1 **Supporting Information for**

2 **Estimation of CH$_4$ emission based on advanced 4D-LETKF assimilation system**

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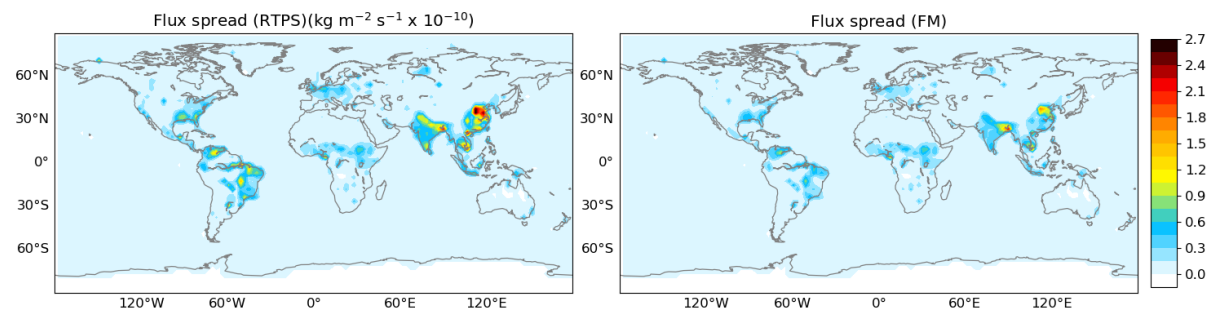8 *corresponding author's e-mail: jagatbisht@jamstec.go.jp

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11 **Figure S1.** Initial analysis ensemble flux spread with RTPS and fixed multiplicative (FM) covariance
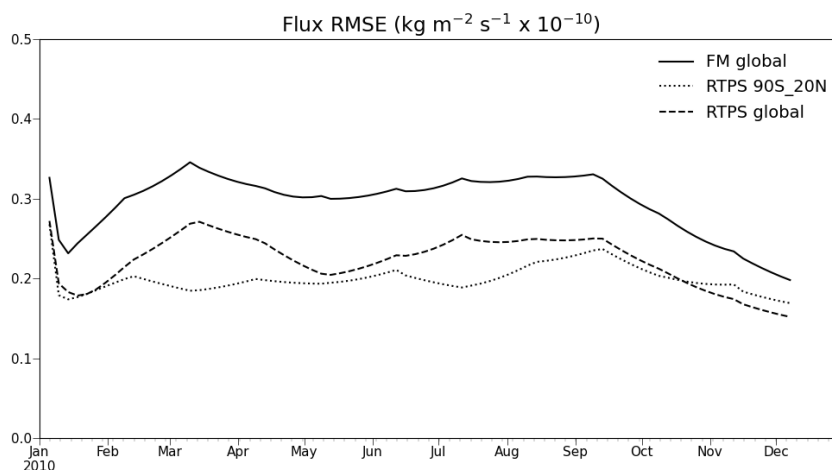
12 inflation methods.

14 **Figure S2.** Time series of RMSE of surface CH4 flux analysis, for 1 year of data assimilation at every

