Model simulated trend of surface carbon monoxide for the 2001-2010 decade

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We thank the reviewers for the constructive and valuable comments, and have revised and improved the manuscript as your comments.

***RC C3736**

General Comments

This manuscript first compares model results from simulations using decadal constant emissions to results using the similar base conditions but including the RCP and GFED time varying emissions. Regional mean CO concentrations using the time-varying sources are found to be more similar to MOPITT than constant emissions. This section could be shortened.

-> Some parts, which do overlap or are unnecessary, will be removed or shortened as suggested by the referee. For example, the Taylor diagram is enough to provide a concise statistical summary of spatial pattern correlation between satellite observation and model simulation, so that the scattering plot (Figure 5 in the manuscript) is not needed and will be removed.

The time varying anthropogenic and biomass burning emissions were used with chemistry to derive a more realistic model response. Model CO distributions give reasonably good agreement with the MOPITT surface product, although there are very large aggregation errors in both data sets. Decadal trends from the model are compared to those determined from a number surface sites, again with good agreement. In addition to model/surface site comparisons in the manuscript, I suggest the model also be compared to trends in the MOPITT products.

-> We are reluctant to compare the trends derived from MOPITT products against the model-simulated trends. In fact, the MOPITT trend has an inevitable error caused by time-varying averaging kernels and a priori CO profiles, as demonstrated in Yoon et al. (2013). The MOPITT surface CO trend is therefore not realistic, with a possible bias, ranging from -10.71 to +13.21 ppbv yr^{-1} (−5.68 to +8.84% yr^{-1}) depending on location. Unfortunately, it is not possible to eliminate the uncertainty of averaging kernels and a priori solely based on satellite observations, as the true state is unknown. This is the reason why in this study we used only the ground-based observations to evaluate the model-simulated trend.

There is a brief discussion on trends in OH, CO and NOx trends from the model and the influence of NOx on OH. This section is relatively weak and could be removed. Some conclusions, such as the observed decreases in concentration over Europe and the US are due to decreased anthropogenic emissions, are not new.

-> We agree that the section referring to the influences of NOx and OH is not enough to identify explicitly the influence on the CO trend. Therefore, we will remove Figure 14 and corresponding discussion.

The simulated deceases in surface CO from anthropogenic emissions over Europe and USA are not new, but confirmed in this study by comparing the simulation with satellite and ground-based observations.

Specific Comments

P. 12410, line 13. It should be noted that the downward trends observed in the 1990s have been attributed primarily to decreases in anthropogenic emissions (Duncan et al. 2007, Novelli et al., 2003, among others).

-> We will additionally mention in the manuscript that the downward trends observed in the 1990s have been attributed primarily to decreases in anthropogenic emissions and cite the relevant references (Duncan et al. 2007, Novelli et al., 2003, among others) in the introduction.

P. 12410-12411. All the satellites which measure CO need not be mentioned. It would be better to say the MOPITT results are used here because of their rigorous evaluation/ validation and their 13 year continuous record. The pros and cons of using satellite retrievals to validate models and to estimate long-term trends should be given.

-> Accordingly to the comment, we will modify the manuscript.

P. 12414, Section 2.3. This section would benefit from more detail on the MOPITT surface product, including the pressure levels that define the surface product, its precision and any bias.

-> We will provide more details about the MOPITT TIR surface product as follows:

"The Measurements of Pollution in the Troposphere (MOPITT) instrument, launched on board the EOS-Terra spacecraft in 1999, has been providing continuous global products of atmospheric profiles of CO volume mixing ratio from 1000 to 100 hPa with 100 hPa interval and CO total column values (Deeter et al., 2003). The global MOPITT retrieved CO data with high accuracy (expected precisions: 10%) has been applied to various researches on its sources, transports, and sinks (e.g., publications at http://www.acd.ucar.edu/mopitt/publications.shtml). In this study, the MOPITT Version 5 (V5) Level 3 (L3) thermal infrared (TIR) surface CO products in daytime are used since they have been improved in the retrieval sensitivity and accuracy for the lower tropospheric CO (Clerbaux et al., 2009; Worden et al., 2010, 2013; Deeter et al., 2007, 2011, 2012, 2013). MOPITT TIR products are based on thermal-infrared radiation at 4.7 μm. Even though a new joint (multispectral) TIR/NIR product features the

maximum sensitivity to near-surface CO, the TIR-based MOPITT can avoid significant random errors in NIR-based MOPITT products (near-infrared radiation at 2.3 µm).”

Section 3.1, P. 12415-12416. I found it hard to follow how the model data were transformed to reflect the pressure and a priori constraints of the MOPITT retrieval. Is there something missing in equation 1? This section needs to be written more clearly (e.g. see Deeter et al., JGR, 2010).

-> To clarify the description of Equation 1, we will rephrase the description of Equation 1 and refer to Deeter et al. (JGR, 2010) as follows:

“The EMAC-simulated surface CO can be transformed into a comparable quantity (so called pseudo-retrieval, $\hat{\mathbf{x}}$), to the MOPITT-retrieved surface CO as follows (Deeter et al., 2003, 2010):

$$\hat{\mathbf{x}} \cong \mathbf{x}_0 + \mathbf{A}\left(\mathbf{x} - \mathbf{x}_0\right) = \mathbf{A}\mathbf{x} + \left(\mathbf{I} - \mathbf{A}\right)\mathbf{x}_0 \quad \textbf{(1)}$$

where $\mathbf{x}_0$, $\mathbf{A}$, $\mathbf{I}$ and $\mathbf{x}$ represent the MOPITT a priori CO, the MOPITT averaging kernels, the identity matrices, and the EMAC-simulated CO profiles from surface to 100 hPa, respectively.”

P. 12416, lines 8-21, Figures 4, 5 and 6. These figures all depict the correlation of the model and MOPITT CO. I think the Taylor diagram (Figure 6) contains the most information and the other 2 can be removed. The authors should briefly say why this type of diagram is useful (e.g. Taylor, JGR, 2001).

-> As noticed by the referee, the Taylor diagram contains all the statistical values (i.e. the spatial correlation coefficient, normalized standard deviation, normalized centred root-mean-square difference, and relative difference) used in this study. Therefore, Figure 5 is unnecessary and will be removed. For Figure 4, we will change it to show the seasonal distribution of MOPITT observation and RG simulation, and therefore provide how different they are at each season as below.

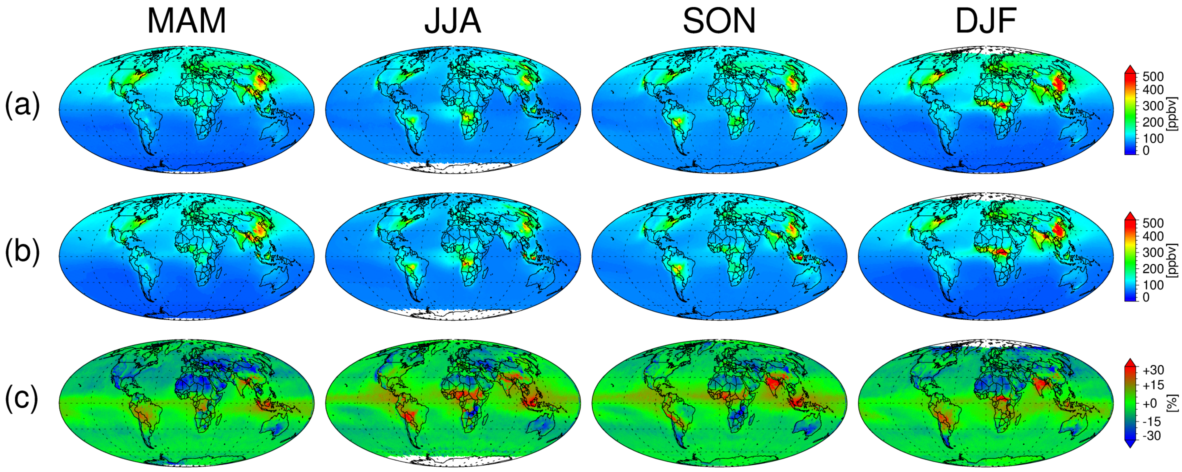

Figure S1. Global distributions of seasonal (a) MOPITT-retrieved surface CO, (b) pseudo-retrievals of EMAC-simulated surface CO based on RG scenarios, and (c) their relative difference from 2001 to 2010.

