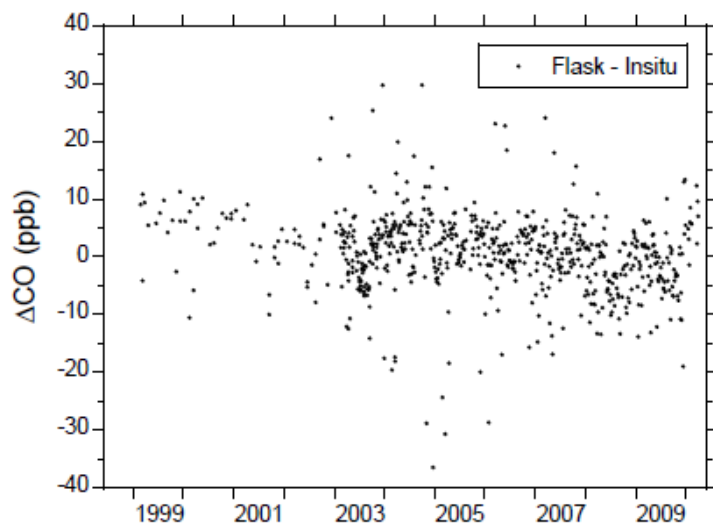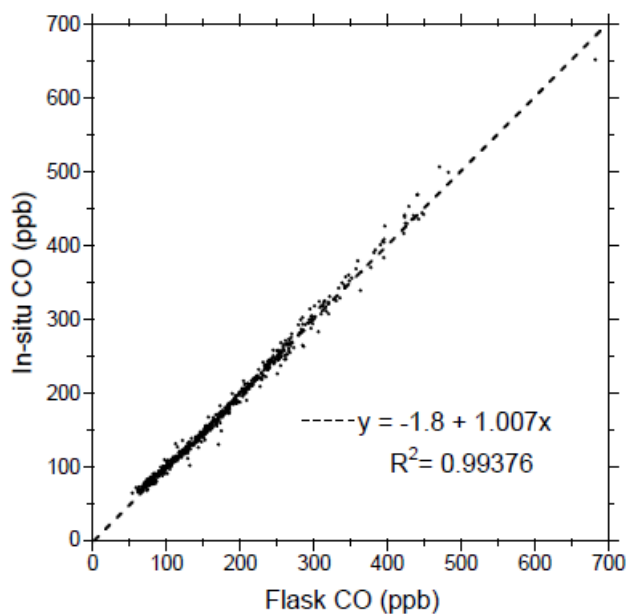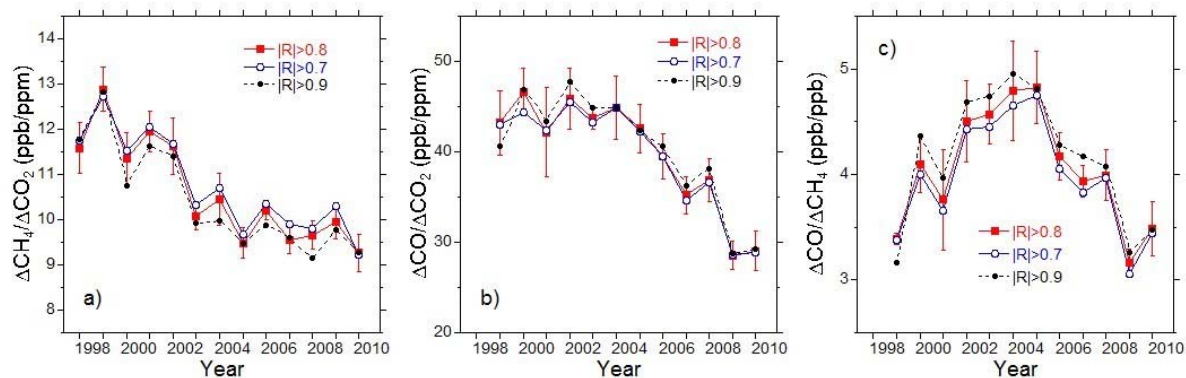1   **Supplementary material**

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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.

