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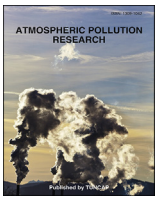

Original article

Effects of convection and long-range transport on the distribution of carbon monoxide in the troposphere over India

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ABSTRACT

Variability in the tropospheric distributions of carbon monoxide (CO) over five selected regions of India has been studied using the MOPITT data for the period of 2001–2014. The average seasonal profiles show highest mixing ratios at 900 hPa in the boreal winter and lowest in the Indian summer monsoon over all the study regions. We observe a slight increase in CO levels from 500 hPa to 200 hPa over all the locations. The CO mixing ratios are found to be higher by about 10–40% around 300–200 hPa as compared to 900 hPa over Ahmedabad, Hyderabad and Trivandrum during monsoon period. This could be due to lifting of polluted air by convection and getting trapped in the anticyclonic winds over the Indian region during the monsoon. Most of the 7 day back trajectories over these regions show transport of the polluted air mass from the major biomass burning regions of central Africa and SE Asia. The results show dominance of the seasonal amplitude at 900 hPa over all the regions, while inter-annual variability dominates mostly over Ahmedabad, Hyderabad and Trivandrum at 300 hPa. In order to check the ability of different models in capturing the observed variability, the results have been compared with simulations from two chemistry transport models (MOZART and EMAC). This comparison shows that both the models perform reasonably well in simulating the basic features such as annual variation as well as increase in CO around 300–200 hPa due to convection during the monsoon season.

1. Introduction

Carbon monoxide (CO) is an important trace constituent of the troposphere due to its role in air quality, atmospheric chemistry and global climate (Duncan and Logan, 2008; Jacob et al., 2013). It removes primary tropospheric oxidant hydroxyl radical (OH) and hence, affects the oxidizing capacity of the atmosphere as well as ozone (O_3) budget (Levy et al., 1997; Warneck, 2000). CO does not absorb the IR radiation emitted by the Earth's surface to be counted as a direct greenhouse gas (GHG), but it affects the atmospheric burden of GHGs such as O_3 and methane (CH_4) and hence, contributes in the positive radiative forcing (~0.23 ± 0.7 W m^{-2}) (Wigley et al., 2002; Myhre et al., 2013). Overall, the tropospheric burden of CO is governed roughly half by the oxidation of hydrocarbons mainly due to the CH_4 oxidation and half by the direct surface emissions (Duncan et al., 2007). Major surface emission sources of CO include the incomplete combustion of fossil fuel, biofuel, wildfires and agricultural biomass burning (Lawrence and Lelieveld, 2010). The chemical lifetime of CO is relatively short, of the order of few weeks to 2 months, depending on the ambient OH concentrations (Lawrence et al., 2003). Due to its lifetime being less than the inter-hemispheric mixing time (~1 year), it does not become evenly mixed in the troposphere, although it can get transported globally from source regions. This makes CO as an excellent tracer to study the transport and circulation of global and regional pollutants from industrial activities and large-scale biomass burning in the troposphere (Edwards et al., 2004; Sodemann et al., 2011).

In the tropical region, deep convective updrafts enhance the vertical transport of CO and create a strong connection between the surface emissions and the upper troposphere. Several modelling and observational studies have highlighted that the deep

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convection coupled with the anticyclonic wind during the Asian summer monsoon in the tropics leads to a plume of enhanced CO levels in the 400–150 hPa range (Kar et al., 2004; Li et al., 2005; Randel and Park, 2006; Park et al., 2009; Ghude et al., 2011). CO in the upper troposphere, can change ozone levels, which in turn can alter the radiative forcing (Forster and Shine, 1997).

India is a major developing country in South Asia with a population of about 1.2 billion. The strong industrial and economic growth contributes to increasing emissions of pollutants (Akimoto, 2003; Pozzer et al., 2012). Hence, the atmosphere over this region is strongly impacted by pollutants. The diurnal and seasonal variations of surface CO have been investigated over limited regions of India using in-situ measurements (Lal et al., 2000, 2012; Naja and Lal, 2002; Sahu and Lal, 2006; Beig et al., 2007; Mallik and Lal, 2014; Sarangi et al., 2014). Several campaigns had been also organized for studying spatial distributions of trace species, including CO over adjacent marine regions of the Arabian Sea and the Bay of Bengal (Lal et al., 2006, 2007; Sahu and Lal, 2006; Srivastava et al., 2012a,b; Mallik et al., 2013). In addition to these in-situ measurements, satellite and model based studies have also been conducted to study dynamical effects and long term trends in the troposphere (Kar et al., 2004, 2008, 2010; Ghude et al., 2011; Kumar et al., 2013; Worden et al., 2013; Girach and Nair, 2014; Sheel et al., 2014; Yoon and Pozzer, 2014). However, a detailed study in terms of long-range transport and seasonal variability in the vertical distribution of CO in the troposphere over the Indian region is still missing.