P. 12417, Table 3. The mean decadal model results for the constant source and time varying source runs are compared. Mean CO over the Eastern USA and Western Europe are greater in the time varying scenario even though emissions have decreased. In Figure 12 a decadal decrease trend is shown for most regions. Transport from Central South America (the only region showing a strong increase in emissions) seems unlikely. Shouldn't have mean CO decreased?

-> The monthly means of surface CO over Eastern USA and Western Europe based RG scenario should be smaller than the monthly means on CE scenario as shown in Figure S2 below (please compare it with b. EUSA and d. WE in Figure 11 in the manuscript) since the RG emissions have decreased.

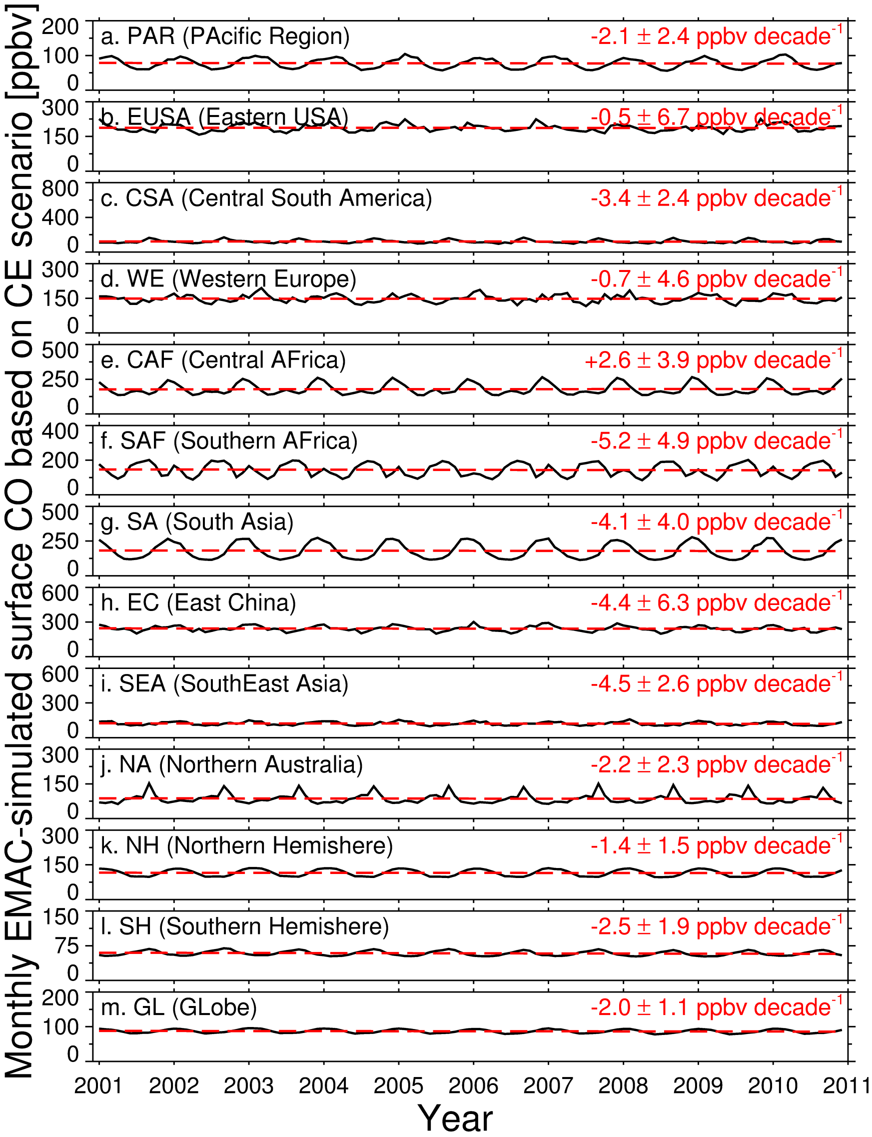

Figure S2. Regional and global trend estimates of monthly EMAC-simulated surface CO based on CE scenario with ±2σ errors from 2001 to 2010.

However, the values in Table 3 and Figure 6 are not the surface CO, but pseudo-retrievals of simulated surface CO. In other words, because the pseudo-retrievals are the weighted mean contributions of the EMAC-simulated CO profiles ($\mathbf{x}_{EMAC}$) and the MOPITT surface a priori CO profiles ($\hat{\mathbf{x}}_{MOPITT}$) in multiple layers from surface to 100 hPa as shown in Equation 1, the pseudo-retrievals of simulated surface CO based on RG scenario can be larger than the pseudo-retrievals on CE scenario over Eastern USA and Western Europe.

P. 12416, line 23. Is 'resume' the correct word?

-> It will be changed into "show".

P. 12418, Table 4. Would the authors comment on why the model gives statistically significant trends at about twice as many sites as the measurements.

-> The simulations on the resolution, 1.875 by 1.875 degrees can diminish extreme values. Therefore, the model simulations can lead to more statistically significant trends.

P. 12418-12419, Figures 7, 8. A majority of the trends determined from the WDCGG surface data and the model (Table 4) fall in the range of 0-20 ppb decade-1. The high statistical agreement between model and measured trends in Figures 7 and 8 appears to be driven by a few locations. Is this the case? Are there commonalities among the sites falling outside of the cluster?

-> The correlation coefficients and linear fittings in Figures 7 and 8 show the statistical correlation between two variables on X and Y axes. We have calculated the statistical dependence between them with the same weights. As you see in Figures 7 and 8, only two points are perfectly on the linear regression line, so the statistical correlation driven by the most of variables except the two points little changes.

P. 12420, Figure 9a. Why do the model results with constant emissions show decreasing trends in the Southern Pacific, Indian and Southern Oceans?

-> The changes in CO in simulation CE are purely due to meteorological changes in the atmosphere. The changes could be due for example to changes in transport patterns and/or increase or decrease in precipitation (which in turn influences the chemistry of CO).

P. 12421, line 9. The oxidation of non-methane hydrocarbons should be included as a major source of CO (Duncan et al., JGR, 2007 and references therein).

-> We will include the oxidation of non-methane hydrocarbons in the manuscript as a major source of CO with relevant references.

P. 12421, Figure 12. GFED 3.1 reports SEAS CO emissions from fires in 2010 were the greatest for the decade but high emissions in Asia are not shown in Figure 12. Would the authors comment on this.

-> You can see higher emissions of biomass burning in South Asia (SA) in 2010 in Figure 12. The SEAS (Southeast Asia) region defined in GFED is partially similar to our South Asia (SA) region as shown in Figure S3 below. Simply we used similar names (here and in GFED v3.1) for different regions.