15 grid using fixed multiplicative inflation and RTPS inflation methods over global landmass region.
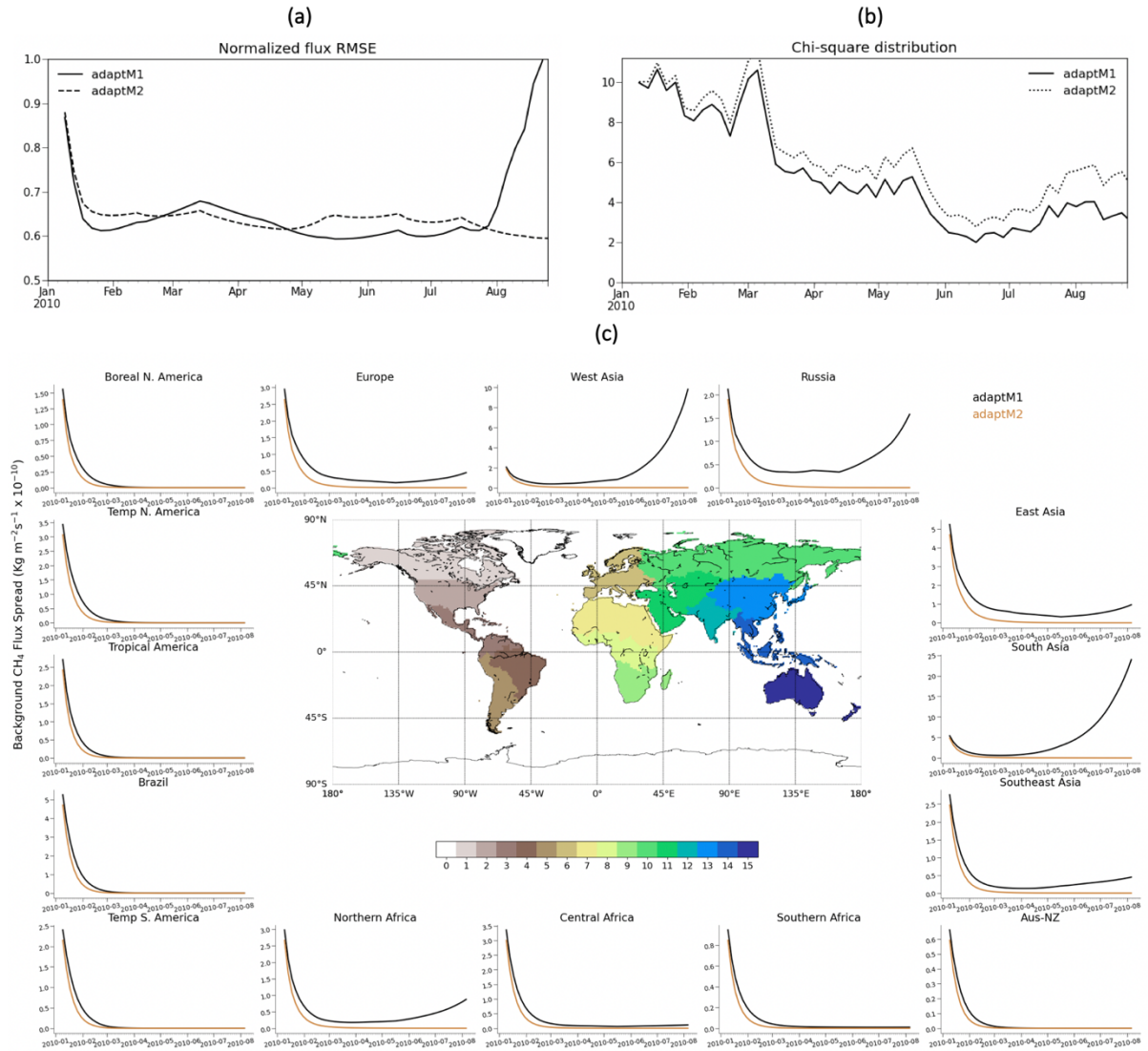
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Figure S3 CH$_4$ LETKF flux estimation time series for; (a) normalized RMSE estimated using
adaptive multiplicative inflation method in two sensitivity experiments (adaptM1; 50% initial
inflation, adaptM2; 40% initial inflation), (b) chi-square distribution comparison for adaptive
multiplicative sensitivity experiments (c) background error spread evolution for adaptive
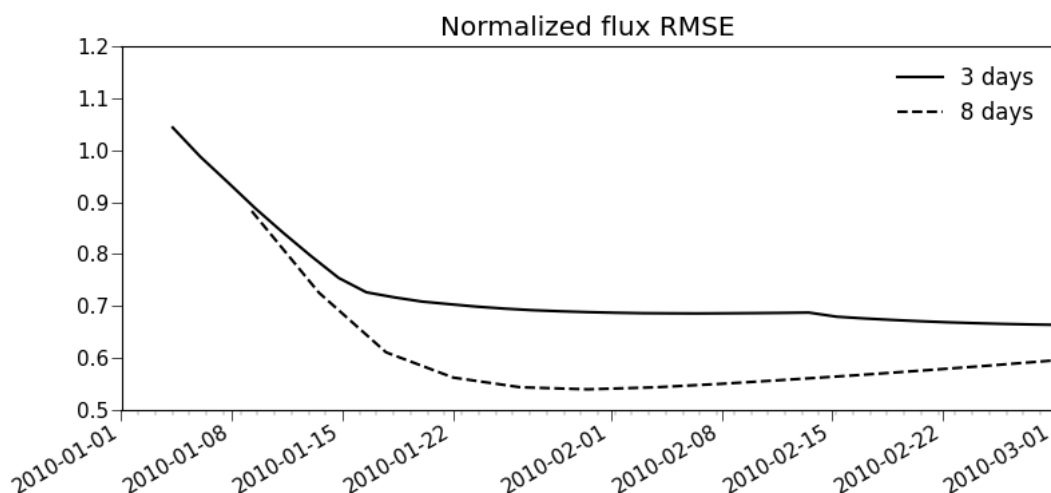multiplicative inflation experiments.

