Supplementary material

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

**Fig. S2.** Scatter plot of the flask and in-situ CO measurements. The broken line represents the linear regression line.
Fig. S3. Temporal changes in the winter average correlation slopes of (a) $\Delta$CH$_4$/ΔCO$_2$, (b) $\Delta$CO/ΔCO$_2$, and (c) $\Delta$CO/ΔCH$_4$ 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.
**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) $\Delta \text{CH}_4/\Delta \text{CO}_2$ and (c, d) $\Delta \text{CO}/\Delta \text{CO}_2$ 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$_4$/ΔCO$_2$ and (b) $\Delta$CO/ΔCO$_2$ 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.