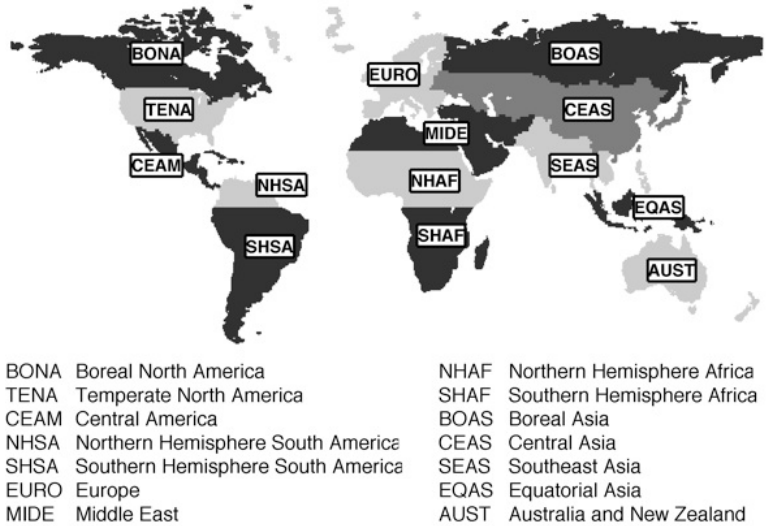

Figure S3. 14 basis regions for GFED annual cumulative emissions [http://www.globalfiredata.org/Tables/index.html].

P. 12422, Section 4.2, lines 5-29. The trends discussed here are given in four or five different units. Can they be normalized to % change per decade? The trends from MOPITT and AIRS instrument come from Worden et al. 2013 and represent the total column. I would like this work to compare model trends with trends calculated from the MOPITT surface product. The model and MOPITT could also be compared in the mid and upper troposphere.

-> It is difficult to unify the unit of the trends from several references due to the missing values for normalization provided in the references. Additionally, as mentioned before, since it is impossible to remove the uncertainty from MOPITT surface data, the

MOPITT trends cannot be used to compare with the model-simulated trends (see Yoon et al., 2013).

Pp. 12422-12423, Figure 13. The changes in CO from the model are compared its emissions. I don't think it is surprising they are similar. This rather long discussion could be shortened. Perhaps say that regional-scale model trends generally reflect trends in the emissions, except for Eastern China. Then examine China more closely.

P. 12423, lines 9-14. Worden et al., 2013 report a strong decrease in MOPITT column CO over E. China during the 2000s. Emissions from Eastern China in the model show a marginal decrease with time however the model results show an increase. The authors suggest this surprising result may come from transport or secondary chemical production. The results from the constant emission model run (Figure 3) don't seem to support transport. The possibility that chemical production from hydrocarbons are referenced to Tohjima et al., 2014 and Anglebratt et al., 2011, but neither of these papers quantitatively examine how reasonable changes in VOCs would effect CO trends. This manuscript should look into the E Asia emissions/surface changes in more detail.

-> As mentioned before, the trend estimates from MOPITT CO data can be biased by the uncertainty from time-varying averaging kernels and a priori, so that we didn't used the MOPITT CO data to evaluate the model-simulated trends. Nevertheless, we agree with the comment. We will more closely examine the trends over Eastern China using Figure S4 as follows;

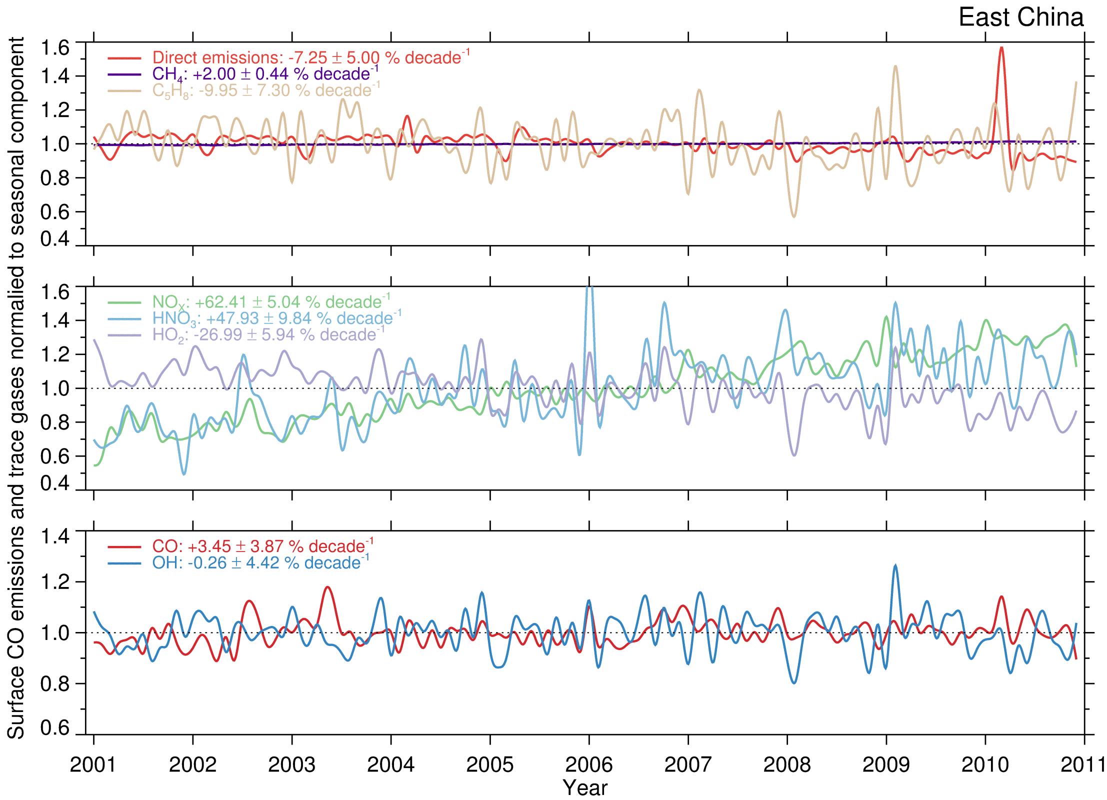

Figure S4. Long-term time series of surface CO emissions and relevant trace gases normalised to seasonal component over East China from 2001 to 2010.

"Notwithstanding a significant decrease in the CO emissions over East China, the simulated trend in surface CO shows an insignificant increase. This is opposite to the results from Worden et al. (2013) that showed a negative trend in MOPITT tropospheric column CO over East China. Figure S3 shows long-term time series of surface CO emissions and trace gases relevant to chemical production of CO and OH over East China from 2001 to 2010. Hydroxyl radical (OH) is the main oxidant of many trace gases and therefore one of the most important species in the atmospheric chemistry (Lawrence et al., 2001; Wallace and Hobbs, 2006). CO removal from the troposphere is almost exclusively by reaction with OH (Hauglustaine et al., 1998; IPCC, 1996) and, on the other hand, CO provides the most important sink for OH (Lelieveld et al., 2002; Thompson et al., 1992). As mentioned, the direct emissions from biomass burning and fossil/domestic fuel has the most influence on the surface CO change, and show significantly negative trend in the East China region (-7.25 ± 5.00 % decade^{-1}). Additionally, biomass burning in 2010 is the greatest for the decade in Asia (Giglio et al., 2010). Oxidation of CH_4 is another primary chemical production of the CO, and the

surface CH_4 significantly increases, +2.00 ± 0.44 % decade^{-1}. In contrast, isoprene (C_5H_8) occupied a majority in biogenic NMHC (Holloway et al., 2000) presents for estimating the change in chemical production of CO by oxidation of NMHC, and it changes by -9.95 ± 7.30 % decade^{-1}. These trends show that both direct emissions and chemical formation of CO over EC region decreased during the decade 2001-2010.

