Atmospheric Environment 167 (2017) 116–128

Contents lists available at ScienceDirect

Atmospheric Environment

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

Source contributions to United States ozone and particulate matter over five decades from 1970 to 2020

Uarporn Nopmongcol [a, *], Yesica Alvarez [a], Jaegun Jung [a], John Grant [a], Naresh Kumar [b], Greg Yarwood [a]

[a] Ramboll Environ, 773 San Marin Drive, Suite 2115, Novato, CA 94945 USA
[b] Electric Power Research Institute, 3420 Hillview Avenue, Palo Alto, CA 94304 USA

HIGHLIGHTS

- U.S. emissions contributions to ozone and $PM_{2.5}$ have reduced over five decades.
- On-road mobile and EGU contributions have declined substantially since 1970.
- Contributions of non-EGU and area sources are declining less since 2005.
- Ozone production efficiency rises from 2000 onwards as NOx emissions are reduced.
- Inter-regional transport is becoming less important in contrast to background ozone.

ARTICLE INFO

Article history:
Received 20 December 2016
Received in revised form
27 July 2017
Accepted 6 August 2017
Available online 8 August 2017

Keywords:
Ozone
Particulate matter
$PM_{2.5}$
Air quality trends
Photochemical modeling
CAMx
Source apportionment
OSAT
PSAT
Emission inventories
Emission trends

ABSTRACT

Evaluating long-term air quality trends can demonstrate effectiveness of control strategies and guide future air quality management planning. Observations have shown that ozone (O_3) and fine particulate matter ($PM_{2.5}$) in the US have declined since as early as 1980 in some areas. But observation trends alone cannot separate effects of changes in local and global emissions to US air quality which are important to air quality planners. This study uses a regional model (CAMx) nested within a global model (GEOS-Chem) to characterize regional changes in O_3 and $PM_{2.5}$ due to the intercontinental transport and local/regional emissions representing six modeling years within five decades (1970–2020). We use the CAMx Source Apportionment Technology (OSAT/PSAT) to estimate contributions from 6 source sectors in 7 source regions plus 6 other groups for a total of 48 tagged contributions. On-road mobile sources consistently make the largest U.S. anthropogenic emissions contribution to O_3 in all cities examined even though they decline substantially from 1970 to 2005 and also from 2005 to 2020. Off-road mobile source contributions increase from 1970 to 2005 and then decrease after 2005 in all of the cities. The boundary conditions, mostly from intercontinental transport, contribute more than 20 ppb to high maximum daily 8-h average (MDA8) O_3 for all six years. We found that lowering NOx emissions raises O_3 formation efficiency (OFE) across all emission categories which will limit potential O_3 benefits of local NOx strategies in the near future. $PM_{2.5}$ benefited from adoption of control devices between 1970 and 1980 and has continued to decline through 2005 and expected to decline further by 2020. Area sources such as residential, commercial and fugitive dust emissions stand out as making large contributions to $PM_{2.5}$ that are not declining. Inter-regional transport is less important in 2020 than 1990 for both pollutants.

1. Introduction

Since the 1970 Clean Air Act, the U.S. Environmental Protection Agency (EPA) has regulated US criteria air pollutants to improve air quality, visibility and acid deposition. Major regulations include passenger vehicle emission controls for nitrogen oxide (NOx), volatile organic compounds (VOCs) and carbon monoxide (CO), which have tightened progressively from Tier 0 through Tier 3 (EPA, 2016a), New Source Performance Standards for industrial sources affecting VOC, NOx, CO, sulfur dioxide (SO_2), and particulate matter

* Corresponding author.
E-mail address: unopmongcol@ramboll.com (U. Nopmongcol).

http://dx.doi.org/10.1016/j.atmosenv.2017.08.009
1352-2310/

(PM) emissions from specific stationary source categories (EPA, 2016b), the Acid Rain Program covering major SOx and NOx sources such as electric utilities (EPA, 2016c), the NOx SIP Call (Federal Register, 1998), to name a few. Previous studies have evaluated long-term trends in air quality and emissions in the US to demonstrate the effectiveness of emission reductions strategies (Simon et al., 2015; Blanchard et al., 2010; Butler et al., 2011; Sather and Cavender, 2012; Hidy and Blanchard, 2015). This information can help in guiding future air quality management plans. The planning in many cases can be complicated by non-controllable factors including intercontinental transport (Jaffe et al., 2003; Lin et al., 2012), meteorology influences (Camalier et al., 2007; Cox and Chu, 1996; Lin et al., 2015), as well as interactions among reacting species.