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Fig. S3. Temporal changes in the winter average correlation slopes of (a) $\Delta CH_4/\Delta CO_2$, (b) $\Delta CO/\Delta CO_2$, and (c) $\Delta CO/\Delta CH_4$ for 3 correlation coefficients that are used in the selection criteria (see text). The red squares represent the correlation coefficient of 0.8, black open circle 0.7, and black closed circle 0.9.
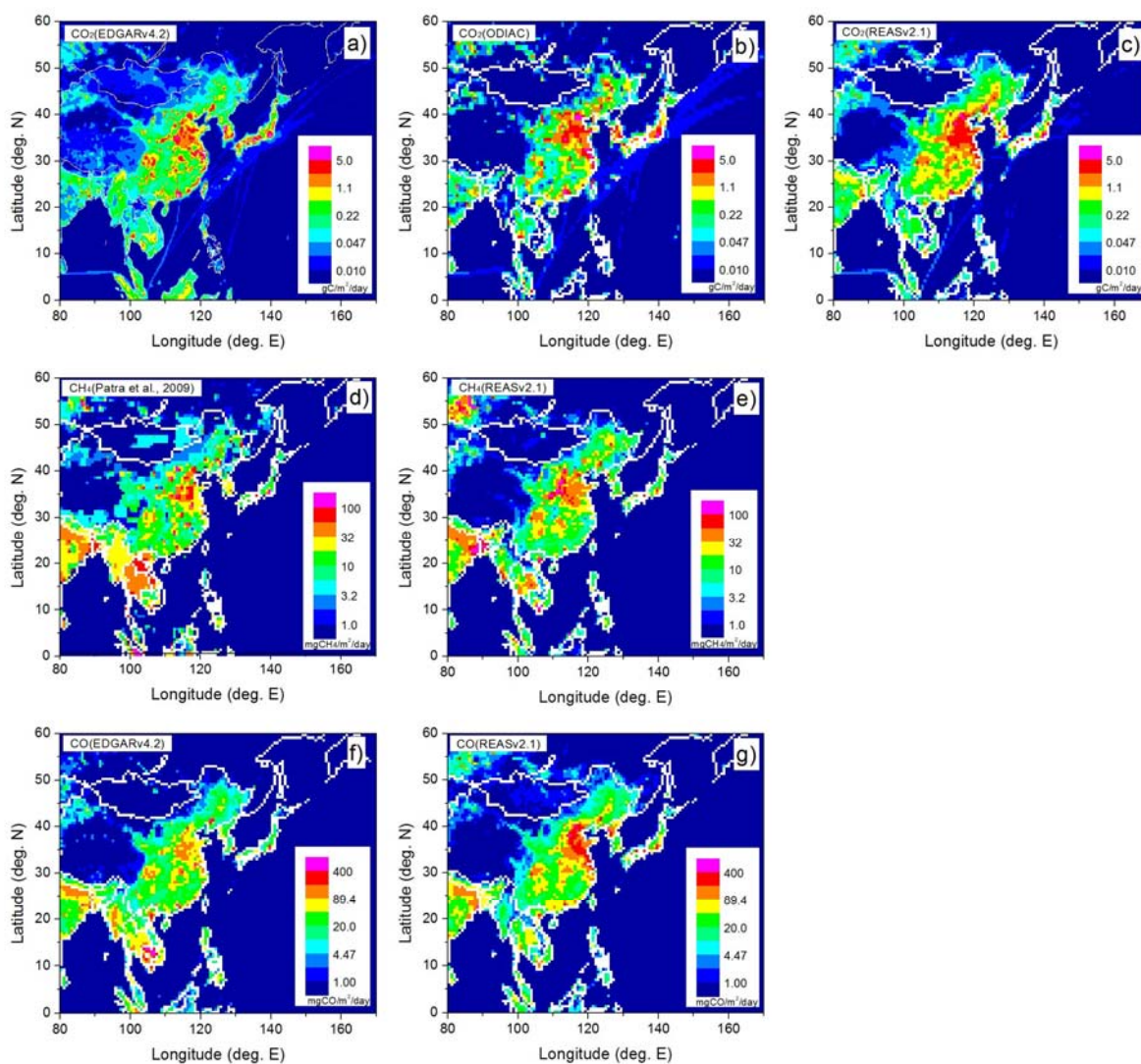
15
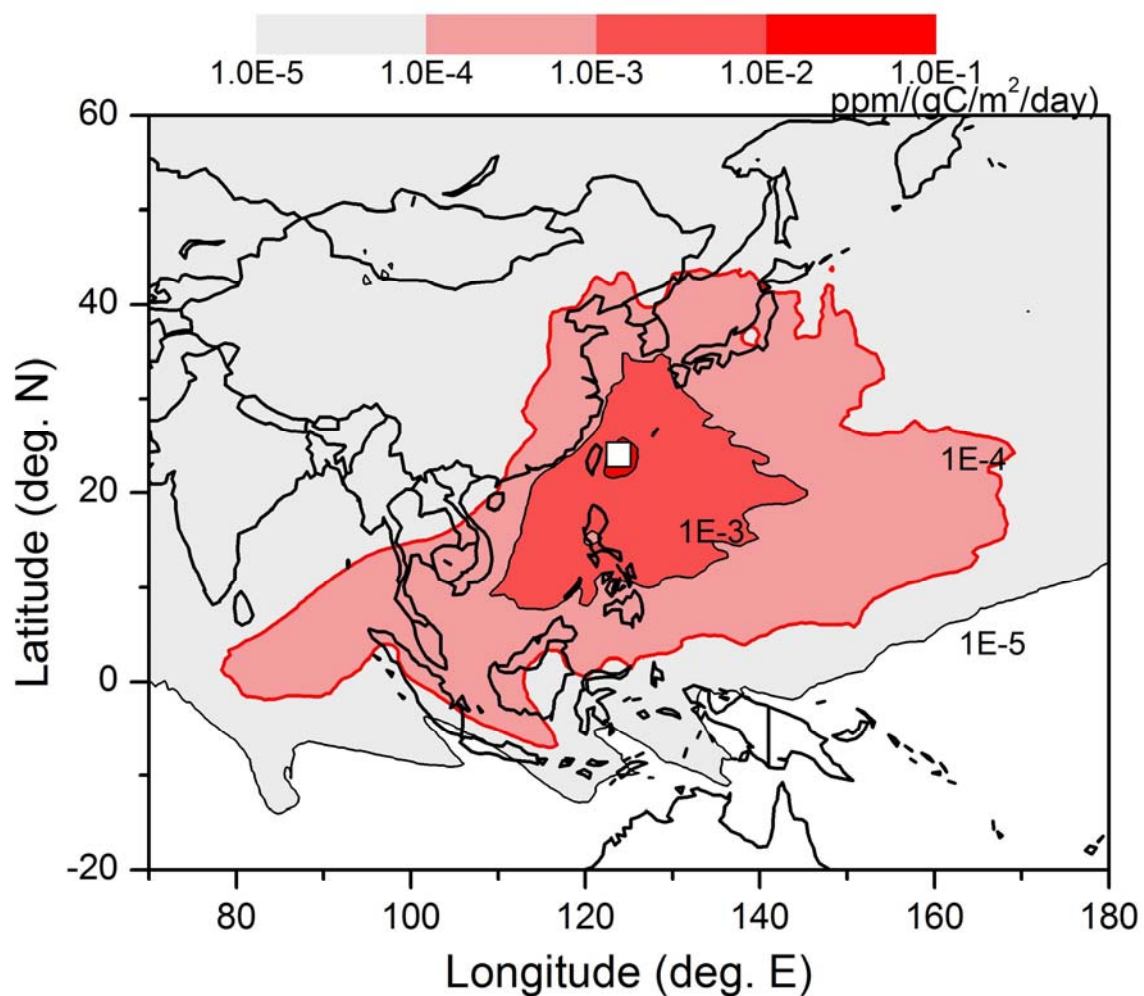
**Fig. S4.** Average footprint (ppm (gC m^{-2} day^{-1})$^{-1}$) for the measurements at HAT during the summer period (May to September). Meteorological data for 2006-2010 are used for the calculation. The location of HAT is indicated by the square.

