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

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## Abstract

We present decadal trend estimates of surface carbon monoxide (CO), simulated using the atmospheric chemistry general circulation model ECHAM5/MESSy (EMAC) based on the emission scenarios, Representative Concentration Pathways (RCP) 8.5 for anthropogenic activity and Global Fire Emissions Database (GFED) v3.1 for biomass burning from 2001 through 2010. The spatial distribution of the modelled surface CO is evaluated with monthly Measurements Of Pollution In The Troposphere (MOPITT) thermal infrared product. The global means of correlation coefficient and relative bias for the 2001–2010 are 0.95 and  $-4.29\%$ , respectively. We also find a reasonable correlation ( $R = 0.78$ ) between the trends of EMAC surface CO and full 10 year monthly records from ground-based observation (World Data Centre for Greenhouse Gases, WDCGG). Over Western Europe, Eastern USA, and Northern Australia, the significant decreases of EMAC surface CO are estimated at  $-35.5 \pm 5.8$ ,  $-59.6 \pm 9.1$ , and  $-13.7 \pm 9.5$  ppbv decade<sup>-1</sup>, respectively, with a 95 % confidence interval. In contrast, the surface CO increases by  $+8.9 \pm 4.8$  ppbv decade<sup>-1</sup> over South Asia. A high correlation ( $R = 0.92$ ) between the significant changes in EMAC-simulated surface CO and total emission flux shows that the significant regional trends are attributed to the changes in primary/direct emissions from both anthropogenic activity and biomass burning. In particular, increasing trends of surface hydroxyl radical (OH) partially contribute to the decreasing trends of surface CO in Western Europe and Eastern USA.

## 1 Introduction

Medium-lived and unevenly-mixed carbon monoxide (CO) in the atmosphere is a key tracer in atmospheric chemistry and climate change (Novelli et al., 1992; Forster et al., 2007, IPCC AR4; Duncan and Logan, 2008; Gomez-Pelaez et al., 2013). Major sources of atmospheric CO are fossil fuel combustion and biomass burning on the Earth's surface (Wallace and Hobbs, 2006). CO leads to the formation of tropospheric ozone (O<sub>3</sub>)

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and carbon dioxide (CO<sub>2</sub>) through photochemical and oxidation reactions (Crutzen and Gidel, 1983; Fishman and Crutzen, 1978; Burrows et al., 1995). It also controls the hydroxyl radical (OH) concentration and distribution in unpolluted and non-forested locations (Levy, 1971; Thompson, 1992; Crutzen, 1974; Logan et al., 1981), which influences the oxidation of most trace gases on the Earth (Khalil and Rasmussen, 1990), such as methane (CH<sub>4</sub>) and other pollutants (Lelieveld et al., 2004; Novelli et al., 1992; Thompson and Cicerone, 1986). Finally it contributes to climate change with direct and indirect radiative forcings around 0.024 and 0.2 Wm<sup>-2</sup>, respectively (Forster et al., 2007, IPCC AR4). Monitoring long-term series of surface CO is therefore important for understanding the influence of the direct CO emissions on atmospheric chemistry and indirectly, on climate. Previous studies showed that CO exhibited an increasing trend (worldwide) before the '90s (Khalil and Rasmussen, 1988) and a decreasing trend (Novelli et al., 1994; Law, 1999) thereafter. Duncan et al. (2007) and Duncan and Logan (2008) reported comprehensive results of the global/regional budget of CO and leading causes of its trends and interannual variability from 1988 to 1997. No studies based on the model-simulations are present, which estimate recent changes in global/regional CO since 2000.

Satellite observations allow scientists and researchers to provide the long-term accumulated data of global CO that are suitable for the global trend analysis (Burrows et al., 2011). Representative satellite instruments are Measurements of Pollution in the Troposphere (MOPITT) on NASA's Terra satellite (Drummond and Mand, 1996; Deeter et al., 2003; Worden et al., 2013), Scanning Imaging Absorption Spectrometer for Atmospheric CHartography (SCIAMACHY) on ESA's Envisat satellite (Buchwitz et al., 2000, 2004, 2005, 2007), Atmospheric Infrared Sounder (AIRS) on NASA's Aqua satellite (Susskind et al., 2003; Warner et al., 2007, 2010), Tropospheric Emission Spectrometer (TES) on NASA's Aura satellite (Luo et al., 2007a, b; Ho et al., 2009; Kopacz et al., 2010), and Infrared Atmospheric Sounding Interferometer (IASI) on EUMETSAT's Metop satellite (Clerbaux et al., 2009; George et al., 2009; Klonecki et al., 2012). However, satellite observations are not a direct sampling method, so it is

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impossible to completely eliminate the uncertainty in the CO retrieval (average errors for individual total column CO estimates:  $\pm 5$ – $6$  % for MOPITT,  $\pm 5$  % for SCIAMACHY,  $\pm 10$  % for AIRS,  $\pm 6$ – $7$  % for TES, and  $\pm 5$ – $7$  % for IASI, de Laat et al., 2007; Worden et al., 2013) due to problems in instrument calibration/stability and lack of complementary information (e.g., atmospheric temperature profile, surface pressure/temperature, and cloud content) (Clerbaux et al., 1999; Deeter et al., 2003). In particular, the retrieval algorithm based on climatology has an inevitable uncertainty in trend estimates (Yoon et al., 2013a). The ground-based observations, in contrast, can provide decades-long and highly accurate records using in situ measurement methods, but only for the ground stations available. Therefore, there is a significant limitation to estimate a reliable trend of global and regional surface CO from the study solely based on satellite-retrieved or ground-based data.

In this study, the ECHAM5/MESSy Atmospheric Chemistry (EMAC) model is used to simulate surface CO trends from 2001 to 2010. The anthropogenic emissions are based on the Representative Concentration Pathways (RCP) 8.5 (Rihai et al., 2007) and the biomass burning emissions on the Global Fire Emissions Database (GFED) v3.1 (Giglio et al., 2010; van der Werf et al., 2010). The main objectives of this study are to analyze the long-term trend of global and regional surface CO, simulated using EMAC model, and to compare them to observationally-derived trends. This paper is organized as follows: in Sect. 2, we describe the EMAC model and emission scenarios used for the global surface CO simulations from 2001 to 2010, and the MOPITT and WDCGG observations for the evaluations of spatial distribution and temporal change in the simulated surface CO. In Sect. 3, the model results are spatially and temporally evaluated through comparison with the observational datasets. In Sect. 4, we estimate the regional and global trends in EMAC-simulated surface CO and explore the major causes for the trends by comparing changes in CO direct emissions. In Sect. 5, we investigate the influence of the surface OH change on the surface CO trend and Sect. 6 summarizes and presents our results and conclusions.

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## 2 Model, emission scenarios, and observational data

### 2.1 ECHAM5/MESSy Atmospheric Chemistry (EMAC) model

The EMAC model is a numerical atmospheric chemistry general circulation model (ACGCM) developed for investigating atmospheric processes and their interaction with ocean, land, and human influences (see Jöckel et al., 2010 and publications at <http://www.messy-interface.org/>). It consists of the fifth generation European Centre Hamburg general circulation model (ECHAM5 version 5.3.02) (Roeckner et al., 2006) and the second version of the Modular Earth Submodel System (MESSy2 version 2.42) (Jöckel et al., 2010). The simulation results have been extensively evaluated with surface, aircraft, and satellite observations in many publications, such as Jöckel et al. (2006) and Pozzer et al. (2007, 2009, 2012a, b). In this study, a T63L31 resolution was used, corresponding to a horizontal resolution of approximately  $1.875^\circ$  by  $1.875^\circ$  in latitude and longitude and a vertical resolution of 31 levels from the surface to 10 hPa.

### 2.2 Emission scenarios, RCP 8.5 and GFED v3.1

Several emission scenarios, e.g. IS92 scenarios (Leggett et al., 1992) and Special Report on Emission Scenarios (SRES) (Nakicenovic et al., 2000), have been broadly used for the research on greenhouse gases, air pollutants, and future climate (e.g. Hogrefe et al., 2004; Jacobson and Streets, 2009). However, since they are the no-climate-policy scenarios, it fails to explore the impact of different climate policies (van Vuuren et al., 2011). The fifth Intergovernmental Panel for Climate Change Assessment Report 5 (IPCC-AR5) gives an account of the concentration of greenhouse gases with respect to atmospheric radiation affected by anthropogenic activities (van Vuuren et al., 2011). The Representative Concentration Pathways (RCPs) were developed by four individual modeling groups (i.e., NIES, IIASA, JGCRI, and PBL) (Riahi et al., 2007; Fujino et al., 2006; Hijjoka et al., 2008; Clarke et al., 2007; Smith and Wigley, 2006; Wise et al., 2009; van Vuuren et al., 2006, 2007, 2011). They consist of four emission



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scenarios, also called RCP 2.6, 4.5, 6.0, and 8.5 representing the radiative forcing of anthropogenic activity from 2.6 to 8.5  $\text{Wm}^{-2}$  in 2100, which depend on the mitigation or emission scenarios (van Vuuren et al., 2011). Among them, the emission RCP 8.5 is used in this study to investigate the influence of anthropogenic activity on the change in surface CO from 2001 to 2010. It assumes that the emissions in greenhouse gases continue to increase till post-2100 and their concentrations are stabilized post-2200 (Riahi et al., 2007; van Vuuren et al., 2011; Meinshausen et al., 2011). The RCP 8.5 has been tested in Granier et al. (2011), which showed that it is a “reasonable” choice for anthropogenic emissions after the year 2000. Figure 1a shows the total mean of monthly emission flux of RCP 8.5 from 2001 and 2010. It illustrates that the high CO emissions due to anthropogenic activities are located in highly-populated regions or the largest urban agglomerations (aka megacities).

Fire is a significant emission source of several trace gases and aerosols, including atmospheric CO (Andreae and Merlet, 2001; Giglio et al., 2010). To consider the influence of CO emission from biomass burning, the Global Fire Emissions Database (GFED) v3.1 is used in this study. It is based on global fire emissions from deforestation, savanna, forest, agricultural and peat fires (van der Werf et al., 2010). The version 3 is updated using the combination of the long-term time series of improved satellite-derived data (e.g. burned area, fire activity, and plant productivity from MODIS, Tropical Rainfall Measuring Mission (TRMM), Visible and Infrared Scanner (VIRS), Along-Track Scanning Radiometer (ATSR), and Advanced Very High Resolution Radiometer (AVHRR)) and model-estimated data (fuel loads and combustion completeness using Carnegie-Ames-Stanford-Approach (CASA) biogeochemical model) from 1997 to 2009 (van der Werf et al., 2010). Figure 1b shows the global mean distribution of monthly GFED v3.1 surface CO emissions from 2001 to 2010, and shows that fire activity in/around tropical rainforests leads to large CO emissions. Direct CO emissions from anthropogenic activity and biomass burning represent around 50 % of the total CO budget (Granier et al., 1999; Duncan et al., 2007; Bergamaschi et al., 2000).

