## Responses to Reviewer #1

Thank you very much for your comments and suggestions on our paper. They were very useful in revising the manuscript. The revised Table and Figures are attached at the end of this text.

**Reply to general comments:**

We accept the reviewer's criticism that the uncertainties of the $CH_4$ and CO emission estimates are possibly underestimated because the uncertainties of the spatial distribution of the estimated emission maps are not considered in the flux estimation in the original manuscript. To respond to the reviewer's question, we have conducted additional simulations using emission maps from ODIAC for $CO_2$ and REAS v2.1 for $CO_2$, $CH_4$, and CO. It should be noted that the national emissions from China, Japan, Korea, and Taiwan from the above emission maps are scaled to those from the emission maps used in the original simulation. Replacing these emission maps with the one of the emission maps used in the original simulation, we have obtained $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes and compared them with the original slopes. The results are plotted in Figure S7. For the ODIAC $CO_2$ emissions, the $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes are 4% and 5% lower than the original slopes, respectively. For the REAS v2.1 $CO_2$ emissions, the $\Delta CH_4/\Delta CO_2$ slopes agree with the original slopes while the $\Delta CO/\Delta CO_2$ slopes are 2% higher than the original slopes. Additionally, we have repeated the $CO_2$ simulation with the $CO_2$ emission map fixed to that of 1998 but scaled to the CDIAC inventories, and have found the differences of the $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes from the original slopes to be less than 1% even in 2010 (see Figure S7). On the other hand, for the REAS v2.1 $CH_4$ emissions, the $\Delta CH_4/\Delta CO_2$ slopes are on average 4% higher than the original slopes, while $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes based on the REAS v2.1 CO emissions are on average 13% lower than the original slopes.

Although the results of the above experimental simulations are very limited, we believe that the above differences in the correlation slopes constitute the lower limits of the uncertainties derived from the uncertain emission distributions: 5% for $CO_2$, 4% for $CH_4$, and 13% for CO. These uncertainties, together with the uncertainties associated with the choice of the criterion for the correlation coefficient (see the reply to the reviewer's comment for Page 22900, Line 8), are included in the uncertainties for the $CH_4$ and CO emission estimates in the revised manuscript. Consequently, we have added one paragraph to explain the additional emission maps to the end of Section 4.2 to read: "In order to examine the influence of different emission databases on the concentration fields in a transport model, we also use anthropogenic $CO_2$ emission distributions from the Open-Source Data Inventory of Anthropogenic $CO_2$ Emission (ODIAC) v3.0, which was originally developed by Oda and Maksyutov (2011) and modified by Maksyutov et al. (2013), and the recently revised REAS v2.1 (Kurokawa et al., 2013, http://www.nies.go.jp/REAS/), and $CH_4$ and CO emission distributions from REAS ver. 2.1. Although both ODIAC v3.0 and REAS v2.1 provide monthly emission maps, annually averaged emission maps for $CO_2$ and CO are used for the simulation comparisons. The $CO_2$ emissions from China, Japan, Korea, and Taiwan contained in the ODIAC and REAS v2.1 databases are scaled to the CDIAC national emission inventories, as was done for the EDGAR $CO_2$ emission maps. Similarly, the annually averaged CO emission maps for 2007 from REAS v2.1 are scaled to match the EDGAR national emissions for China, Japan, Korea, and Taiwan for 2007. The REAS v2.1 monthly $CH_4$ emission maps for 2007 are also scaled to match the monthly national emissions of Patra et al. (2009) for China, Japan, Korea, and Taiwan. The influence of these different emission maps on the correlation slopes are discussed in Section 5.2."

In addition, we have added one paragraph to the end of Section 5.2 to read as: "In order to examine the influence of different emission maps on the simulated $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes, we repeat the above calculation by replacing one emission map with the ODIAC or REAS emission map (see Section 4.2). The results are depicted in Fig. S7. Note that since the emission maps are scaled to match the national emissions from the East Asian countries, the differences in the correlation slopes should be attributed to the differences in the emission distributions. The ODIAC $CO_2$ emission maps systematically produce about 4% and 5% lower $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes, respectively, than the EDGAR $CO_2$ emission maps. The $\Delta CH_4/\Delta CO_2$ slopes based on the REAS and EDGAR emission maps agree with each other to within ±1% while the $\Delta CO/\Delta CO_2$ slopes based on the REAS emission maps are systematically higher by about 2% than those based on the EDGAR emission maps. Additionally, we simulate $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes with the 1998 EDGAR $CO_2$ emission map, although the national emissions had increased according to the CDIAC inventories. This caused the simulated slopes to gradually depart from those for the normal case but the differences are less than +1% even in 2010. The REAS $CH_4$ emission map systematically produces about 4% higher $\Delta CH_4/\Delta CO_2$ on average while the REAS CO emission map produces about 13% lower $\Delta CO/\Delta CO_2$ slopes. From these limited results, we take these differences in the correlation slopes as the uncertainties caused by the uncertain emission distributions: 5% for $CO_2$, 4% for $CH_4$, and 13% for CO." Furthermore, we have changed the 3rd sentence of the 3rd paragraph in Section 5.3 to "The error bars in the figure represent estimated uncertainties, which consist of uncertainties associated with the observed and simulated average correlation slopes, the uncertainties associated with the correlation coefficient criteria (2.5% for $\Delta CH_4/\Delta CO_2$ and 2.3% for $\Delta CO/\Delta CO_2$, see Section 3), the uncertainty of the fossil fuel-derived $CO_2$ emissions from China (15%, Gregg et al., 2008), and the uncertainty derived from the uncertain emission distributions (5% for $CO_2$, 4% for $CH_4$, and 13% for CO, see Section 5.2)".

Following these modifications, we have also changed the uncertainties of the $CH_4$ and CO emission estimates listed in Table 1 and redrawn the error bars for the plots of Fig. 8, Fig. 9, and Fig. 10.

As the reviewer suggested, there are large discrepancies between observations and model simulations as shown in Fig. 3. These discrepancies are of critical nature when emission estimates are based on the direct observation-model comparison of atmospheric variations. However, we estimate the regional emissions by comparing the correlation slopes between the observation and the model simulation under the assumption that the fossil fuel-derived $CO_2$ emissions are correct. Therefore, the model simulation doesn't necessarily reconstruct exactly the observations as long as the regional contributions are well evaluated. Since the 8-day backward footprints are used for the simulation in our study, the variations caused by the large-scale air mass mixing are not included in the simulation. Therefore, the lack of such mid-range variations (with a several-day period) might contribute partially to the apparently smaller variations of the simulation than the observation because the filtering procedure adopted in this study cannot effectively remove such mid-range variations. To mention this clearly, we have added the sentence "A lack of variations of several days due to the time scale of large-scale air mass mixing might partially contribute to the apparent smaller-than-observed simulated variations because the filtering procedure mentioned in

Section 3 cannot effectively remove such mid-range variations." after the 4th sentence of the 2nd paragraph in Section 4.3.

We wish to thank the reviewer for its useful information on the recently revised REAS inventory (v2.1). As mentioned above, we have conducted additional simulations by using REAS v2.1 emission maps, and compared the resulting correlation slopes with the original results. Generally, these different inventories, EDGAR v4.2, REAS v2.1 and ODIAC, provide similar variations by using the FLEXPART LPDM, but there are small differences in the amplitude and shape from peak to peak when examined closely. These differences, reflecting the differences in the geographical emission distributions of the inventories, could be used to constrain a much finer regional relative emission strength. But these discussions are beyond this study, and would be discussed in future work.

**Reply to specific comments:**

Page 22894, Line 10ff: We accept the reviewer's criticism concerning the changes in the decreasing $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes. As the reviewer mentioned, the several-year data are required to determine the trend, so it was inaccurate to state that the decreasing trends showed abrupt changes in 2004-2005. Accordingly, we have changed the relevant sentence "Although the ratios …" to "The observed ratios $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ both show an overall gradual decrease over the study period due to a recent rapid increase in fossil fuel consumption in China. We note, however, that the decreasing rates of $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ show gradual decrease and increase, respectively, during the entire observation periods used in this study."

We have edited the whole text for consistency: The 6th and 7th sentences of 2nd paragraph in Section 5.1 have been changed to "But the rate of decrease is gradually getting smaller for the $\Delta CH_4/\Delta CO_2$ slope, while it is gradually getting larger for the $\Delta CO/\Delta CO_2$ slope during the whole observation periods". We have deleted the 3rd sentence, "But the observed decreasing … the simulated decreasing rate.", of the 2nd paragraph in Section 5.2. The 1st sentence of 4th paragraph in Section 5.3 has been changed to "The estimated annual $CH_4$ emissions are relatively stable during the first several years but increase thereafter; for CO, it seems to increase and decrease during the early part and the later part of the observation period, respectively." The 3rd sentence of the 1st paragraph in Conclusion has been changed to "However, there are differences in the decreasing trends between $\Delta CH_4/\Delta CO_2$ and $\Delta CO/\Delta CO_2$ slopes; the rate of decrease for the $\Delta CH_4/\Delta CO_2$ slope is decreasing while that for the $\Delta CO/\Delta CO_2$ slope is increasing during the study period".

Page 22894, Line 12-14: Here, we simply intended to mention that the short-term variations at HAT are attributed to the regional emissions from East Asia. Therefore, we have removed the misleading word "variations" from the relevant sentences (line 14). To examine the spatial distribution of the emissions from East Asia, we compared the results based on the EDGAR v4.2 emission maps with those based on the ODIAC and REAS v2.1 emission maps as is mentioned in the reply to the general comments.