Nevertheless the surface NOx drastically increased during the same decade (+62.41 ± 5.04 % decade^{-1}), which contributed to the decrease of the HO_2 (-26.99 ± 5.94 % decade^{-1}) via HNO_3 formation (+47.93 ± 9.84 % decade^{-1}) (see also Lelieveld et al, 2002, 2004).

The decrease in OH concentration (-0.26 ± 4.42 % decade^{-1}) implies a reduce oxidation of CO, and therefore the presence over the EC region of a slightly positive trends of CO. It must be underline that this trend is not significant, and it is calculated only for the surface. The total tropospheric column of CO is strongly influenced by the long-range transport of CO, which has a lifetime of around 1 month. The results of simulation CE, where the pure CO transport induce a slight negative trends in the CO concentration over EC, are therefore in agreement with the results of Worden et al. (2013).”

P. 12423, Section 5. From model calculated trends of OH, CO and NOx, the authors conclude OH trends are largely controlled by NOx. The discussion would benefit from a description of CO-OH-CH4-NOX-O3 chemistry. (e.g. Tables 1 and 2 in Lelieveld et al., ACP, 2004). The authors should temper this conclusion, e.g. ‘These results suggest that more than just the CO trend effects trends in OH’.

-> Thanks for your valuable suggestion. As the section will be removed, we do not think it is necessary to described CO-OH-CH_4-NO_X-O_3 chemistry, as this is done in many publications and textbooks. Nevertheless we will add the sentence suggested by the referee, enhancing the fact that CO is controlled by multiple factors.

The changes in OH and NOx shown in Figure 14 are very small and contain large uncertainties. A more robust conclusion would require a multivariate analysis of the important species in the OH cycle.

-> We agree your comment and will remove Figure 14. As your previous comments, we will more closely examine the trends over Eastern China using Figure S4 and temper the conclusion.

References

I suggest the number of citations for each reference be limited to no more than 3-4 carefully chosen papers.

-> We will choose 3 or 4 references for each citation.

Tables and Figures

Table 2. Note that ‘GC-HgO’ is the method, RGD is the instrument.

-> The information about instrument and analyses measurement method is from GAW Report No. 188 (WMO, 2009). We will change the label, “Measurement Method” into “Instrument or Analyses Measurement Method”.

Table 3. Mean MOPITT CO for PAR is reported as 91.78 ± 33.4 with standard deviation of 7.32 ± 5.28. Are these values the mean and aggregation errors? Please clarify this in the caption

-> They are the mean values of monthly means, standard deviations, spatial correlation coefficient, centred root-mean-square (RMS) difference, and relative bias from 2001 to 2010 with ±2σ.

Table 4. As for Table 3. Also add in the caption that (ω/ωσ) > 2 is significant.

-> We will add it in the caption.

Figures 1a, b. The maps of global mean CO emissions show the general distribution of the fossil fuel and biomass burning emission strengths but say little about changing emissions. Figures showing model emissions by region over time would better serve in discussions of trends.

-> The model emissions by region over time have been shown already in Figure 12.

Figure 3. The names of the regions cannot be read. The dots showing the site locations are hard to see these should all be larger.

-> As your comment, the figure will be modified as below.

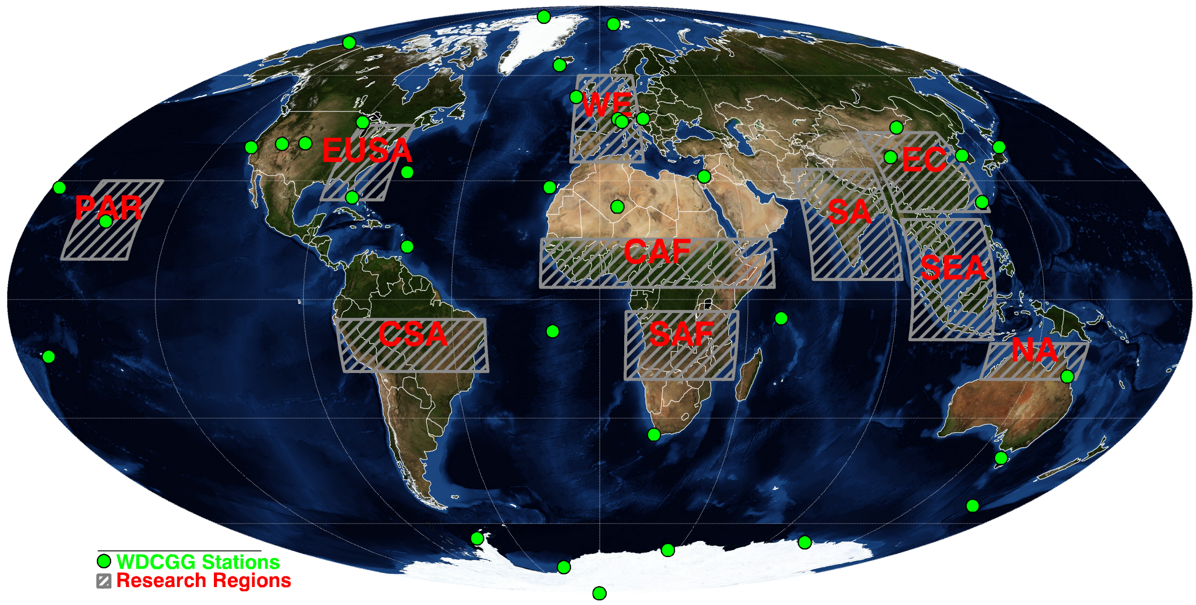

Figure S5. Research region domains and geolocations of WDCGG stations listed on Tables 1 and 2, respectively.

Figure 5. What do the yellow, blue and black dots show?

-> Black, blue, and yellow respectively indicate larger points than 1, 10, and 100 falling into each of bins (bin size: 5 ppbv).

Figure 6. What are the standard deviations normalized to?

-> The simulated standard deviations are normalized to the corresponding observed standard deviation (σ_r).

Figure 8. There are 4-5 sites which fall outside of the general cluster. Which these are they? Perhaps they can be labelled as in Figures 13 and 14 or defined in the figure caption.

-> As your suggestion, they will be labelled in Figure 8 as below.

