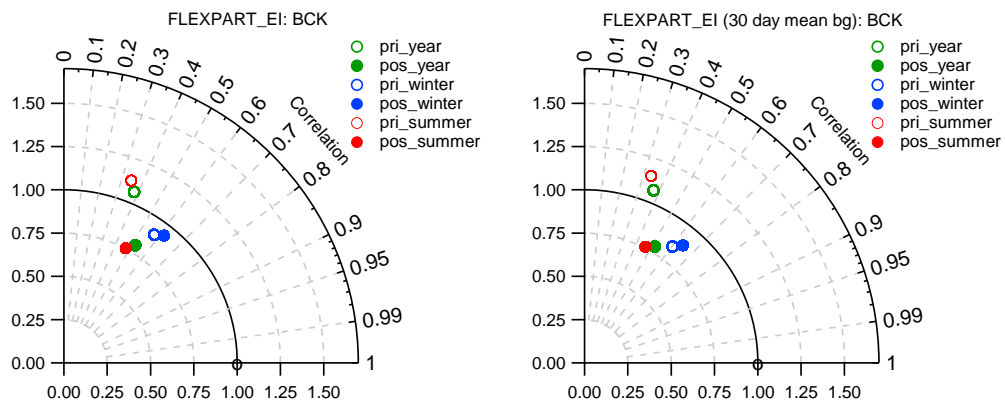
(a) Background



(b) Estimated Fluxes



(c) Taylor diagrams for prior and posterior concentrations



**Figure S9: Impact of background concentrations.** (a) the background concentrations for Behchoko (BCK) with no smoothing (black), 5-day (green), 10-day (blue) and 30-day (red) running mean. (b) Estimated total fluxes for the entire Canadian Arctic with background with no smoothing (left) and with 30-day running mean (right) in the experiments with Mask B and Prior emission C3 by FLEXPART\_EI (red), FLEXPART\_JRA55 (green) and WRF-STILT (blue). The prior fluxes are indicated by pink histograms. (c) Taylor diagram for comparison of prior (open circles) and posterior (closed circles) concentrations by FLEXPART\_EI for the entire period and winter/summer seasons between the non-smoothed background (left) and smoothed background (right).