Figure S4. Time series of normalized RMSE estimated using 3 days and 8 days assimilation window for CH_4 LETKF data assimilation (inflation method: RTPS).
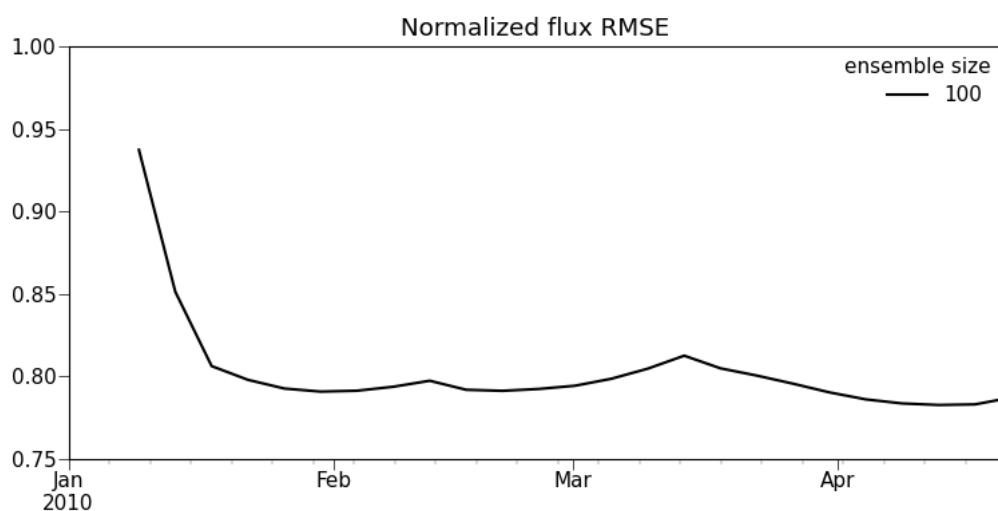

