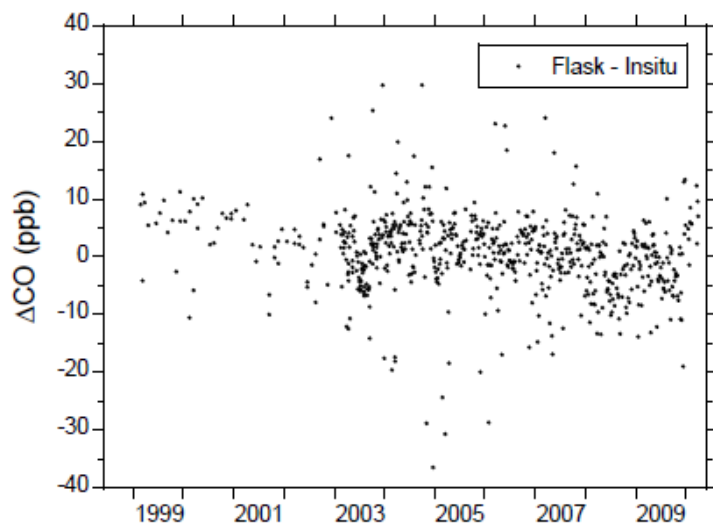
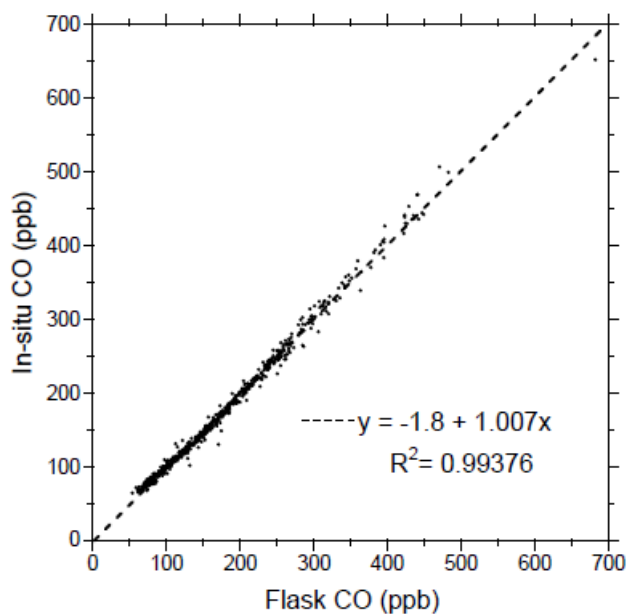
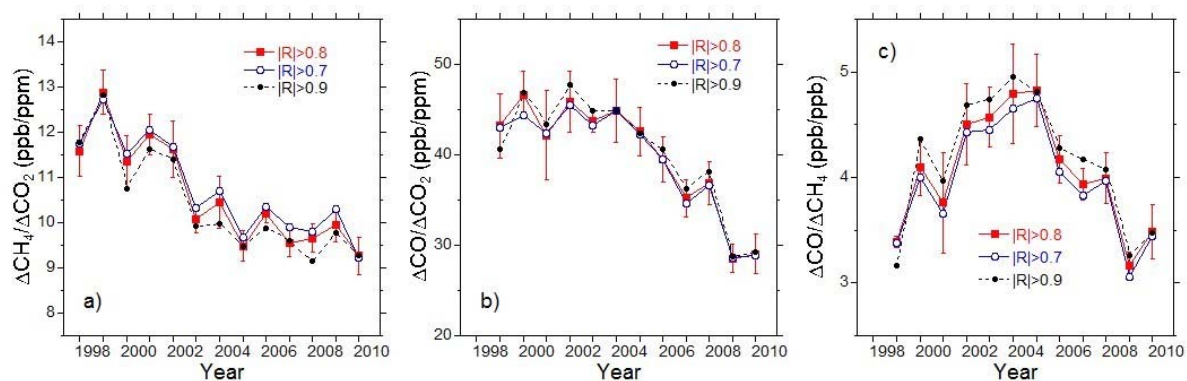


1 **Supplementary material**

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3 **Fig. S1.** Time series of the differences between flask and in-situ measurements of the
4 atmospheric CO mixing ratios at HAT during the period from 1999 to 2010.