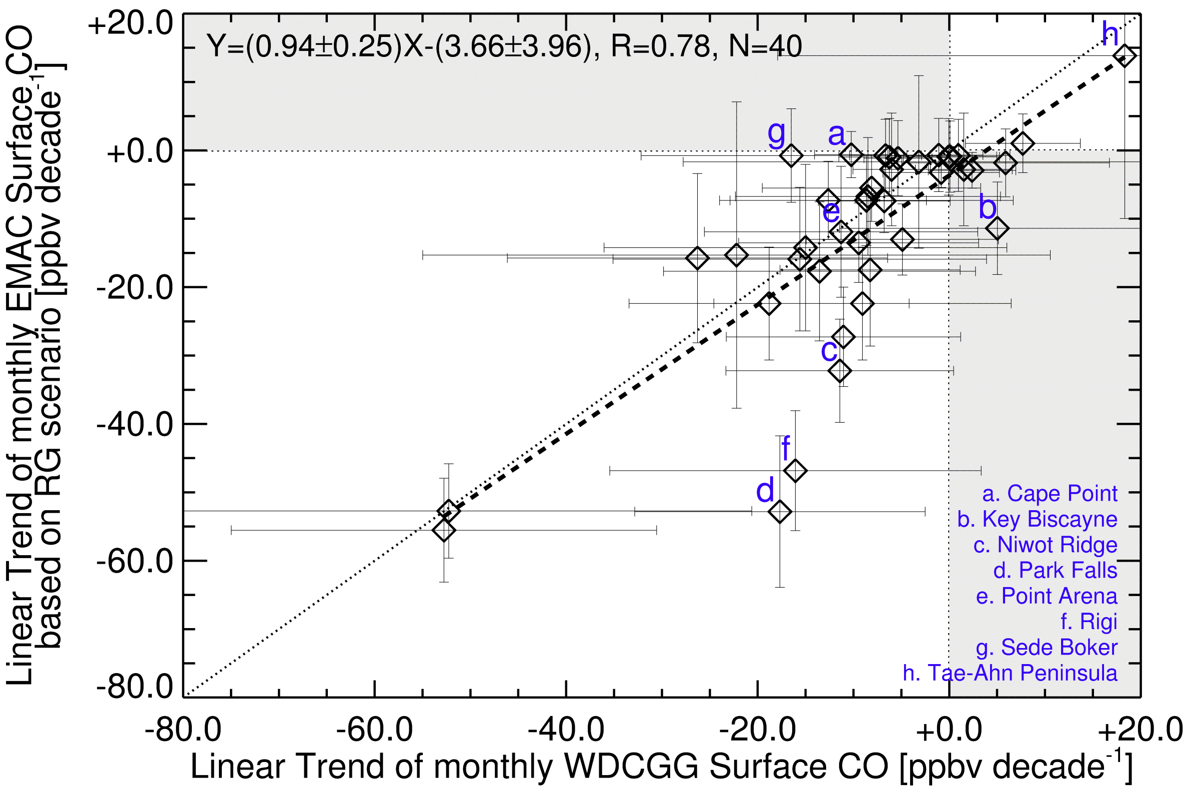

Figure S6. As in Figure 7 (b), but the trends of monthly EMAC-simulated surface CO from a model grid-box to the upwind direction at the stations (i.e. Cape Point, Key Biscayne, Niwot Ridge, Park Falls, Point Arena, Rigi, Sede Boker, and Tae-ahn Peninsula).

Figure 9. The site symbols should be larger.

-> They will be enlarged as below.

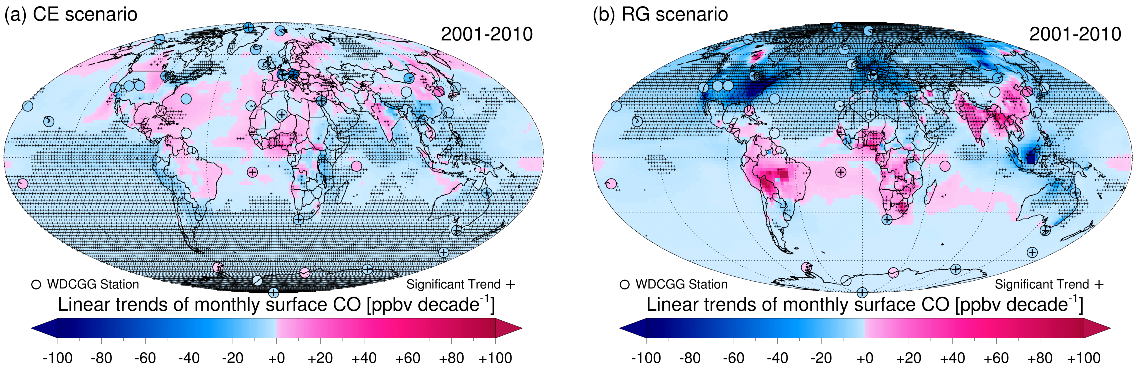

Figure S7. Global trend estimates of monthly WDCGG-archived and EMAC-simulated surface CO based on (a) CE and (b) RG scenarios from 2001 to 2010. The significant trends are shown as a plus symbol (+).

Figure 10. The colored panels are not very useful. The trend data are given in several other places. These figures could be removed.

-> We agree with your comment. Figure 10 will be removed.

***RC C3922**

Main Comments

1: Trend analysis: Fig 9, 10, 11, I suggest that the authors should add MOPITT data and do a cross comparison among model, station observation and MOPITT.

-> As reply to referee #1, we are reluctant to compare the trends derived from MOPITT products against the model-simulated trends. As shown in Yoon et al. (2013), the trend estimated from MOPITT data does contain an inevitable error caused by time-varying averaging kernels. Yoon et al. (2013) also showed that the MOPITT surface CO trend could be biased, ranging from −10.71 to +13.21 ppbv yr^{-1} (−5.68 to +8.84%yr^{-1}) depending on location. Unfortunately, it is not possible to eliminate such uncertainty solely based on satellite observations, as the true state of the atmosphere is unknown.

This is the reason why in this study we used only the ground-based observations to evaluate the model-simulated trend.

2: MOPITT data: According to MOPITT science team, "the new joint (multispectral) TIR/NIR products, featuring the maximum sensitivity to near surface CO are fundamentally much more capable of characterizing surface-level CO than either purely TIR- or NIR-based products". The authors may want to compare the model simulations with TIR/NIR joint product since the manuscript is looking into the trend near surface.

-> Actually, a new joint (multispectral) TIR/NIR product features the maximum sensitivity to near-surface CO. Nonetheless, since the NIR-based MOPITT products can contain significant random errors, so it may require significant spatial and/or temporal averaging.

Additionally as your suggestion, we have compared the model simulation with the TIR/NIR product on March 2007 as shown in Figure S1 (b) and found a good correlation like in Figure S1 (a) (i.e. R=0.97 in Figure S1 (a) and R=0.95 in Figure S1 (b)). Since there is no significant difference in the correlations of Figures S1 (a) and (b), we would therefore keep the part of spatial comparison between the MOPITT TIR products and model simulations in the manuscript as it is.

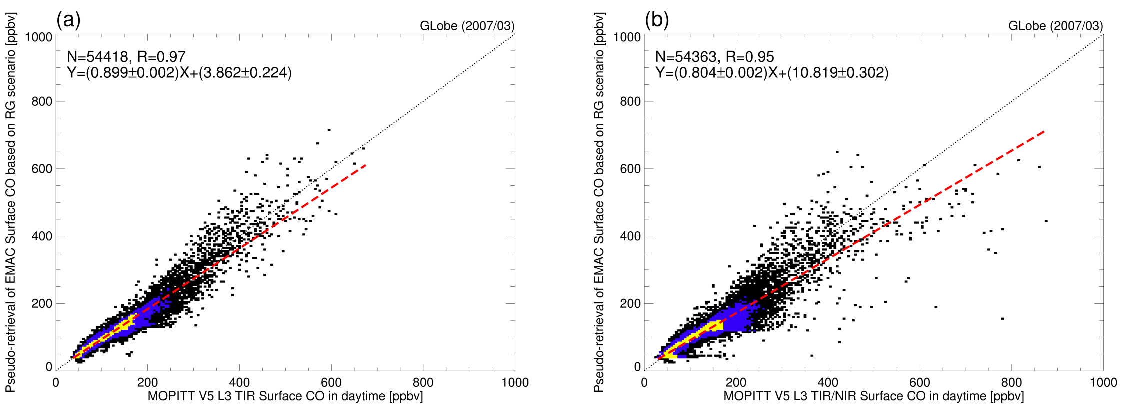

Figure S1. Spatial comparisons of global MOPITT-retrieved (a) TIR and (b) TIR/NIR surface CO with the pseudo-retrievals of EMAC simulations based on RG scenarios on March 2007. Black, blue, and yellow respectively indicate larger points than 1, 10, 100 falling into each of bins (bin size: 5 ppbv).

