Distribution and Variation of Carbon Monoxide in the Tropical Troposphere and Lower Stratosphere

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Abstract  The authors examine the distribution and variation of carbon monoxide (CO) in the tropics from the surface to the lower stratosphere. By analyzing space-borne microwave limb sounder (MLS) measurements, measurements of pollution in the troposphere (MOPITT) and modern-era retrospective analysis for research and applications (MERRA) meteorological products, and atmospheric chemistry and climate model intercomparison project (ACCMIP) surface emission inventories, the influences of atmospheric dynamics and surface emissions are investigated. The results show that there are four centers of highly concentrated CO mixing ratio over tropical areas in different seasons: two in the Northern Hemisphere and another two in the Southern Hemisphere. All of these centers correspond to local deep convective systems and monsoons/anticyclones. The authors suggest that both deep convections and anticyclones affect CO in the tropical troposphere and lower stratosphere—the former helping to transport CO from the lower to the middle troposphere (or even higher), and the dynamical uplift and isolation effects of the latter helping to build up highly concentrated CO in the upper troposphere and lower stratosphere (UTLS). Similarly, there are two annual surface emission peaks induced by biomass burning emissions: one from the Northern Hemisphere and the other from the Southern Hemisphere. Both contribute to the highly concentrated CO mixing ratio and control the seasonal variabilities of CO in the UTLS, combining the effects of deep convections and monsoons. Results also show a relatively steady emission rate from anthropogenic sources, with a small increase mainly coming from Southeast Asia and India. These emissions can be transported to the UTLS over Tibet by the joint effort of surface horizontal winds, deep convections, and the Asian summer monsoon system.

Keywords: carbon monoxide, tropics, troposphere, lower stratosphere, surface emissions


1 Introduction

The atmospheric distribution and variation of carbon monoxide (CO) has a potential influence on the global environment and climate change (Crutzen, 1995). Approximately 1000–1400 Tg/year of CO is emitted from surface sources and enters the atmosphere (Intergovernmental Panel on Climate Change (IPCC), 2001), eventually being converted into CO₂. Furthermore, CO plays an important role affecting the production of ozone in the troposphere and lower stratosphere (Seiler and Fishman, 1981; Bregman et al., 1997). Through chemical reactions, CO also causes non-negligible reductions in the atmosphere of the important hydroxyl radical (OH) (Levy, 1971).

The major sources of atmospheric CO are surface emissions (Andreae and Merlet, 2001). Considering its relatively long lifetime (a few weeks to several months), CO becomes a good tracer to study the transport of atmospheric pollutants and their impacts on local environments (Liang et al., 2004; Schoeberl et al., 2006). It is well known that surface pollution can be transported from the troposphere to the stratosphere and induce further effects there (Chen et al., 2006). In particular, deep convective activity in tropical regions (Liu and Zipser, 2005) plays an important role. Moreover, dynamical effects of monsoon systems (Li et al., 2005; Randel et al., 2010; Bian et al., 2011) can contribute to the transportation of atmospheric pollutants from the surface to the upper troposphere and lower stratosphere (UTLS).

In this paper, we conduct a study of the detailed features of CO distribution and variation from the surface to the UTLS. We also examine the individual role and combined influences of deep convections, monsoons systems, and surface emissions. Recent analysis has proved that satellite CO measurements from American National Aeronautics and Space Administration (NASA)’s Aura Microwave Limb Sounder (MLS) (Schoeberl et al., 2006; Waters et al., 2006) and Terra Measurements Of Pollution In The Troposphere (MOPITT) product (Emmons et al., 2004; Kar et al., 2004) can supply distinct illustrations of the temporal and spatial distribution of atmospheric CO. Meanwhile, the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Lamarque et al., 2013) provides comprehensive information on surface CO emissions. Analysis of ACCMIP datasets can help to improve our understanding of activities on the Earth’s surface and their impacts on the atmosphere above.
The remainder of the paper is organized as follows. In the following section, we describe the space-borne measurements of CO and the existing surface emissions inventories. In section 3, we explore the distribution and variation of CO in the tropical troposphere and lower stratosphere. Roles of deep convections, monsoon systems, and surface emissions are also discussed. Finally, a summary of the key findings of the study is presented in section 4.

2 Data

2.1 CO mixing ratio

We analyze MLS retrievals of the CO mixing ratio in the UTLS. The MLS instrument (Waters et al., 2006) was launched aboard NASA’s Earth Observing System (EOS) Aura spacecraft in July 2004. A single day of MLS observations consists of 3495 scans across the Earth’s limb and covers all latitudes between 82°S and 82°N with a resolution of approximately 1.5° of latitude. MLS makes daily, global measurements of 14 trace species using 32 spectrometers distributed across five different regions of the spectrum. The CO mixing ratio is retrieved from a spectrometer centered on the 240-GHz spectral line with a vertical resolution of 4 km in the UTLS region.

