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Regional and global modeling estimates of policy relevant background ozone over the United States

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ABSTRACT

Policy Relevant Background (PRB) ozone, as defined by the US Environmental Protection Agency (EPA), refers to ozone concentrations that would occur in the absence of all North American anthropogenic emissions. PRB enters into the calculation of health risk benefits, and as the US ozone standard approaches background levels, PRB is increasingly important in determining the feasibility and cost of compliance. As PRB is a hypothetical construct, modeling is a necessary tool. Since 2006 EPA has relied on global modeling to establish PRB for their regulatory analyses. Recent assessments with higher resolution global models exhibit improved agreement with remote observations and modest upward shifts in PRB estimates. This paper shifts the paradigm to a regional model (CAMx) run at 12 km resolution, for which North American boundary conditions were provided by a low-resolution version of the GEOS-Chem global model. We conducted a comprehensive model inter-comparison, from which we elucidate differences in predictive performance against ozone observations and differences in temporal and spatial background variability over the US. In general, CAMx performed better in replicating observations at remote monitoring sites, and performance remained better at higher concentrations. While spring and summer mean PRB predicted by GEOS-Chem ranged 20–45 ppb, CAMx predicted PRB ranged 25–50 ppb and reached well over 60 ppb in the west due to event-oriented phenomena such as stratospheric intrusion and wildfires. CAMx showed a higher correlation between modeled PRB and total observed ozone, which is significant for health risk assessments. A case study during April 2006 suggests that stratospheric exchange of ozone is underestimated in both models on an event basis. We conclude that wildfires, lightning NO_x and stratospheric intrusions contribute a significant level of uncertainty in estimating PRB, and that PRB will require careful consideration in the ozone standard setting process.

1. Introduction

Policy Relevant Background (PRB) ozone is the metric that the US Environmental Protection Agency (EPA) uses in its standard setting process to define uncontrollable "background" concentrations (EPA, 2006, 2007). Specifically, PRB is the surface ozone concentration that would be present across the US in the absence of all anthropogenic emissions from North America (US, Canada and Mexico). It includes contributions from natural sources globally (e.g., biogenic, wildfires, lightning NO_x, and stratosphere-troposphere exchange) and from anthropogenic emissions outside of North America.

In 2008, EPA promulgated a reduction in the 8-h ozone National Ambient Air Quality Standard (NAAQS) from 0.08 ppm to 0.075 ppm (Federal Register, 2008), and has begun the next review for the 2013 ozone standard. PRB is critically important in the standard setting process because it establishes the baseline in the comparison of health risks at alternate ozone levels being evaluated for the NAAQS. Health benefits derived for different levels of the ozone standard can be overestimated when PRB is set too low, as outlined by Lefohn (2007) using data from the EPA's Risk Assessment Technical Support Document (Abt Associates, 2007). PRB also has a significant impact on the feasibility and cost of compliance; as the ozone standard approaches the zero emission PRB level, the probability of practicably achieving the NAAQS is greatly diminished.

Prior to 2006, EPA based estimates of background ozone on observational evidence from data at remote monitoring sites on

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"clean" days. EPA first considered global modeling as a means to establish the range of PRB over the US when preparing for the 2008 ozone NAAQS. EPA (2006) specifically cited the work of Fiore et al. (2003), who applied the GEOS-Chem global model at $2 \times 2.5°$ grid size (>200 km) for the year 2001. GEOS-Chem estimated a mean PRB range of 15–35 ppb, with a 2–7 ppb mean stratospheric influence and a 4–12 ppb global anthropogenic contribution. While GEOS-Chem performed well in replicating seasonal mean rural ozone observations, it did not replicate the frequency of the highest western US ozone events (>60 ppb) in winter and spring when global transport and stratospheric–tropospheric exchange (STE) peak (Yienger et al., 1999; Lefohn et al., 2001). EPA (2006) discusses the technical issues associated with global models, including coarse spatial/temporal resolution, highly uncertain global emission inventories (most notably for Asia), and simplifications of some important processes such as STE.

Observational research by Lefohn et al. (2001) suggests higher background ozone (often exceeding 50 ppb) with more natural short-term variability and more evidence of transport from the stratosphere (points which were directly countered by Fiore et al., 2003). Subsequent observational studies have continued to present evidence for higher background ozone, particularly with respect to STE influences (e.g., Cooper et al., 2005; Hocking et al., 2007; Oltmans et al., 2008; Langford et al., 2009). Lefohn et al. (2011) describe statistical and trajectory modeling analyses over 2006–2008 that suggest spring and summer STE events are well correlated with multi-day surface ozone enhancements reaching 50–65 ppb at remote sites in the western and northern US. Furthermore, Parrish et al. (2009) present compelling evidence that ozone entering the US west coast between 1980 and 2008 is increasing at 3–5 ppb per decade, signifying that long-term PRB ozone trends need to be addressed.

