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Validation of lower tropospheric carbon monoxide inferred from MOZART model simulation over India

Y. Yarragunta, S. Srivastava *, D. Mitra

Marine & Atmospheric Sciences Department, Indian Institute of Remote Sensing, Dehradun 248001, India

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ABSTRACT

In the present study, MOZART-4 (Model for Ozone and Related chemical Tracers-Version-4) simulation has been made from 2003 to 2007 and compared with satellite and in-situ observations with a specific focus on Indian subcontinent to illustrate the capabilities of MOZART-4 model. The model simulated CO have been compared with latest version (version-6) of MOPITT (Measurement Of Pollution In The Troposphere) carbon monoxide (CO) retrievals at 900, 800 and 700 hPa. Model reproduces major features present in satellite observations. However model significantly overestimates CO over the entire Indian region at 900 hPa and moderately overestimates at 800 hPa and 700 hPa. The frequency distribution of all simulated data points with respect to MOZART error shows maximum in the error range of 10–20% at all pressure levels. Over total Indian landmass, the percentage of gridded CO data that are being overestimated in the range of 0–30% at 900 hPa, 800 hPa and 700 hPa are 58%, 62% and 66% respectively. The study reflects very good correlation between two datasets over Central India (CI) and Southern India (SI). The coefficient of determination (r^2) is found to be 0.68–0.78 and 0.70–0.78 over the CI and SI respectively. The weak correlation is evident over Northern India (NI) with r^2 values of 0.1–0.3. Over Eastern India (EI), Good correlation at 800 hPa ($r^2 = 0.72$) and 700 hPa ($r^2 = 0.66$) whereas moderately weak correlation at 900 hPa ($r^2 = 0.48$) has been observed. In contrast, Over Western India (WI), strong correlation is evident at 900 hPa ($r^2 = 0.64$) and moderately weak association is found to be present at 800 hPa and 700 hPa. Model fairly reproduces seasonal cycle of CO in the lower troposphere over most of the Indian regions. However, during June to December, model shows overestimation over NI. The magnitude of overestimation is increasing linearly from 900 hPa to 700 hPa level. During April–June months, model results are coinciding with observed CO concentrations over SI region at 900 hPa. Model simulation has been compared with surface in-situ observations over ten Indian locations. Model performance is found to be moderate to good over various observational locations. However, over highly polluted megacities, model underestimates observed CO concentration by up to 3500 ppbv. A case study over the forest fire prone area reveals the clear increase of modeled and retrieved CO in February–March and a decrease in May which is coinciding with biomass burning emissions and fire counts. Model performance is found to be relatively poor over this region with r^2 of 0.29 and slope of 0.56.

1. Introduction

Carbon monoxide (CO) is one of the pernicious air pollutants in the troposphere. High level CO concentration in the boundary layer can lead to serious health effects (Ukpebor et al., 2010). It plays an important role in the atmospheric chemistry by removing hydroxyl radical (OH) (Novelli et al., 1998), which defines the lifetime of several greenhouse gases like methane and ozone. Thus CO indirectly contributes to the global warming by affecting the concentration of key greenhouse gases (Wigley et al., 2002).

CO is a useful tracer for tracking the transport of air masses on regional and global scales due to its mean tropospheric lifetime of 2–3 months (Xiao et al., 2007; Yashiro et al., 2009). The transport of CO from the source region can significantly degrade the air quality at the downwind locations (Lelieveld, 2001; Cooper et al., 2004; Srivastava and Sheel, 2013). The main emission sources of CO are from both natural and anthropogenic origins which include forest fire, agricultural wastage burning, biofuel burning, incomplete combustion of fossil fuel as well as oxidation of hydrocarbons (Holloway et al., 2000). Moreover, man-made emissions (~500–600 Tg/yr) are contributing >50% to the total global CO budget (Granier et al., 2000, 2011; van der Werf et al., 2010). Anthropogenic emissions are increasing over Asia particularly over the Indian region due to the rapid industrialization and urbanization. Indian anthropogenic CO emissions have increased from 50 Tg/yr to 80 Tg/yr during a period of 1998–2010 (Granier et al., 2011).

* Corresponding author at: Marine and Atmospheric Sciences Department, Indian Institute of Remote Sensing, Indian Space Research Organisation, Kalidas Road, Dehradun, India.
E-mail addresses: shuchita@iirs.gov.in, srivastava.shuchita@gmail.com (S. Srivastava).

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Hence, it is important to study and understand the processes that control the variability and distribution of CO over the Indian subcontinent.

Numerous studies have been made using in-situ measurements all over the globe to address the spatial and temporal variation of CO over different chemical environments (Jacob, 2003; Emmons, 2004; Drummond, 1992). However, such measurements are sparse over the Indian subcontinent and available over few Indian sites only (Lal et al., 2000; Naja et al., 2003). Measurements of CO have also been made over the Bay of Bengal and Arabian Sea during few ship-based campaigns but these measurements are limited over specific region and for very short duration (Naja, 1999; Sahu et al., 2006; Srivastava et al., 2012; Mallik et al., 2013; Girach and Nair, 2010, 2014). Satellite based observations can provide CO distribution with good spatial and temporal coverage. Several satellite based instruments like Measurement of Air Pollution from Space (MAPS), Atmospheric Infrared Sounder (AIRS), Tropospheric Emission Spectrometer (TES), and Measurements of Pollution in the Troposphere (MOPITT) provide global distribution of CO. However, satellite observations cannot provide information about the various atmospheric processes responsible for pollution episodes. This gap can be fulfilled using chemistry transport models. Satellite observations along with chemical transport model simulation, would enhance the understanding of CO emission, distribution, transport and transformation in the troposphere (Pfister et al., 2004; Kar et al., 2008, 2010; Kumar et al., 2013; Ghude et al., 2011).

