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Global budget of tropospheric ozone: Evaluating recent model advances with satellite (OMI), aircraft (IAGOS), and ozonesonde observations

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HIGHLIGHTS

- GEOS-Chem updates over past decade have led to more active model ozone chemistry.
- V10-01 improves tropospheric ozone simulation relative to earlier model versions.
- A prominent flaw occurs at high northern latitudes, likely due to insufficient STE.
- GEOS-Chem ozone burden, production, & lifetime are on high side of other models.
- OMI data maintained persistent high quality and no significant drift over 2006–2013.

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ABSTRACT

The global budget of tropospheric ozone is governed by a complicated ensemble of coupled chemical and dynamical processes. Simulation of tropospheric ozone has been a major focus of the GEOS-Chem chemical transport model (CTM) over the past 20 years, and many developments over the years have affected the model representation of the ozone budget. Here we conduct a comprehensive evaluation of the standard version of GEOS-Chem (v10-01) with ozone observations from ozonesondes, the OMI satellite instrument, and MOZAIC-IAGOS commercial aircraft for 2012–2013. Global validation of the OMI 700-400 hPa data with ozonesondes shows that OMI maintained persistent high quality and no significant drift over the 2006–2013 period. GEOS-Chem shows no significant seasonal or latitudinal bias relative to OMI and strong correlations in all seasons on the $2^\circ \times 2.5^\circ$ horizontal scale ($r = 0.88$–0.95), improving on previous model versions. The most pronounced model bias revealed by ozonesondes and MOZAIC-IAGOS is at high northern latitudes in winter-spring where the model is 10–20 ppbv too low. This appears to be due to insufficient stratosphere-troposphere exchange (STE). Model updates to lightning NO_x, Asian anthropogenic emissions, bromine chemistry, isoprene chemistry, and meteorological fields over the past decade have overall led to gradual increase in the simulated global tropospheric ozone burden and more active ozone production and loss. From simulations with different versions of GEOS meteorological fields we find that tropospheric ozone in GEOS-Chem v10-01 has a global production rate of 4960–5530 Tg a^{-1}, lifetime of 20.9–24.2 days, burden of 345–357 Tg, and STE of 325–492 Tg a^{-1}. Change in the intensity of tropical deep convection between these different meteorological fields is a major factor driving differences in the ozone budget.

1. Introduction

Tropospheric ozone (O_3) is an important greenhouse gas, a

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surface pollutant, and the primary precursor of the hydroxyl radical (the main atmospheric oxidant). Understanding the factors controlling tropospheric ozone is a central problem in atmospheric chemistry. Ozone is transported to the troposphere from the stratosphere, and is also produced within the troposphere by photo-oxidation of volatile organic compounds (VOCs) and carbon monoxide (CO) in the presence of nitrogen oxides ($NO_x \equiv NO + NO_2$). These precursors have both anthropogenic and natural sources, and lifetimes ranging from minutes to years. Ozone has a lifetime of a few weeks and is eventually removed by photochemical loss and dry deposition. Here we use an ensemble of satellite, aircraft, and sonde measurements to test our understanding of the tropospheric ozone budget as simulated by the GEOS-Chem global 3-D chemical transport model (CTM), and we examine the role of major new GEOS-Chem developments in affecting the ozone simulation.

Simulation of global tropospheric ozone has long been a target of atmospheric chemistry models. A large database of observations is available from the ozonesonde network (Logan, 1999; Thompson et al., 2007). Observations are also made from commercial aircraft (Brenninkmeijer et al., 2007; Volz-Thomas et al., 2009; Nédélec et al., 2015). Global mapping of tropospheric ozone from space began in the 1980s (Fishman et al., 1986; Fishman and Larsen, 1987), and has since expanded with direct retrievals in both the infrared (IR) (Worden et al., 2007) and the ultraviolet (UV) (Liu et al., 2005, 2006). Current state-of-science CTMs typically show a successful simulation of global tropospheric ozone including its large-scale gradients and seasonal variations (Stevenson et al., 2006; Fiore et al., 2009; Young et al., 2013). However, the global ozone production and loss rates can vary by a factor of 2 or more between models (Wild, 2007; Wu et al., 2007). Models are also unable to reproduce observed centurial and multi-decadal trends (Mickley et al., 2001; Parrish et al., 2014).

Simulation of global tropospheric ozone has been a major driver in the development of the GEOS-Chem CTM, starting from the original model described by Bey et al. (2001). Wu et al. (2007) showed the evolution of the global tropospheric ozone budget in successive GEOS-Chem versions pre-2006 and Fig. 1 shows the effect of more recent developments. These include in particular updating Asian anthropogenic emissions, improving lightning NO_x emissions by using satellite data for individual years (Murray et al., 2012), improving biogenic VOC chemistry (Paulot et al., 2009a, 2009b), implementing tropospheric bromine chemistry (Parrella et al., 2012), and using newer-generation assimilated meteorological data. These changes have overall led to a gradual upward creep in the global tropospheric ozone burden and production rate.