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6 **Fig. S2.** Scatter plot of the flask and in-situ CO measurements. The broken line represents the
7 linear regression line.
8

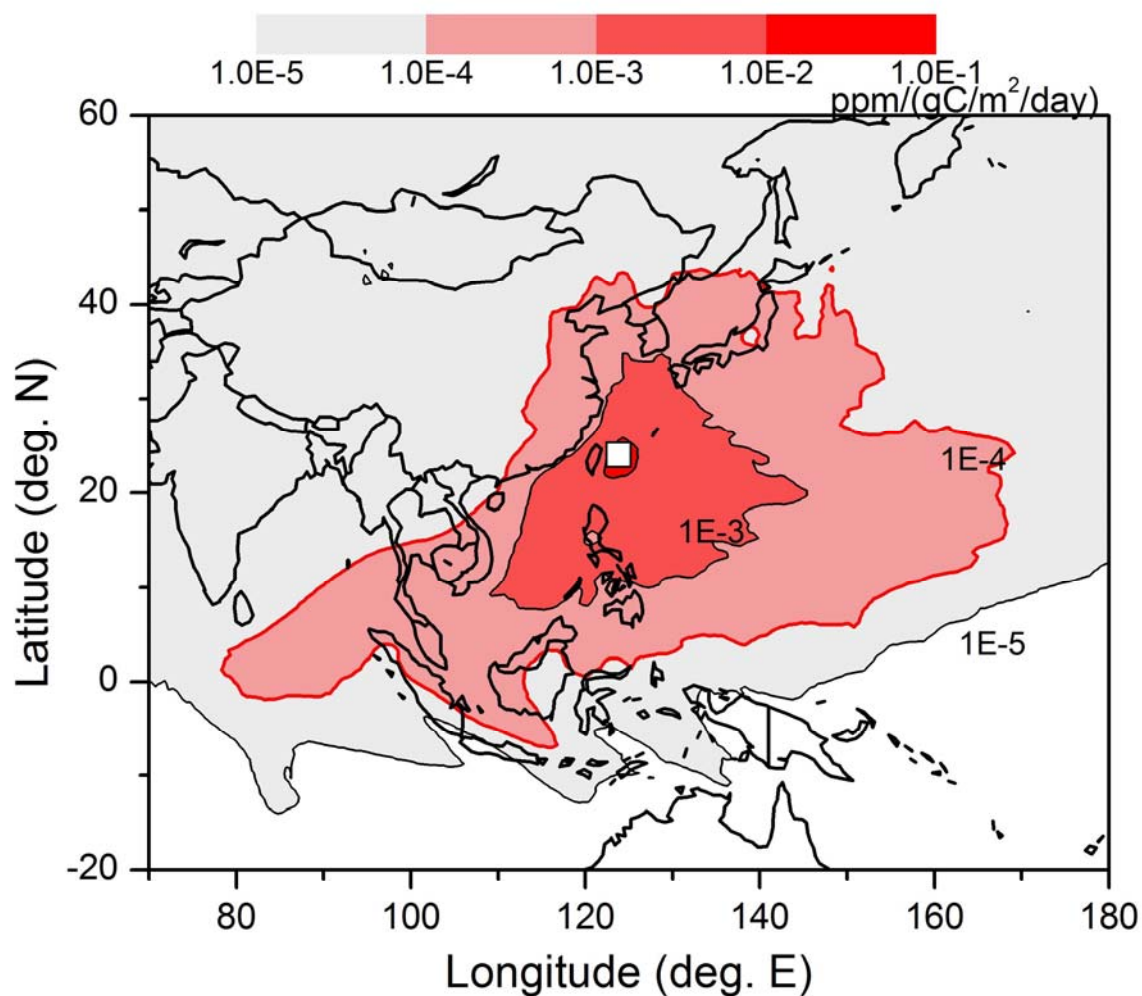


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11 **Fig. S3.** Temporal changes in the winter average correlation slopes of (a) $\Delta\text{CH}_4/\Delta\text{CO}_2$, (b)
 12 $\Delta\text{CO}/\Delta\text{CO}_2$, and (c) $\Delta\text{CO}/\Delta\text{CH}_4$ for 3 correlation coefficients that are used in the selection
 13 criteria (see text). The red squares represent the correlation coefficient of 0.8, black open circle
 14 0.7, and black closed circle 0.9.