In this paper, we use the version 3.3 data of Level 2 MLS products in the altitudes above 215 hPa (Liversey et al., 2008). For analysis of the CO concentration in the troposphere, we apply the MOPITT instrument (Deeter et al., 2003; Emmons et al., 2004), which was launched aboard NASA’s EOS Terra spacecraft in December 1999. MOPITT observations include nadir-viewing channels for monitoring CO, and it became operational in March 2000. The CO mixing ratio profiles in the troposphere and total column amounts can be retrieved from the radiances. In nadir channels, the MOPITT instantaneous field of view (or "pixel size") is 22 × 22 km over all latitudes and measures the global distribution of CO every three days. For our study, we analyze the MOPITT version 5 data of Level 3 products.

2.2 Meteorological fields

The Modern-Era Retrospective Analysis for Research and Applications (MERRA) (Rienecker et al., 2011) data undertaken by NASA’s Global Modeling and Assimilation Office (GMAO) are analyzed to show the horizontal wind fields at each level (Figs. 1 and 2). The MERRA meteorological products provide information on global wind fields at horizontal resolutions of 1.25° × 1.25° and with 42 vertical levels up to 0.1 hPa. In order to explore the influence of deep convections on CO distribution, we investigate NOAA monthly data of Outgoing Longwave Radiation (OLR) (Liebmann and Smith, 1996). These data are available in 2.5° × 2.5° horizontal grids. We choose the value OLR ≤ 220 W m⁻² as a proxy to represent regions where deep convective systems generally happen (Camara et al., 2011).

2.3 CO surface emissions

By using monthly mean datasets of global emissions from ACCMIP (Lamarque et al., 2013), we investigate CO surface emissions (including biomass burning (BB) emissions from anthropogenic sources; Fig. 3). ACCMIP datasets provide information on global and regional emissions in 0.5° × 0.5° grids.

3 Results and discussion

3.1 CO concentration in the troposphere and lower stratosphere

Figure 1 shows the distribution of the CO mixing ratio in the troposphere and lower stratosphere in different seasons (indicated as January, April, July, and October, respectively). It can be seen that high values of the CO mixing ratio are mainly located in the tropics in the UTLS, although some can be observed at mid-high latitudes in the middle troposphere (500 hPa), especially in January and April. Meanwhile, most of the deep convective systems, which are indicated by contours of OLR = 220 W m⁻² (Fig. 1), also occur in the tropics. This implies that deep convections may contribute to the highly concentrated CO in the tropical UTLS.

However, Fig. 1 also reveals that not all of the centers of highly concentrated CO in the UTLS exactly correspond to deep convective systems. For example, high values of the CO mixing ratio at 100 hPa in January and July can be observed over the Northwest Pacific and Tibet, and both of these two centers depart slightly from the deep convective areas of the Indo-Pacific in January and Southeast Asia in July. On the other hand, each of these two centers corresponds well to a local anticyclone system, which can be exhibited by the distribution of horizontal wind fields (Fig. 1). Furthermore, there are another two centers of highly concentrated CO mixing ratio (Fig. 1)—at 100 hPa over Central Africa in April, and at 146 hPa over South America in October. These two centers sit more closely to local deep convective systems; but meanwhile, the wind field pattern in Fig. 1 shows two weaker anticyclones in the same regions.

As indicated above, deep convections may help to transport CO from the surface to the middle troposphere in a short period. However, dynamical uplift and occlusion by anticyclones can contribute by carrying air rich in CO further into the UTLS and isolate the air there for a longer term. These influences can be observed not only in the well-known Asian summer monsoon region (Park et al., 2008; Randel et al., 2010), but also in other regions (Barret et al., 2008), where anticyclones/monsoons occur in different seasons (Fig. 1).

3.2 Surface emissions and their potential impacts on CO

Using the ACCMIP products, we also explore the features of surface emissions to investigate their influences on the CO concentration in the tropical troposphere and lower stratosphere. Figure 2 shows that the major emission regions might be different in individual seasons, although the most significant emissions take place at low latitudes. In January, the highest emission rates can be...
observed in Central Africa and Southeast Asia/India. A similar pattern of emission rates also appears over the Southeast Asia/India region in April, July, and October, suggesting a stable CO emission rate coming from this region. There also exists an ensemble of lower emission rates in Central Africa in April. We find that the highest rates on the African continent shift to southern Africa in July and might be reduced in October. Some other high rates over South America can be observed in July and October. It is also worth mentioning that high emissions appear in July and October (Figs. 2c and 2d) over Indonesia/Australia, where deep convections constantly dominate.

