Atmospheric Environment 109 (2015) 282–296

Contents lists available at ScienceDirect

Atmospheric Environment

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

Comparison of background ozone estimates over the western United States based on two separate model methodologies

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HIGHLIGHTS

- Two modeling approaches have been applied to estimate background ozone levels.
- Background ozone estimates are generally independent of the model methodology used.
- Seasonal mean background ozone can approach 40–45 ppb at sites in the western U.S.
- On high days, ozone in the rural western U.S. can average 60–80 percent background.

ARTICLE INFO

Article history:
Received 2 September 2014
Received in revised form
31 December 2014
Accepted 3 January 2015
Available online 3 January 2015

Keywords:
Ozone
Background ozone
U.S. background
Zero-out modeling
Source apportionment
CMAQ
CAMx

ABSTRACT

Two separate air quality model methodologies for estimating background ozone levels over the western U.S. are compared in this analysis. The first approach is a direct sensitivity modeling approach that considers the ozone levels that would remain after certain emissions are entirely removed (i.e., zero-out modeling). The second approach is based on an instrumented air quality model which tracks the formation of ozone within the simulation and assigns the source of that ozone to pre-identified categories (i.e., source apportionment modeling). This analysis focuses on a definition of background referred to as U.S. background (USB) which is designed to represent the influence of all sources other than U.S. anthropogenic emissions. Two separate modeling simulations were completed for an April–October 2007 period, both focused on isolating the influence of sources other than domestic manmade emissions. The zero-out modeling was conducted with the Community Multiscale Air Quality (CMAQ) model and the source apportionment modeling was completed with the Comprehensive Air Quality Model with Extensions (CAMx). Our analysis shows that the zero-out and source apportionment techniques provide relatively similar estimates of the magnitude of seasonal mean daily 8-h maximum U.S. background ozone at locations in the western U.S. when base case model ozone biases are considered. The largest differences between the two sets of USB estimates occur in urban areas where interactions with local NO_x emissions can be important, especially when ozone levels are relatively low. Both methodologies conclude that seasonal mean daily 8-h maximum U.S. background ozone levels can be as high as 40–45 ppb over rural portions of the western U.S. Background fractions tend to decrease as modeled total ozone concentrations increase, with typical fractions of 75–100 percent on the lowest ozone days (<25 ppb) and typical fractions between 30 and 50% on days with ozone above 75 ppb. The finding that estimates of background ozone are not strongly dependent on the technique applied lends credibility to this and earlier modeling work.

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1. Introduction

It is well-established that surface ozone levels measured across the United States can be influenced by background ozone

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http://dx.doi.org/10.1016/j.atmosenv.2015.01.005
1352-2310/Published by Elsevier Ltd.

concentrations (Fiore et al., 2002, 2003; Zhang et al., 2011). The main sources of background ozone include natural emissions of ozone precursors (e.g., biogenic methane and volatile organic compounds [VOCs], as well as oxides of nitrogen [NO_x] from lightning and other natural processes), wildfires, transport of naturally occurring ozone from the stratosphere to the troposphere, and transport of anthropogenic ozone and ozone precursors from upwind regional and/or international locations. The first step in implementing an efficient plan for improving air quality is to develop a conceptual model of all the processes that lead to high concentrations of atmospheric pollutants within an airshed. As such, accurate estimates of the relative contribution of background ozone to observed ozone levels will be a key element in the development of air quality management plans for attainment of the ozone National Ambient Air Quality Standards (NAAQS), especially in the western U.S. where background influences tend to be greater.

The first ozone NAAQS was established in 1971 by the Environmental Protection Agency pursuant to its authority in the Clean Air Act to protect against the health and ecosystem risks associated with ambient oxidants (36 FR 8186). The earliest attempts to mitigate the ozone problem in the U.S. focused on local hydrocarbon controls in highly urbanized cities. While there was progress in reducing ozone, many of these urban areas failed to meet their NAAQS attainment goals in the mid-1970s. This prompted questions about the contribution of “background” processes (i.e., any sources outside the local region) to high ozone observations and how their impacts should be considered in the design of effective air quality control plans (Stasiuk and Coffey, 1974; Singh et al., 1977). Observational studies during the 1970s and 1980s began to quantify the local background that was associated with regional transport of ozone and ozone precursors in the eastern U.S. (Wolff et al., 1977; Clarke and Ching, 1983). By the early 1990s, most ozone control planning efforts were aimed at reducing regional NO_x emissions and local VOC emissions. Over the intervening decades, peak ozone concentrations improved considerably in California and the eastern U.S., but less so in rural portions of the western U.S. (Cooper et al., 2012; Simon et al., 2014), where ozone levels were generally lower. The increasing stringency of the ozone NAAQS, as needed to ensure adequate protection of public health, combined with the relatively unchanged ozone levels in the inter-mountain western U.S. have prompted increased concern about the role of background ozone levels in this region.

