



Supplement of

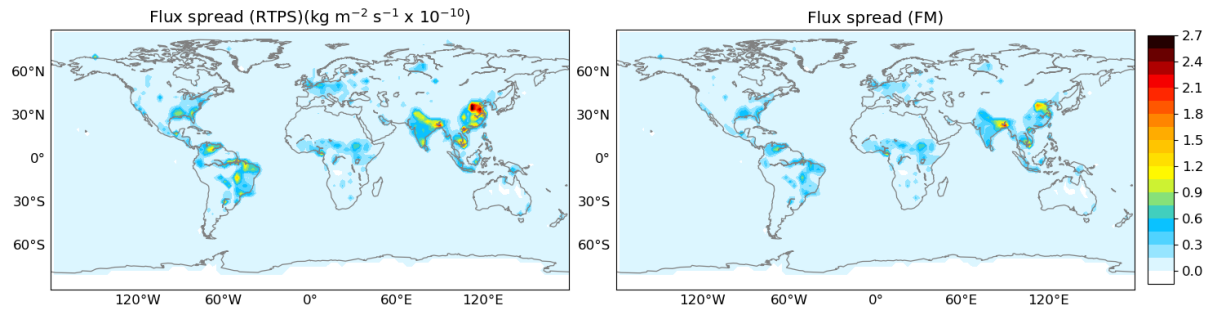
Estimation of CH₄ emission based on an advanced 4D-LETKF assimilation system

Jagat S. H. Bisht et al.

Correspondence to: Jagat S. H. Bisht (jagatbisht@jamstec.go.jp, jshbisht@gmail.com)

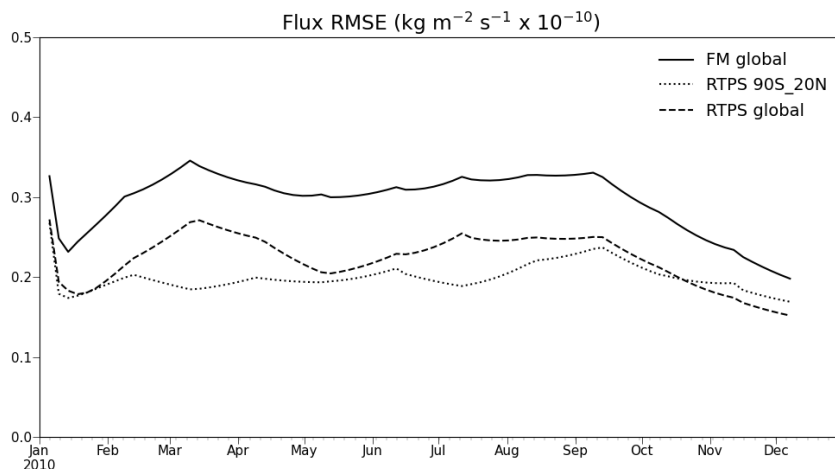
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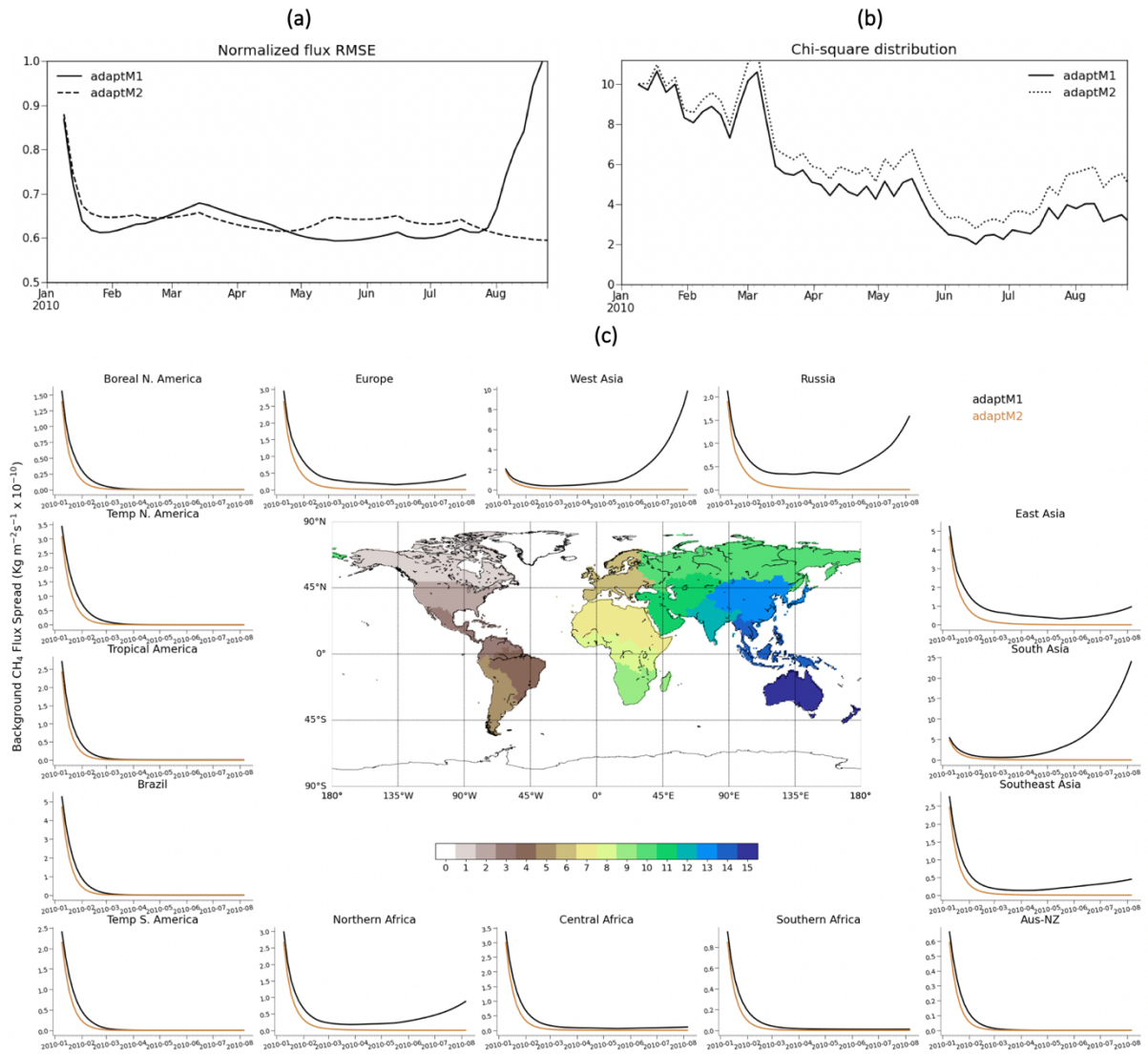
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3 **Figure S1.** Initial analysis ensemble flux spread with RTPS and fixed multiplicative (FM) covariance
4 inflation methods.