3: The sensitivity run with constant emissions: The purpose of CE simulation is highlighting the influence of emission/transport on the trend and spatial distribution of surface CO. Therefore, I don't think it is necessary to discuss the comparison between CE simulation and MOPITT data during the evaluation section (e.g. Fig 4). Instead, it is more important to evaluate the model during different burning seasons. Therefore, I suggest the author removing CE simulation in Fig4 and add another season comparison (e.g. September, the SH biomass burning season).

->As suggested, we will remove Figure 4 and add seasonal distributions of the MOPITT TIR products and model RG simulations with their relative difference from 2001 to 2010 as shown in Figure S2 below.

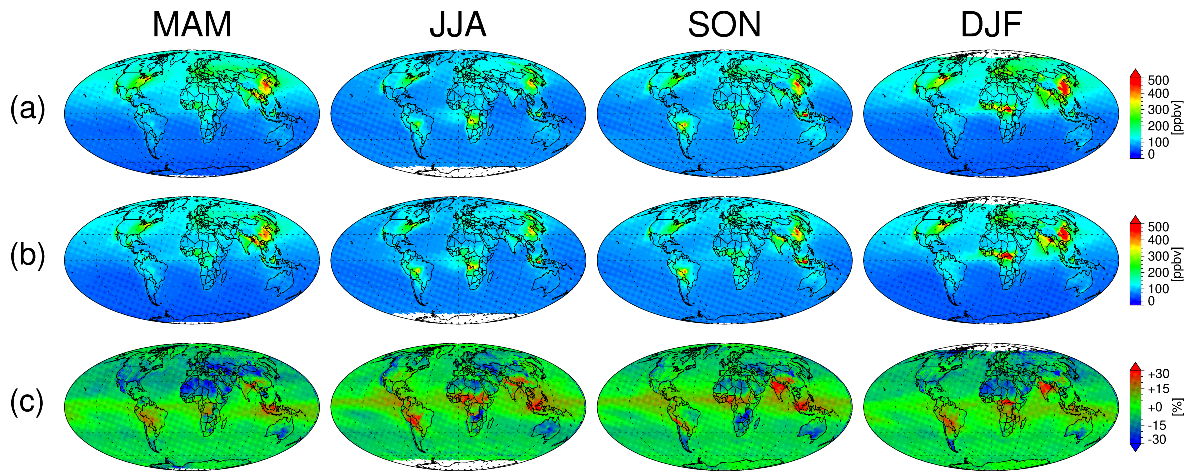

Figure S2. Global distributions of seasonal (a) MOPITT-retrieved surface CO, (b) pseudo-retrievals of EMAC-simulated surface CO based on RG scenarios, and (c) their relative difference from 2001 to 2010.

4: Trend in East Asian: the surface CO trends are positive but not significant, while the emission is negatively significant. And Worden et al 2013 showed a negative trend in MOPIT tropospheric column CO. The authors argue this is influenced by transport or chemistry. From Fig9, the transport actually produced a negative trend over EC. So the authors should remove the transport in Line 13 and discuss more about other reasons with evidences for the positive trend over EC at surface.

-> We will extend the analysis of the CO trend over East China as follow.

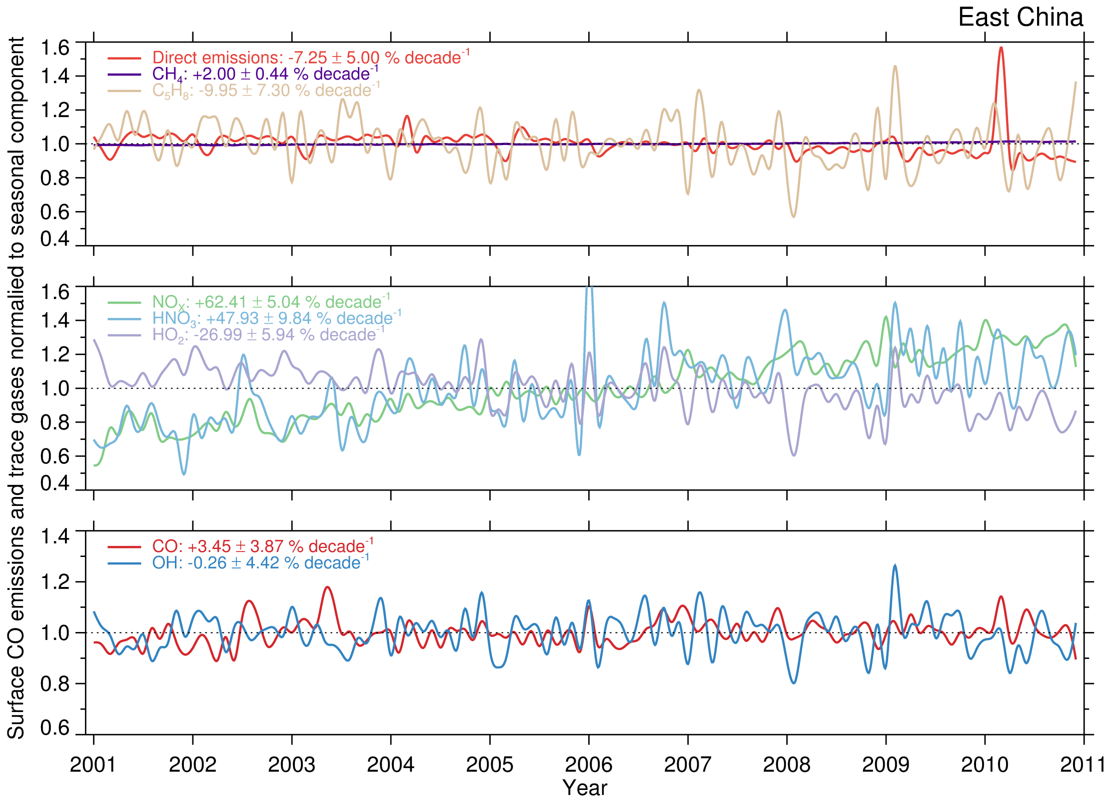

Figure S3. Long-term time series of surface CO emissions and relevant trace gases normalised to seasonal component over East China from 2001 to 2010.

"Notwithstanding a significant decrease in the CO emissions over East China, the simulated trend in surface CO shows an insignificant increase. This is opposite to the results from Worden et al. (2013) that showed a negative trend in MOPITT tropospheric column CO over East China. Figure S3 shows long-term time series of surface CO emissions and trace gases relevant to chemical production of CO and OH over East China from 2001 to 2010. Hydroxyl radical (OH) is the main oxidant of many trace gases and therefore one of the most important species in the atmospheric chemistry (Lawrence et al., 2001; Wallace and Hobbs, 2006). CO removal from the troposphere is almost exclusively by reaction with OH (Hauglustaine et al., 1998; IPCC, 1996) and, on the other hand, CO provides the most important sink for OH (Lelieveld et al., 2002; Thompson et al., 1992). As mentioned, the direct emissions from biomass burning and fossil/domestic fuel has the most influence on the surface CO change, and show significantly negative trend in the East China region (-7.25 ± 5.00 % decade^{-1}). Additionally, biomass burning in 2010 is the greatest for the decade in Asia (Giglio et al., 2010). Oxidation of CH_4 is another primary chemical production of the CO, and the

surface CH_4 significantly increases, +2.00 ± 0.44 % decade^{-1}. In contrast, isoprene (C_5H_8) occupied a majority in biogenic NMHC (Holloway et al., 2000) presents for estimating the change in chemical production of CO by oxidation of NMHC, and it changes by -9.95 ± 7.30 % decade^{-1}. These trends show that both direct emissions and chemical formation of CO over EC region decreased during the decade 2001-2010.