As a hypothetical construct, PRB is not directly measureable and so modeling is a necessary tool, but modeled estimates must be informed by and evaluated based on measurement data from remote sites. More recently, Wang et al. (2009) re-estimated 2001 PRB levels using GEOS-Chem with 1° (~100 km) resolution over North America and reported little difference from PRB estimates of Fiore et al. (2003). Mueller and Mallard (2011) evaluated 2002 North American background ozone at 36 km resolution using EPA's regional Community Multiscale Air Quality (CMAQ) model, with lateral boundary conditions provided by a $2 \times 2.5°$ degree GEOS-Chem run. Most recently, Zhang et al. (2011) employed GEOS-Chem with improved estimates of Asian emissions, a revised stratospheric ozone treatment, and North American resolution of $0.5 \times 0.625°$ (~50 km) to simulate PRB over 2006–2008. These enhancements incrementally improved model performance in replicating the high end of the observed ozone frequency distribution, particularly at high elevation sites, while marginally increasing PRB estimates. However, Zhang et al. (2011) state that GEOS-Chem is unable to replicate event-oriented phenomena such as wildfires and STE. Global models continue to be driven by meteorological analyses of low temporal resolution (6 h), which can severely limit the models' capacity to replicate rapid deep circulations at relatively small scales, such as often occur in the intermountain western US.

Whereas the majority of PRB modeling in the literature to date has employed global models, this paper summarizes a comprehensive ozone modeling analysis for the year 2006 using both low-resolution global ($2 \times 2.5°$) and very high-resolution regional (12 km) chemical transport models. We compare differences in model predictive performance against ozone observations and differences in temporal and spatial background variability over the US. Regional modeling over the North American continent was conducted using the Comprehensive Air quality Model with extensions (CAMx; ENVIRON, 2010). Following the nesting approach of Mueller and Mallard (2011), lateral boundary conditions were determined from the global modeling component using a contemporary version of GEOS-Chem.

2. Methodology

2.1. Global modeling

GEOS-Chem version 8-03-01 was used to derive ozone estimates over the US and to provide boundary condition inputs for CAMx. This version of GEOS-CHEM includes several important updates as described by Zhang et al. (2011), including several chemistry and solver updates, revised treatment of stratospheric chemistry and stratosphere–troposphere exchange ("LINOZ"), and global emission updates. GEOS-Chem was run on a $2 \times 2.5°$ latitude/longitude grid with 47 vertical layers, using 3-hourly surface and 6-hourly aloft GEOS-5 global meteorological analyses produced and distributed by the National Aeronautics and Space Administration (NASA) Global Modeling and Assimilation Office (GMAO, 2011). Standard and default settings, solvers, algorithms, and datasets were used to treat emissions, chemistry, transport, and removal. Gases and aerosols were resolved with 43 chemical species, and LINOZ was invoked. Additional information on GEOS-Chem structure, inputs and algorithms is available at http://acmg.seas.harvard.edu/geos/index.html.

The following anthropogenic emission inventories were employed and internally adjusted to the 2006 simulation year:

- Europe: 2005 European Monitoring and Evaluation Programme (EMEP, 2011);
- Asia: Streets 2006 Inventory (Zhang et al., 2009);
- Mexico: 1999 Big Bend Regional Aerosol and Visibility Observation Study (BRAVO; Kuhns et al., 2005);
- Canada: 2002 Criteria Air Contaminants (CAC) inventory (Environment Canada, 2011);
- US: 2005 National Emission Inventory (NEI; EPA, 2010);
- Remaining world: Emissions Database for Global Atmospheric Research (EDGAR, 2011).

The 2006 Streets inventory for Asia reflects a doubling of anthropogenic NO_x emissions in China relative to the previous 2001 Streets inventory, based on comparisons of earlier GEOS-Chem results against satellite measurements (Zhang et al., 2009). To be consistent with satellite evidence, and following the approach from Zhang et al. (2011), we scaled NO_x emissions in Japan and Korea upward by a factor of two. Natural sources include biogenic emissions derived from the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006), monthly fire emissions from the Global Fire Emissions Database version 2 (GFED2, 2005), internally calculated lightning NO_x according to GEOS-5 meteorology, and soil NO_x from both natural bacterial activity and agricultural fertilizer application.

GEOS-Chem was first run for the year 2006 in two ways: (1) with all global anthropogenic emissions included for the purposes of assessing model performance against US observational data (the "Base Case"); and (2) with North American anthropogenic emissions from US, Mexico, and Canada removed (the "PRB Case").

2.2. North American regional modeling

CAMx version 5.30 was run for the entire year of 2006 on a single large North American domain with 36 km grid spacing. CAMx was subsequently run on two smaller nested domains with 12 km grid spacing that split the US into western and eastern halves