Page 22894, Line 25: In response to the reviewer's comment, we have changed "the second most important greenhouse gas" to "the second most important anthropogenic greenhouse gas".

Page 22898, Line 1: To explain "cleanup air", we have added the sentence "The cleanup air is made from natural air by passing it through a cold trap to reduce water vapor, a heated Pt catalyst to oxidize CO and hydrocarbons into $CO_2$ and water vapor, and a Molecular Sieve 5A column to remove $CO_2$ and residual water vapor." after the 3rd sentence of the relevant paragraph (the 3rd paragraph in Section 2).

Page 22899, Line 24: Yes, we tested the cut-off frequencies of 2.3 and 9.2 cycles $yr^{-1}$ and found there were no significant differences in the resulted correlation slopes. To mention this clearly, we have added "We have also calculated the best-fit smooth curves by using cut-off frequencies of 2.3 and 9.2 cycles $yr^{-1}$ and have found that the average $\Delta CH_4/\Delta CO_2$, $\Delta CO/\Delta CO_2$, and $\Delta CO/\Delta CH_4$ slopes (see Section 5.1) for the 3 different cut-off frequencies agree to within 1%." after the last sentence of the 1st paragraph in Section 3.

Page 22900, Line 8: Unfortunately, we arbitrarily determine the threshold of $|R|>0.8$. So, we examined the influence in the choice of R (0.7 and 0.9) on the average correlation slopes, and found that the resulting correlation slopes depend only slightly on the choice of R, and the differences are relatively small. Consequently, we added "Although we arbitrarily choose the correlation coefficient of 0.8 as a rough criterion for selecting significant correlation slopes, it should be noted that the average correlation slopes of $\Delta CH_4/\Delta CO_2$, $\Delta CO/\Delta CO_2$, and $\Delta CO/\Delta CH_4$, as discussed in Section 5.1, do depend slightly on the value of the criterion. These average slopes for $|R|<0.7$, $|R|<0.8$ and $|R|<0.9$ are plotted in Fig. S3. The root mean squares of the overall differences in the average regression slopes for $|R|<0.7$ and $|R|<0.9$ from those for $|R|<0.8$ are 2.5%, 2.3%, and 3.4% for $\Delta CH_4/\Delta CO_2$, $\Delta CO/\Delta CO_2$, and $\Delta CO/\Delta CH_4$, respectively." after the end of the 2nd paragraph in Section 3.
In addition, we believe that both increasing and decreasing changes in the short-term variations are explained by changes in the contributions from the emissions and clean or background air. In fact, in the FLEXPART simulations, the decreasing changes are expressed as decreases in the contributions from the regional emissions. Therefore, the decreasing changes in the short-term variations observed at HAT also reflect the regional emissions.

Page 22900, Line 21: As Reviewer #2 pointed out, the seasonal change in the $\Delta CO/\Delta CH_4$ slope is mainly attributed to the seasonality in the air mass transport. To explain the air mass transport during summer, we have added an average footprint for the measurements at HAT during the summer period (May to September) in Figure S4. So, the effect of the seasonally changing CO emissions is relatively small. Consequently, we have changed the 4th and 5th sentences of the relevant paragraph (the 3rd paragraph in Section 3) to "We also note that the seasonal variation in the average $\Delta CO/\Delta CH_4$ slope may be attributable mainly to the seasonality in the air mass transport. During the summer, air masses arriving at HAT are predominantly transported from the Pacific region and the contributions of the South East Asian emissions show a relative increase. (Figure S4 shows the average footprint, which is discussed in Section 4.1, during the summer period (May to September)). Thus the average $\Delta CO/\Delta CH_4$ slope is low in summer because the $CO/CH_4$ emission ratios for the South East Asian countries are lower than those for China, Japan and Korea (e.g. Kurokawa et al., 2013). However, it is possible that the seasonality in the emissions from East Asia, the maximum $CH_4$ emissions in summer (Yan et al.,, 2003) and the maximum CO emissions in winter (Streets et al., 2003; Zhang et al., 2009), and the significantly faster CO reaction with

OH in summer could partially contribute to the seasonality in the average $\Delta CO/\Delta CH_4$ slope."

Page 22902, Line 20: "atmopsheric" has been changed to "atmospheric".

Page 22902, Line 22: Patra et al. (2009) confirmed that the simulation based on their $CH_4$ monthly emission maps and an atmospheric transport model (ACTM) well reproduces the observed atmospheric $CH_4$ variations. On the other hand, the EDGAR v4.2 database only provides annual emission maps of the anthropogenic $CH_4$ sources, so we need more work to prepare monthly $CH_4$ emission maps that would include natural sources. Therefore, we use the $CH_4$ emission maps of Patra et al. (2009) to save the effort of the preparation. To mention this clearly, we have added the following 2 sentences "Simulated atmospheric $CH_4$ mixing ratios in a transport model using these $CH_4$ emission maps generally agree with the observed global distribution and seasonality (Patra et al., 2009). We use the above $CH_4$ emission maps due to their technical ease-of-use and suitability, and not the $CH_4$ emission maps from the EDGAR v4.2 database because it only provides the annual flux maps." after 1st sentence of the relevant paragraph (the 3rd paragraph in Section 4.2).

Page 22903, Line 14: Please see the reply to the general comments.

Page 22903, Line 24: In response to the reviewer's question, we have compared the spatial distributions of the $CO_2$, $CH_4$, and CO flux maps prepared in this study (EDGAR v4.2, REAS v2.1, ODIAC, and Patra et al., 2009). The emission maps are depicted in Figure S5. As shown in the figures, the strong emissions are primarily confined to the land areas in the southern part of North China, East China, the Korean Peninsula, and Japan in either flux maps. Therefore, we think that the $CO_2$, $CH_4$, and CO fluxes have roughly similar spatial distributions within EFA. To state this clearly, we have added the sentence "To examine the robustness of this assumption, we compare the flux maps prepared in Section 4.2 (see Figure S5). Since the strong emissions, primarily confined to the land areas, are generally distributed in the southern part of North China, East China, the Korean Peninsula, and Japan, we are confident that the spatial distributions of the $CO_2$, $CH_4$, and CO fluxes within EFA are roughly similar to each other." to the end of the relevant paragraph (the 2nd paragraph in Section 4.3).

Page 22904, Line 14: In response to the review's comment, we have changed our explanations, which gave a false impression that the changes in the emission trend occurred in 2004/2005, in the revised manuscript. Therefore, we have not changed Fig. 6.

Page 22905, Line 16: In the simulation, we use repeatedly the monthly $CH_4$ emission maps for 2007 while the fossil fuel $CO_2$ emissions are changed in accordance with the CDIAC emission inventories. Therefore, the discrepancy in the temporal change in the decreasing trend of the $\Delta CH_4/\Delta CO_2$ slope may be attributed to the change in the $CH_4$ emissions from EFA. To mention this clearly, we have changed the relevant sentence of 1st paragraph in Section 5.2 to "However, the temporal decrease in the decreasing rate is not well simulated for the latter period. This discrepancy may be explained by the previously noted increase in the $CH_4$ emissions from EFA, since the model $CH_4$ field is driven by the fixed 2007 $CH_4$ monthly emission maps while the $CO_2$ field is driven by the temporal varying CDIAC emission inventories."

Page 22908, Line 4: In response to the reviewer's comment, we have added the comparisons of our $CH_4$ estimate with the results from REAS v2.1, the TranCom-$CH_4$ experiment (Patra et al., 2011), and the recent inversion study using satellite and ground observations (Bergamaschi et al., 2013). Consequently, Section 5.4.1 has been changed to "Bottom-up estimates of the $CH_4$ emission from Chinese anthropogenic sources without the rice fields taken from the inventory databases EDGAR v4.2, REAS v1.1 (Ohara et al., 2007, http://www.jamstec.go.jp/frcgc/research/p3/emission.htm) and REAS v2.1 are plotted for comparison with our results in Fig. 9. The EDGAR v4.2 emission estimates show good agreement with our estimates for the period 1998 to 2002. However, after 2002 the EDGAR v4.2 data show a much faster increase of 3.1 ± 0.1 $TgCH_4\ yr^{-2}$ (2002-2008), which is about 3 times larger than our estimates of 1.1 ± 0.2 $TgCH_4\ yr^{-2}$ (2002-2010). The REAS v2.1 estimates, being higher than our estimates, also show a faster increase of 3.6 ± 0.2 $TgCH_4\ yr^{-2}$ (2000-2008). About 70% and 90% of the increases in the Chinese emissions in the EDGAR v4.2 and REAS v2.1 estimates, respectively, are attributed to the emissions related to coal mining (fugitive emissions from solid fuels), and occurs mostly within EFA. Note that the REAS v1.1 estimates are lower than our estimates and the differences from the REAS v2.1 estimate for 2000 are attributed to the fugitive emissions from fossil fuels (73%) and the emissions from land disposal of solid waste (24%).

