Atmospheric Environment xxx (2015) 1–13

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Simulation of the interannual variations of tropospheric ozone over China: Roles of variations in meteorological parameters and anthropogenic emissions

Sijia Lou, Hong Liao*, Yang Yang, Qing Mu

State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

HIGHLIGHTS

- We quantify the interannual variations (IAVs) of tropospheric O_3 concentrations in China.
- The roles of variations in meteorological parameters and anthropogenic emissions are examined.
- Surface-layer O_3 concentrations over NC, SC, and SCB have IAVs of 0.7–3.9%, 1.4–3.7%, and 2.7–3.8%, respectively.
- Variations in winds are found to have the largest impact on the IAVs of O_3 over NC, SC, and SCB.

ARTICLE INFO

Article history:
Received 10 March 2015
Received in revised form
24 August 2015
Accepted 26 August 2015
Available online xxx

Keywords:
Tropospheric ozone
Interannual variation
Process analysis
China

ABSTRACT

We quantify the interannual variations (IAVs) of tropospheric O_3 over China for the years 2004–2012 by using the one-way nested-grid version of the global three-dimensional Goddard Earth Observing System chemical transport model (GEOS-Chem). The roles of variations in meteorological fields and anthropogenic emissions of O_3 precursors are examined separately and together through sensitivity simulations. With variations in both meteorological parameters and emissions, simulated seasonal mean surface-layer O_3 concentrations over North China (NC, 110–120°E, 32–42°N) exhibit the largest IAVs in June–July–August (JJA). The regionally averaged absolute percent departure from the mean (APDM) values over NC are 0.7%, 3.2%, 3.9%, and 2.1% in December–January–February (DJF), March–April–May (MAM), and September–October–November (SON), respectively. Over South China (SC, 110–120°E, 22–32°N), the IAVs of O_3 are found maximum in MAM, and minimum in JJA; the APDM values are 2.7%, 3.7%, 1.4%, and 2.6% in DJF, MAM, JJA, and SON, respectively. With respect to the IAVs of O_3 over the Sichuan Basin (SCB, 102–110°E, 27–33°N), the APDM values are simulated to be 2.7–3.8% throughout the year. The IAVs in surface-layer O_3 by variations in meteorological fields are simulated to be larger than those by variations in anthropogenic emissions throughout the year in NC and SC except for JJA in SC. The relatively more important role of variations in anthropogenic emissions is simulated in SCB in all seasons. Process analyses are performed to identify key meteorological parameters that influence the IAVs of O_3 over NC, SC, and SCB. Over all of these regions, variations in winds are found to have the largest impact on the IAVs of O_3, followed by those in temperature and specific humidity. Considering that the APDM values represent the IAVs averaged over 2004–2012, the magnitudes of IAVs of O_3 for specific years can be more significant than the numbers reported here. Our results have important implications for the effectiveness of short-term air quality control strategies in China.

1. Introduction

Tropospheric O_3 is a major air pollutant in the atmosphere that has adverse effects on human health and ecosystem productivity (UNEP, 2006; Shindell et al., 2012). It is also an important greenhouse gas with a global mean radiative forcing of 0.4 (0.2–0.6) W

* Corresponding author.
E-mail address: hongliao@mail.iap.ac.cn (H. Liao).

http://dx.doi.org/10.1016/j.atmosenv.2015.08.081

m^{-2} (Intergovernmental Panel on Climate Change (IPCC, 2013)). High O_3 concentrations have been observed in China, with seasonal mean concentrations of 20–60 ppbv (parts per billion by volume; Xu et al. (2008); Yang et al. (2008); Zhang et al. (2008); Wang et al. (2011)) and episodic O_3 concentrations of exceeding 100 ppbv (Wang et al., 2006; Duan et al., 2008; Ge et al., 2012). Ozone concentrations exhibit variations on different time scales, from days to decades (Pozzoli et al., 2011; Fu et al., 2012; Yang et al., 2014). In this work we aim to quantify the interannual variations (IAVs) of tropospheric O_3 and examine the drivers (either variations in meteorological parameters or in emissions) of the IAVs. Such studies can help with in interpretation of year-by-year variations in O_3 measurements and understanding of the effectiveness of short-term air quality control strategies. For example, if the IAVs of O_3 driven by variations in meteorological parameters are larger than those caused by variations in emissions, the short-term air quality control may need extra efforts to reduce emissions if the meteorological parameters are not favorable for O_3 air quality.

The absolute IAVs of O_3 can be quantified by standard deviation (SD) and mean absolute deviation (MAD), defined as $SD = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(C_i - \frac{1}{n}\sum_{i=1}^{n}C_i\right)^2}$ and $MAD = \frac{1}{n}\sum_{i=1}^{n}\left|C_i - \frac{1}{n}\sum_{i=1}^{n}C_i\right|$, respectively, while the IAVs relative to the multi-year average of concentrations can be quantified by relative standard deviation (RSD) and absolute percent departure from the mean (APDM), defined as $RSD = 100\% \times SD/\left(\frac{1}{n}\sum_{i=1}^{n}C_i\right)$ and $APDM = 100\% \times MAD\Big/\left(\frac{1}{n}\sum_{i=1}^{n}C_i\right)$, where C_i is the observed or simulated O_3 concentration in year i, and n represents the number of years examined.