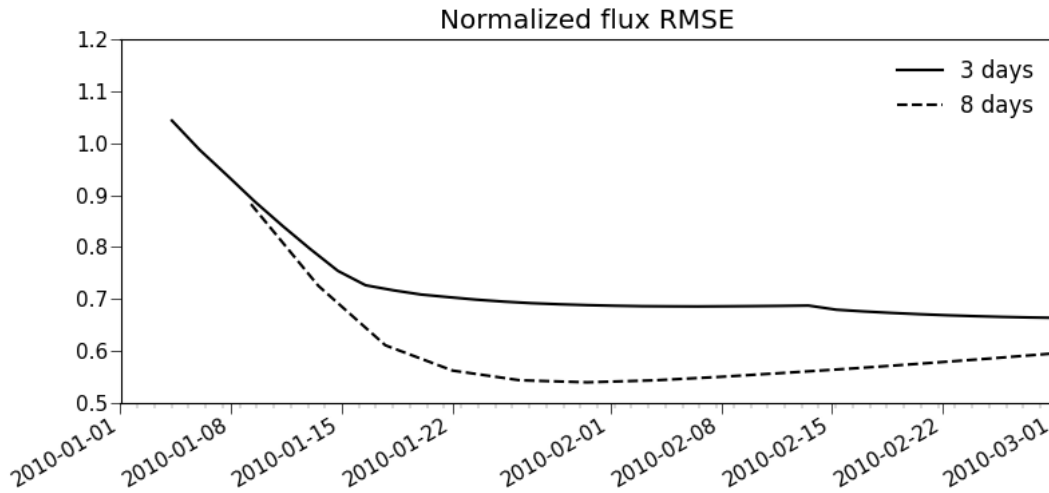
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6 **Figure S2.** Time series of RMSE of surface CH₄ flux analysis, for 1 year of data assimilation at every
7 grid using fixed multiplicative inflation and RTPS inflation methods over global landmass region.



