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

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We thank the reviewer for the constructive and valuable comments, and will revise and improve the manuscript soon as your comments.

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.

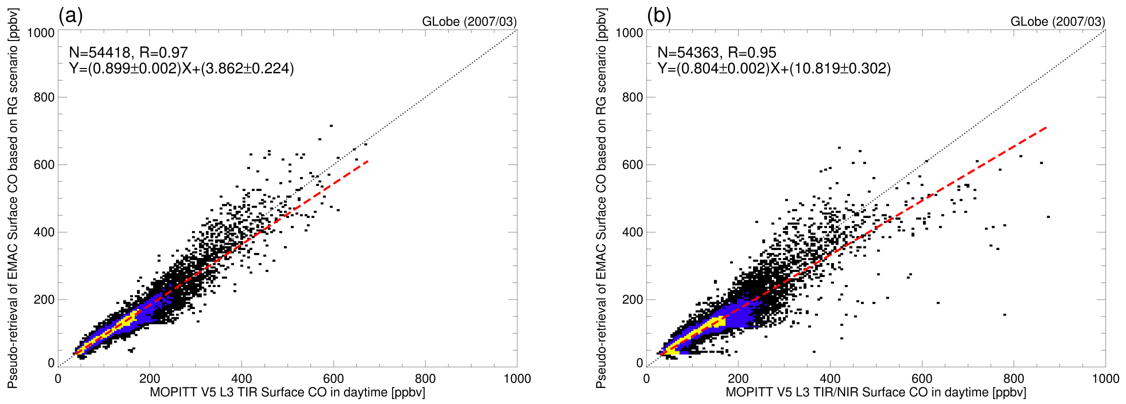

Figure S1. Spatial comparisons of global MOPITT-retrieved (a) TIR and (b) TIR/NIR surface CO with the pseudo-retrievals of EMAC simulations based on RG scenarios on March 2007. Black, blue, and yellow respectively indicate larger points than 1, 10, 100 falling into each of bins (bin size: 5 ppbv).

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.

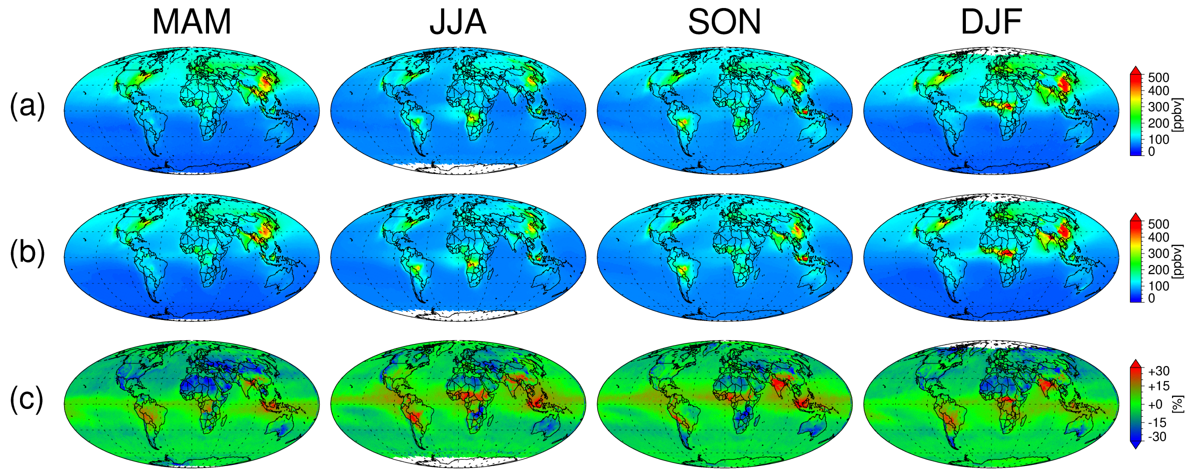

Figure S2. Global distributions of seasonal (a) MOPITT-retrieved surface CO, (b) pseudo-retrievals of EMAC-simulated surface CO based on RG scenarios, and (c) their relative difference from 2001 to 2010.

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.