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A simulation with constant emission (hereafter, called *CE* scenario) is performed to assess only the possible influence of the meteorological transports on the surface CO trend as shown in Fig. 2a. Emissions in the model simulation CE are kept equal to the year 2000 of RCP 8.5 and GFED v3.1 for all 10 years of the simulation (2001–2010). In addition to the simulation CE, the combination of RCP 8.5 and GFED v3.1 (hereafter, called *RG* scenario) in Fig. 2b is used for simulating a realistic surface CO concentration. It should be noted that in the RCP 8.5, CO emission does slightly decrease (globally) from the beginning of the 21st century (Butler et al., 2012). Finally, chemistry and transport are fully decoupled, so that both simulations have binary identical meteorology (i.e. transport). Additionally, the model has been weakly nudged towards analysis data of the European Centre for Medium-Range Weather Forecast (ECMWF) (Jeuken et al., 1996) up to 100 hPa to obtain realistic model dynamics.

### 2.3 MOPITT Version 5 Level 3 thermal infrared CO

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 tropospheric CO (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). The grey line-shaded regions depicted in Fig. 3 and listed in Table 1, including the globe, Northern, and Southern Hemispheres, are selected for the spatial evaluation and trend estimates. These regions are important to monitor the surface CO released from anthropogenic and fire activities (see Fig. 1).

## 2.4 WDCGG surface CO

The World Data Centre for Greenhouse Gases (WDCGG) under the World Meteorological Organization/Global Atmosphere Watch (WMO/GAW; [http://www.wmo.int/pages/prog/arep/gaw/gaw\\_home\\_en.html](http://www.wmo.int/pages/prog/arep/gaw/gaw_home_en.html)) was established in 1990 by the Japan Meteorological Agency (JMA) to assist in more reliable monitoring and analysing of greenhouse (CO<sub>2</sub>, CH<sub>4</sub>, CFCs, N<sub>2</sub>O, surface ozone, etc.) and related gases (CO, NO<sub>x</sub>, SO<sub>2</sub>, VOC, etc.) (<http://ds.data.jma.go.jp/gmd/wdcgg/introduction.html>). The WDCGG-archived CO data are categorized according to the observation platforms or analytical methods (see more details in GAW Report No. 188, WMO, 2009). The full 10 year monthly records of air sampling observations at the stationary platforms (shown as green dots in Fig. 3) were used to evaluate the temporal trend of EMAC-simulated surface CO. Detailed information about the station's geolocations, measurement methods, and contributors are listed in Table 2.

## 3 Evaluation of EMAC-simulated surface CO

### 3.1 Evaluation of spatial distribution using MOPITT V5 L3 TIR surface CO

Using mostly passive remote sensing instruments (including MOPITT), it is quite challenging to retrieve tropospheric CO profiles because of a significant dependence on atmospheric temperature profile, surface pressure, and surface temperature in the retrieval algorithms (Deeter et al., 2003). In particular, without proper additional information, it is difficult to avoid the systematic error in the retrieved profiles from the algorithm developed that is based on climatology (i.e. a priori CO profiles) (Eskes and Boersma, 2003). This is why the averaging kernels, reflecting the relation between the retrieved and true profiles (Pan et al., 1998; Rodgers, 2000; Deeter et al., 2003), are important for a proper comparison between satellite-retrieved and model-simulated profiles (Rodgers, 2000; Rodger and Connor, 2003; Eskes and Boersma, 2003). The

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EMAC-simulated surface CO can be transformed into a comparable quantity (so called pseudo-retrieval,  $\hat{x}_{EMAC}^{surface}$ ), to the MOPITT-retrieved surface CO as follows:

$$\hat{x}_{EMAC}^{surface} \equiv \hat{x}_{MOPITT}^{surface} + A_{MOPITT}^{surface} \left( x_{EMAC} - \hat{x}_{MOPITT} \right) \quad (1)$$

5 where  $\hat{x}_{MOPITT}^{surface}$ ,  $A_{MOPITT}^{surface}$ ,  $\hat{x}_{MOPITT}$ , and  $x_{EMAC}$ , and represent the MOPITT surface a priori CO, surface averaging kernels matrix, a priori CO profiles, and EMAC-simulated CO profiles from surface to 100 hPa, respectively.

Figure 4 presents the global distributions of the MOPITT surface CO and the pseudo-retrievals based on the EMAC results (CE and RG). Both pseudo-retrievals are similar  
10 to the distribution of the remote-sensed MOPITT surface CO: the high concentration of surface CO emanating from the source regions over the Eastern USA, Western Europe, Central Africa, and South/East Asia is due to the combustion of fossil fuels and biomass burning (Wallace and Hobbs, 2006; Worden et al., 2013), while the transported CO by the atmospheric circulation can be detected over neighbouring areas. To further  
15 analyse a spatial correlation between the global distributions of MOPITT-retrieved and EMAC-simulated surface CO, we compared them directly. The EMAC-simulated surface CO based on the realistic RG scenario obviously shows better agreement with MOPITT surface CO; for example, as in December 2008 (see Fig. 5) (i.e., the spatial correlation coefficient ( $R$ ) and the slope of the linear best-fit line ( $A$ ) are 0.97 and  
20  $1.072 \pm 0.003$ , respectively) than the surface CO based on the CE scenario ( $R = 0.92$  and  $A = 0.771 \pm 0.003$ ).

The Taylor diagrams (Taylor, 2001; Forster et al., 2007; Meehl et al., 2007) in Fig. 6 graphically resume the spatial correlation coefficient ( $R$ ), normalized standard deviation (STD), and normalized centred root-mean-square (RMS) difference for the different  
25 regions (see Fig. 3 and Table 1) and for the globe. These statistical quantities are listed in Table 3. In Fig. 6, the more closely the simulated pattern is located to the “Obs.” on x-axis, the more closely it matches up with the observed spatial pattern. Additionally, the relative bias ( $B$ ) is included in Fig. 6, allowing a more effective comparison

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between the spatial patterns of the monthly EMAC-simulated and MOPITT-observed CO. As mentioned already and shown again in Fig. 6b, the RG simulation results are more consistent with the MOPITT observations (i.e. in most regions,  $R$  is greater than about 0.9, less than about  $\pm 0.25$  of normalized STD, less than about 0.5 of normalized centred RMS difference, and less than about  $\pm 10\%$  of B) than the one based on CE scenario in Fig. 6a. Therefore, we can conclude that the simulation RG agrees well with the MOPITT-observed surface CO in the spatial distribution. Rather poor agreement at the Pacific Region (PAR) can be explained by the biases in the MOPITT CO surface retrievals at cleaner locations, such as over the Pacific Ocean (Emmons et al., 2004) and 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.

### 3.2 Evaluation of temporal change using WDCGG surface CO

In this section, the WDCGG surface CO observations at the stationary sites (see Fig. 3 and Table 2) are used to evaluate the temporal changes of EMAC simulations. We applied the monthly time series ( $Y_t$ ) of EMAC-simulated and WDCGG-archived data to a linear trend model. The following form of a typical linear model has been adopted in various studies (Zhao et al., 2008; Hsu et al., 2012; de Meij et al., 2012; Yoon et al., 2011, 2012, 2013a, b) for estimating climatological changes in the atmospheric system (Weatherhead et al., 1998, 2002).

$$Y_t = \mu + \omega X_t + S_t + N_t \quad (2)$$

where  $\mu$ ,  $\omega$ , and  $X_t$  denote the constant, the magnitude of the trend per year, and the time index term ( $t/12$ ), respectively.  $S_t$  is a seasonal component fitted using Fourier analysis as follows:

$$S_t = \sum_{j=1}^4 [\beta_{1,j} \sin(2\pi jt/12) + \beta_{2,j} \cos(2\pi jt/12)] \quad (3)$$

$N_t$  is the unexplained noise term, which is often assumed to be autocorrelated with one time lag (Weatherhead et al., 1998, 2002) as follows:

$$N_t = \varphi N_{t-1} + \varepsilon_t \quad (4)$$

where  $\varphi$  is the autocorrelation coefficient ( $-1 < \varphi < 1$ ) and  $\varepsilon_t$  is an independent random variable. If  $\hat{\omega}$  denotes the trend estimate in Eq. (2), determined by minimizing the chi-square error statistic, the standard deviation ( $\sigma_{\hat{\omega}}$ ) of the trend estimate can be quite accurately approximated as follows:

$$\sigma_{\hat{\omega}} \approx \frac{\sigma_{\varepsilon}}{(1-\varphi)} \frac{1}{n^{3/2}} = \frac{\sigma_N}{n^{3/2}} \sqrt{\frac{1+\varphi}{1-\varphi}} \quad (5)$$

where  $\sigma_{\varepsilon}$ ,  $\sigma_N$ , and  $n$  denote the standard deviation of  $\varepsilon$  and  $N$ , and the number of years, respectively. In this study, we define  $\hat{\omega}$  as a statistically significant trend at a 95% confidence level when  $|\hat{\omega}/\sigma_{\hat{\omega}}|$  is larger than two (Tiao et al., 1990; Weatherhead et al., 1998). This method is strictly applied to the full 10 year monthly records to minimize statistical biases from data inconsistencies in the trend estimates. In the same manner, the trends are derived from the EMAC-simulated surface CO data based on CE and RG scenarios at the closest grid to the WDCGG stations (see Table 2).

The trends of EMAC-simulated surface CO based on the RG scenario show better agreement and higher correlation (i.e., the correlation coefficient ( $R$ ) and the slope of linear best-fit line ( $A$ ) are 0.60 and  $0.93 \pm 0.40$  in Fig. 7b) with the WDCGG trend than the ones based on the CE scenario ( $R = -0.32$  and  $A = -0.06 \pm 0.06$  in Fig. 7a). The difference is more clearly shown in the comparison between only significant trends ( $R = 0.94$  and  $A = 0.20 \pm 0.11$  in Fig. 7a, and  $R = 0.70$  and  $A = 1.02 \pm 0.93$  in Fig. 7b). The specific values of trend estimates and statistical quantities are summarized in Table 4. At 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, the trends of EMAC-simulated surface CO based on the RG scenario

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are considerably different to the WDCGG trends. It is attributed to the WDCGG flask sampling method to minimize the contamination from local pollution (Haas-Laurse and Hartley, 1997) and the rather low resolution of the EMAC model grid, which cannot discriminate the local sources (Pozzer et al., 2007). Therefore, for such stations, we have not used the closest grid of EMAC-simulated surface CO to the WDCGG stations, but a model grid-box to the upwind direction as suggested in Pozzer et al. (2007). Again, Fig. 8 presents the comparison between the trends of EMAC-simulated surface CO based on the RG scenario and WDCGG-archived surface CO. A better correlation coefficient and slope of linear best-fit line are obtained ( $R = 0.78$  and  $A = 0.94 \pm 0.25$ ). Therefore, we can conclude that the long-term simulations based on the RG scenario provide statistically reliable trend estimates of global surface CO.