The possibility that the $CH_4$ emissions in the EDGAR v4.2 inventory are overestimated was also suggested by the following model studies. In a chemistry-transport model intercomparison experiment of $CH_4$ (TransCom-$CH_4$), the forward simulations of atmospheric $CH_4$ were conducted using several transport models and various sets of surface $CH_4$ emission scenarios (Patra et al., 2011). The forward $CH_4$ simulation based on the EDGAR v4.0 emissions, which are almost same as the EDGAR v4.2 emissions, shows a significantly faster growth rate during 2003-2007 than the observations. The Chinese emission increase contributes nearly 40% to the global $CH_4$ emission increase in the EDGAR inventory. Recently, Bergamaschi et al. (2013) estimated global $CH_4$ emissions during the 2000s based on an inverse modeling constrained by atmospheric $CH_4$ data from the global air sampling network and satellite sensor. The inversion result shows a significant increase in the anthropogenic $CH_4$ emissions from China but a smaller increase than that indicated by the EDGAR inventory. The increasing rate of 1.1 ± 0.3 $TgCH_4\ yr^{-2}$ estimated by Bergamaschi et al. (2013) for the period of 2000-2010 is in excellent agreement with our estimation. Therefore, we suspect that the EDGAR v4.2 and REAS v2.1 inventories are overestimating the recent increase in the $CH_4$ emissions related to the coal mining."

Page 22908, Line 10: "EDGARR" has been changed to "EDGAR".

Page 22909, Line 17: As the reviewer mentioned, we meant to say "underestimate" here in the original manuscript. However, we have accepted the comment of Reviewer #2 that the discrepancy of CO emission estimates between EDGAR and other studies could be attributed to the secondary CO production (oxidation products from VOCs), of which values are included in the top-down estimates but not explicitly included in EDGAR inventories. Therefore, we have changed the 2nd paragraph of Section 5.4.2 to "The top-down estimates including ours reflect not only the primary CO emissions but also the secondary CO production from the oxidation of NMVOC. However, we consider the contribution of the $CH_4$ oxidation to the top-down estimates of CO emissions based on the atmospheric observations in the downwind regions from China to be negligible because of the much longer life time of atmospheric $CH_4$ (about 10 yr, e.g. Patra et al., 2011) compared to its transit time. It is to be noted that the EDGAR database reports only the primary CO emissions. Duncan et al. (2007) estimated that the oxidation of NMVOC contributes nearly 50% of the total primary CO emissions to the global CO emission. If this ratio is valid and can be applied to the EDGAR estimate for China, then the resulting net CO emissions with both primary and secondary sources can be applied to our top-down estimates. In addition, our winter emission estimates would of course be biased if the CO emission has a noticeable seasonality. For example, using monthly data for power generation and industry, as well as residential energy consumption, Zhang et al. (2009) developed a dataset of monthly CO emissions from China. The result shows a significant seasonality, with 17% larger average monthly emission for our 5-month winter than for an entire year. If in fact there is a strong seasonal variation in the CO emission, then our winter estimate needs to be reduced by 17%, which also brings our estimate close to the EDGAR v4.2 estimate. Above discussion points to the importance of correct evaluation of the secondary CO emissions when comparing top-down and bottom-up emission estimates. Note that the REAS v2.1 estimates, in which the secondary CO emissions are not explicitly included, agree well with the top-down estimate. Kurokawa et al. (2013) attribute the differences in the CO emissions between REAS v2.1 and EDGAR v4.2 to the emission factors used in the estimations; the emission factors for REAS v2.1 might implicitly include the secondary productions."

Figure 1: Reviewer #2 recommend remove USA line from Fig. 1a to be consistent. Therefore, we have removed USA line from Fig. 1.

Figure 3: We understand the reviewer's comment that the observed and simulated variations should be plotted in the same scales for ease of the direct comparison. However, plotting the simulated variations in the same scales to the observations makes it hard to see the variations because the simulated variations are significantly underestimated. Therefore, we have redrawn Fig. 3b with the ranges of the y-axis set to half of the corresponding y-axis of Fig. 3a. To mention this clearly, we have added the sentence "The range of the y-axis for the simulation plot for each chemical species is half of that for the corresponding observation plot." in the figure caption.

Figure 7c: In response to the reviewer's comment, we have changed the wordings "Sim." and "Sim. (estimated emissions)" to "Sim. (initial emissions)" and "Sim. (corrected emissions)", respectively.

Figure 9: "READ" has been changed to "REAS"

**Revised Table and Figures are shown below:**

**Table 1**. Summary of the estimated $CH_4$ and CO emissions from China[a]

| Year | Fossil fuel $CO_2$[b] | $CH_4$[c, d] | CO[d] |
|---|---|---|---|
| 1997/1998 | 0.93 | 38.4 ± 6.8 | |
| 1998/1999 | 0.91 | 40.5 ± 7.0 | 134 ± 32 |
| 1999/2000 | 0.92 | 37.3 ± 6.6 | 149 ± 34 |
| 2000/2001 | 0.94 | 39.4 ± 6.8 | 140 ± 35 |
| 2001/2002 | 0.98 | 39.1 ± 6.9 | 153 ± 36 |
| 2002/2003 | 1.12 | 37.3 ± 6.4 | 158 ± 36 |
| 2003/2004 | 1.34 | 40.7 ± 7.2 | 179 ± 42 |
| 2004/2005 | 1.51 | 39.4 ± 6.7 | 182 ± 42 |
| 2005/2006 | 1.66 | 44.0 ± 7.4 | 176 ± 40 |
| 2006/2007 | 1.80 | 43.3 ± 7.3 | 169 ± 38 |
| 2007/2008 | 1.88 | 44.7 ± 7.6 | 181 ± 41 |
| 2008/2009 | 1.98 | 46.5 ± 7.9 | 150 ± 33 |
| 2009/2010 | 2.14 | 45.8 ± 7.9 | 159 ± 36 |

[a]Values for $CO_2$ are given in PgC $yr^{-1}$, for $CH_4$ in $TgCH_4$ $yr^{-1}$, and for CO in TgCO $yr^{-1}$.
[b]Fossil $CO_2$ emissions are taken from the CDIAC database. Each value is the average of the emissions for the consecutive two years described in the first column. The uncertainty is assumed to be 15%, which is the lower limit of the estimation of Gregg et al. (2008).
[c]Values represent the emissions from non-seasonal $CH_4$ sources (see text).
[d]Uncertainties are calculated from the uncertainties of the fossil fuel-derived $CO_2$ emissions in China, of the observed correlation slopes including the influence of the correlation coefficient criteria selection, and of the simulated correlation slopes including the influence of the uncertain emission distributions used in the simulation (see text).

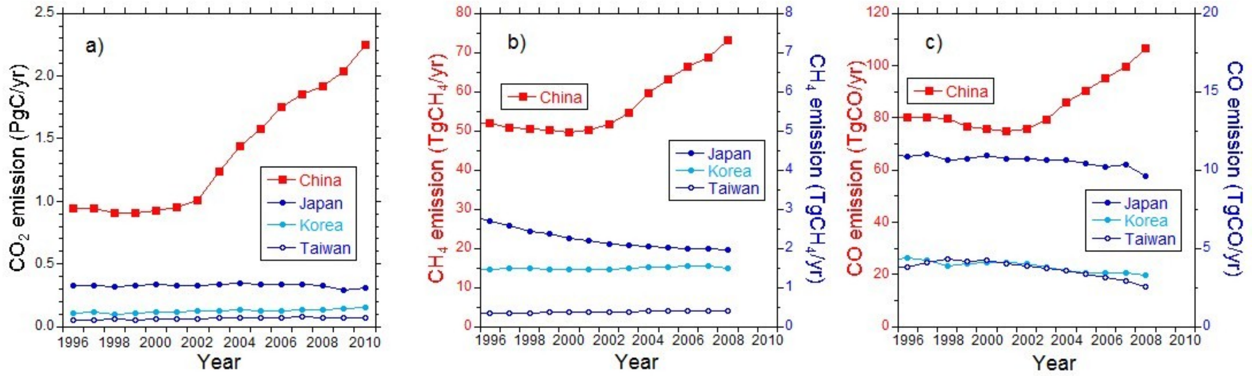


**Fig. 1.** Temporal changes in the estimated emissions of (a) fossil fuel-derived $CO_2$, (b) $CH_4$, and (c) CO from China, Japan, Korea, and Taiwan. The $CO_2$ emissions are taken from the CDIAC database. $CH_4$ and CO emissions are taken from EDGAR v4.2. $CH_4$ and CO emissions from Japan, Korea and Taiwan are plotted against the right-Y axis.