Figure S5. Time series of normalized RMSE of surface CH_4 flux analysis after assimilating GOSAT synthetic CH_4 observations shown for the period of 4 months using initial spread that is generated by considering the initial perturbation at each model grid by considering horizontal spatial error correlation between grid points among ensemble members, with a global mean correlation of 20%.
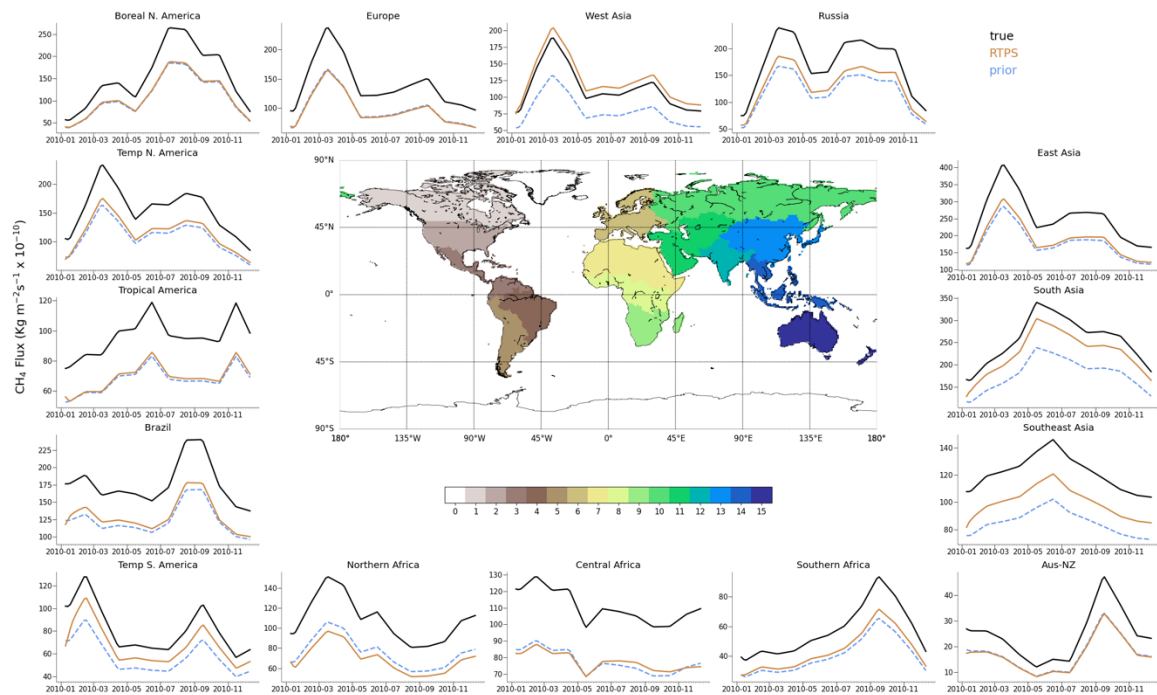
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Figure S6. The 1-year CH4 total flux seasonal cycles of true (black), prior (gray), and estimated from the LETKF (blue) in 15 regions using FM inflation method after assimilating synthetic GOSAT CH$_4$ observations (Fig. 6).
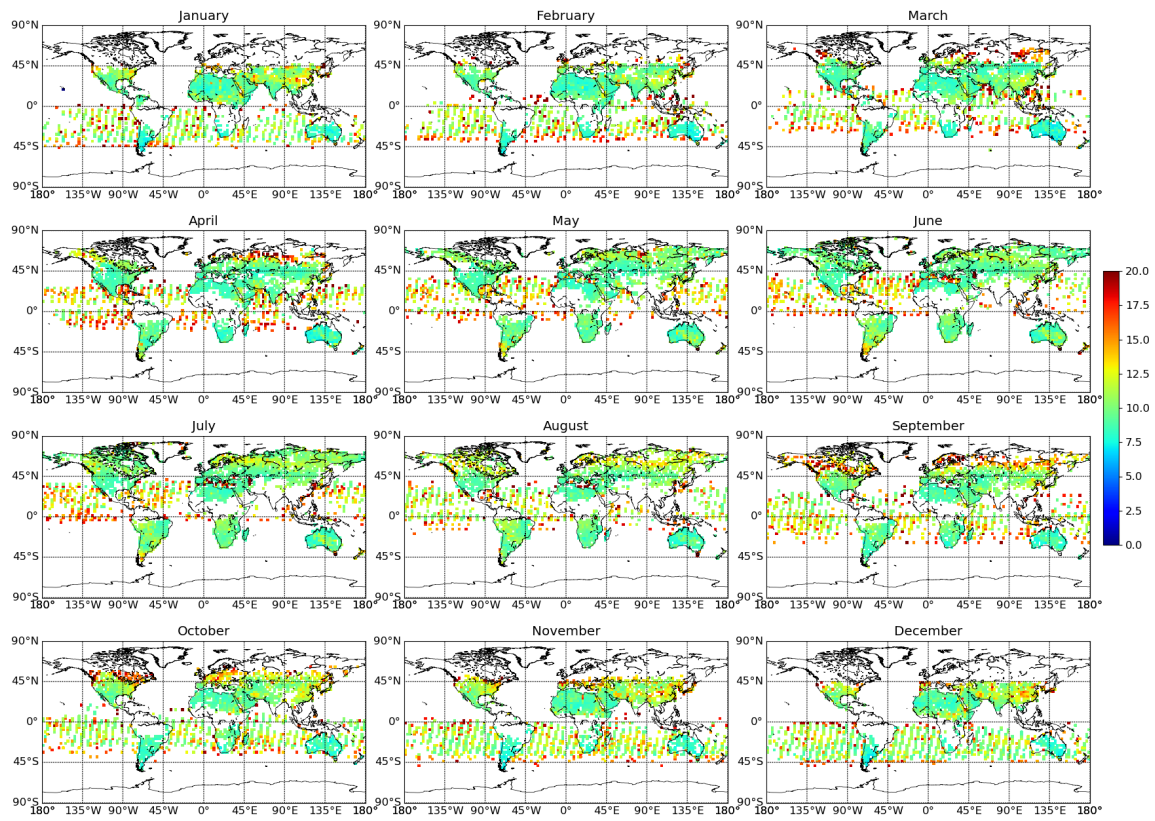
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Figure S7. Monthly mean of real GOSAT observations retrieval error during the year 2010.

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