In view of the above perspectives, the present manuscript is based on the analysis of CO measurements using the new version (V6) of MOPITT (Measurement of pollution in the troposphere) satellite data at five urban regions representing northern, eastern, central and southern parts of India, having different physical and climatic conditions, to study its vertical distribution, seasonal cycle and effects of long range transport on its distribution in the troposphere. This study begins with a brief discussion about the dataset used in Section 2 and about the study regions in Sections 3. The vertical distributions of CO during different seasons are discussed in Section 4.1. Annual variations at lower (900 hPa) and upper (300 hPa) tropospheric heights are discussed in Section 4.2 and the effects of long range transport over the study regions at upper tropospheric height (300 hPa) are discussed in Section 4.3. Furthermore, the climatological mean annual distributions of CO over all the study regions have been compared with the annual mean climatology from two chemistry transport models (MOZART and EMAC) in Section 4.4. The summary and conclusions are given in Section 5.

2. Data sources

2.1. CO, meteorological and fire count data

This study uses Level 3 V6-TIR monthly mean gridded ($1^\circ \times 1^\circ$resolution) CO products from Measurements of Pollution in the Troposphere (MOPITT) for a period of 14 years (January, 2001–December, 2014) at 9 pressure levels between 900 hPa and 100 hPa. The "a priori" profile plays an important role to constrain the retrieved profile to fall within the range of physically realistic solutions and represents the geographical and seasonal variation of "background" concentration of CO. These profiles are based on the monthly climatologies of CO for the period of 2000–2009, simulated by the Community Atmosphere Model with Chemistry (CAM-Chem) (Deeter et al., 2014). MOPITT CO retrievals at different pressure levels have been validated against NOAA aircraft CO profiles and are found to be positively biased, which varies from 3.5% at the surface to 0.8% at 200 hPa (Deeter et al., 2014). Low bias exhibits minimal influence of a variety of potential bias sources, including errors in instrumental specifications, forward model errors, spectroscopy errors, and geophysical errors in the retrieved CO data. Therefore, the choice of V6-TIR products is best for our study which is focused on the transport effects on CO distributions in the upper troposphere. For ensuring the best quality satellite retrievals, each grid of the retrievals is screened to have Degrees of Freedom for Signal (DFS) value greater than 1.2. As a result of applying this filter, we did not get monthly profiles for one or two months over some of the study regions. Furthermore, it is also important to analyse the MOPITT averaging kernels (AK), because they quantify the sensitivity of the retrieved profiles to the true profiles (from in situ measurements). For instance, if we look the AK profile at 900 hPa, it shows a broad peak at 500 hPa level with very low value at 900 hPa, which means the retrieval is not much sensitive to CO at 900 hPa, but is correlated to the CO at 500 hPa. On the other hand, if the 900 hPa AK has a sharp peak at 900 hPa itself, then it shows that it has good sensitivity to the CO at 900 hPa. Hence, for checking the sensitivity of CO profiles over the study regions, we took the climatological mean of averaging kernel profiles at different pressure levels for the four different seasons from January, 2001 to December, 2014 (Fig. S1a and b, in Supplementary part). The details about MOPITT data and averaging kernel studies are given in the Supplementary Section S1. As suggested by Rodgers (2000), the "area" of the averaging kernel rows (i.e., the sum of all elements of a particular row in the AK matrix) can be used as a rough measure of the fraction of the retrieval which comes from the data, rather than the a priori. In other words, if the AK row sum for a particular retrieval level is much less than 1, the retrieval is dominated by the a priori. As the AK rows sum approaches 1, the a priori contribution becomes smaller and smaller. Our main focus of this study is on the seasonal cycle of CO at 900 hPa and 300 hPa. Hence, we calculate the fraction of the retrieval using this method at 900 hPa and 300 hPa during different seasons and observed smaller contributions from the a priori at these levels over all the study regions (Table S1 in Supplementary part).

Monthly gridded fire-count data at $1^\circ \times$ 1 spatial resolution corrected for cloud overpasses, taken from the MODerate resolution Imaging Spectro-radiometer (MODIS) on the Terra spacecraft for the period of January 2002 to December 2012, have been used to identify the fire activities. Details of MODIS are given in Justice et al. (2002). The monthly averaged wind and vertical velocity (omega) data for different seasons are taken from National Centers for Environmental Prediction (NCEP) reanalysis dataset over India and surrounding marine regions for the period of January 2001–December 2014. The monthly average rainfall data are based on the Tropical Rainfall Measuring Mission (TRMM) satellite, while monthly averaged surface temperature and RH data have been taken from Wunderground (http://www.wunderground.com) for the period of January 2001 to December 2014.

2.2. Model simulation results

This study uses the average monthly climatology of CO for the period of January, 2001–December, 2007 from an off-line three dimensional global chemical transport model MOZART-4 (Model for Ozone And Related Tracers, version 4) at a horizontal resolution of 2.8° latitude by 2.8° longitude with 28 sigma pressure levels extending from surface up to 2 hPa. The input data for anthropogenic and biomass burning emissions have been taken from Precursors of Ozone and their Effects in the Troposphere (POET) (Olivier et al., 2003; Granier et al., 2005) and Global Fire Emission Database, version 2 (GFEDv2.0) respectively. More details about the MOZART and its simulation can be found in Sheel et al. (2014). Additionally, this study also uses the average monthly climatology of CO for the period of January, 2001–December, 2007 from

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