## 4 Global, latitudinal, and regional trend estimates of EMAC-simulated surface CO

### 4.1 Global and latitudinal scales

After the Mt. Pinatubo eruption on June 1991, the global surface CO from 1991 to 2001 decreased by  $-0.52 \text{ ppbvyr}^{-1}$  (Novelli et al., 2003). It is attributed to the aftereffect of the eruption (Dlugokencky et al., 1996) and the subsequent increase in surface atmospheric OH (Bekki et al., 1994). Novelli et al. (2003) reported the decrease of CO ( $-1.4 \text{ ppbvyr}^{-1}$ ) in the northern extra-tropics caused by the decrease in anthropogenic emissions in the Northern Hemisphere from 1991 to 2001, but there was no significant trend in the Southern Hemisphere. Similarly, Duncan and Logan (2008) found that the decrease in European emissions leads to a decreasing trend ( $-0.85 \text{ \%yr}^{-1}$ ) in northern extra-tropics from 1988 to 1997. Worden et al. (2013) showed that the entire CO column still has a small negative trend in recent years (below  $1 \text{ \%yr}^{-1}$ ), from 2000 through 2011, with a strong decrease of CO over the US, Europe and China, although it is not clear what is behind such trends.

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Figure 9 presents the global trend of EMAC-simulated surface CO based on the CE and RG scenarios. The trends estimated in simulation CE could be purely attributed to dynamical changes as emissions and long-lived species are constant throughout the entire simulation. These trends (Fig. 9a) are statistically significant in the Southern Hemisphere and their magnitudes are relatively small. These are nevertheless not significant when changing emissions are included (simulation RG, Fig. 9b). In the realistic simulation RG (Fig. 9b), generally decreasing trends are predominant, in particular over Europe and the Eastern USA as a result of strict environmental regulation (Streets et al., 2003; Yoon et al., 2011, 2012, 2013b; Hilboll et al., 2013), and Southeast Asia due to less fire activity (Giglio et al., 2010). They are consistent with the WDCGG-archived trends (see Sect. 3.2) as well as the decreasing trends in total CO columns retrieved from MOPITT and AIRS satellite instruments (MOPITT:  $-1.44 \pm 0.44 \text{ \% yr}^{-1}$  for Europe and  $-1.42 \pm 0.40 \text{ \% yr}^{-1}$  for the Eastern USA, AIRS:  $-1.00 \pm 0.66 \text{ \% yr}^{-1}$  and  $-0.96 \pm 0.36 \text{ \% yr}^{-1}$ , with  $\pm 2\sigma$  errors, respectively) (Worden et al., 2013). In contrast, upward trends are estimated over the quickly developing countries and the forested regions (i.e., around South Asia, East China, Central South America, Central/Southern Africa). They can be attributed to the increases of anthropogenic and fire activities (Burrrows et al., 1995; Bovensmann et al., 1999; Richter et al., 2005; Giglio et al., 2010). More significant signals in the trend estimates are found in the Northern Hemisphere than Southern Hemisphere. These differences are clearly shown in Fig. 10, which depicts the latitudinal means and corresponding trends of the EMAC-simulated surface CO from  $90^\circ \text{ S}$  to  $90^\circ \text{ N}$  at  $1.875^\circ$  intervals. The mixing ratio of surface CO is definitely higher in the Northern Hemisphere where about 90 % of the human population resides. Finally, in simulation CE (Fig. 10a), almost no change in the EMAC-simulated surface CO is estimated over all latitudinal ranges. Contrariwise, in simulation RG, the surface CO shows a significant decreasing trend (Fig. 10b), especially between  $30^\circ \text{ N}$  to  $60^\circ \text{ N}$  the mean trend with  $\pm 2\sigma$  errors is  $-19.46 \pm 11.79 \text{ ppbv decade}^{-1}$ . Novelli et al. (2003) and Duncan and Logan (2008) also reported a similar tendency in northern extra-tropics influenced by the decrease in anthropogenic emissions in the North-

ern Hemisphere for the decade 1988–2001. At lower latitudes (i.e. less than 30° N), the same simulation shows no significant trends. These latitudinal tendencies generally show good agreement with the significant trends of WDCGG-archived data except the one at the stations Hegyhatsal and Payerne located in Eastern Europe where the downward trend (i.e.  $-52.26 \pm 31.63$  and  $-52.76 \pm 22.21$  ppbvdecade<sup>-1</sup>, respectively) is relatively high compared to the same latitudinal trends (see Table 4).

## 4.2 Regional scale

The sources of tropospheric CO are in situ oxidations and direct emissions: primarily oxidation of CH<sub>4</sub> ( $\sim 800$  Tg(CO)yr<sup>-1</sup>) and emissions from biomass burning ( $\sim 700$  Tg(CO)yr<sup>-1</sup>) and fossil/domestic fuel ( $\sim 650$  Tg(CO)yr<sup>-1</sup>) (Bergamaschi et al., 2000; WMO, 1999; IPCC, 1996; Ehhalt et al., 2001). In particular, only the direct emissions account for around 50% of the total CO budget (Ehhalt et al., 2001). In this section, we estimate the surface CO trends by the region and compare them with the changes in the direct emissions.

Figure 11 shows the regional trends of EMAC-simulated surface CO for the selected regions (see Table 1 and Fig. 3), and Fig. 12 shows the corresponding changes in the emissions from anthropogenic activity and the biomass burning dataset (i.e. RCP 8.5 and GFED v3.1).

The PACific Region (PAR) is a remote area over open oceans and is the focus of many studies on climate change (e.g. Trenberth et al., 2002; Latif and Keenlyside, 2009; Rieder et al., 2013) since it is sensitive to El Niño and La Niña-Southern Oscillation (Rasmusson and Carpenter, 1982). Although PAR is almost not influenced by human activity, shipping transport in this region still plays a role on the CO concentration. The significant trend of EMAC surface CO is estimated ( $-5.8 \pm 5.5$  ppbvdecade<sup>-1</sup>) and consistent with significant trend of monthly anthropogenic emission ( $-0.007 \pm 0.003$  MtCOdecade<sup>-1</sup>).

The Eastern USA (EUSA) and Western Europe (WE) are highly industrialized countries/regions (Zhang et al., 2012; Yoon et al., 2011), where anthropogenic CO

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emissions are predominant (see Fig. 1a). An occasional influence from biomass burning is found, as shown in Fig. 12. As a result of environmental regulations over these regions during past decades (Streets et al., 2003; Yoon et al., 2011, 2012, 2013b), the decreasing trends of atmospheric aerosol and short-lived trace gases have been reported in many studies (e.g. Smith et al., 2001; Streets et al., 2006; Richter et al., 2005; Hilboll et al., 2013). The dramatic decreases of EMAC surface CO are estimated ( $-59.6 \pm 9.1$  ppbvdecade<sup>-1</sup> for EUSA and  $-35.5 \pm 5.8$  ppbvdecade<sup>-1</sup> for WE) and are compatible (see Fig. 12) with the significant change in monthly emissions ( $-2.101 \pm 0.328$  MtCOdecade<sup>-1</sup> for EUSA and  $-0.856 \pm 0.036$  MtCOdecade<sup>-1</sup> for WE). These results are comparable to Worden et al. (2013) and Angelbratt et al. (2011), which derived the downward trends in MO-PITT total CO column ( $(-3.14 \pm 0.88) \times 10^{16}$  moleculescm<sup>-2</sup>yr<sup>-1</sup> for Eastern USA and  $(-3.03 \pm 0.92) \times 10^{16}$  moleculescm<sup>-2</sup>yr<sup>-1</sup> for Europe) and solar FTIR partial CO column (i.e.  $\sim 0$ –15 km) ( $-0.45 \pm 0.16$  %yr<sup>-1</sup>,  $-1.00 \pm 0.24$  %yr<sup>-1</sup>,  $-0.62 \pm 0.19$  %yr<sup>-1</sup> and  $-0.61 \pm 0.16$  %yr<sup>-1</sup> at the ground-based stations Jungfraujoch, Zugspitze, Harestua, and Kiruna over Europe).

Central South America, Central/Southern Africa, SouthEast Asia, and Northern Australia (i.e. CSA, CAF, SAF, SEA, and NA) are representative for the tropical rainforests (Ahlm et al., 2009; Eltahir and Bras, 1996; Li and Fu, 2004; Nepstad et al., 1999; Held et al., 2005; Chung and Ramanathan, 2006; Facchini et al., 2000; McFiggans et al., 2005, 2006). Over these regions, a large amount of surface CO has been emitted by biomass burning (see Figs. 1b and 12) through deforestation caused by land use, subsistence agriculture, and spontaneous combustion in warm and dry seasons (Reeves et al., 2010; Johnson et al., 2008; Kirby et al., 2006; Davidson and Artaxo, 2004). Insignificant trends in EMAC-simulated surface CO are estimated over these regions ( $+30.0 \pm 53.4$  ppbvdecade<sup>-1</sup> for CSA,  $+2.2 \pm 7.4$  ppbvdecade<sup>-1</sup> for CAF,  $+7.2 \pm 10.5$  ppbvdecade<sup>-1</sup> for SAF, and  $-21.3 \pm 39.1$  ppbvdecade<sup>-1</sup> for SEA) except in NA ( $-13.7 \pm 9.5$  ppbvdecade<sup>-1</sup>) because of the strong interannual variability of biomass burning emission as shown in Fig. 12.

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China and India are highly-populated and developing countries occupying about 36.5 % of the world population in 2013 ([http://en.wikipedia.org/wiki/List\\_of\\_countries\\_by\\_population](http://en.wikipedia.org/wiki/List_of_countries_by_population)) and 14.1% of world GDP in 2012 (World Bank Data, <http://data.worldbank.org/>). As a consequence, large amounts of anthropogenic pollutants (e.g. aerosols, nitrogen dioxide, sulfur dioxide, and surface ozone) that lead to environmental and health problems are emitted into the atmosphere (Ohara et al., 2007; Pozzer et al., 2012a, 2012b; Lelieveld et al., 2013). The long-term change in these pollutants has been a key issue in many studies (e.g. Yoon et al., 2012, 2013b; Richter et al., 2005; Hilboll et al., 2013; Lu et al., 2010; Xu et al., 2008). The increases of EMAC surface CO are estimated in both regions ( $+8.9 \pm 4.8$  ppbvdecade<sup>-1</sup> for SA and  $+9.1 \pm 9.7$  ppbvdecade<sup>-1</sup> for EC). In the case of EC, since the trend is not statistically significant and is opposite to the decreasing trend in the emission ( $-0.116 \pm 0.094$  MtCOdecade<sup>-1</sup>), it is influenced by the changes in transports or secondary chemical production (Tohjima et al., 2014; Angelbratt et al., 2011).