Nevertheless the surface NOx drastically increased during the same decade (+62.41 ± 5.04 % decade^{-1}), which contributed to the decrease of the HO_2 (-26.99 ± 5.94 % decade^{-1}) via HNO_3 formation (+47.93 ± 9.84 % decade^{-1}) (see also Lelieveld et al, 2002, 2004).

The decrease in OH concentration (-0.26 ± 4.42 % decade^{-1}) implies a reduce oxidation of CO, and therefore the presence over the EC region of a slightly positive trends of CO. It must be underline that this trend is not significant, and it is calculated only for the surface. The total tropospheric column of CO is strongly influenced by the long-range transport of CO, which has a lifetime of around 1 month. The results of simulation CE, where the pure CO transport induce a slight negative trends in the CO concentration over EC, are therefore in agreement with the results of Worden et al. (2013).”

Detailed Comments

We will modify the manuscript following your comments and suggestions.

P12410 Line 12: Western Europe, Eastern USA, and Northern Australia. Should be de-capitalized for “western, eastern, and northern”. Please make same changes in the rest of manuscript for the similar situation.

P12410, line 16: remove “significant” or change into another word

P12411, line 3: remove in unpolluted and non-forested locations. Change the CO-OH and the lifetime of CO description like: The main sink of CO is oxidation by OH and results in a ~2-month mean lifetime. Because of this relatively short lifetime, CO is not well-mixed in the troposphere

P12411, line 7: replace finally with therefore.

P12411, line 15: Using GEOS-Chem model, Liu et al. 2010 (Analysis of CO in the tropical troposphere using Aura satellite data and the GEOS-Chem model: insights into transport *characteristics of the GEOS meteorological product) looked into the interannual variation of tropical tropospheric CO in 2005 and 2006. In her 2013 paper, she looked into the IAV of tropical CO in UTLS during the Aura period also with GEOS-Chem model.*

P12411 line18: remove "allow scientists and researchers to"

P12411 line 19: remove the 2nd global

P12412 line8: remove in contrast.

P12412 line10: change into available ground stations

P12413 line 13: specify the vertical resolution.

P12414 line 12: change into urban megacities.

P12415 line8: remove finally

P12416 line 16: Change into"It is quite challenging to retrieve tropospheric CO profiles based on mostly passive remote sensing instruments (including MOPITT) because ..

P12418 line 7: Pacific

P12419 Line 10: make n and N consistent in the equation.

P12422 line2: replace tendencies into trends

P12410, line 22, define Medium-lived.

-> It means that the CO lifetime ranges from weeks to months.

P12411 line 20: The main purpose of this paragraph is showing the limitation of using satellite data or ground station solely to explain the CO trends. The authors should not just list all the satellite names. The authors should provide more detailed discussion of using satellite data (here MOPITT) to evaluate model simulation and to do the IAV and trend analysis. Another point is: the last sentence of this paragraph seems indicating that in the rest of paper, the authors will combine the satellite data and ground based data. So the introducing of model in the next paragraph seems unexpected. Please make sure the logical is smooth.

-> As your comment, the paragraph will be improved by providing more details on the MOPITT surface product. Additionally, we will add pros and cons of model simulations to make the paragraph more logical.

P12417 line19: add "in December 2008". It is obvious that PG simulation agree better than CE simulation with the observations. So remove the comparison to CE simulation and corresponding discussion and Fig 5. Also in Fig 4 add another burning season (e.g. September) and corresponding discussion of spatial distribution of surface CO. The authors should also consider adding one panel showing the difference between model and MOPITT in these two months.

-> We will remove Figure 5 since the Taylor diagram in Figure 6 is enough to provide a concise statistical summary of spatial pattern correlation between satellite observation and model simulation in Figure 6. For Figure 4, we have changed it to show the seasonal distribution of MOPITT observation and RG simulation as shown in Figure S2 and therefore provide how different they are at each season.

P12418 line 10: Not clear and please explain this sentence. "the failings to consider significant influences of natural sources (e.g. effects of the El Niño on tropospheric CO, Chandra et al., 2009) in the EMAC model"

-> We agree with the referee that the sentence is not well formulated. In general El Niño can induce drought and therefore spread rapidly forest fires. This effect increases atmospheric CO concentration due to the enhanced biomass burning (Chandra et al., 2009). However, in the EMAC model the biomass burning is emitted always at 140 m, without higher injection level. Therefore EMAC could easily underestimate the large-scale transport of the enhanced biomass burning during such events.

P12418 line 15: WDCGG-archived data (Xt)

-> No, it is not X_t, but Y_t.

P12418 line 22: what does (t/12) stands for?

-> It means years (X_t=t/12).

P12419 line 22: for the correlation between only significant trends, r increased to 0.7, but n drops to 7 (Fig 7). What is the p-value for this correlation? Does the model significantly capture the trends in observations?

-> The pairs of significant trends are only 7 and the P value for the correlation is 0.0799 that is interpreted as low presumption against null hypothesis based on a significance level of about 10%. Therefore, we will remove the comparison between only the significant trends, but calculate the correlations between all the trends as shown in Figure S4 below. The P values for the correlations are very small (~0.000) that is interpreted as very strong presumption against null hypothesis.

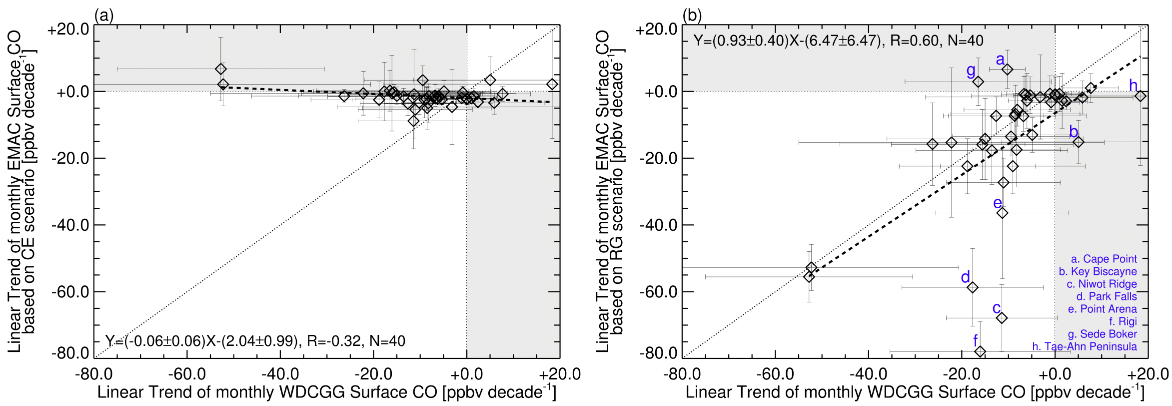

Figure S4. Comparisons of the trends of monthly EMAC-simulated surface CO based on (a) CE and (b) RG scenarios against the trend of monthly archived surface CO with ±2σ errors for selected WDCGG stations listed on Table 2. Detailed values are summarized in Table 4. Some stations (i.e. Cape Point, Key Biscayne, Niwot Ridge, Park Falls, Point Arena, Rigi, Sede Boker, and Tae-ahn Peninsula) influenced by local pollution or its transports are labelled.