Previous studies have reported the IAVs of tropospheric O_3 for different regions. Liu et al. (2011) found that simulated July O_3 concentrations at 400 hPa over the Middle East over 1987–2006 had SD and RSD values of 6 ppbv and 7%, respectively, by using the GEOS-Chem model with a horizontal resolution of 4° latitude by 5° longitude. Pozzoli et al. (2011) examined the global mean concentrations of pollutants simulated for years of 1980–2005 from the coupled aerosol-chemistry-climate model ECHAM5-HAMMOZ with a horizontal resolution of 2.8° × 2.8°. They found that the global mean surface-layer O_3 exhibited SD and RSD values of 0.83 ppbv and 2.3% with changes in anthropogenic emissions and of 0.63 ppbv and 1.7% with fixed anthropogenic emissions. Kumar et al. (2013) compared O_3 concentrations observed at the Pico Mountain Observatory (PMO) (at an altitude of 2225 m) with those simulated from the GEOS-Chem model for years of 2004–2010, and found that the averaged O_3 concentrations over May–August from observations and simulations exhibited MAD (APDM) values of 1.1 ppbv (2.7%) and 1.1 ppbv (2.4%), respectively. Sahu et al. (2014), by analyzing Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) datasets during 2006–2008 over Hyderabad and India, reported that O_3 concentrations averaged over the premonsoon season (March–May) had large IAVs, with values of 58 ppbv in 2006 and 44 ppbv in 2008 in the lower troposphere (2–4 km) depending on the transport of marine and continental air and values of 72 ppbv in 2006 and 50 ppbv in 2008 in the upper troposphere (8–11 km) depending on deep convections.

Observational and modeling studies have also illustrated that surface-layer O_3 have large IAVs over China. Tang et al. (2009) showed that, on the basis of measurements at 6 sites in downtown Beijing for years of 2001–2006, the average O_3 concentrations over July–September in Beijing exhibited APDM value of 3.6%. Zhang et al. (2013) analyzed O_3 measurements in Hong Kong over the period of 1999–2011, and reported that the seasonal mean O_3 concentrations had MAD and APDM values of 3–6 ppbv and 9–18%, respectively. They also reconstructed the time series of O_3 by taking into account the IAVs in frequency and intensity of circulation patterns. Such reconstruction captured up to 50% of the observed IAV of O_3, suggesting that the changes in meteorological parameters have large impacts on the IAVs of surface O_3 levels in Hong Kong.

The concentrations of O_3 are dependent on both precursor emissions and meteorological conditions. Meteorological parameters influence O_3 concentrations by altering cross stratosphere–-troposphere flux of O_3 (Voulgarakis et al., 2011; Hess and Zbinden, 2013), biogenic emission of volatile organic compounds (Fu and Liao, 2012), and the chemical production and loss, transport, and deposition of O_3 (Lin et al., 2001; Camalier et al., 2007; Ramsey et al., 2014). Olsen et al. (2013) estimated the extratropical stratosphere–troposphere exchange of O_3 from 2005 to 2010 by combining Microvave Limb Sounder (MLS) ozone observations with Modern Era Retrospective-Analyses for Research and Applications (MERRA) of meteorological fields, and found that the difference between the highest annual flux in 2006 and the lowest annual flux in 2008 was about 15% of the multiyear mean STE (Strat-Trop exchange) in the Northern Hemisphere. Neu et al. (2014) used Aura satellite measurements (from Tropospheric Emission Spectrometer (TES) and MLS instruments) of stratospheric water and tropospheric O_3 levels for years of 2005–2010 to quantify the impact of changes in the stratospheric circulation on tropospheric O_3. They reported that a 25% increase in stratospheric O_3 results in a 2% increase in 500 hPa O_3 in the northern mid-latitudes, approximately half of the interannual tropospheric O_3 variability, based on CAM-Chem simulation. Variations in temperature and water vapor in the atmosphere also influence production and loss of O_3. Dawson et al. (2007) found that the daily maximum 8-h O_3 concentrations increase with temperature by about 0.34 ppbv K^{-1} and decrease by 0.025 ppbv for each percent increase in absolute humidity over eastern United States during July 12–21, 2001, by using the PMCAMx model with perturbations of individual meteorological parameters. Furthermore, climate patterns, such as monsoon and El Nino, play important roles in the IAVs of O_3 concentrations (Neu et al., 2014; Sahu et al., 2014; Sekiya and Sudo, 2014; Yang et al., 2014).

The major processes that influence the concentrations of O_3 include horizontal and vertical transport, chemical production and loss, dry and wet deposition. The role of each physical or chemical process can be quantified by the Integrated Process Rate (IPR) analysis. Such approach has been applied to episodic events (Jose et al., 2002; Goncalves et al., 2009; Im et al., 2011), and yearly to decadal simulations (Zhang et al., 2009b; Civerolo et al., 2010). For example, Goncalves et al. (2009) found that the formation of surface O_3 was dominated by the horizontally advected flows and gas-phase chemical reactions occurring aloft in the morning, and by vertical advective flows in the afternoon and dusk, during a photochemical pollution episode over southern Mediterranean region in summer. Similar process analyses were also implemented in other models, such as the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem, Jiang et al., 2012; Jiang et al., 2013) and the GEOS-Chem model (Mu and Liao, 2014), to understand the key processes that influence the concentrations or variations of O_3 and aerosols.

This work aims to (1) quantify the IAVs of tropospheric O_3 concentrations in China due to the variations in meteorological fields and/or anthropogenic emissions during 2004–2012 using the GEOS-Chem model driven by the assimilated meteorological fields, and (2) identify the dominant meteorological parameters that

Please cite this article in press as: Lou, S., et al., Simulation of the interannual variations of tropospheric ozone over China: Roles of variations in meteorological parameters and anthropogenic emissions, Atmospheric Environment (2015), http://dx.doi.org/10.1016/j.atmosenv.2015.08.081