On a global scale, the simulation RG shows a significant trend only in the NH ( $-13.5 \pm 11.1$  ppbvdecade<sup>-1</sup> for the NH,  $-0.8 \pm 6.7$  ppbvdecade<sup>-1</sup> for the SH, and  $-7.2 \pm 7.8$  ppbvdecade<sup>-1</sup> for the GL). Notably, the evident change in NH CO is attributed to the significant change in emissions ( $-6.940 \pm 3.805$  MtCOdecade<sup>-1</sup>). Figure 13 clearly shows a high correlation between the trends in EMAC-simulated surface CO and total emission flux ( $R = 0.88$  between all trends and  $R = 0.92$  between significant trends). Since the trends of WDCGG-archived surface CO are highly correlated with the EMAC-simulated trends as shown in Fig. 8, we can confirm that the changes in surface CO over the past decade are mostly due to changes in the emissions.

## 5 Influence of surface OH trend

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



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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). It is hence important to investigate the potential influence of change in surface CO and OH and their mutual relationship. Figure 14 shows the comparisons between the EMAC-simulated trends of surface OH and CO based on RG scenario by regions. Any changes in surface CO would cause also some OH trend, as they are correlated with each other. However, Fig. 14 shows that the slope of the correlation does depend on the  $\text{NO}_x$  (nitric oxide and nitrogen dioxide) concentration and trends. In fact, with the exception of regions with very low  $\text{NO}_x$  concentration (i.e. (j) Northern Australia), CO trends and OH trends are highly correlated (see blue and red dashed lines). The blue line shows the regression line for regions with simultaneous CO and  $\text{NO}_x$  decrease (i.e., (b) Eastern USA, (d) Western Europe, and (k) Northern Hemisphere); In this case, the CO decrease is connected with a OH decrease, as expected. Additionally, the decrease of  $\text{NO}_x$  also limits further the potential recycling of  $\text{HO}_2$  to OH, hence enhancing the influence of CO changes on the OH levels. On the hand, the regions (e) Central Africa, (f) Southern Africa, (g) South Asia, and (h) East China have significant increasing trend in surface  $\text{NO}_x$ . These increasing trends enhance the OH recycling from  $\text{HO}_2$  to OH so that the increase of CO does not correspond to a decrease of OH (Lelieveld et al., 2002). Rather, in this case the OH levels are mostly controlled by the OH recycling and its reaction with other pollutants (e.g.,  $\text{NO}_x$ , sulphur dioxide, methane, and ammonia) (Lelieveld et al., 2002, 2004; Novelli et al., 1992; Thompson and Cicerone, 1986). Therefore, we can conclude that the CO trends in the regions (b) Eastern USA, (d) Western Europe, and (k) Northern Hemisphere do also influence surface OH only because of the simultaneous reduction of  $\text{NO}_x$ . On the other side, the increase of CO in developing countries such as (g) South Asia and (h) East China is also accompanied by an increase of other pollutants, such as  $\text{NO}_x$  which, in turn, provide an efficient channel for the OH recycling and therefore a reduction of OH is not observed, but rather an increase.

## 6 Summary and conclusion

The global and regional changes in surface CO have been estimated using the EMAC model from 2001 to 2010. The spatial distributions and temporal changes in the EMAC-simulated surface CO based on CE and RG scenarios have been extensively evaluated with results derived from MOPITT-retrieved and WDCGG-archived data. We have shown that the spatial distribution and temporal change of EMAC-simulated surface CO based on the RG scenario are consistent with the observational datasets. Significant trends of the EMAC surface CO have been found in the Northern Hemisphere (confirming the decreasing trends already reported in the northern extra-tropics in the decade 1988–2001), in particular, a decreasing trend over the Eastern USA, Western Europe, and an increasing trend over South Asia, mostly due to the changes of anthropogenic emission. Additionally, the increasing trend of surface OH contributes to the decrease of surface CO in these regions. In contrast, over the regions influenced by biomass burning (i.e. Central South America, Central Africa, Southern Africa, and Southeast Asia), no significant trend has been detected because of a high interannual variability of fire activity.

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**Table 1.** Geolocation and abbreviation of the regions for evaluating the spatial distribution and estimating the regional/global trend of EMAC-simulated surface CO.

Region	Abbreviation	Latitude range	Longitude range
a. Pacific Region	PAR	10–30° N	165–145° W
b. Eastern USA	EUSA	25–45° N	90–70° W
c. Central South America	CSA	18–5° S	80–35° W
d. Western Europe	WE	35–60° N	10–15° E
e. Central Africa	CAF	3–15° N	18–53° E
f. Southern Africa	SAF	20–3° S	8–42° E
g. South Asia	SA	5–33° N	65–92° E
h. East China	EC	22–43° N	95–124° E
i. SouthEast Asia	SEA	10° S–20° N	95–120° E
j. Northern Australia	NA	20–11° S	120–150° E
k. Northern Hemisphere	NH	0–90° N	180° W–180° E
l. Southern Hemisphere	SH	90° S–0° N	180° W–180° E
m. GLOBE	GL	90° S–90° N	180° W–180° E

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**Table 2.** WDCGG stations providing full 10 year monthly records of surface CO from 2001 to 2010. The WDCGG-archived data are used for the evaluation of trend estimates in this study.

Station name	Latitude	Longitude	Country/Territory	Contributor <sup>a</sup>	Measurement Method <sup>b</sup>
Alert	82.5° N	62.5° W	Canada	CSIRO	RGD
Ascension Island	7.9° S	14.4° W	UK	NOAA/ESRL	GC-HgO
Assekrem	23.3° N	5.6° E	Algeria	NOAA/ESRL	GC-HgO
Barrow	71.3° N	156.6° W	USA	NOAA/ESRL	GC-HgO
Cape Ferguson	19.3° S	147.1° E	Australia	CSIRO	RGD
Cape Grim	40.7° S	144.7° E	Australia	CSIRO	RGD
Cape Grim	40.7° S	144.7° E	Australia	NOAA/ESRL	GC-HgO
Cape Point	34.4° S	18.5° E	South Africa	SAWS	GC-other
Casey Station	66.3° S	110.5° E	Australia	CSIRO	RGD
Halley Bay	75.6° S	26.5° W	UK	NOAA/ESRL	GC-HgO
Hegyhatsal	47.0° N	16.7° E	Hungary	NOAA/ESRL	GC-HgO
Heimaey	63.4° N	20.3° W	Iceland	NOAA/ESRL	GC-HgO
Izafña (Tenerife)	28.3° N	16.5° W	Spain	NOAA/ESRL	GC-HgO
Key Biscayne	25.7° N	80.2° W	USA	NOAA/ESRL	GC-HgO
Mace Head	53.3° N	9.9° W	Ireland	AGAGE	GC-MD
Mace Head	53.3° N	9.9° W	Ireland	NOAA/ESRL	GC-HgO
Macquarie Island	54.5° S	159.0° E	Australia	CSIRO	RGD
Mahe Island	4.7° S	55.2° E	Seychelles	NOAA/ESRL	GC-HgO
Mauna Loa	19.5° N	155.6° W	USA	NOAA/ESRL	GC-HgO
Mt. Waliguan	36.23° N	100.9° E	China	NOAA/ESRL, CMA	GC-HgO

<sup>a</sup> CSIRO: Commonwealth Scientific and Industrial Research Organisation (<http://www.csiro.au/>), NOAA/ESRL: National Oceanic and Atmospheric Administration/Earth System Research Laboratory (<http://www.esrl.noaa.gov/>), SAWS: South African Weather Service (<http://www.weathersa.co.za/web/index.php>), AGAGE: Advanced Global Atmospheric Gases Experiment (<http://agage.eas.gatech.edu/>), CMA: China Meteorological Administration (<http://www.cma.gov.cn/en/>), JMA: Japan Meteorological Agency (<http://www.jma.go.jp/jma/indexe.html>), Empa: Swiss Federal Laboratories for Materials Science and Technology (German acronym for Eidgenössische Materialprüfungs- und Forschungsanstalt) (<http://www.empa.ch/>)

<sup>b</sup> RGD: Reduction Gas Detector, GC-HgO: Gas Chromatography – Mercuric Oxide Reduction Detection, GC-MD: Gas Chromatography – MultiDetector, GC-other: Gas Chromatography (other), NDIR: Non-Dispersive InfraRed gas analyzer

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**Table 2.** Continued.

Station name	Latitude	Longitude	Country/Territory	Contributor <sup>a</sup>	Measurement Method <sup>b</sup>
Niwot Ridge (T-van)	40.1° N	105.6° W	USA	NOAA/ESRL	GC-HgO
Park Falls	45.9° N	90.3° W	USA	NOAA/ESRL	GC-HgO
Palmer Station	64.9° S	64.0° W	USA	NOAA/ESRL	GC-HgO
Payerne	46.8° N	7.0° E	Switzerland	Empa	NDIR
Point Arena	39.0° N	123.7° W	USA	NOAA/ESRL	GC-HgO
Ragged Point	13.2° N	59.4° W	Barbados	NOAA/ESRL	GC-HgO
Rigi	46.1° N	8.5° E	Switzerland	Empa	NDIR
Ryori	39.0° N	141.8° E	Japan	JMA	RGD
Sand Island	28.2° N	177.4° W	USA	NOAA/ESRL	GC-HgO
Sede Boker	31.1° N	34.9° E	Israel	NOAA/ESRL	GC-HgO
South Pole	90.0° S	24.8° W	USA	CSIRO	RGD
South Pole	90.0° S	24.8° W	USA	NOAA/ESRL	GC-HgO
Syowa Station	69.0° S	39.6° E	Japan	NOAA/ESRL	GC-HgO
Tae-ahn Peninsula	36.7° N	126.1° E	Republic of Korea	NOAA/ESRL	GC-HgO
Tudor Hill	32.3° N	64.9° W	UK	NOAA/ESRL	GC-HgO
Tutuila (Cape Matatula)	14.2° S	170.6° W	USA	NOAA/ESRL	GC-HgO
Ulaan Uul	44.5° N	111.1° E	Mongolia	NOAA/ESRL	GC-HgO
Wendover	39.9° N	-113.7° W	USA	NOAA/ESRL	GC-HgO
Yonagunijima	24.5° N	123.0° E	Japan	JMA	RGD
Zeppelinfjellet (Ny-Alesund)	79.0° N	11.9° E	Norway	NOAA/ESRL	GC-HgO

<sup>a</sup> CSIRO: Commonwealth Scientific and Industrial Research Organisation (<http://www.csiro.au/>), NOAA/ESRL: National Oceanic and Atmospheric Administration/Earth System Research Laboratory (<http://www.esrl.noaa.gov/>), SAWS: South African Weather Service (<http://www.weathersa.co.za/web/index.php>), AGAGE: Advanced Global Atmospheric Gases Experiment (<http://agage.eas.gatech.edu/>), CMA: China Meteorological Administration (<http://www.cma.gov.cn/en/>), JMA: Japan Meteorological Agency (<http://www.jma.go.jp/jma/indexe.html>), Empa: Swiss Federal Laboratories for Materials Science and Technology (German acronym for Eidgenössische Materialprüfungs- und Forschungsanstalt) (<http://www.empa.ch/>)

<sup>b</sup> RGD: Reduction Gas Detector, GC-HgO: Gas Chromatography – Mercuric Oxide Reduction Detection, GC-MD: Gas Chromatography – MultiDetector, GC-other: Gas Chromatography (other), NDIR: Non-Dispersive InfraRed gas analyzer

**Table 3.** Means ( $\overline{\text{CO}}$ ) and corresponding statistic quantities\* used in the comparison of spatial patterns (i.e. Taylor diagram in Figure 6.) between MOPITT-retrieved surface CO and pseudo-retrievals of EMAC-simulated surface CO.