Figure 3a reveals that there are two annual emission peaks mainly induced by BB emissions (Fig. 3c). The peak that occurs in July to October comes from surface BB emissions in the Southern Hemisphere (Fig. 3d). As indicated above, the results of Fig. 2 suggest a contribution by southern Africa and South America to this emission peak. The other peak, which occurs in December to April, is caused by BB emissions in the Northern Hemisphere (Fig. 3e), including from the regions of North Africa, Central Africa, and Central America. On the other hand, anthropogenic emissions remain rather stable, with only a slight increase (≤ 5%) in six years (Fig. 3b). This corresponds to steady emission patterns (Fig. 2) in regions including Southeast Asia/India in particular.

Figure 2 also presents those regions where deep convections tend to occur. We find that there is correspondence between deep convections and surface emissions over Central Africa in April (Fig. 2b) and over South
America in October (Fig. 2d). This implies that both of these two factors contribute to the observed highly concentrated CO in the middle and upper troposphere (e.g., 146 hPa; Fig. 1). Similarly, the deep convection system covering Southeast Asia and the Indian Ocean (Fig. 2c) can help to transport CO emitted from Southeast Asia/India to the middle troposphere (or even higher (Liu and Zipser, 2005)). Then, via the control of the Asian summer monsoon system, CO is able to concentrate over Tibet in the UTLS (Fig. 1). Furthermore, the distribution of surface wind fields in Fig. 2a reveals that horizontal winds help to blow CO emitted from Southeast Asia/India to tropical regions, where deep convections generally occur. Therefore, CO has the potential to become highly concentrated in January over the Northwest Pacific (Fig. 1).

Figure 4 presents the “tape recorder” of CO (Schoeberl et al., 2006), which represents the seasonal and interannual variabilities of the CO concentration in the troposphere and lower stratosphere over the tropics. It can be seen that there is a clear correlation between the seasonal changes of surface emissions and the variation of CO, not only in the troposphere but also in the UTLS. The two annual peaks of surface CO emissions correspond well to the positive anomalies of CO near to 146 hPa, with a delay of two to three months. This implies that surface emissions have a strong controlling effect on the variation of CO in the tropical UTLS. However, we still find strong interannual variations in CO anomalies. For example, Fig. 4 shows large values of positive CO anomalies in late 2006 and 2010. Earlier studies suggested that both regional emissions (Li et al., 2009) and monsoon systems (Randel et al., 2010) have impacts on this interannual variation regime.

4 Summary

We analyze MLS and MOPITT satellite observations to investigate the CO distribution and variation in the tropical troposphere and lower stratosphere. By applying the MERRA meteorological fields and ACCMIP emission products, the influences of deep convections, monsoon systems, and surface emissions are discussed.

In the UTLS, highly concentrated CO is mainly distributed over the tropics, where most deep convections (indicated by OLR \( \leq 220 \text{ W m}^{-2} \)) tend to occur. In the Northern Hemisphere, there are two centers of highly concentrated CO mixing ratio: one in January over the northern Pacific, and another in July over Tibet. Both are situated not far from deep convective regions. Moreover, both centers correspond well to local anticyclone systems in the UTLS. In the Southern Hemisphere, another two centers of large values of CO mixing ratio are also found: one in April over Central Africa, and another in October over South America. Compared to the two centers in the Northern Hemisphere, these two centers are located closer to local deep convective regions, and also sit in a relatively weaker anticyclone area.

Deep convections and anticyclones jointly affect the distribution and variation of CO in the troposphere and lower stratosphere—the former helping to transport CO from the lower to the middle troposphere (or even
Figure 3 Variation of the averaged ACCMIP CO emission rate over 30°S–30°N: (a) total emission rate (anthropogenic + biomass burning); (b) anthropogenic emission rate; (c) biomass burning emission rate; (d) biomass burning emission rate over 30°S–0°; (e) biomass burning emission rate over 0°–30°N.

Figure 4 CO (ppbv) anomalies (colored shading) as a function of pressure (hPa) from MLS (above 215 hPa) and MOPITT (below 215 hPa) data, and the mean CO emission rate (black line) from ACCMIP data over 30°S–30°N.

...higher), and the latter helping to carry air rich in CO to the UTLS and isolate the air there for a longer period of time. Through the combined effects of deep convections and anticyclones, surface emissions also induce impacts on CO. Analysis revealed that there are two annual emission peaks induced by BB emissions: one from the Northern Hemisphere in December to April, and another from the Southern Hemisphere in July to October. Both may contribute to highly concentrated CO, and control the seasonal variation of CO in the UTLS. On the other hand, steady emissions from anthropogenic sources in Southeast Asia/India can be transported to the UTLS over Tibet through the joint effort of surface horizontal winds, deep convections, and the Asian summer monsoon. We also find interannual variation of CO in the tropical UTLS. Earlier studies suggested that regional emissions and the dynamical control of monsoon systems might induce such interannual variation. However, the detailed processes and significance of each factor remain a puzzle. Further work will be carried out in the future using chemical transport model simulations.

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