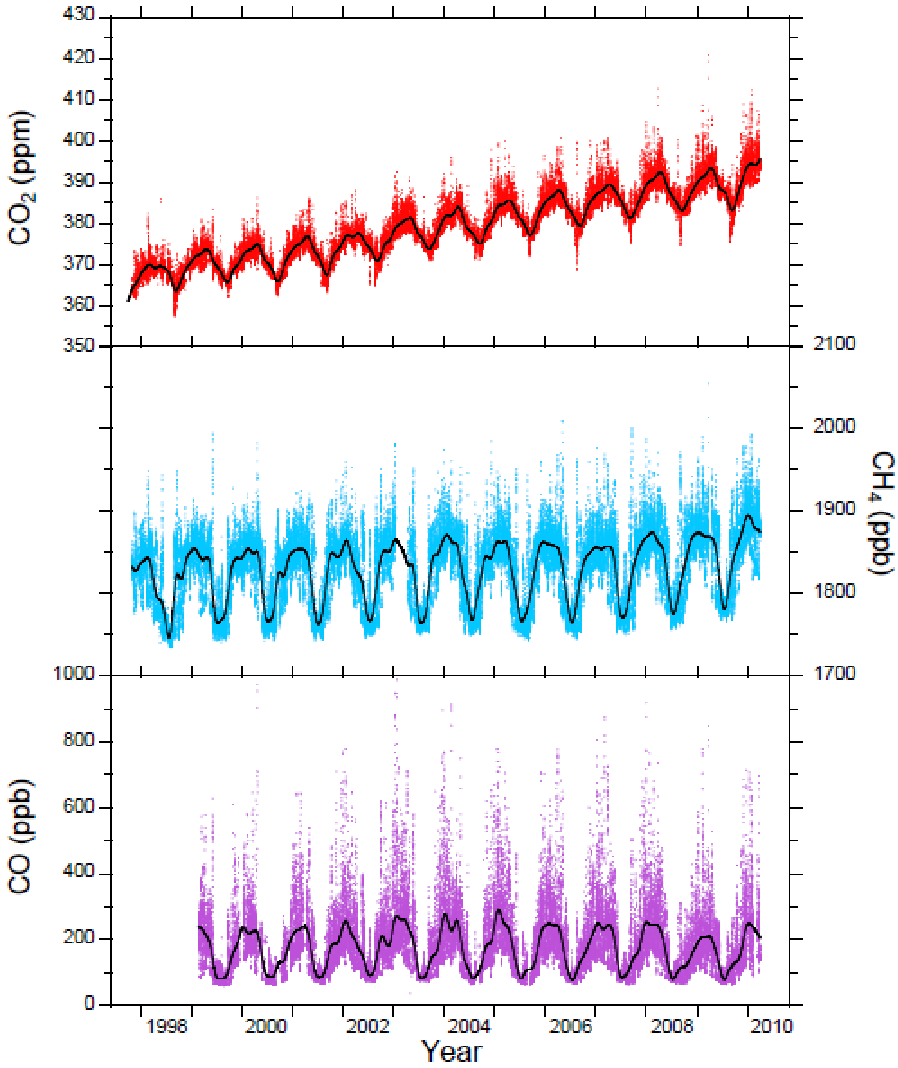


**Fig. 2.** Time series of atmospheric (top) $CO_2$, (middle) $CH_4$, and (bottom) CO mixing ratios observed at HAT. Each dot represents hourly average. Black lines represent the smooth curve fits to the data.

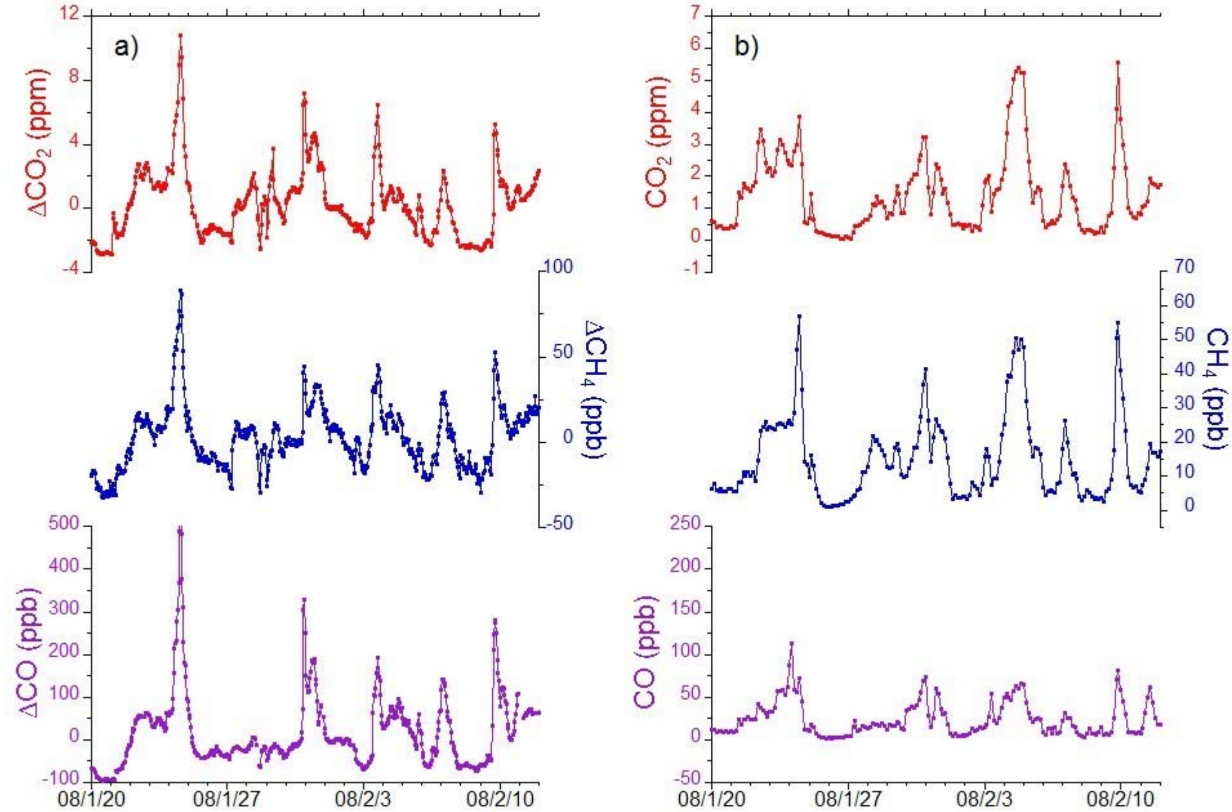


**Fig. 3.** Synoptic scale variations in hourly (top) $CO_2$, (middle) $CH_4$, and (bottom) CO based on (a) the observation and (b) the model simulation for the period from January 20 to February 12, 2008. The range of the y-axis for the simulation plot for each chemical species is half of that for the corresponding observation plot.

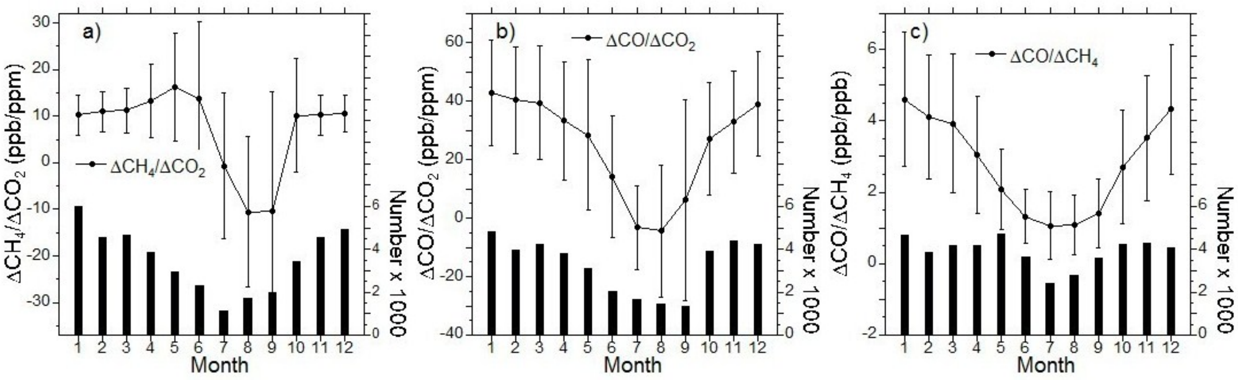


**Fig. 4.** Average seasonal variation of (a) $\Delta CH_4/\Delta CO_2$, (b) $\Delta CO/\Delta CO_2$, and (c) $\Delta CO/\Delta CH_4$ slopes observed at HAT. The error bars represent the standard deviations from the monthly averages. The vertical bars represent the data number.