Region	MOPITT-retrieved surface CO		Pseudo-retrieval of EMAC-simulated surface CO based on CE scenario				
	$\overline{\text{CO}}$ [ppbv]	$\sigma$ [ppbv]	$\overline{\text{CO}}$ [ppbv]	$\sigma$ [ppbv]	$R$ [unitless]	RMS [ppbv]	$B$ [%]
PAR	91.78 ± 33.49	7.32 ± 5.28	78.05 ± 15.88	3.28 ± 2.82	0.45 ± 0.63	6.53 ± 5.86	-14.97 ± 15.72
EUSA	212.59 ± 83.20	68.62 ± 20.38	156.62 ± 24.81	56.16 ± 23.11	0.83 ± 0.23	37.81 ± 30.95	-26.33 ± 23.90
CSA	113.29 ± 78.23	38.85 ± 66.01	111.38 ± 56.94	35.71 ± 50.32	0.92 ± 0.12	15.59 ± 23.14	-1.68 ± 16.39
WE	159.28 ± 47.53	30.35 ± 18.35	127.78 ± 25.46	27.74 ± 22.21	0.67 ± 0.42	23.56 ± 16.71	-19.78 ± 12.39
CAF	144.80 ± 89.62	48.61 ± 66.40	145.30 ± 88.66	63.57 ± 102.14	0.89 ± 0.15	30.55 ± 41.63	+0.35 ± 18.07
SAF	131.09 ± 83.03	45.83 ± 49.46	129.09 ± 61.08	44.95 ± 46.05	0.77 ± 0.36	30.86 ± 34.40	-1.53 ± 23.09
SA	148.30 ± 97.91	47.01 ± 30.33	125.09 ± 63.00	37.44 ± 25.01	0.87 ± 0.12	23.52 ± 20.34	-15.65 ± 19.05
EC	251.16 ± 97.49	105.37 ± 51.19	176.03 ± 49.78	77.18 ± 39.77	0.74 ± 0.14	70.42 ± 39.08	-29.91 ± 14.74
SEA	132.01 ± 52.60	61.51 ± 50.05	133.78 ± 33.34	57.43 ± 44.83	0.89 ± 0.12	27.85 ± 29.05	+1.34 ± 19.27
NA	90.60 ± 38.02	17.15 ± 14.18	89.12 ± 15.11	13.32 ± 7.23	0.82 ± 0.19	9.84 ± 11.35	-1.64 ± 26.81
NH	131.86 ± 40.93	48.95 ± 22.27	111.97 ± 24.17	39.46 ± 23.05	0.81 ± 0.13	28.90 ± 16.72	-15.08 ± 10.53
SH	67.89 ± 21.01	22.62 ± 19.06	73.08 ± 13.31	23.83 ± 13.33	0.86 ± 0.10	12.45 ± 6.67	+7.64 ± 13.67
GL	99.80 ± 13.54	50.98 ± 22.70	92.30 ± 6.52	38.52 ± 16.30	0.87 ± 0.06	25.99 ± 13.24	-7.51 ± 7.05

Region	Pseudo-retrieval of EMAC-simulated surface CO based on RG scenario				
	$\overline{\text{CO}}$ [ppbv]	$\sigma$ [ppbv]	$R$ [unitless]	RMS [ppbv]	$B$ [%]
PAR	86.66 ± 25.64	4.68 ± 4.47	0.69 ± 0.56	5.28 ± 3.23	-5.58 ± 10.06
EUSA	197.96 ± 48.06	66.63 ± 16.25	0.96 ± 0.04	19.97 ± 12.95	-6.88 ± 17.85
CSA	121.76 ± 82.84	45.99 ± 74.00	0.91 ± 0.13	19.14 ± 25.80	+7.48 ± 12.93
WE	146.77 ± 36.01	28.22 ± 17.63	0.90 ± 0.12	13.15 ± 6.85	-7.85 ± 7.67
CAF	154.74 ± 89.55	63.99 ± 96.87	0.93 ± 0.09	26.33 ± 32.73	+6.86 ± 18.10
SAF	127.93 ± 67.17	47.06 ± 53.73	0.89 ± 0.12	21.34 ± 24.41	-2.41 ± 17.74
SA	164.62 ± 107.02	64.56 ± 46.64	0.94 ± 0.05	25.36 ± 21.47	+11.00 ± 16.06
EC	244.31 ± 83.70	107.22 ± 57.33	0.91 ± 0.06	45.32 ± 21.73	-2.73 ± 14.23
SEA	144.45 ± 47.70	72.66 ± 76.01	0.94 ± 0.06	25.68 ± 46.89	+9.42 ± 15.39
NA	91.07 ± 31.94	16.06 ± 13.43	0.84 ± 0.22	9.43 ± 6.72	+0.51 ± 17.14
NH	125.14 ± 33.00	48.63 ± 27.50	0.93 ± 0.03	18.43 ± 8.82	-5.10 ± 7.94
SH	66.16 ± 19.04	25.44 ± 21.77	0.94 ± 0.06	9.03 ± 9.26	-2.54 ± 5.23
GL	95.52 ± 8.53	49.72 ± 20.84	0.95 ± 0.03	15.49 ± 5.17	-4.29 ± 5.58

\* The statistic quantities are the mean standard deviation ( $\sigma$ ), spatial correlation coefficient ( $R$ ), centred root-mean-square (RMS) difference, and relative bias ( $B$ ) with a 95% confidence interval.

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**Table 4.** Trend estimate ( $\hat{\omega}$ ) and corresponding significance value ( $|\hat{\omega}/\sigma_{\hat{\omega}}|$ ) of the WDCGG-archived and EMAC-simulated surface CO at a 95 % confidence level.

Station	Contributor	WDCGG-archived surface CO		EMAC-simulated surface CO on CE scenario		EMAC-simulated surface CO on RG scenario	
		$\hat{\omega} \pm 2\sigma_{\hat{\omega}}$ [ppbv decade <sup>-1</sup> ]	$ \hat{\omega}/\sigma_{\hat{\omega}} $ [unitless]	$\hat{\omega} \pm 2\sigma_{\hat{\omega}}$ [ppbv decade <sup>-1</sup> ]	$ \hat{\omega}/\sigma_{\hat{\omega}} $ [unitless]	$\hat{\omega} \pm 2\sigma_{\hat{\omega}}$ [ppbv decade <sup>-1</sup> ]	$ \hat{\omega}/\sigma_{\hat{\omega}} $ [unitless]
Alert	CSIRO	-26.29 ± 19.85	2.65	-1.40 ± 2.01	1.40	-15.75 ± 12.37	2.55
Ascension Island	NOAA/ESRL	+7.68 ± 5.99	2.56	-0.74 ± 2.19	0.68	+1.03 ± 4.29	0.48
Assekrem	NOAA/ESRL	-6.79 ± 6.89	1.97	-1.23 ± 3.30	0.75	-7.39 ± 4.64	3.19
Barrow	NOAA/ESRL	-15.01 ± 21.03	1.43	-1.32 ± 2.65	1.00	-14.22 ± 12.18	2.34
Cape Ferguson	CSIRO	-12.64 ± 10.26	2.47	-3.61 ± 3.58	2.01	-7.32 ± 5.71	2.56
Cape Grim	CSIRO	-6.02 ± 4.04	2.98	-1.40 ± 2.2	1.27	-2.79 ± 8.23	0.68
Cape Grim	NOAA/ESRL	+1.52 ± 5.06	0.60	-1.40 ± 2.2	1.27	-2.79 ± 8.23	0.68
Cape Point	SAWS	-10.23 ± 3.84	5.33	-3.35 ± 3.56	1.88	+6.62 ± 5.77	2.30
Casey Station	CSIRO	-6.26 ± 5.28	2.37	-2.25 ± 1.83	2.45	-0.98 ± 5.63	0.35
Halley Bay	NOAA/ESRL	-1.10 ± 4.61	0.47	-1.98 ± 1.76	2.26	-0.68 ± 5.34	0.25
Hegyhatsal	NOAA/ESRL	-52.26 ± 31.63	3.30	+2.12 ± 6.46	0.66	-52.73 ± 6.90	15.28
Heimaey	NOAA/ESRL	-13.54 ± 16.28	1.66	-1.51 ± 2.45	1.23	-17.68 ± 10.17	3.48
Izaña (Tenerife)	NOAA/ESRL	-4.89 ± 10.04	0.97	+0.02 ± 3.29	0.01	-13.02 ± 5.20	5.01
Key Biscayne	NOAA/ESRL	+5.03 ± 16.49	0.61	+3.40 ± 6.94	0.98	-15.14 ± 6.50	4.66
Mace Head	AGAGE	-9.059 ± 15.53	1.17	-2.51 ± 5.41	0.93	-22.39 ± 8.26	5.42
Mace Head	NOAA/ESRL	-18.81 ± 14.63	2.57	-2.51 ± 5.41	0.93	-22.39 ± 8.26	5.42
Macquarie Island	CSIRO	-5.35 ± 3.55	3.01	-2.38 ± 1.50	3.18	-1.15 ± 5.50	0.42
Mahe Island	NOAA/ESRL	+5.89 ± 10.85	1.08	-3.40 ± 3.43	1.98	-1.83 ± 4.96	0.74
Mauna Loa	NOAA/ESRL	-8.11 ± 11.40	1.42	-1.40 ± 2.45	1.14	-5.49 ± 4.89	2.25
Mt. Waliguan	NOAA/ESRL, CMA	-3.18 ± 24.61	0.26	-4.64 ± 11.22	0.83	-1.68 ± 12.61	0.27

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**Table 4.** Continued.