Fig. S5. Comparison of the flux distributions of (a) fossil CO_2 from EDGAR v4.2, (b) fossil CO_2 from ODIAC, (c) fossil CO_2 from REAS v2.1, (d) CH_4 from Patra et al., (2009), (e) CH_4 from REAS v2.1, (f) CO from EDGAR v4.2, and (g) CO from REAS v2.1. The flux maps for 2007 are shown. Annual mean fluxes are depicted for CO_2 and CO, while monthly mean fluxes in January are depicted for CH_4.
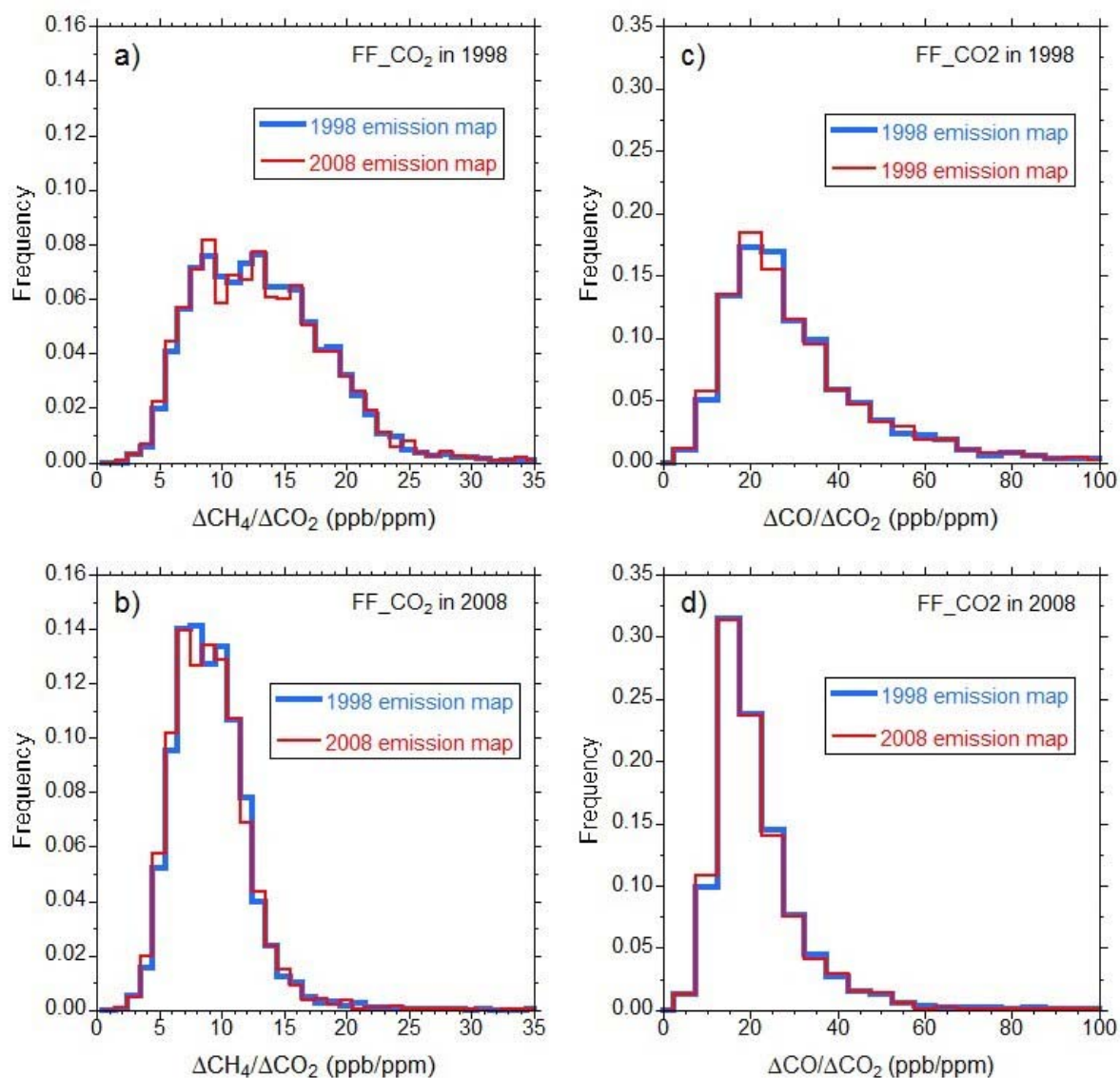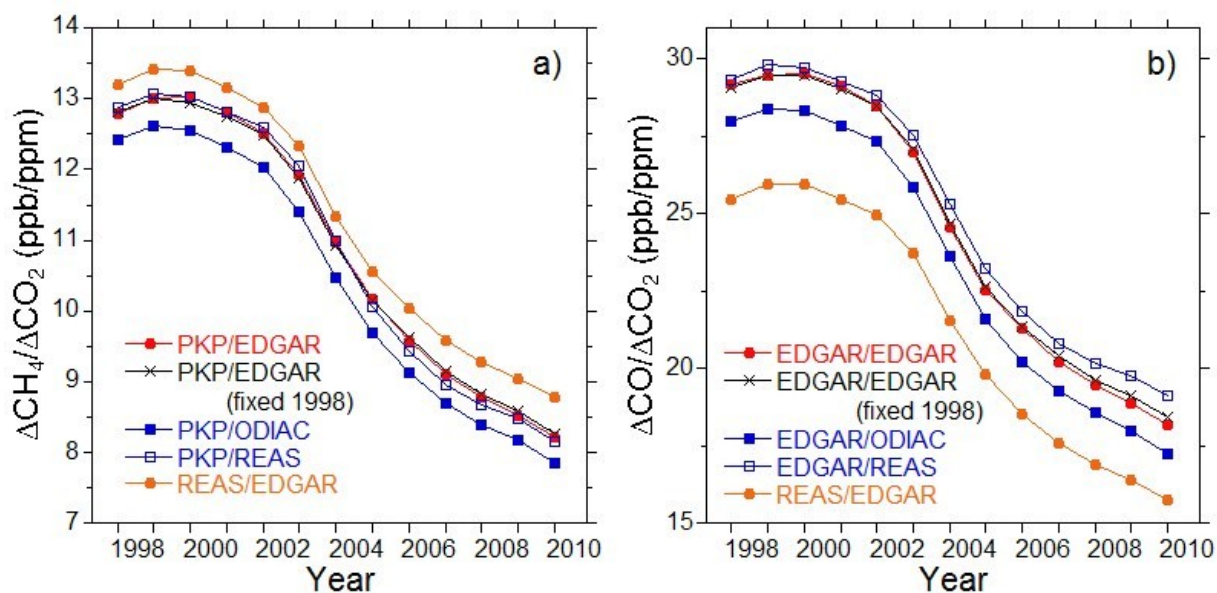
Fig. S6. Histograms of the simulated correlation slopes of (a, b) $\Delta CH_4/\Delta CO_2$ and (c, d) $\Delta CO/\Delta CO_2$ for fossil CO_2 emissions in (a, c) 1998 and (b, d) 2008. The correlation slopes all meet the selection criteria ($|R|>0.8$). The simulated results based on the fossil fuel-derived CO_2 emission maps for 1998 and 2008 are depicted as blue and red lines, respectively.

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