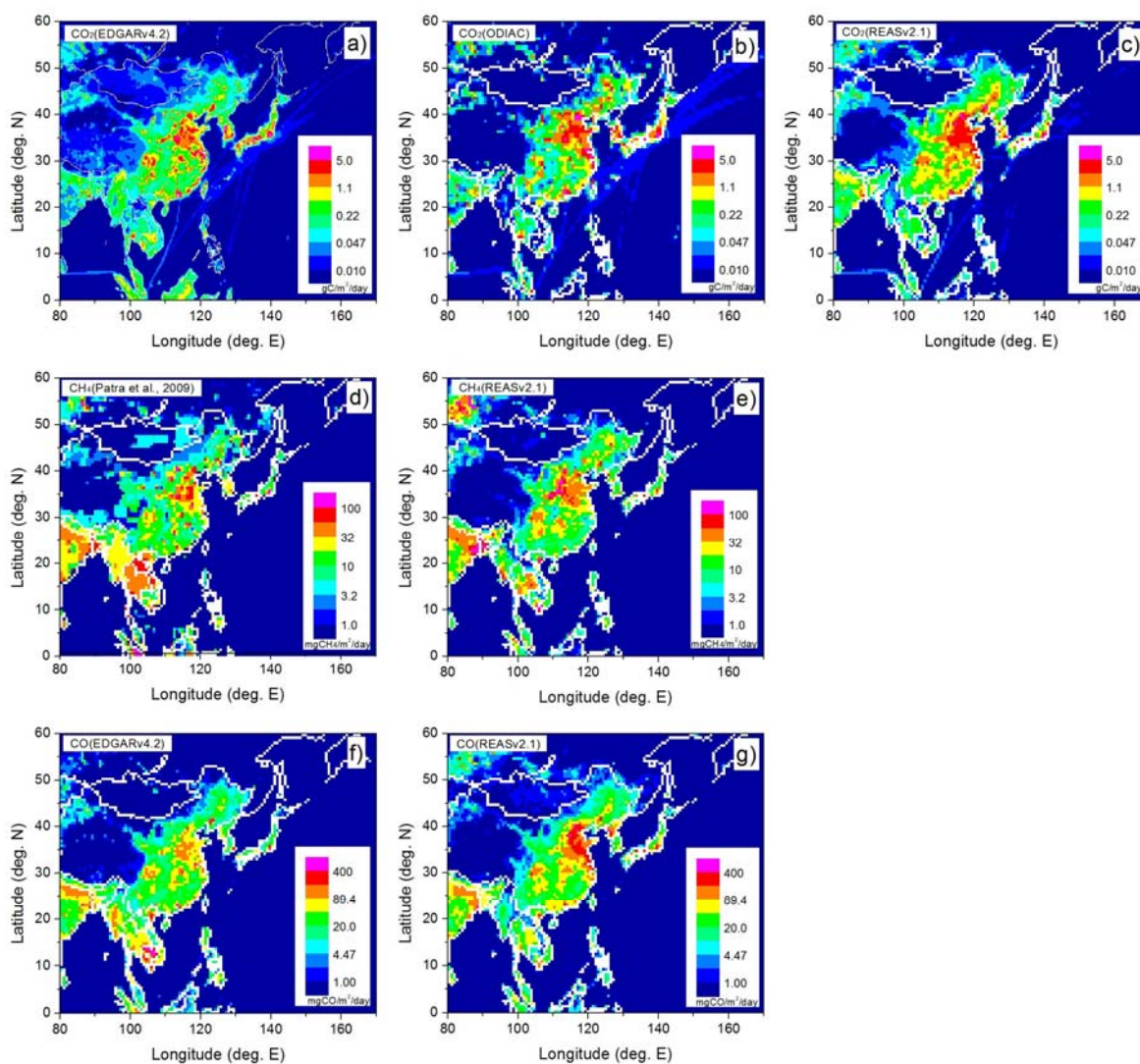
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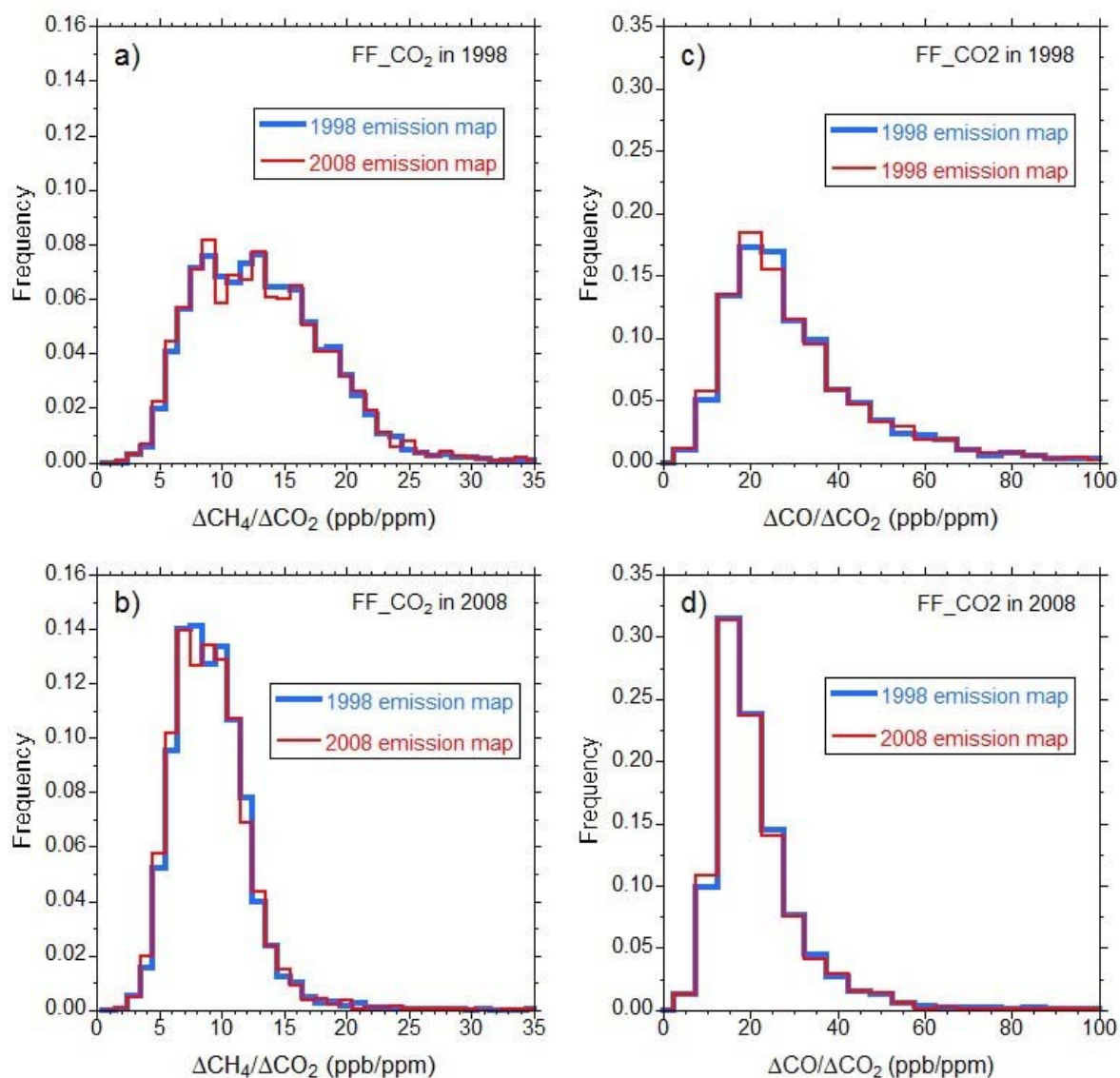
18 **Fig. S4.** Average footprint ($\text{ppm} (\text{gC m}^{-2} \text{ day}^{-1})^{-1}$) for the measurements at HAT during the
19 summer period (May to September). Meteorological data for 2006-2010 are used for the
20 calculation. The location of HAT is indicated by the square.