Station	Contributor	WDCGG-archived surface CO		EMAC-simulated surface CO on CE scenario		EMAC-simulated surface CO on RG scenario	
		$\dot{\omega} \pm 2\sigma_{\dot{\omega}}$ [ppbvdecade <sup>-1</sup> ]	$ \dot{\omega}/\sigma_{\dot{\omega}} $ [unitless]	$\dot{\omega} \pm 2\sigma_{\dot{\omega}}$ [ppbvdecade <sup>-1</sup> ]	$ \dot{\omega}/\sigma_{\dot{\omega}} $ [unitless]	$\dot{\omega} \pm 2\sigma_{\dot{\omega}}$ [ppbvdecade <sup>-1</sup> ]	$ \dot{\omega}/\sigma_{\dot{\omega}} $ [unitless]
Niwot Ridge (T-van)	NOAA/ESRL	-11.42 ± 11.89	1.92	-8.85 ± 8.33	2.12	-67.85 ± 9.97	13.61
Park Falls	NOAA/ESRL	-17.68 ± 15.16	2.33	-0.03 ± 8.77	0.01	-58.70 ± 11.67	10.06
Palmer Station	NOAA/ESRL	-0.05 ± 5.30	0.02	-2.21 ± 1.76	2.51	-0.94 ± 5.59	0.34
Payerne	Empa	-52.76 ± 22.21	4.75	+6.73 ± 9.53	1.41	-55.53 ± 7.60	14.61
Point Arena	NOAA/ESRL	-11.30 ± 14.28	1.58	-0.84 ± 13.39	0.13	-36.40 ± 19.73	3.69
Ragged Point	NOAA/ESRL	-0.84 ± 6.08	0.28	-0.35 ± 1.87	0.37	-3.24 ± 2.19	2.96
Rigi	Empa	-16.06 ± 19.39	1.66	-0.54 ± 11.34	0.10	-77.92 ± 9.02	17.27
Ryori	JMA	-8.26 ± 9.40	1.76	-3.54 ± 5.85	1.21	-17.47 ± 11.15	3.13
Sand Island	NOAA/ESRL	-8.65 ± 15.33	1.13	-2.21 ± 3.46	1.28	-7.31 ± 6.60	2.22
Sede Boker	NOAA/ESRL	-16.49 ± 15.68	2.10	+0.23 ± 6.14	0.08	+2.91 ± 7.11	0.82
South Pole	CSIRO	-6.65 ± 3.77	3.53	-2.04 ± 1.73	2.36	-0.74 ± 5.28	0.28
South Pole	NOAA/ESRL	+0.96 ± 5.38	0.36	-2.04 ± 1.73	2.36	-0.74 ± 5.28	0.28
Syowa Station	NOAA/ESRL	+0.08 ± 4.88	0.03	-2.21 ± 1.67	2.65	-0.91 ± 5.20	0.35
Tae-ahn Peninsula	NOAA/ESRL	+18.32 ± 36.25	1.01	+2.18 ± 16.26	0.27	-1.37 ± 20.77	0.13
Tudor Hill	NOAA/ESRL	-9.45 ± 12.53	1.51	+3.36 ± 4.34	1.55	-13.51 ± 5.81	4.65
Tutuila (Cape Matatula)	NOAA/ESRL	+2.39 ± 4.57	1.04	-3.15 ± 1.29	4.89	-2.93 ± 2.60	2.25
Ulaan Uul	NOAA/ESRL	-22.21 ± 32.76	1.36	-0.55 ± 6.57	0.17	-15.30 ± 22.40	1.37
Wendover	NOAA/ESRL	-11.05 ± 12.24	1.81	-5.43 ± 4.83	2.25	-27.26 ± 7.27	7.51
Yonagunijima	JMA	-8.50 ± 13.84	1.23	-4.97 ± 6.52	1.52	-6.73 ± 8.61	1.56
Zeppelinfjellet (Ny-Alesund)	NOAA/ESRL	-15.61 ± 19.50	1.60	-0.21 ± 2.33	0.18	-15.90 ± 10.49	3.03

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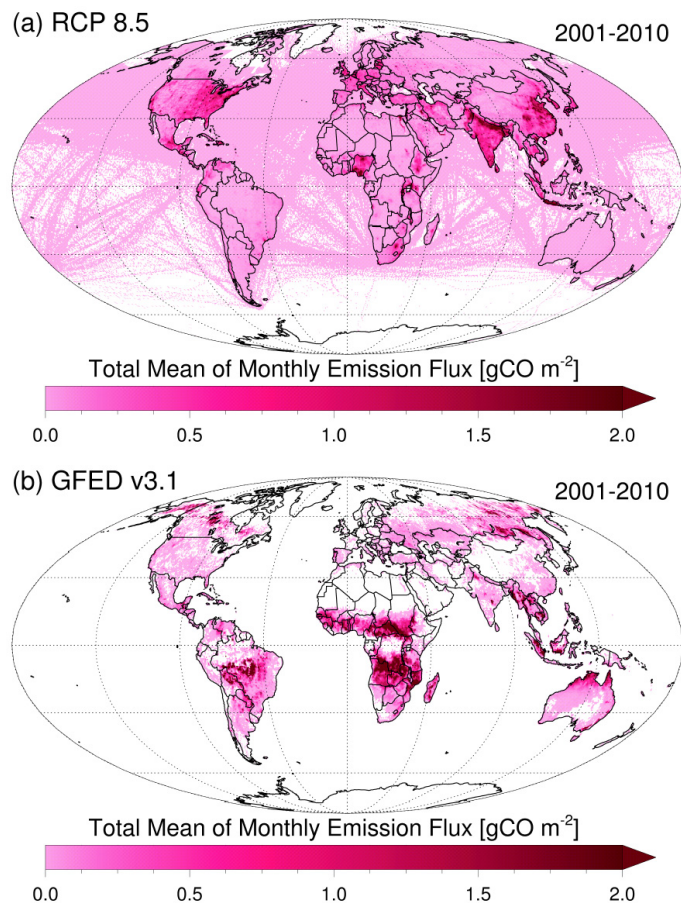
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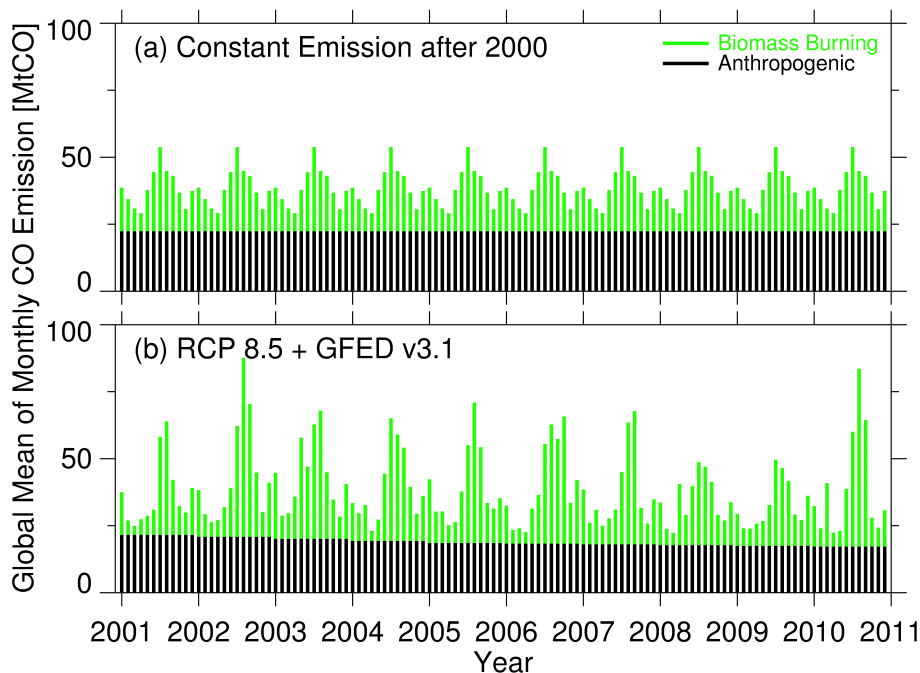


**Fig. 1.** Global distributions of the total mean of monthly CO emission fluxes of (a) RCP 8.5 and (b) GFED v3.1 from 2001 and 2010.

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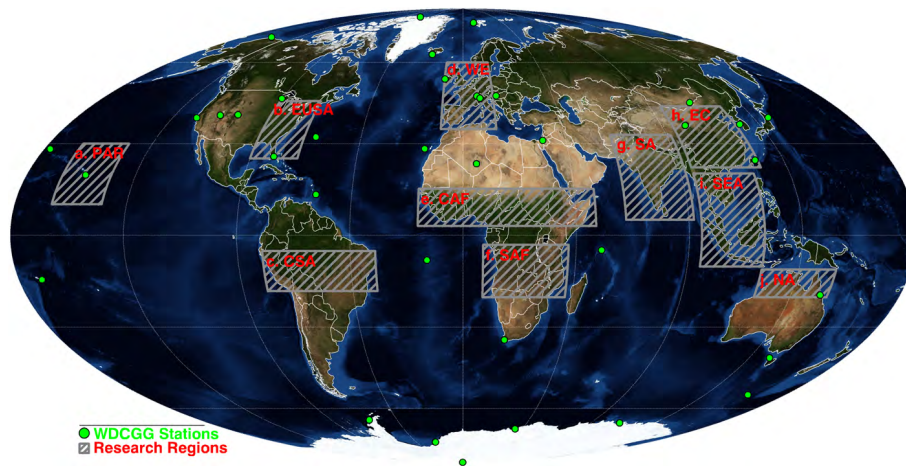
**Fig. 2.** Time series of the global mean of monthly CO emissions, **(a)** a constant emission of RCP 8.5 and GFED v3.1 after 2000 and **(b)** the combination of RCP 8.5 and GFED v3.1.

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**Fig. 3.** Research region domains and geolocations of WDCGG stations listed on Tables 1 and 2, respectively.