The last global evaluation of the GEOS-Chem simulation of tropospheric ozone was presented by Zhang et al. (2010) using v8-01-04 of the model released in March 2009. That work used worldwide ozonesonde data as well as satellite observations from the TES and OMI instruments. It found that GEOS-Chem systemically underestimated ozone in the tropics and overestimated ozone in the northern subtropics and southern mid-latitudes. Here we revisit the global evaluation of the GEOS-Chem tropospheric ozone simulation using ozonesonde, satellite, and aircraft data, to examine in particular if the flaws previously identified by Zhang et al. (2010) have been corrected and if new flaws have developed. We focus on the global ozone distribution. Other recent GEOS-Chem studies have examined the capability of the model to simulate ozone precursors for individual regions (Wang et al., 2011; Marais et al., 2012; Hu et al., 2015; Jiang et al., 2015; Johnson et al., 2016; Sofen et al., 2016; Yan et al., 2016) and the correlation of ozone with its precursors regionally and globally (Kim et al., 2013; Travis et al., 2016).

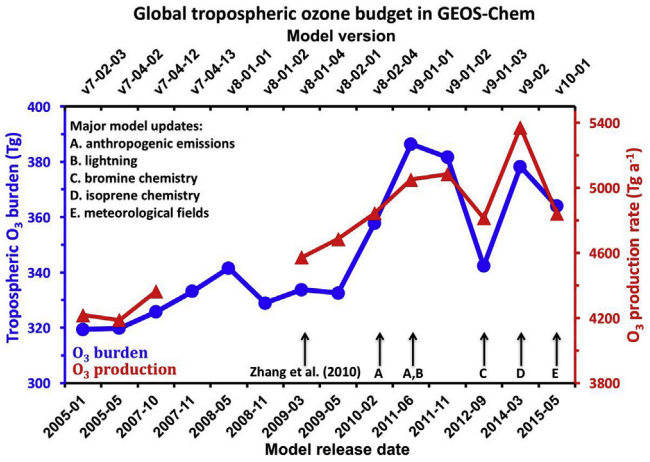

Fig. 1. Evolution of the annual mean global tropospheric ozone burden and production rate in successive versions of the GEOS-Chem model. Data are from benchmark simulations conducted when new versions of the model are released. All benchmark simulations are done for the same meteorological and emission year of 2005, except for the first two (2001 for v7-02-03 and v7-04-02) and the last one (2013 for v10-01). All benchmark simulations are conducted at 4° × 5° horizontal resolution with 12 months of spin-up. Major model developments and the version previously evaluated by Zhang et al. (2010) are identified with vertical arrows. Ozone production rate is for the odd oxygen (O_x) chemical family to account for fast cycling between ozone and short-lived reservoirs. Documentation for all versions and benchmarking procedure are at http://www.geos-chem.org. A similar figure for pre-2006 versions of the GEOS-Chem ozone budget is shown in Wu et al. (2007).

2. GEOS-chem chemical transport model

2.1. General description

We use GEOS-Chem v10-01 (http://www.geos-chem.org) driven by assimilated meteorological data from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). Operational GEOS data are produced by GMAO with a grid resolution of 0.25° × 0.3125° (latitude by longitude) and 72 vertical levels extending up to 0.01 hPa. They are archived every 3 h (1 h for surface data). Here we conduct GEOS-Chem simulations at 2° × 2.5° resolution for 2011–2013 by averaging the native-resolution meteorological data, with time steps of 15 min for transport and 30 min for chemistry (Philip et al., 2016). We use 12 months for initialization and report results for June 2012–May 2013. This time period is chosen because of the overlap of two versions of operational GEOS-5 meteorological data produced by GMAO, allowing us to examine the sensitivity to meteorology. Hereinafter we refer to the latest version of meteorological data (GEOS-5.7.2 and later versions) as GEOS-5 Forward-Processing or GEOS-FP, and to the previous version (GEOS-5.2.0) as GEOS-5. We will also show results from a sensitivity simulation with older-generation GEOS-4 meteorological fields available through 2006.

Emissions in GEOS-Chem are computed by the Harvard-NASA Emission Component (HEMCO) (Keller et al., 2014), which combines and regrids ensembles of user-selected regional and global emission inventories. Global emissions used here are given in Table 1. The model includes detailed HO_x-NO_x-VOC-ozone-BrO_x-aerosol tropospheric chemistry with JPL and IUPAC recommendations for chemical kinetics (Sander et al., 2011; IUPAC, 2013). BrO_x chemistry is from Parrella et al. (2012) and isoprene oxidation chemistry is from Mao et al. (2013). Photolysis frequencies are calculated with the Fast-JX scheme (Bian and Prather, 2002), as implemented in GEOS-Chem by Mao et al. (2010) and Eastham et al. (2014). Stratospheric chemistry is represented using the Linoz