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Changes in US background ozone due to global anthropogenic emissions from 1970 to 2020

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HIGHLIGHTS

- CAMx regional model used to calculate US background ozone (USB) over five decades.
- Estimated contributions from neighboring countries in 2020 exceed 4 ppb near the Canadian border.
- Summer H4MDA8 USB across major 22 cities increased by 0.8 ppb/decade from 1970 to 2000.
- Increasing USB in the western US for all MDA8 metrics examined; top 30 MDA8 rose faster than H4MDA8 USB in most cities.
- Declining H4MDA8 USB after 2000 in the northeast due to reduction of Canadian emissions.

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ABSTRACT

Estimates of North American and US Background (NAB and USB) ozone (O_3) are critical in setting and implementing the US National Ambient Air Quality Standards (NAAQS) and therefore influence population exposure to O_3 across the US. NAB is defined as the O_3 concentration in the absence of anthropogenic O_3 precursor emissions from North America whereas USB excludes anthropogenic emissions inside the US alone. NAB and USB vary geographically and with time of year. Analyses of O_3 trends at rural locations near the west coast suggest that background O_3 is rising in response to increasing non-US emissions. As the O_3 NAAQS is lowered, rising background O_3 would make attaining the NAAQS more difficult. Most studies of changing US background O_3 have inferred trends from observations whereas air quality management decisions tend to rely on models. Thus, it is important that the models used to develop O_3 management strategies are able to represent the changes in background O_3 in order to increase confidence that air quality management strategies will succeed. We focus on how changing global emissions influence USB rather than the effects of inter-annual meteorological variation or long-term climate change. We use a regional model (CAMx) nested within a global model (GEOS-Chem) to refine our grid resolution over high terrain in the western US and near US borders where USB tends to be higher. We determine USB from CAMx simulations that exclude US anthropogenic emissions. Over five decades, from 1970 to 2020, estimated USB for the annual fourth highest maximum daily 8-h average O_3 (H4MDA8) in the western US increased from mostly in the range of 40–55 ppb to 45–60 ppb, but remained below 45 ppb in the eastern US. USB increases in the southwestern US are consistent with rising emissions in Asia and Mexico. USB decreases in the northeast US after 1990 follow declining Canadian emissions. Our results show that the USB increases both for the top 30 MDA8 days and the H4MDA8 (the former at a faster rate in most areas). Our USB increases in the western US are lower by about a factor of four than trends inferred from analyses of rural ozone near the west coast which is consistent with meteorology also influencing the observed ozone trends. Comparing H4MDA8 NAB and USB for 2020 shows that contributions from neighbouring countries can exceed 4 ppb near the Canadian border and 2 ppb near the Mexican border.

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1. Introduction

The ground-level ozone (O_3) is among the six criteria pollutants regulated by the US Environmental Protection Agency (EPA), which

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sets National Ambient Air Quality Standards (NAAQS) for these pollutants. The current O_3 NAAQS are based on a three-year average of the annual fourth highest maximum daily 8-h average (H4MDA8) ambient concentration. The EPA recently lowered the primary O_3 NAAQS from 75 parts per billion (ppb) to 70 ppb (EPA, 2015). Both the primary and secondary standards were set to the same level as has been the case earlier. Background O_3 levels are considered in setting the O_3 NAAQS and critically important in attaining those standards because the US background (USB) O_3 is the lowest O_3 achievable by reducing anthropogenic emissions within the US. Therefore, it is important to estimate USB as accurately as possible and to understand how USB may have changed over time. It has been shown that pollution from Asia can be transported to North America (Duce et al., 1980; Jaffe et al., 1999, 2003; Lin et al., 2012) and recent evidence suggests an increase in this transport (Parrish et al., 2009, 2012; Cooper et al., 2012; Gratz et al., 2015).

Definitions of background O_3 used in context of the O_3 NAAQS have changed over time. Prior to 2014, EPA defined "Policy-Relevant Background" (PRB) as ground-level O_3 that would exist in the absence of any anthropogenic emissions from North America (US, Canada, and Mexico). PRB, which is now termed "North American Background" (NAB), includes contributions from global natural sources (e.g., wildfires, biogenic, lightning NOx, and stratosphere-troposphere exchange). More recently, EPA (2014) has defined USB as similar to NAB but including influence of anthropogenic emissions from Canada and Mexico. USB is more relevant to attaining the O_3 NAAQS than NAB because USB includes influence of all anthropogenic emissions outside the domain of US regulations.