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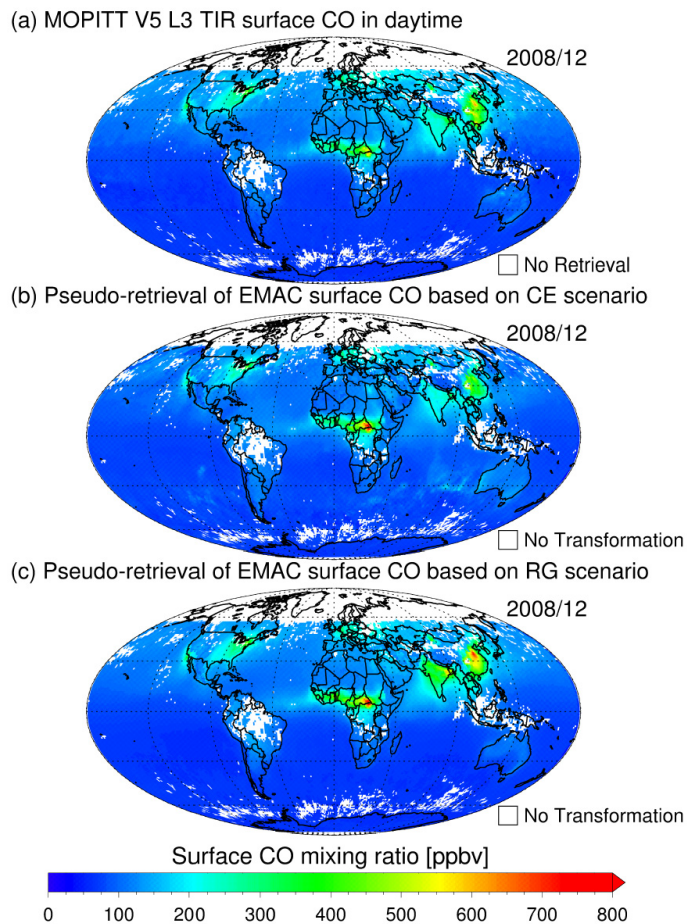
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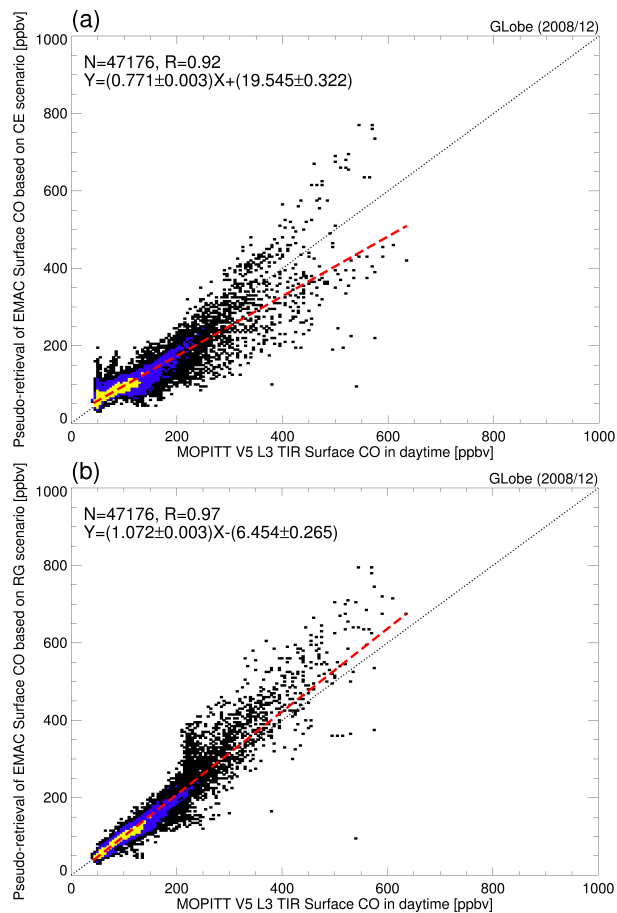
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**Fig. 4.** Global distributions of (a) the MOPITT-retrieved surface CO and pseudo-retrievals of EMAC-simulated surface CO based on (b) CE and (c) RG scenarios on December 2008.

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**Fig. 5.** Spatial comparisons of global MOPITT-retrieved surface CO with the pseudo-retrievals of EMAC simulations based on (a) CE and (b) RG scenarios on December 2008.

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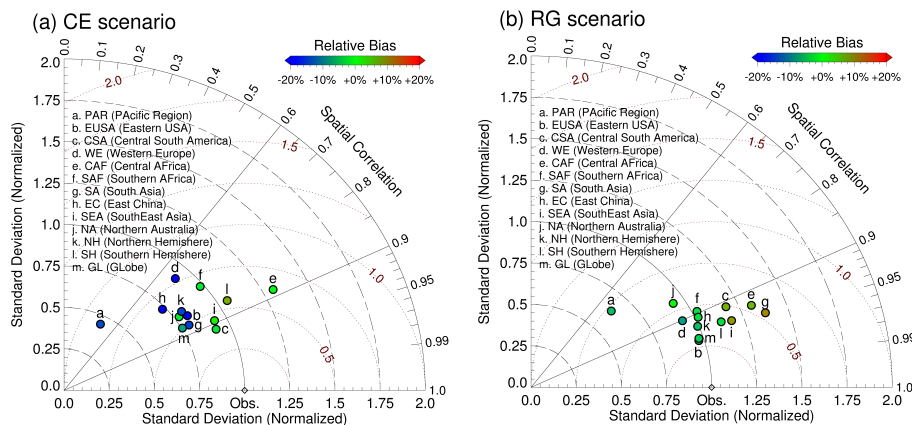
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**Fig. 6.** Spatial pattern analyses of the pseudo-retrievals of EMAC simulations based on (a) CE and (b) RG scenarios against MOPITT-retrieved surface CO for selected region domains listed on Table 1. Detailed statistical quantities are summarized in Table 3.

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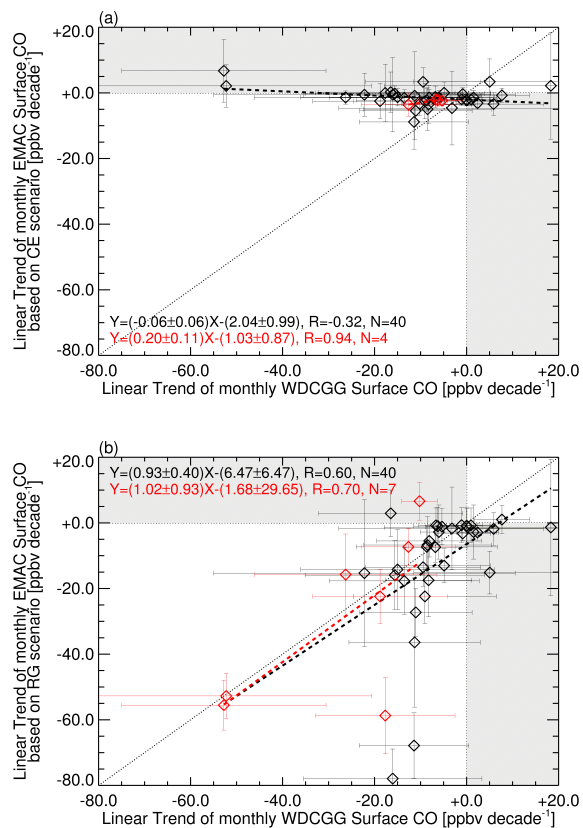
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**Fig. 7.** Comparisons of the trends of monthly EMAC-simulated surface CO based on (a) CE and (b) RG scenarios against the trend of monthly WDCGG-archived surface CO with  $\pm 2\sigma$  errors for selected WDCGG stations listed on Table 2. Black and red fonts indicate the results between all available trends and between only significant trends, respectively. Detailed values are summarized in Table 4.

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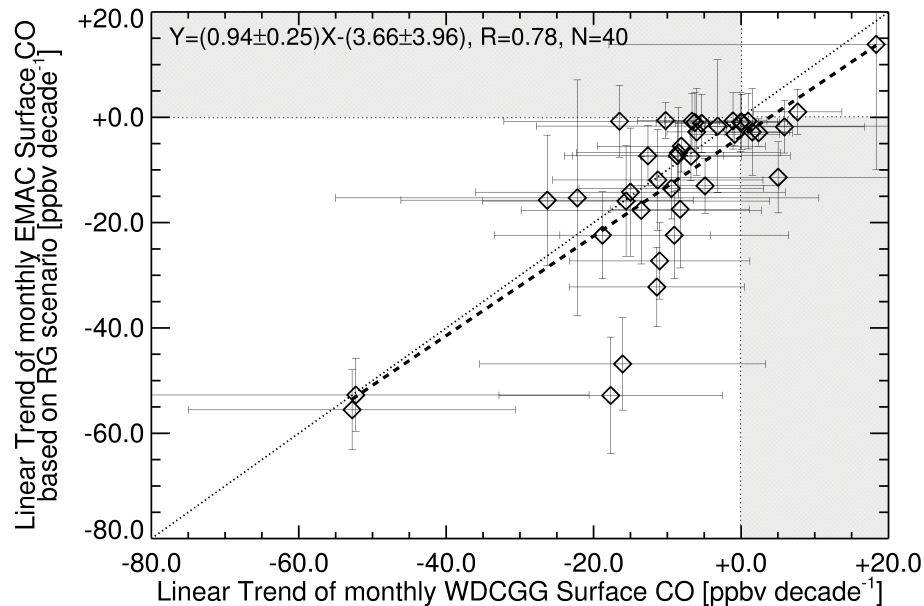
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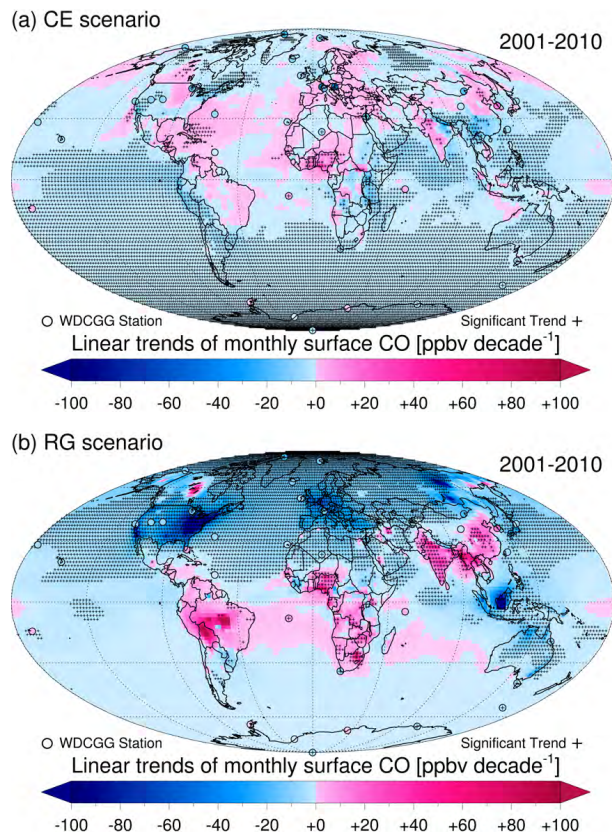
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**Fig. 8.** As in Fig. 7b, but the trends of monthly EMAC-simulated surface CO from a model grid-box to the upwind direction at the stations (i.e. Cape Point, Key Biscayne, Niwot Ridge, Park Falls, Point Arena, Rigi, Sede Boker, and Tae-ahn Peninsula).