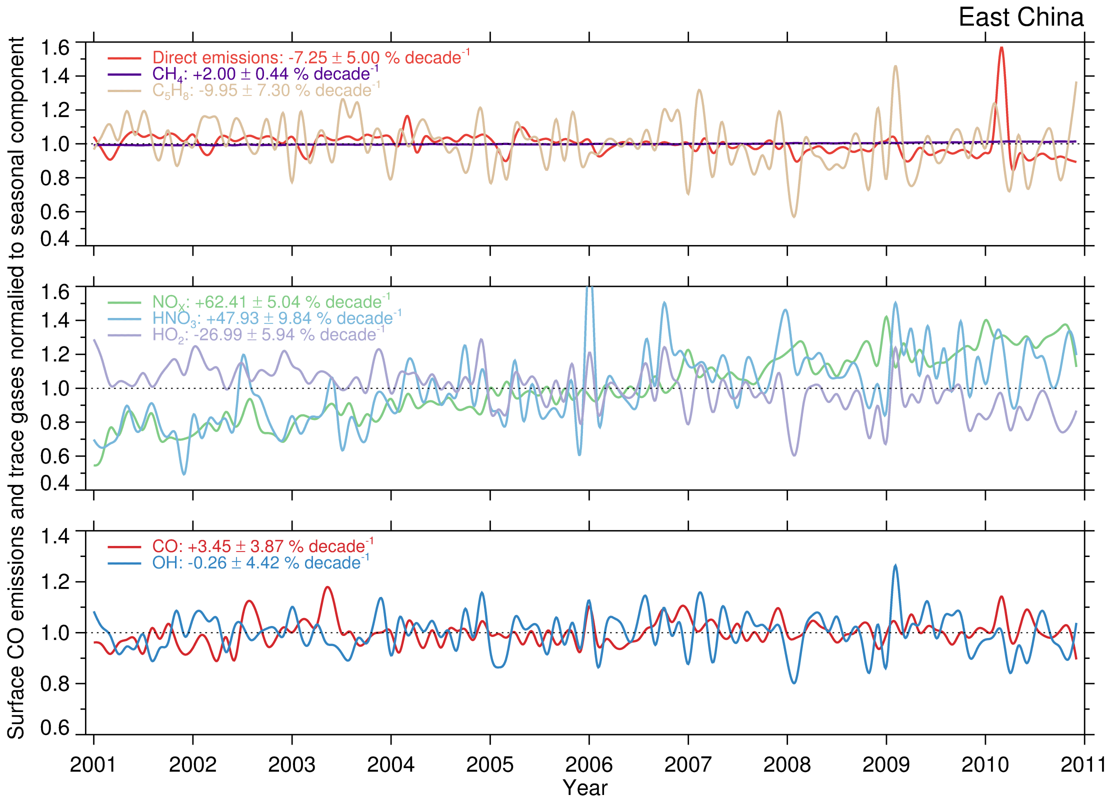

Figure S3. Long-term time series of surface CO emissions and relevant trace gases normalised to seasonal component over East China from 2001 to 2010.

"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.