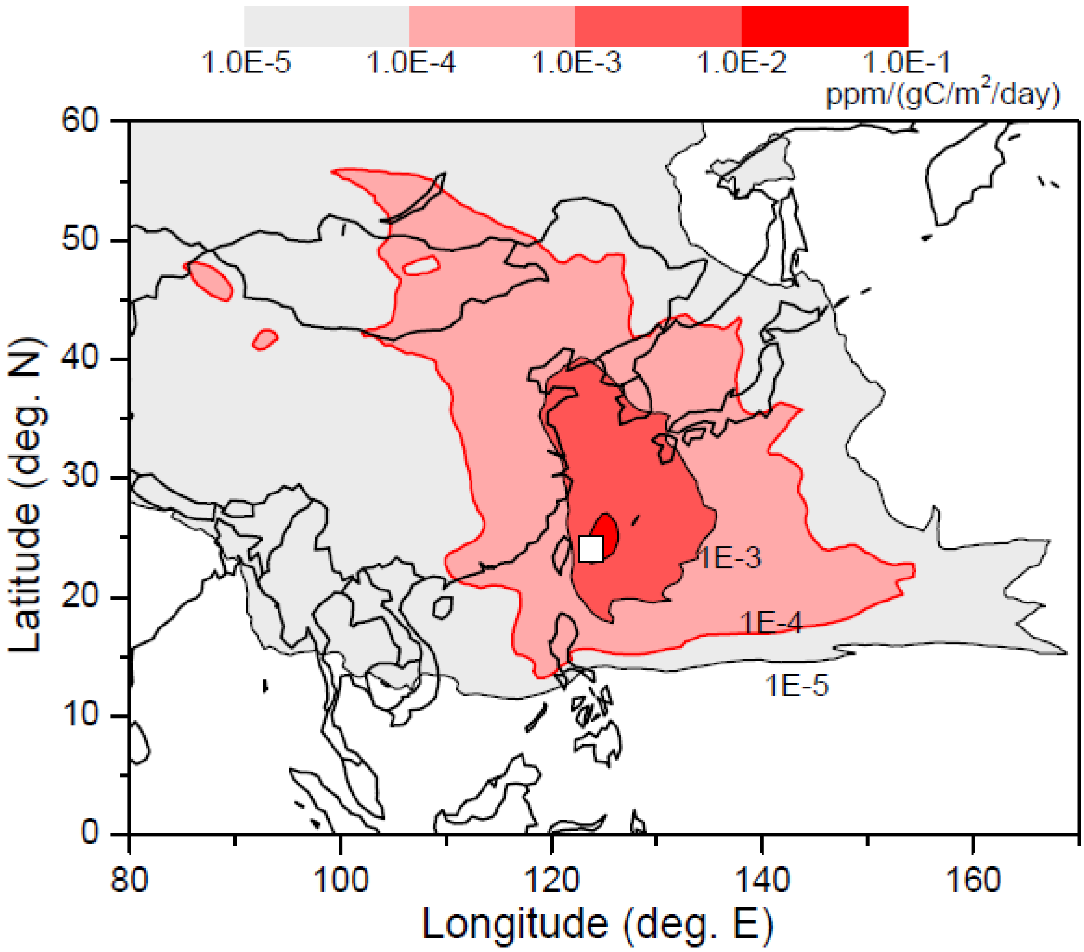


**Fig. 5.** Average footprint (ppm (gC m$^{-2}$ day$^{-1}$)$^{-1}$) for the measurements at HAT during the winter period (November to March). Meteorological data for 2006-2010 are used for the calculation. The location of HAT is indicated by the square. The area surrounded by the red thick contour lines of $1\times10^{-4}$ ppm (gC m$^{-2}$ day$^{-1}$)$^{-1}$ is defined as an effective footprint area (EFA).

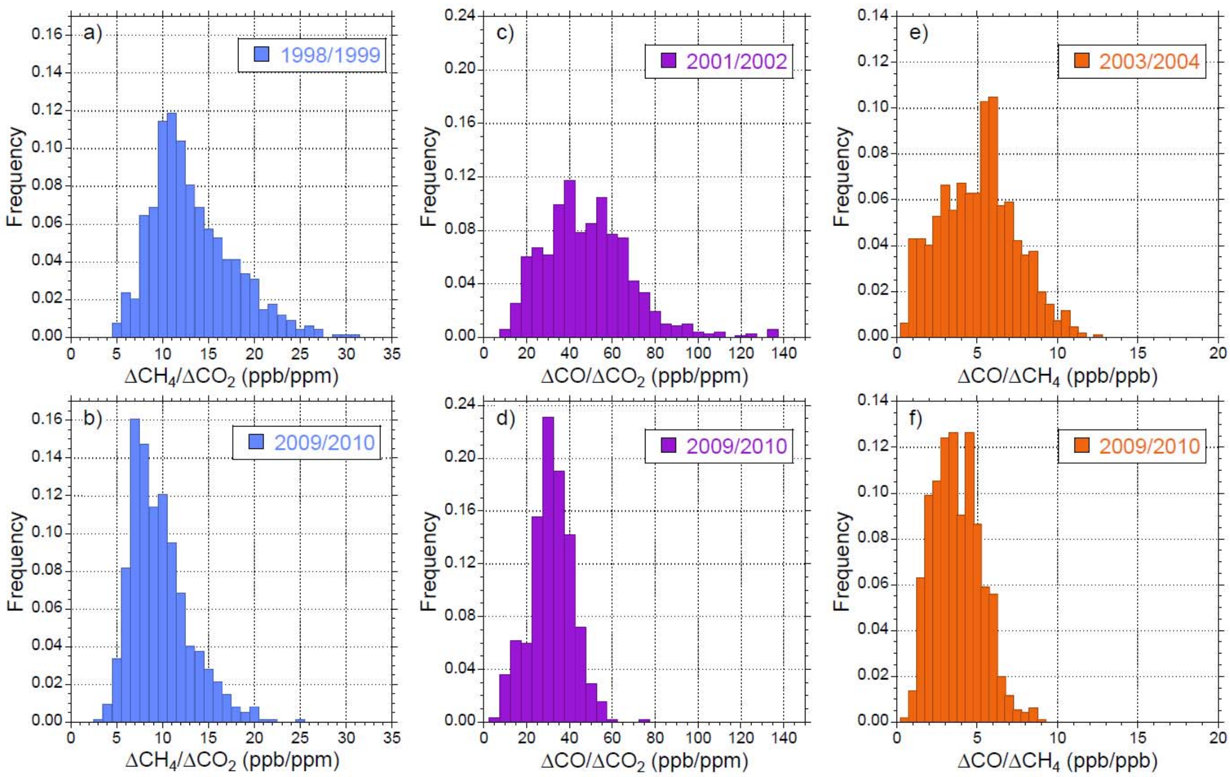


**Fig. 6.** Histograms of the correlation slopes of (a, b) $\Delta CH_4/\Delta CO_2$, (c, d) $\Delta CO/\Delta CO_2$, and (e, f) $\Delta CO/\Delta CH_4$ for the selected two periods. The correlation slopes all meet the selection criteria (see text).

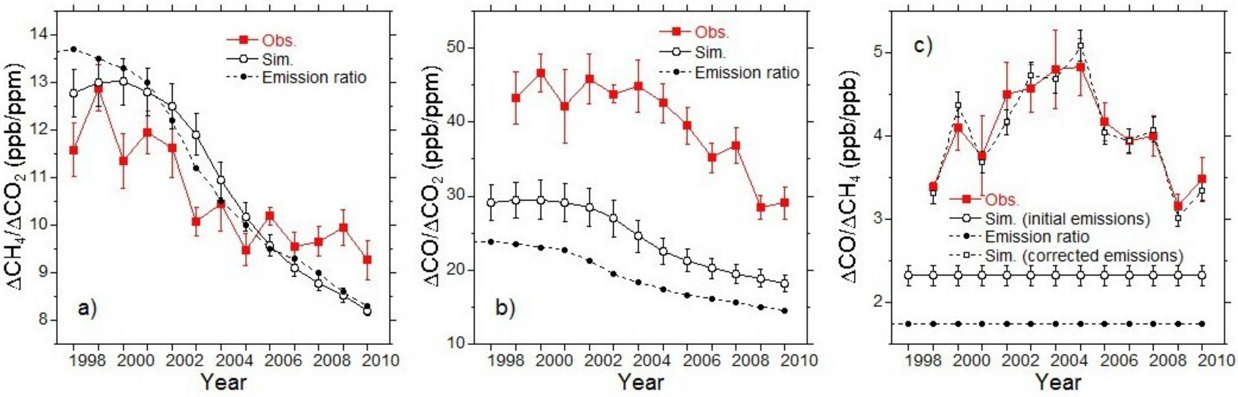


**Fig. 7.** Temporal changes in the winter average correlation slopes of (a) $\Delta CH_4/\Delta CO_2$, (b) $\Delta CO/\Delta CO_2$, and (c) $\Delta CO/\Delta CH_4$. The red closed squares represent the observation and the open circles represent the simulation. The error bars represent the standard errors. The ratios of the emissions within EFA are also depicted as closed circles. The black open squares in **Fig. 7c** represent the $\Delta CO/\Delta CH_4$ slopes based on the optimized $CH_4$ and CO emissions from China within EFA (see text).

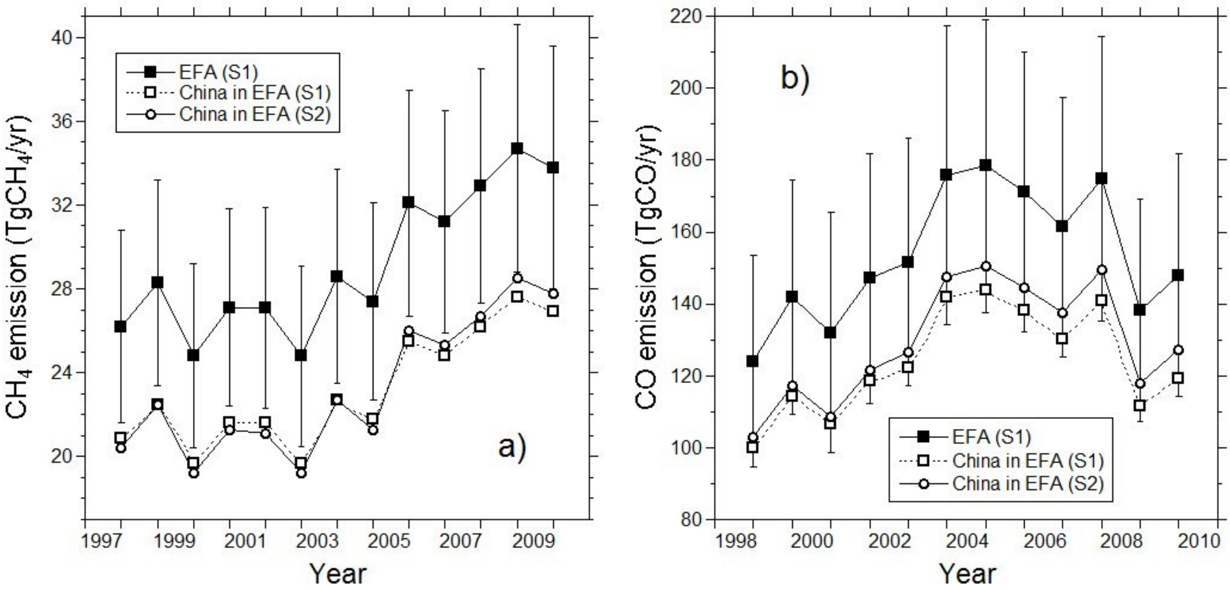


**Fig. 8.** Temporal changes in the estimated (a) $CH_4$ and (b) CO emissions from EFA. The emissions from EFA for S1 are depicted by closed squares with uncertainties. The emissions from China in EFA are depicted for S1 by open squares and for S2 by open circles (see text).