Initially, background ozone estimates were derived entirely from measurement studies, either by using direct measurements at relatively remote monitoring sites or via inference according to co-located measurements of ozone, NO_x, NO_y, and CO (Trainer et al., 1993; Altshuller and Lefohn, 1996). While some rural monitoring locations are affected substantially by background sources and may be suitable for comparisons with model results (McDonald-Buller et al., 2011), several analyses have shown that even the most remote ozone monitoring locations in the United States are periodically (or persistently) affected by U.S. manmade emissions (Parrish et al., 2009; Wigder et al., 2013). Further, most routine ozone monitoring sites across the U.S. do not have the co-located NO_x, NO_y, and CO measurements that are considered necessary to support inferential, observation-based, determinations of background contributions and monitoring coverage can be sparse in some locations. Because background contributions cannot be easily quantified using monitoring data, photochemical grid models have been widely used to characterize the contribution of background ozone to observed concentrations.

Over the past 10–15 years, several photochemical modeling analyses have been conducted to estimate the role of background sources on U.S. ozone levels. Many of these modeling analyses estimated a specific background metric which was intended to represent the ozone levels that would remain after North American anthropogenic emissions have been removed, or “zeroed out”. This metric is generally referred to as North American background (NAB), although older analyses referred to it as policy-relevant background. One of the first analyses to attempt to quantify NAB was conducted by Fiore et al. (2003) using a zero-out methodology and the GEOS-Chem global model (Bey et al., 2001) with a relatively coarse horizontal grid resolution ($2.0 \times 2.5°$). The modeling concluded that springtime afternoon ozone concentrations (1300–1700 local time) could occasionally reach 40–50 ppb at high altitude sites in the western U.S. without any contribution from North American anthropogenic emissions. Background concentrations in the eastern U.S. were substantially lower. Summer season average NAB concentrations were lower, generally ranging from 15 to 35 ppb. Subsequent zero-out GEOS-Chem modeling (Wang et al., 2009), conducted with a finer horizontal grid resolution ($1.0 \times 1.0°$), essentially confirmed the NAB estimates from the earlier study and separately estimated that anthropogenic emissions from Canada and Mexico could contribute an additional 3 ppb to mean ozone levels across the U.S., with larger impacts at near-border sites. A third set of zero-out modeling was conducted by Zhang et al. (2011) using an updated version of the GEOS-Chem model and still finer horizontal grid resolution ($0.5 \times 0.67°$). This analysis concluded that seasonal mean North American background levels generally ranged from approximately 20–35 ppb at low-altitude U.S. sites, but could exceed 45 ppb at certain high-altitude locations in the western U.S. Lin et al. (2012a,b) utilized a separate global high-resolution model (GFDL AM3, approximately 50×50 km^2) to estimate springtime NAB levels at high-elevation western U.S. sites and concluded that April–June mean NAB values could be approximately 50 ppb at these sites. The Lin et al. (2012a,b) study was one of the first background assessments to use a bias-correction technique to adjust the background estimates for ozone biases inherent in the base case model predictions. Fiore et al. (2014) compared background estimates between the GEOS-Chem and GFDL AM3 models and concluded that the varying model estimates of background magnitude resulted primarily from differences in the treatment of stratospheric–tropospheric exchange, wildfire emissions, lightning NO_x emissions, and isoprene oxidation chemistry between the modeling systems. This finding highlighted the need for targeted, process-level analyses to reduce error in model estimates of the contributions of these source categories when developing estimates of background ozone.

Emery et al. (2012) extended the traditional methodology for estimating North American background by conducting zero-out simulations with a coupled system, which used both global GEOS-Chem modeling and regional scale (12 km) CAMx modeling simulations informed by the boundary conditions derived from the coarser-scale global simulation. The coupled global-regional modeling yielded slightly higher estimates of North American background (25–50 ppb) than what had been previously been estimated by comparable stand-alone global modeling. These increases were partially attributed to the higher resolution in the regional modeling. Lefohn et al. (2014) further advanced the traditional approach for characterizing background ozone by utilizing a coupled global-regional modeling system that included the CAMx ozone source apportionment technique (Dunker et al., 2002; ENVIRON, 2013) to track the contribution of background sources to total ozone within the simulation. Because historically the NAB definition had been inherently linked to zero-out modeling, Lefohn et al. (2014) introduced a new metric called “emissions-influenced background” (EIB) which represented the combined influence of natural sources and sources of ozone from outside the modeling domain on total modeled ozone, as well as combined chemical interactions between the manmade and background sources. The