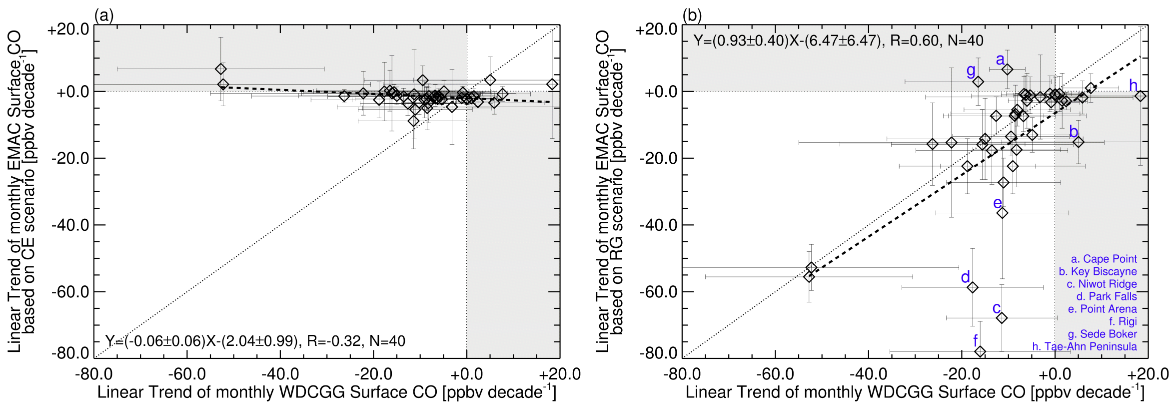

Figure S4. Comparisons of the trends of monthly EMAC-simulated surface CO based on (a) CE and (b) RG scenarios against the trend of monthly archived surface CO with ±2σ errors for selected WDCGG stations listed on Table 2. Detailed values are summarized in Table 4. Some stations (i.e. Cape Point, Key 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 ±2σ.

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.

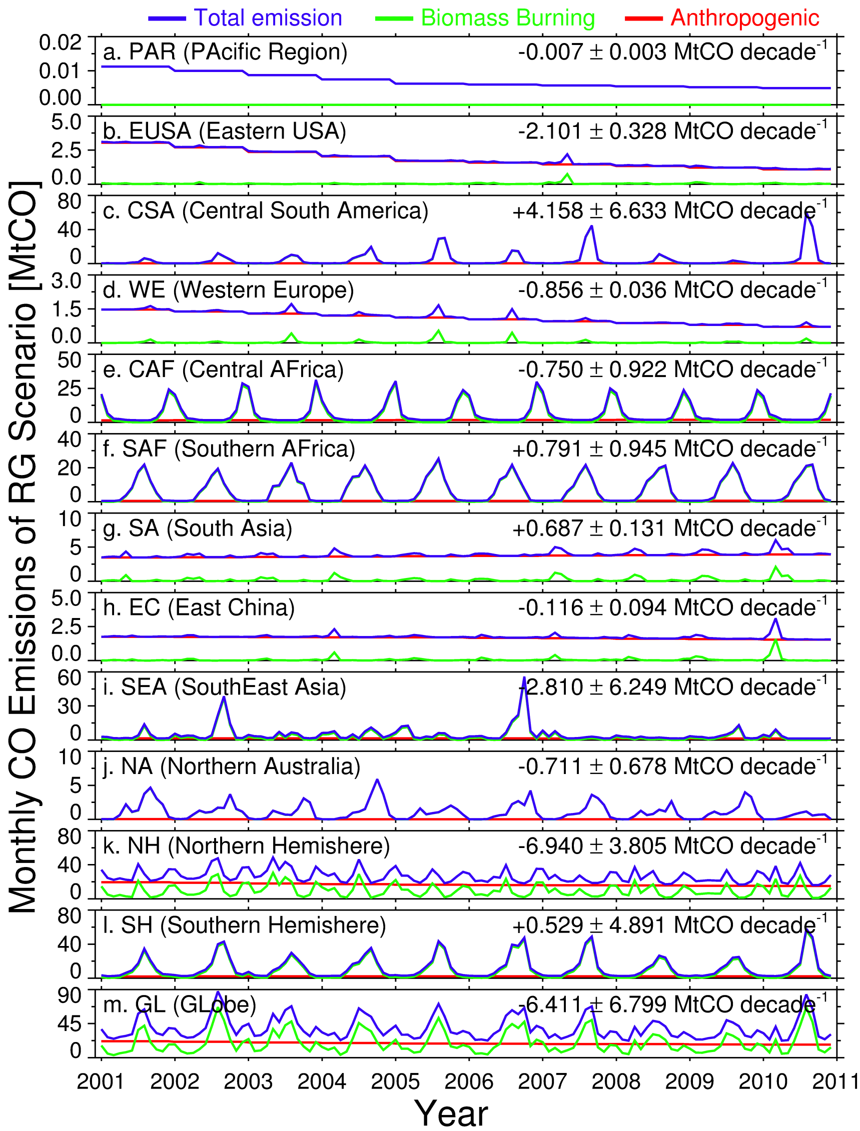

Figure S5. Regional and global trend estimates of monthly RG CO emissions with ±2σ errors from 2001 to 2010.

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.