A chemistry transport model can be taken as reliable if it reproduces the observed features well. In the present work, MOZART-4, a global chemistry transport model, has been validated over the Indian subcontinent using MOPITT retrieved CO and surface measurements of CO in the lower troposphere. The lower tropospheric region is chosen as all natural and anthropogenic sources of CO are located at the Earth's surface and emitted CO is largely trapped in the lower troposphere due to boundary layer dynamics. In addition, MOPITT shows maximum sensitivity in the lower troposphere (Deeter et al., 2014). The model simulation has been systematically compared with MOPITT CO over different Indian sub-regions for five years. In order to access the ability of model in terms of temporal scale, the seasonal variability of CO has been compared with satellite and in-situ observations.

2. Model and data description

2.1. The MOZART-4 model

The Model for Ozone and Related chemical Tracers-Version4 (MOZART-4) is a global chemistry transport model developed jointly by the National Center for Atmospheric Research (NCAR), the NOAA Geophysical Fluid Dynamics Laboratory (GFDL) and the Max Planck Institute for Meteorology (MPI-Met). MOZART-4 includes chemical mechanism of 85 gas-phase species, 12 bulk aerosol compounds, 39 photolysis and 157 gas phase reactions (Emmons et al., 2010). This offline chemical transport model requires meteorological fields from either climate models or assimilations of meteorological observations. In the present study, the model is driven by meteorological fields from the National Centers for Environmental Prediction/Global Forecast System (NCEP/ GFS) for the time period of 2003–2007. The model has horizontal resolution of 2.8° latitude by 2.8° longitude and the vertical coverage from the surface to about 2.7 hPa. The global anthropogenic CO emissions included in the present study are based on the POET (Precursors of Ozone and their Effects in the Troposphere) inventory for 2000. The anthropogenic emissions (i.e., emissions from fossil fuel combustion, biofuel combustion) over Asia have been updated with the REAS (Regional Emission inventory in ASia) inventory (Ohara et al., 2007) during the years of simulation. The monthly average biomass burning emissions of trace gases have been taken from the Global Fire Emissions Database version-2 (GFED-2) (van der Werf et al., 2006). The tropospheric aerosol interactions and their related chemical properties are included based on the work done by Tie et al. (2001, 2005). The online simplified radiative transfer model FTUV (fast Tropospheric Ultraviolet Visible) is used to calculate the photolysis rates which is the extended version of TUV model (Tie, 2003). Dry deposition velocities of atmospheric gases and aerosols are included in the model based on the Wesley (1989). Boundary layer transport for long lived species like CH4 (Methane), H2 (Hydrogen) are included based on the measurement of NOAA Climate Monitoring and Diagnostics Laboratory (CMDL) (Dlugokencky et al., 2005). Emissions of isoprene and monoterpenes from vegetation are also included based on the online calculations of the MEGAN (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006). The complete description of MOZART-4 model is given by Emmons et al. (2010).

2.2. The MOPITT satellite data

The MOPITT (Measurement Of Pollution In The Troposphere) instrument is an eight-channel gas correlation radiometer onboard the NASA Earth Observing System (EOS) -Terra spacecraft launched in December 1999 (Drummond, 1992). It was the first sensor to observe the tropospheric Carbon Monoxide (CO) which measures the terrestrial radiation in the thermal and near-IR region of the electro-magnetic spectrum. The retrievals of CO are available from March 2000 to present at10 pressure levels (uniform pressure levels from surface to 100 hPa) and total column amount for each individual pixel of 22 km × 22 km. This spacecraft orbits in a polar sun synchronous orbit at an altitude of 705 km and completes the global coverage in ~3 days. The instrument details, measurement techniques and algorithm of retrievals are well descried in several research papers (Drummond, 1992; Edwards et al., 1999; Deeter, 2003; Deeter et al., 2004a, 2004b).

The latest version (version-6) of monthly retrieved CO data has been used for the present study. We have preferred to use daytime level-3 data having both thermal and near infrared spectral bands (MOP03JM). This version does not require any additional filtering and any geolocation correction. In addition it contains updated CO a priori information which is based on the climatology for 2000–2009 simulated with the CAM-Chem model (Deeter et al., 2014). The V6 MOPITT retrieval algorithm provides CO data into three different formats namely, 'Thermal IR (TIR only)', 'Near IR (NIR only)'and 'Thermal and Near IR Joint (TIR/NIR)'. These products are having the higher horizontal and vertical resolution in meteorological fields due to the use of MERRA data over the land which improves the retrieval quality. The 'TIR only' products are available for both daytime/nighttime over the both land and ocean regions whereas 'NIR only' products are available for clear-sky, daytime and land regions only. The V6 Joint product 'TIR/NIR' provides more sensitivity over lower troposphere and greatest vertical resolution as compared to the other products (Deeter et al., 2014). Geographical regions are more sensitive to the degree of freedom of the signal. However, daytime observations possess greater degree of freedom than their nighttime counterpart over tropical regions. Due to these reasons only daytime retrievals are used for the present study.

2.3. Methodology

The model results are compared with satellite observations by linearly interpolating the model simulation to the geographical grids of MOPITT. Satellite retrievals cannot be compared directly with the model output. This is because retrieval of a trace gas by satellite depends on the relative sensitivity of the retrieval to different altitudes in the atmosphere and on the a priori information of retrieved trace gas profile. The spatially and temporally matched model profiles are transformed using the averaging kernels and a priori profiles used in the satellite retrievals to obtain model profiles that satellite instrument would measure for the modeled state of the atmosphere in the absence of other errors. Thus, to compare properly the vertical profile of simulated CO with satellite observation, each model CO profile has gone through the same degree of smoothing as corresponding to satellite CO profile