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**Fig. 9.** 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 (+).

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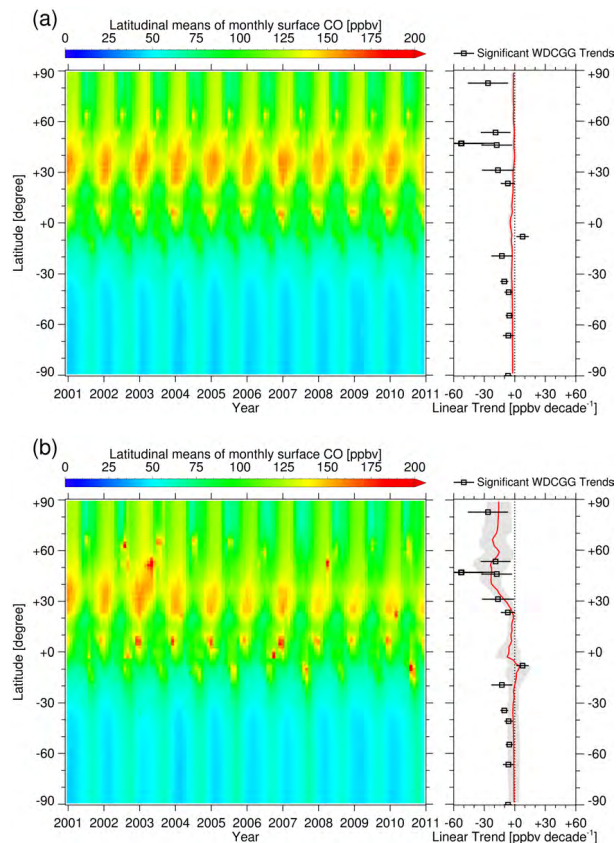
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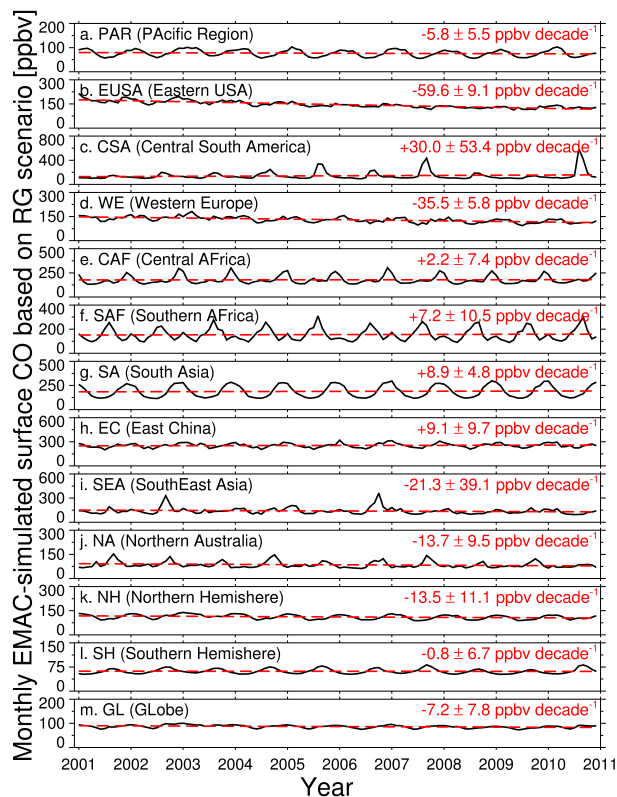
**Fig. 10.** Latitudinal means and trend estimates of monthly EMAC-simulated surface CO based on **(a)** CE and **(b)** RG scenarios from 2001 to 2010. The significant trends of WDCGG-archived surface CO at a 95 % confidence level are over-plotted to compare with the EMAC latitudinal trends.

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**Fig. 11.** Regional and global trend estimates of monthly EMAC-simulated surface CO based on RG scenario with  $\pm 2\sigma$  errors from 2001 to 2010.

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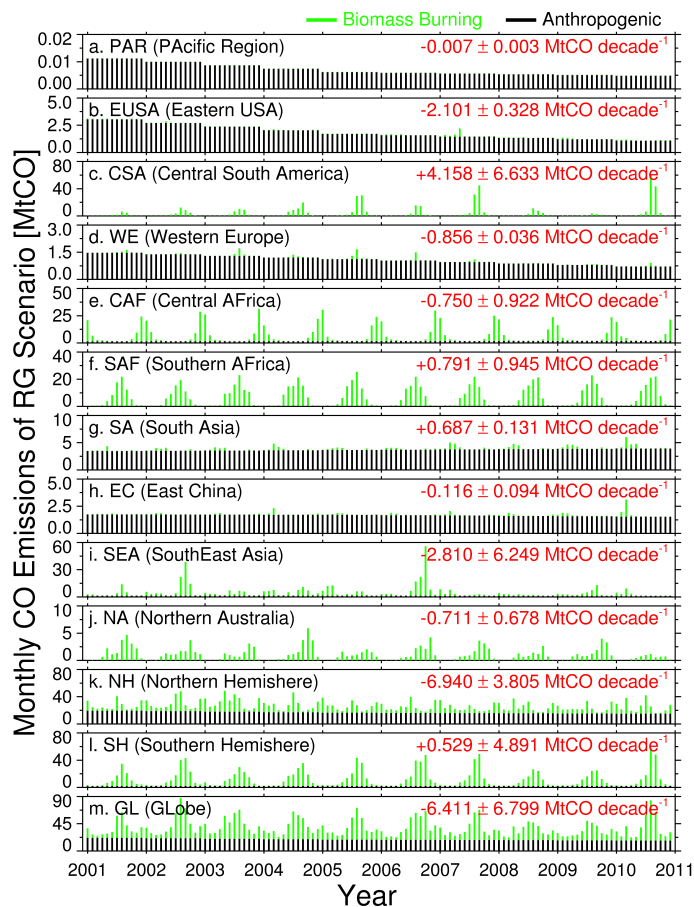


Fig. 12. As in Fig. 11, but ones of monthly RG CO emissions.

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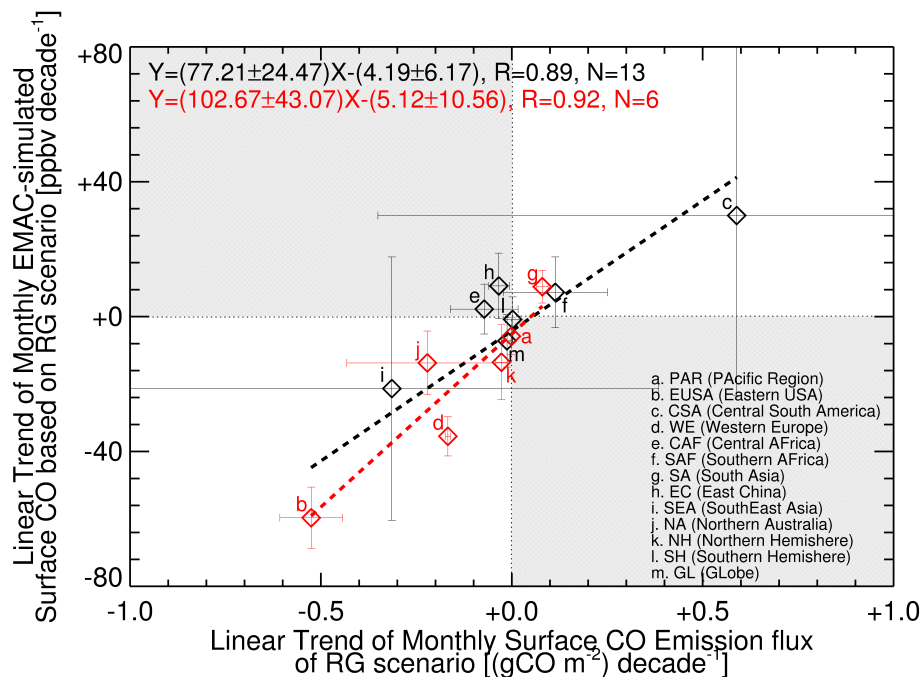
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**Fig. 13.** Comparisons of the trends of monthly RG CO emission flux against the trend of monthly EMAC-simulated surface CO with  $\pm 2\sigma$  errors for the selected regions listed on Table 1. Black and red fonts indicate the results between all available trends and between only significant trends, respectively.

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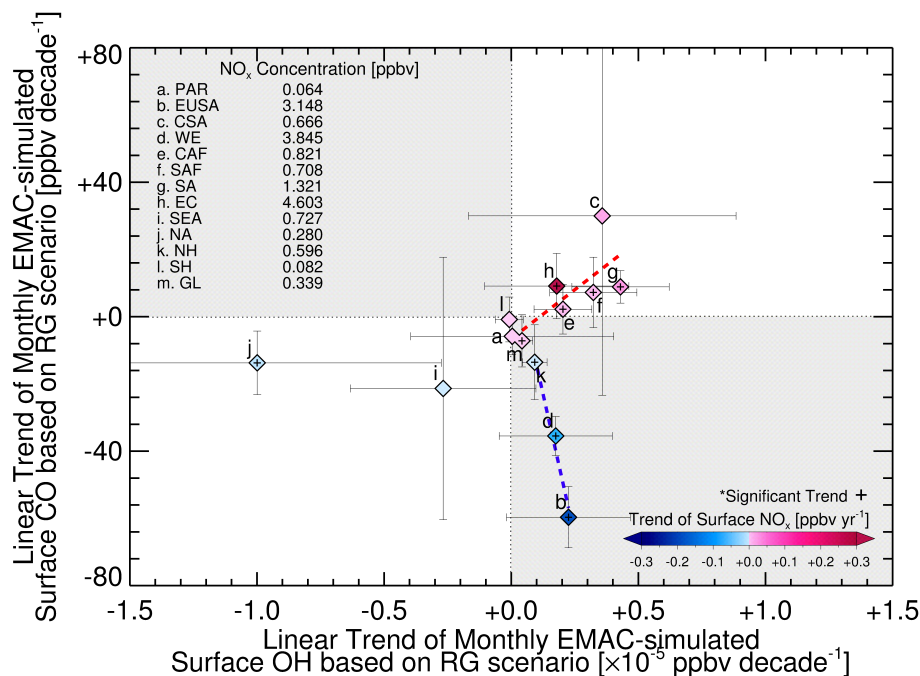
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**Fig. 14.** As in Fig. 13, but trends of monthly EMAC-simulated surface OH with surface NO<sub>x</sub> (nitric oxide and nitrogen dioxide) concentration and trend for the selected regions listed on Table 1. The significant trends in surface NO<sub>x</sub> are shown as a plus symbol.

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