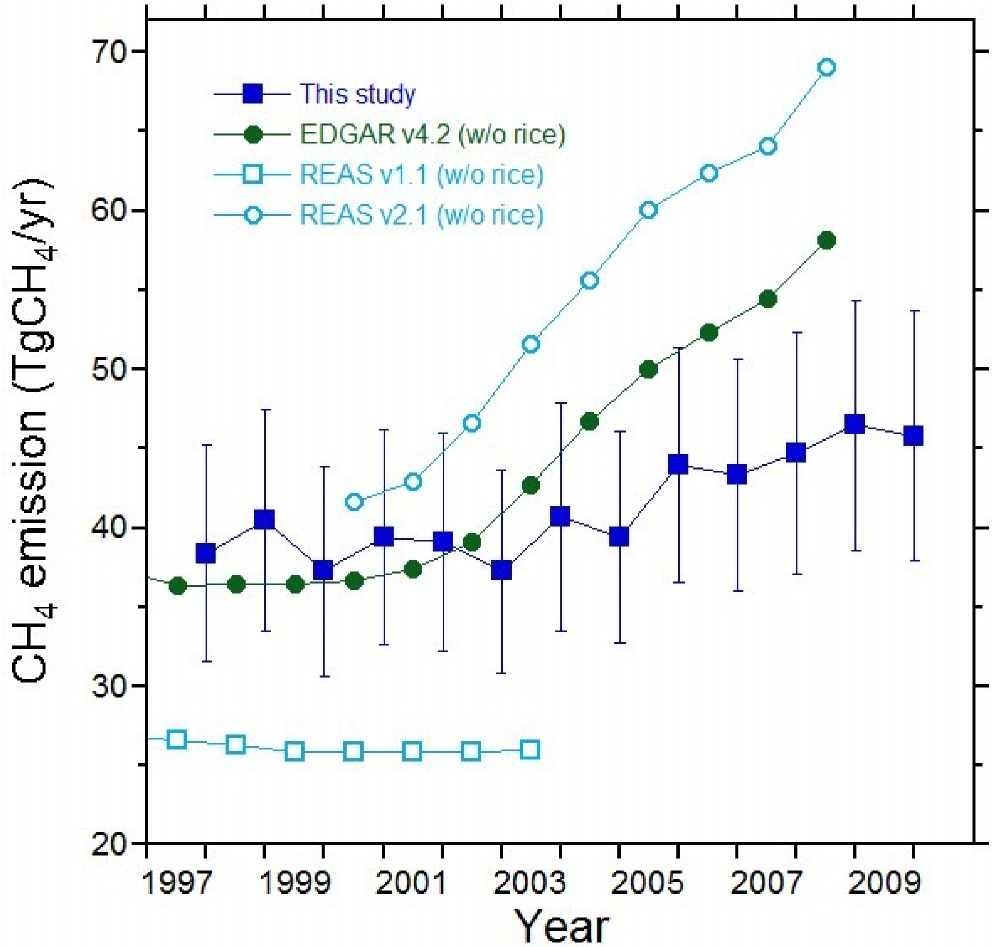


**Fig. 9.** Comparison of estimated non-seasonal $CH_4$ emissions from China. The values are expressed as annual emissions. Closed blue squares are the estimated emissions of this study. Green circles, light blue squares, and light blue circles represent the $CH_4$ emissions from anthropogenic sources (excluding rice fields) in China based on the emission inventories from EDGAR v4.2 (http://edgar.jrc.ec.europa.eu/), REAS v1.1 (Ohara et al., 2007) and REAS v2.1 (Kurokawa et al., 2013), respectively.

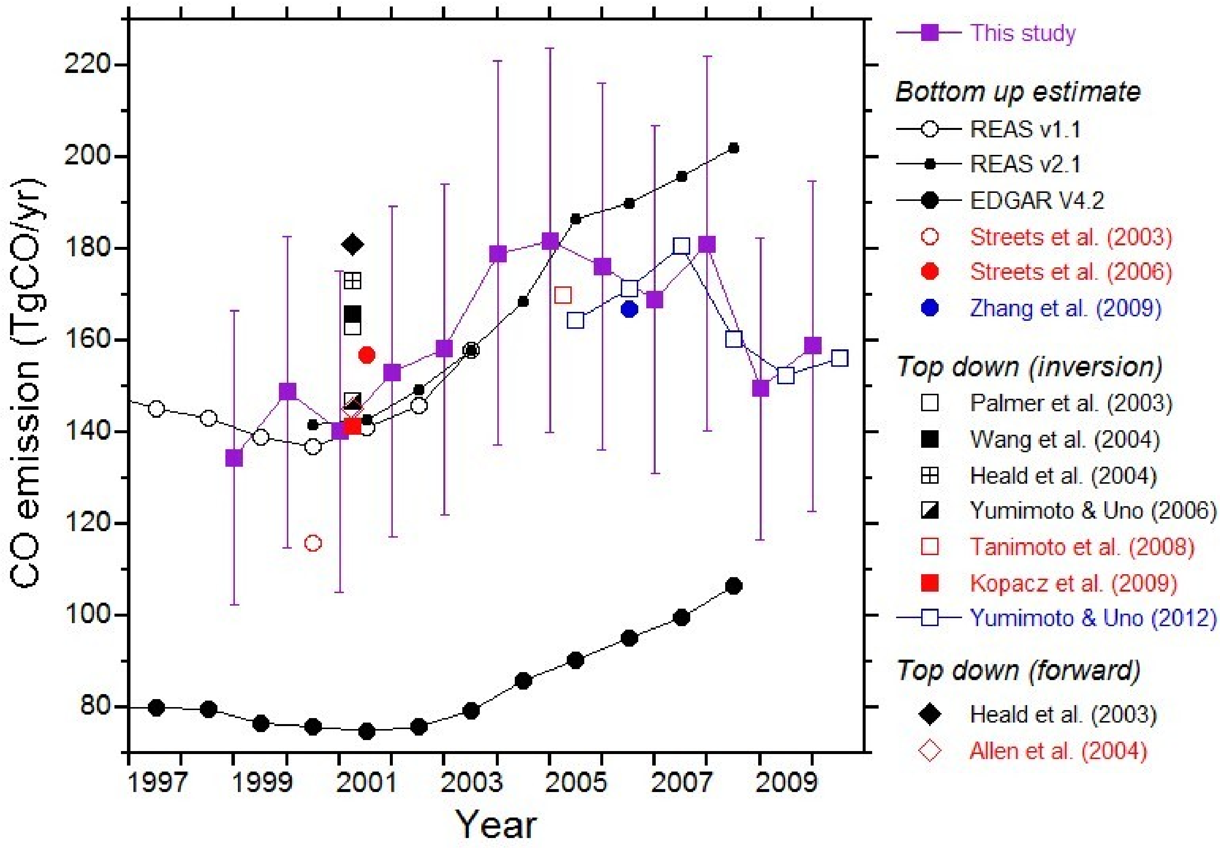


**Fig. 10.** Comparison of estimated CO emissions from China. The values are expressed as annual emissions. Closed blue squares are the estimated emissions of this study. Circles, squares, and diamonds represent the bottom up estimates, top down (inversion), and top down (forward) estimates, respectively.

## Supplementary material

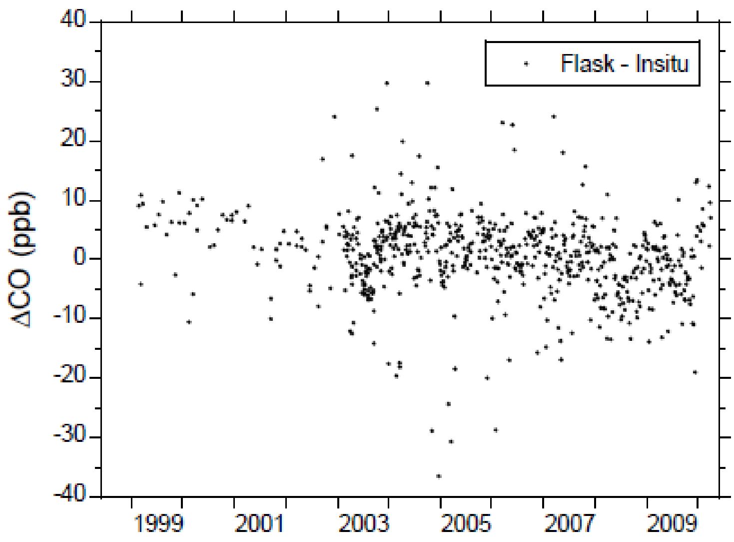


**Fig. S1.** Time series of the differences between flask and in-situ measurements of the atmospheric CO mixing ratios at HAT during the period from 1999 to 2010.