In the past decade, several techniques have been used to estimate background O_3. Prior to 2006, EPA determined background O_3 based on measurements at remote monitoring sites. In the previous O_3 NAAQS review, EPA (2006) estimated NAB in the mean range of 15–35 ppb using the GEOS-Chem global chemical transport model (Fiore et al., 2003). Since then, regional chemical transport models have been used to estimate NAB and USB, although global models continue to be widely used. While most NAB and USB modeling studies focused on one modeling year (Zhang et al., 2014; Lin et al., 2012; Emery et al., 2012; Mueller and Mallard, 2011; Dolwick et al., 2015) a few have examined multi-decade variation. A 27-year simulation (1981–2007) using a global climate model found year-to-year variation in mean MDA8 NAB to be higher in spring than summer and the largest variability was estimated over high elevation in the western US and New Mexico, with standard deviations of over 2 ppb (Fiore et al., 2014). Wild et al. (2012) considered the impacts of changing anthropogenic emissions over time by using parameterized responses of mean surface O_3 to multiple sources, based on 14 global chemistry transport models with 2001 meteorology. They found that for North America, external sources raised the annual mean surface O_3 by 2 ppb during 1960–1990 (~0.67 ppb/decade). These contributions then levelled off through 2000. Wang et al. (2009) used GEOS-Chem to simulate NAB in 2001 and 2020 and found that the mean MDA8 NAB in summer increased by 2 ppb, but decreased by 3–7 ppb in the northeast. This study also provided the first estimates of USB and the contributions from Canada and Mexico. Using ambient measurements, Parrish et al. (2012) analysed multi-year O_3 measurements at remote marine boundary layer (MBL) sites along the west coast and derived upward trends of 0.41 ppb/year in spring and 0.23 ppb/year in summer over two decades (up to 2010). Parrish et al. (2014) compared observation-derived trends with baseline model results from three different climate models, finding that the models under-predicted the rate of O_3 increase by about a factor of four (see their Fig. 9). Despite quantitative differences, the model-based and observational-based studies point to increasing USB.

We focus on how changing global emissions influence USB rather than the effects of inter-annual meteorological variation or long-term climate change. For this reason the meteorology is held constant (for 2005) and there are no changes to meteorology-dependent emissions (e.g., from fires, vegetation, lightning). Air quality planners in the US use the same approach to assess how changes in local emissions influence O_3. Because modelled background O_3 is influenced by grid resolution (Emery et al., 2012; Zhang et al., 2011), this study employs a regional model (CAMx) nested within a global model to refine spatial resolution within the US. Emery et al. (2012) found that a 12 km resolution regional model simulated US O_3 better than a global model, especially in regions with complex terrain. We simulate USB with global emissions representing six years within five decades (1970–2020) and characterize regional changes in USB. We compare our modelled USB changes in response to anthropogenic emissions to observation-derived trends for the west coast (Parrish et al., 2014).

2. Methodology

We use CAMx version 6.1 (ENVIRON, 2014) with the 2005 version of the Carbon Bond chemical mechanism (CB05; Yarwood et al., 2005) to simulate USB with anthropogenic emissions for 1970, 1980, 1990, 2000, 2005, and 2020 (See Table 1S for model configurations). The meteorology and all natural emissions, including wild fires, are held constant for 2005 to isolate the effect of changing non-US anthropogenic emissions on USB. The 2005 US anthropogenic emissions are used to evaluate model performance but are not needed to simulate USB. The modeling domain has 36 km resolution and covers the lower 48 states (Figure S1). The vertical domain has 14 layers, which span the entire troposphere and the lower stratosphere to a pressure altitude of 100 mb. The CAMx 2005 Base Case emissions (except for biogenic) and meteorology are from the EPA's Final Transport Rule modeling platform (referred to as CSAPR2005 database; EPA, 2011) which adopts the 2005 National Emissions Inventory (NEI). Biogenic emissions are from the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006).

The emissions inventory is a crucial component of any air quality modeling study and this study of long-term background estimates needs consistent inventory methods and underlying data over extended time periods. Our approach projects finely resolved modeling inventories to other years using national-level inventories that are available for many years. The CSAPR2005 database includes Canada's 2006 inventory (Environment Canada, 2011) and Mexico's Phase III 1999 inventory (Eastern Research Group Inc., 2006). The Mexico 1999 emissions are projected to 2005 and other historical years using the Emissions Database for Global Atmospheric Research (EDGAR) inventory (EDGAR, 2011). We adopt a similar approach for Canada by assuming that 2006 emissions are representative of 2005 and then adjusting from 2005 to other years. The Representative Concentration Pathway (RCP) 8.5 scenario (RCP8.5; Riahi et al., 2007) is the basis for our 2020. Among four available RCP scenarios, RCP8.5 has the-least aggressive emission reductions over the period 2000–2100; therefore it is most likely to represent actual emissions for the time horizon of this study (out to 2020). The resulting emission (Table 1) show increasing Mexican emissions but declining Canadian emissions after 2000. Mexican and Canadian emissions were projected at a national level and so spatial distributions remained unchanged from the respective base years.

Contributions from outside of North America are provided to CAMx through lateral boundary conditions (BCs) obtained from our global $2 \times 2.5^{\circ}$ degree GEOS-Chem model simulations. GEOS-Chem

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