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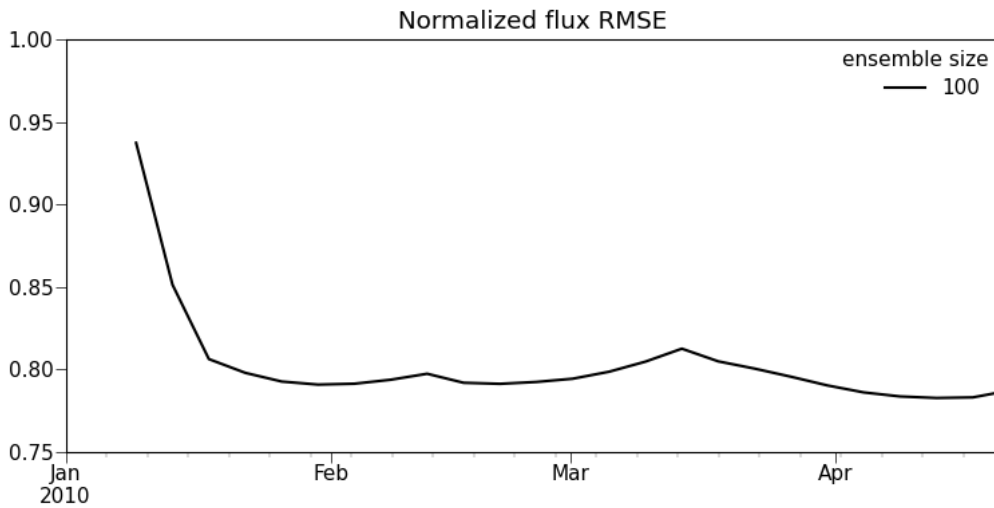
9 **Figure S3** CH₄ LETKF flux estimation time series for; (a) normalized RMSE estimated using
 10 adaptive multiplicative inflation method in two sensitivity experiments (adaptM1; 50% initial
 11 inflation, adaptM2; 40% initial inflation), (b) chi-square distribution comparison for adaptive
 12 multiplicative sensitivity experiments (c) background error spread evolution for adaptive
 13 multiplicative inflation experiments.



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15 **Figure S4.** Time series of normalized RMSE estimated using 3 days and 8 days assimilation window
 16 for CH₄ LETKF data assimilation (inflation method: RTPS).