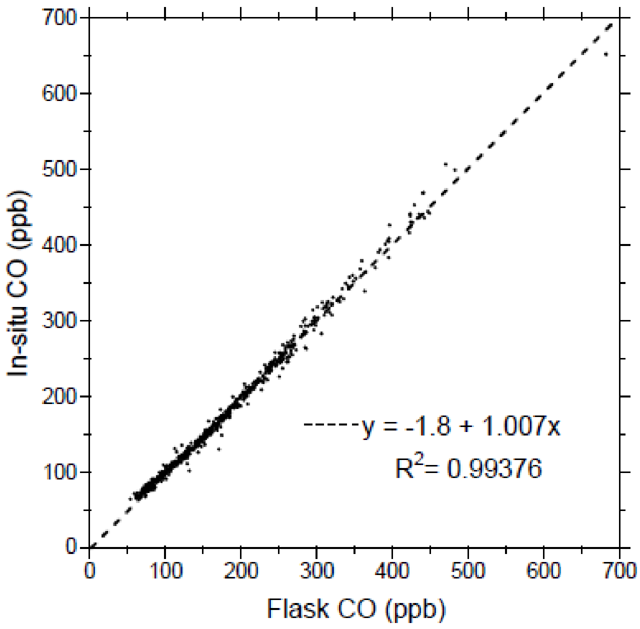


**Fig. S2.** Scatter plot of the flask and in-situ CO measurements. The broken line represents the linear regression line.

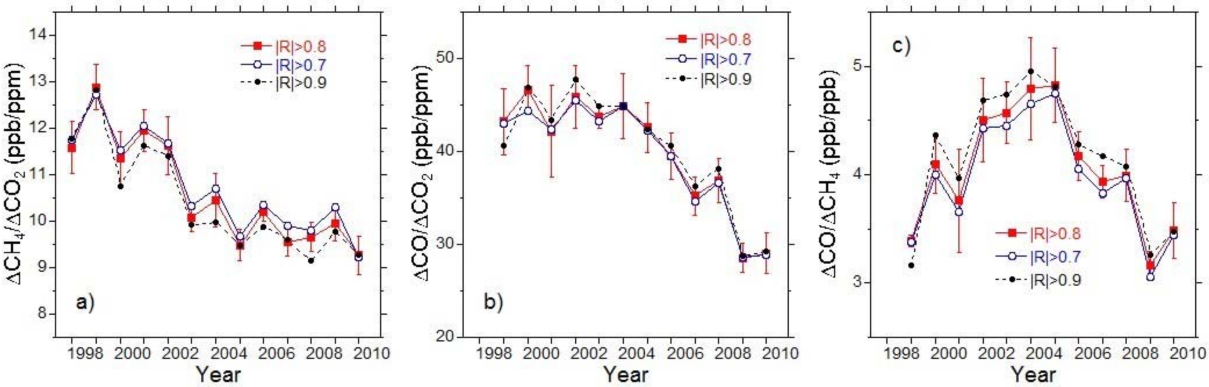


**Fig. S3.** Temporal changes in the winter average correlation slopes of (a) $\Delta CH_4/\Delta CO_2$, (b) $\Delta CO/\Delta CO_2$, and (c) $\Delta CO/\Delta CH_4$ for 3 correlation coefficients that are used in the selection criteria (see text). The red squares represent the correlation coefficient of 0.8, black open circle 0.7, and black closed circle 0.9.

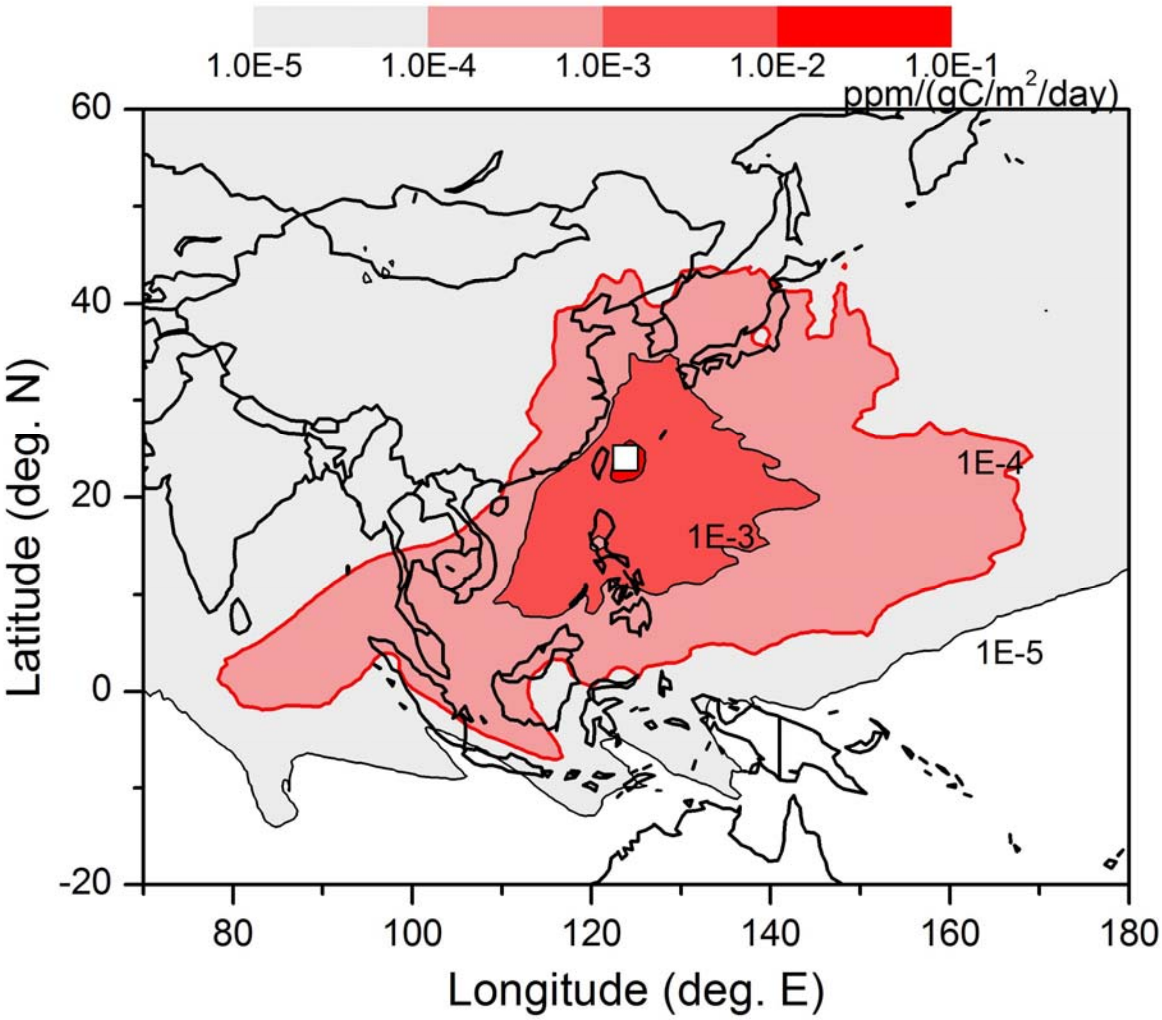


**Fig. S4.** Average footprint (ppm (gC m$^{-2}$ day$^{-1}$)$^{-1}$) for the measurements at HAT during the summer period (May to September). Meteorological data for 2006-2010 are used for the calculation. The location of HAT is indicated by the square.