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23 **Fig. S5.** Comparison of the flux distributions of (a) fossil CO₂ from EDGAR v4.2, (b) fossil CO₂
 24 from ODIAC, (c) fossil CO₂ from REAS v2.1, (d) CH₄ from Patra et al., (2009), (e) CH₄ from
 25 REAS v2.1, (f) CO from EDGAR v4.2, and (g) CO from REAS v2.1. The flux maps for 2007 are
 26 shown. Annual mean fluxes are depicted for CO₂ and CO, while monthly mean fluxes in January
 27 are depicted for CH₄.

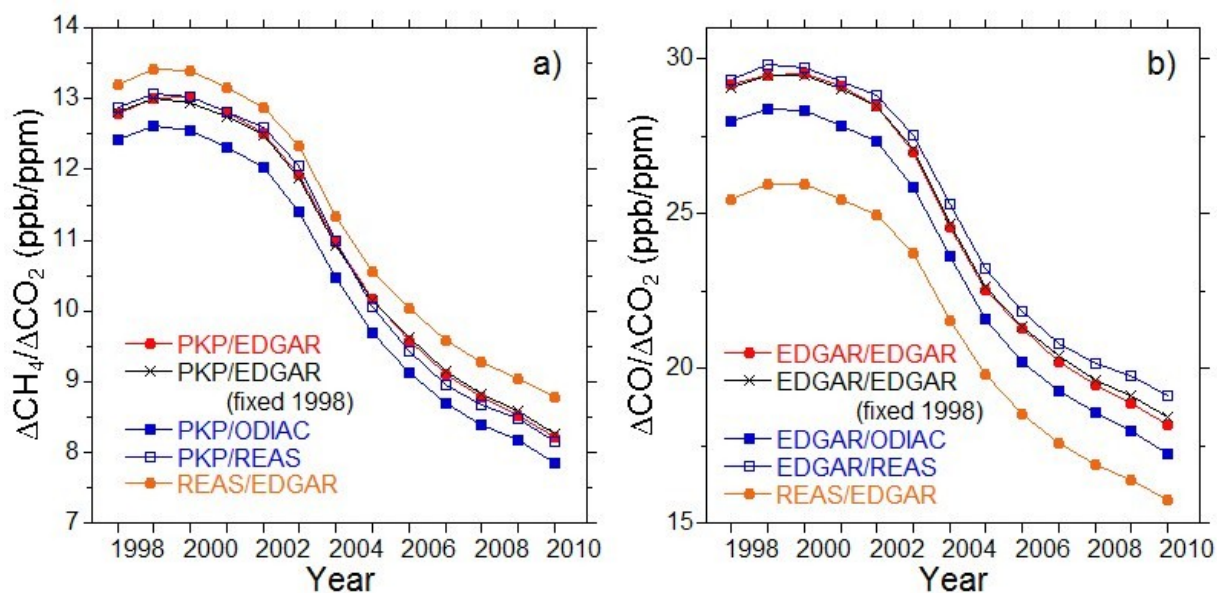


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30 **Fig. S6.** Histograms of the simulated correlation slopes of (a, b) $\Delta\text{CH}_4/\Delta\text{CO}_2$ and (c, d)
 31 $\Delta\text{CO}/\Delta\text{CO}_2$ for fossil CO₂ emissions in (a, c) 1998 and (b, d) 2008. The correlation slopes all
 32 meet the selection criteria ($|R| > 0.8$). The simulated results based on the fossil fuel-derived CO₂
 33 emission maps for 1998 and 2008 are depicted as blue and red lines, respectively.

34



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36

37 **Fig. S7.** Comparison of the winter average correlation slopes of simulated (a) $\Delta\text{CH}_4/\Delta\text{CO}_2$ and
 38 (b) $\Delta\text{CO}/\Delta\text{CO}_2$ for different combinations of the emission maps described in the legend. PKP in
 39 the legend represents the CH_4 emissions from Patra et al., (2009). The simulated correlation
 40 slopes for the 1998 EDGAR CO_2 emission map are also depicted as crosses.

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