P12419 line 10: I am confused with the method to calculate the standard deviation of the trends. The standard way of doing this is first calculate the autocorrelation coefficients, then infer the effective degree of freedom then calculate the standard deviation of the trends. Line 11: I guess in equation (5) σ_N is the standard deviation of the x time series. If so, please correct in your definition.

-> No. σ_N denotes the standard deviation of N (noise). n is the number of years. As your description, the standard deviation of the trends calculated by autocorrelation (ϕ), σ_N, and n as Equation (5).

P12420 line 15: wrong reference. Novelli et al 2003 was examine the effect of 1997-1998 fire on tropospheric CO. They didn't mention the perturbation of Pinatubo. Furthermore, the influence of Pinatubo should only last for a few years (its effect on stratospheric and tropospheric ozone lasted until 1994). How should this contribute to the decrease trend of CO from 1991 to 2001?

-> Novelli et al. (2003) reported that "Between 1991 and 2001 global average CO decreased at a rate of 0.52 ± 0.10 ppb yr^{-1}. About 30% of the decline may be attributed to the sharp decrease in CO that followed the eruption of Mt. Pinatubo [Bekki et al., 1994; Dlugokencky et al., 1996].". We will rephrase the misleading sentences.

P12421 Line 27: keep the unit of trends uniform @@y-1?

-> This study reports the surface CO trends based on 10-year simulations and observations. We would keep the results in the unit of trends, ppbv decade^{-1}.

P12424 Line11: for east China, the surface CO trends is positive but not significant, while the emission is negatively significant. And Worden et al 2013 showed a negative trend in MOPIT tropospheric column CO. The authors argue this is influenced by transport or chemistry. From Fig9, the transport actually produced a negative trend over EC. So the authors should remove the transport in Line 13 and discuss more about other reasons caused the positive trend over EC at surface. The authors could put the discussion in the conclusion part, since the summary and conclusion part is quite short and nothing new in this section.

-> We agree that the section referring to the influences of NOx and OH is not enough to identify explicitly the influence on the CO trend. Therefore, we will remove Figure 14 and corresponding discussion. Instead, by including additional analysis of surface OH, CO, NOx, CH_4, HNO_3, HO_2, C_5H_8 trends over East China, we will discuss their chemical influences on the CO trend as Figure S3.

Tables and Figures

Table 3: please clarify the the definition of mean and corresponding statistcs. For example, for PAR mean MOPITT CO is 91.78+-33.49. My understanding is 33.49 is the 1 or 2 σ *of the mean CO. But* σ *in the table is 7.32+-5.28.*

-> They are the mean values of monthly means, standard deviations, spatial correlation coefficient, centred root-mean-square (RMS) difference, and relative bias from 2001 to 2010 with $\pm 2\sigma$**.**

Fig 2 and Fig 12: Is there any specific reason of using bar plots for emission time series? It is better to change them into line plots.

-> Bar plot can show each time series of anthropogenic and biomass burning emissions as well as total emission at one time. For Figure 12, because some regions are influenced by only one emission source, the line plots of biomass burning or anthropogenic can be overlapped with the line of total emission as figure below, so it is difficult to distinguish each other. Therefore, only Figure 2 will be changed into a line plot as suggested.

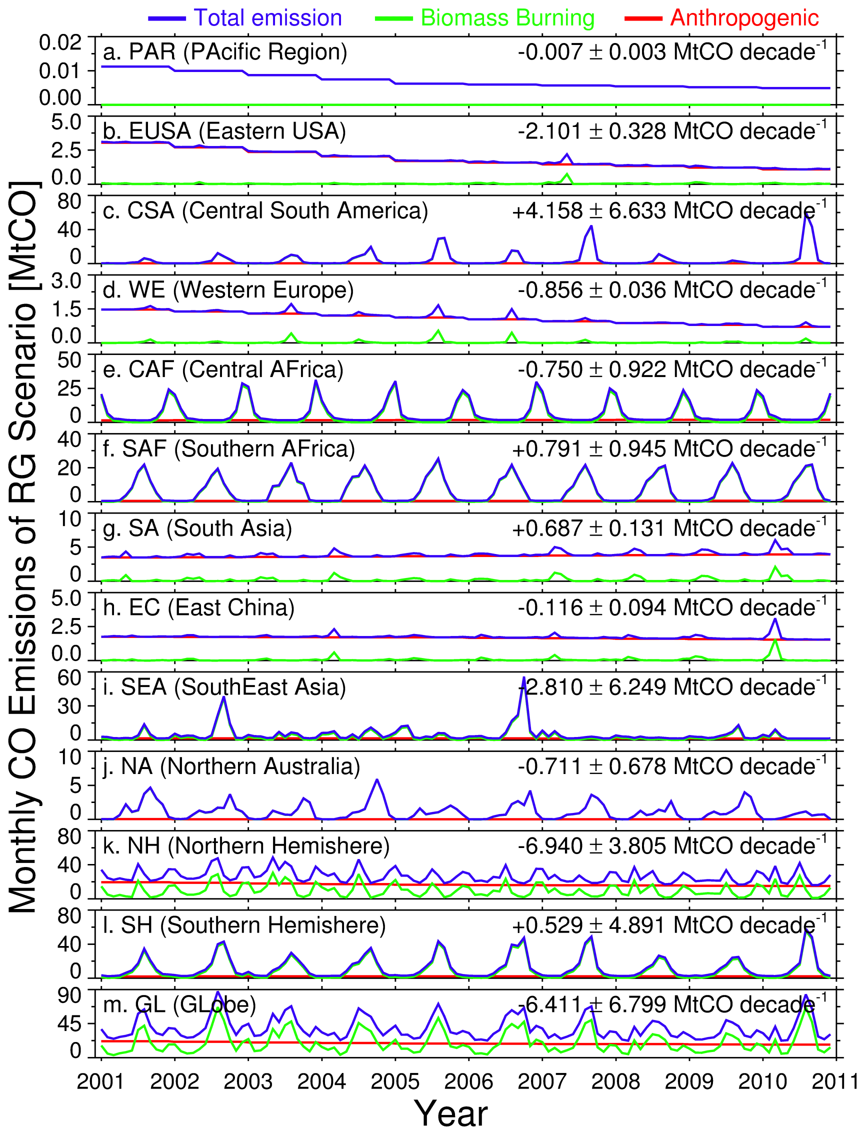

Figure S5. Regional and global trend estimates of monthly RG CO emissions with ±2σ errors from 2001 to 2010.

Fig4: I suggest removing CE results and adding a 3rd panel for difference. Also adding another set of results in SH burning season (e.g. September).

-> As your suggestion, we will show the seasonal distribution of MOPITT product and RG model simulation, and their relative difference on 3rd panel as shown in Figure S2.

Fig5: Suggesting removing Fig5. But what are the blue and yellow color?

-> Black, blue, and yellow respectively indicate larger points than 1, 10, 100 falling into each of bins (bin size: 5 ppbv).

Fig11: It is hard to see the trends. I suggest putting all the line plots into two horizontal panels.

-> Thanks for your suggestion. However, since each time series of surface CO by region is so variable with different intensity, it is not effective to show the seasonal cycles and decadal trends of surface CO for all the regions in two horizontal panels. Therefore we would keep the figure as it is.