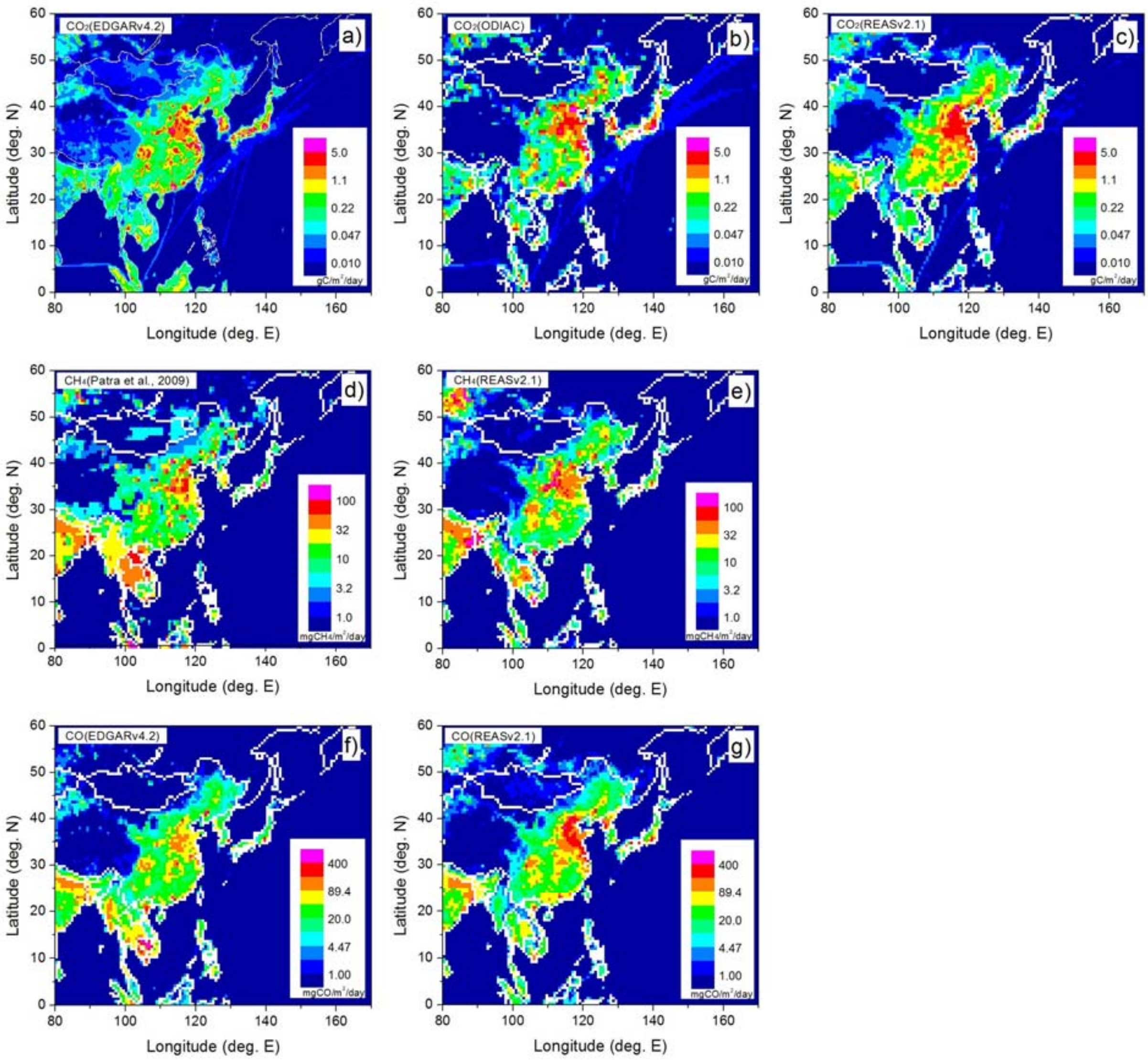


**Fig. S5.** Comparison of the flux distributions of (a) fossil $CO_2$ from EDGAR v4.2, (b) fossil $CO_2$ from ODIAC, (c) fossil $CO_2$ from REAS v2.1, (d) $CH_4$ from Patra et al., (2009), (e) $CH_4$ from REAS v2.1, (f) CO from EDGAR v4.2, and (g) CO from REAS v2.1. The flux maps for 2007 are shown. Annual mean fluxes are depicted for $CO_2$ and CO, while monthly mean fluxes in January are depicted for $CH_4$.

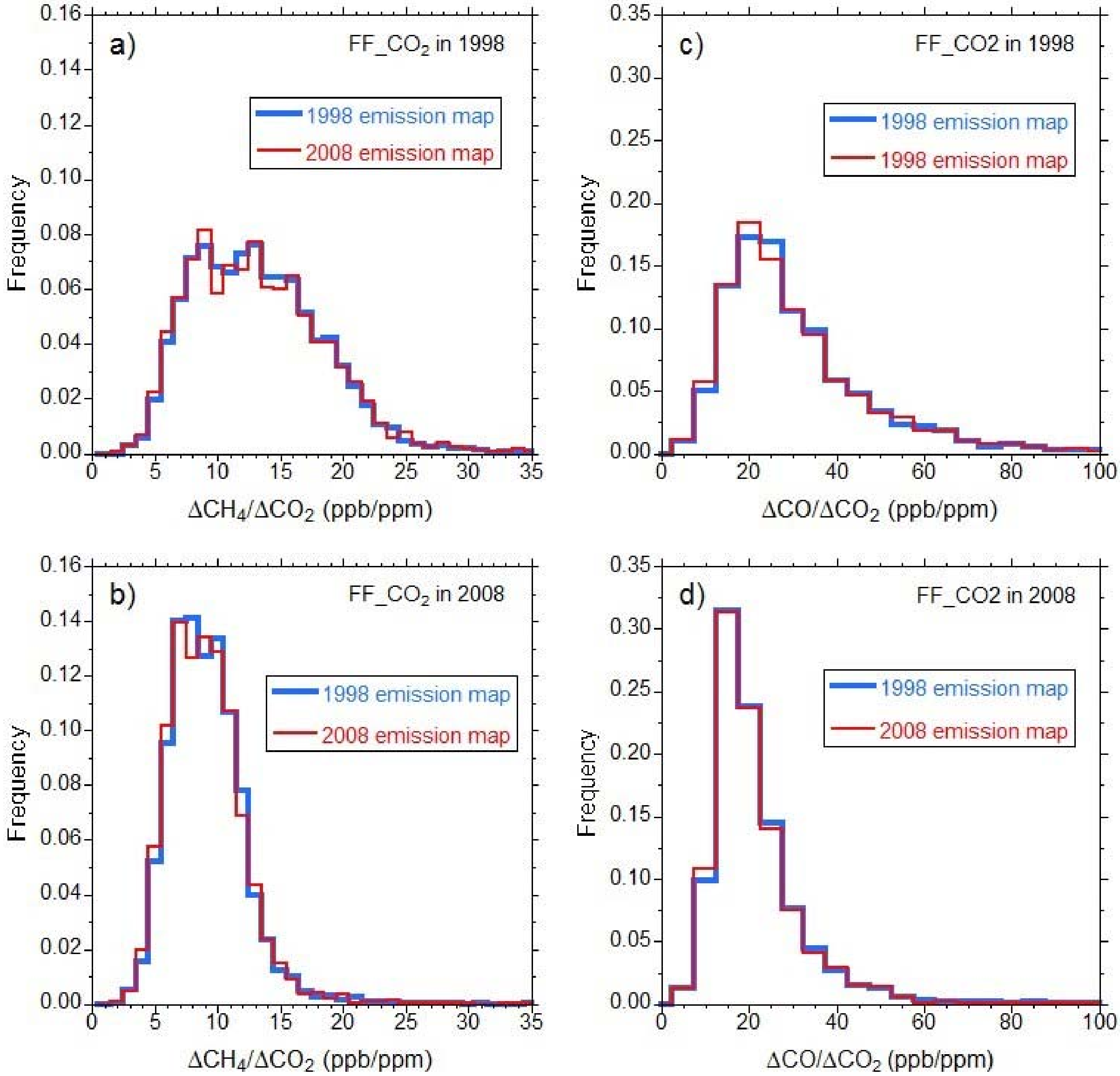


**Fig. S6.** Histograms of the simulated correlation slopes of (a, b) $\Delta CH_4/\Delta CO_2$ and (c, d) $\Delta CO/\Delta CO_2$ for fossil $CO_2$ emissions in (a, c) 1998 and (b, d) 2008. The correlation slopes all meet the selection criteria (|R|>0.8). The simulated results based on the fossil fuel-derived $CO_2$ emission maps for 1998 and 2008 are depicted as blue and red lines, respectively.

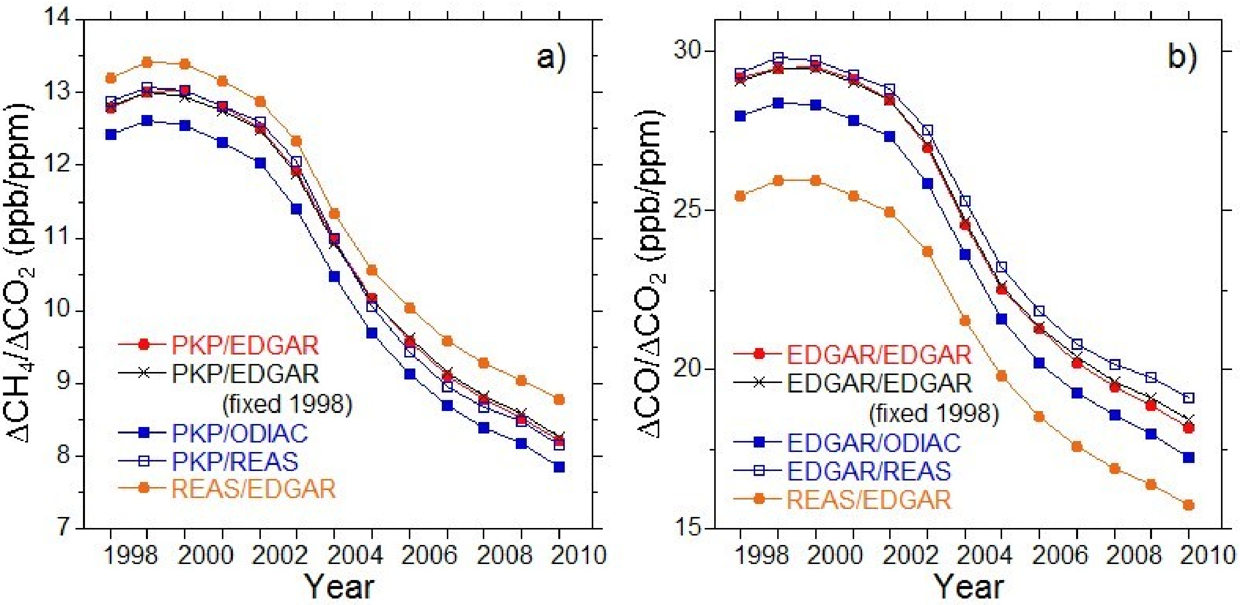


**Fig. S7.** Comparison of the winter average correlation slopes of simulated (a) $\Delta CH_4/\Delta CO_2$ and (b) $\Delta CO/\Delta CO_2$ for different combinations of the emission maps described in the legend. PKP in the legend represents the $CH_4$ emissions from Patra et al., (2009). The simulated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