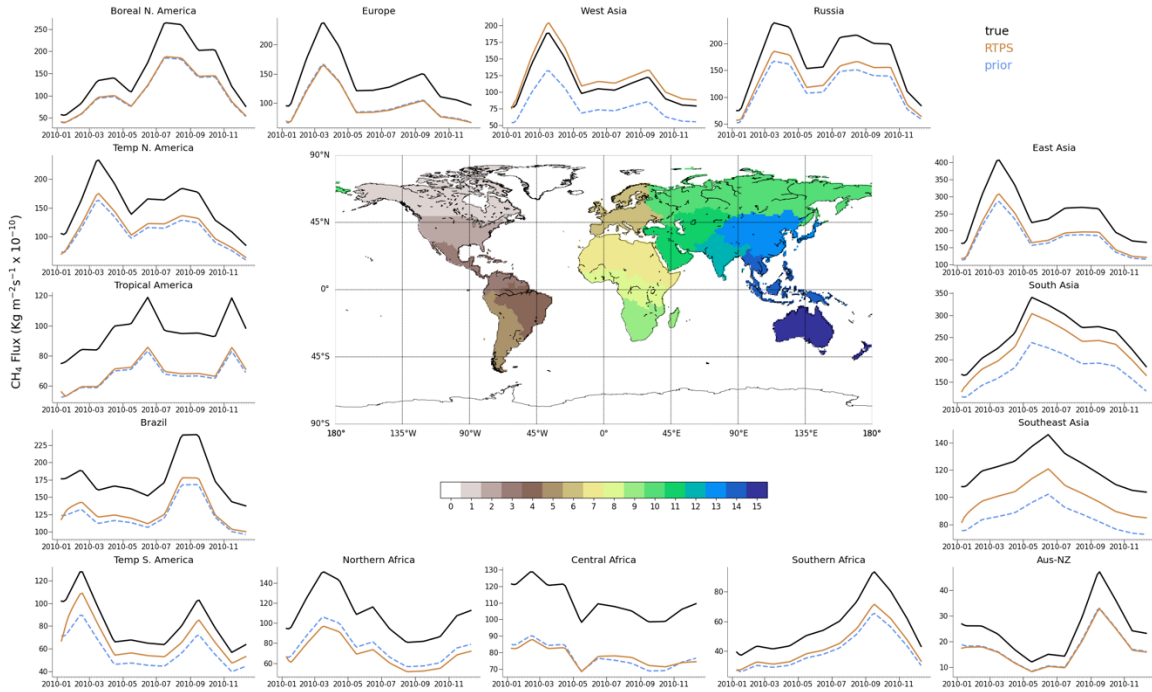
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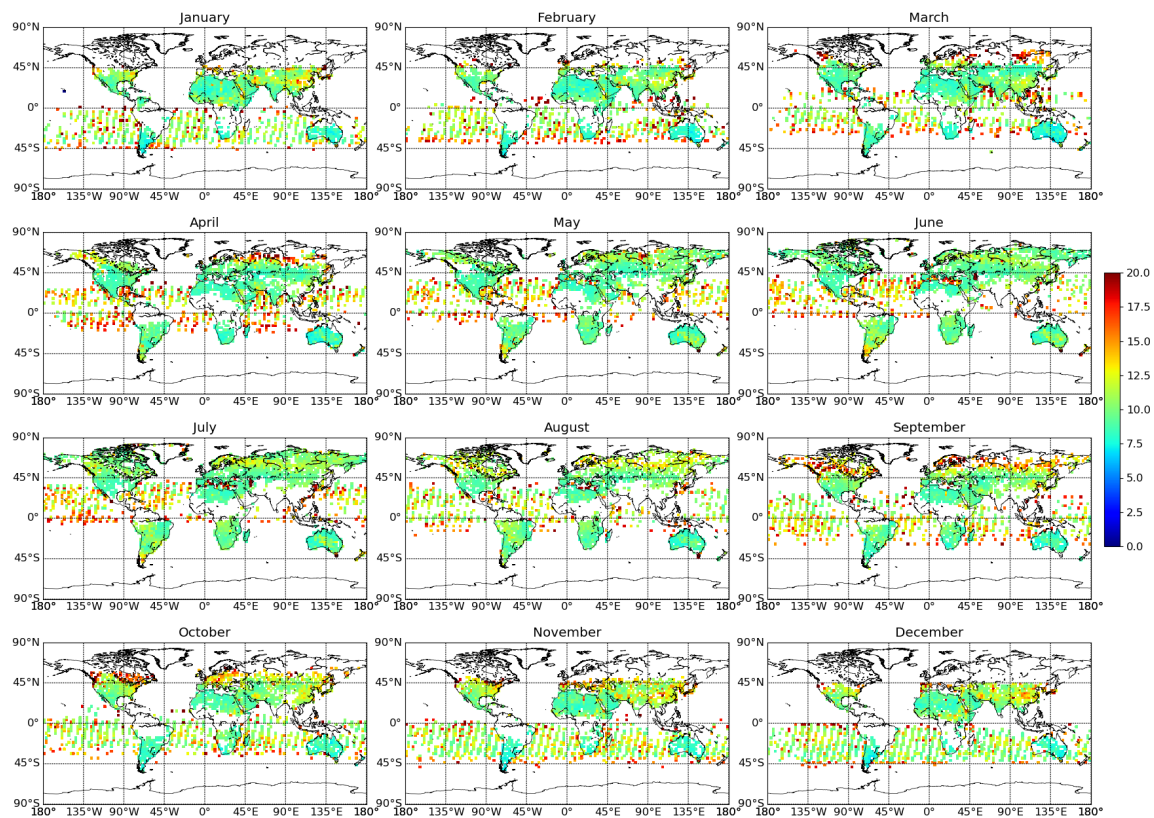
20 **Figure S5.** Time series of normalized RMSE of surface CH₄ flux analysis after assimilating GOSAT
 21 synthetic CH₄ observations shown for the period of 4 months using initial spread that is generated by
 22 considering the initial perturbation at each model grid by considering horizontal spatial error
 23 correlation between grid points among ensemble members, with a global mean correlation of 20%.



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25 **Figure S6.** The 1-year CH₄ total flux seasonal cycles of true (black), prior (gray), and estimated from
 26 the LETKF (blue) in 15 regions using FM inflation method after assimilating synthetic GOSAT CH₄
 27 observations (Fig. 6).

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30 **Figure S7.** Monthly mean of real GOSAT observations retrieval error during the year 2010.

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