Impacts from intercontinental transport to U.S. air quality depend on emissions in other regions in the world which have changed significantly in the last five decades. Europe has taken extensive measures to reduce emissions which have resulted in a reduction of pollution recorded between 1980 and 2000 (Lovblad et al., 2004). However, fast growing economies contribute to increased emissions in parts of Asia (Streets and Waldhoff, 2000; Akimoto, 2003; Richter et al., 2005). Such spatial changes in global emissions can potentially change background pollution levels in different regions of US, thus they need to be considered when evaluating U.S. trends. In fact, evidence has suggested recent increase of foreign contributions in the U.S. (Parrish et al., 2009, 2012; Cooper et al., 2012; Gratz et al., 2015). Therefore, an accurate description of temporal and spatial variations at regional and global scales in long-term emissions inventories is crucial in trend studies.

Air quality models (AQMs) have been increasingly used to demonstrate effectiveness of control scenarios and long-range transport. AQMs can overcome some deficiencies of spatial and temporal data of ambient monitoring networks. For example, Hogrefe et al. (2009) simulated $PM_{2.5}$ over the northeastern United States for 1988–2005 and integrated with observations to provide greater spatial coverage and speciation information in addition to total mass. AQMs can also help separate the effects of intercontinental transport and domestic contributions (Dolwick et al., 2015; Wild et al., 2012; Hogrefe et al., 2004). To date, most AQM applications in the U.S. are applied for short episodes or single years (Goldberg et al., 2016; Dolwick et al., 2015; Nopmongcol et al., 2014 to name a few). Only a few have extended to multi-years (Bouchet et al., 1999; Pierce et al., 2010; Godowitch et al., 2010; Hogrefe et al., 2011; Xing et al., 2015). Most recent air quality modeling by Xing et al. (2015) applied a regional model with 108 × 108 km resolution across the northern hemisphere over 1990–2010 period. Their trends represent sub-grid variability as well as changes in local emissions. While such an approach allows a direct comparison to observations, it cannot separate effects of changes in local and global emissions to US air quality, which are important to air quality planners.

We focus on how changing global and local emissions influence air quality rather than the effects of inter-annual meteorological variation or long-term climate change. A clear focus on the influence of emission trends can provide useful information to motivate needed inventory improvements. Meteorological changes can mask the trend attributable to changes in precursor emissions (Lin et al., 2015). For this reason the meteorology is held constant (for 2005) and there are no changes to meteorology-dependent emissions (e.g., vegetation, from fires). Air quality planners in the U.S. use the same approach to assess how changes in local emissions influence Ozone (O_3) and $PM_{2.5}$. We simulate intercontinental transport with global emissions and characterize regional changes in O_3 and $PM_{2.5}$ due to the intercontinental transport and local emissions representing six years within five decades (1970–2020). We track contributions from each source sector and each US region throughout the five decades.

2. Methodology

2.1. Air quality modeling

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 O_3 and $PM_{2.5}$ with anthropogenic emissions for 1970, 1980, 1990, 2000, 2005, and 2020 using the model configurations described in Nopmongcol et al. (2016). The meteorology and all natural emissions, including wild fires, are held constant for 2005 to isolate the effect of changing anthropogenic emissions outside the US. The modeling domain has 36 km resolution and covers the lower 48 states. Gan et al. (2016) compared simulations with 36 km and 12 km grid resolution and concluded that 36 km resolution can describe long term trends, but we consider limitations of using 36 km grid when analysing results. 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 and meteorology are from the EPA's Final Transport Rule modeling platform (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) with 2005 meteorology. Boundary conditions (BCs) are from global 2 × 2.5° degree GEOS-Chem model simulations described in a companion study (Nopmongcol et al., 2016). Briefly, we use GEOS-Chem version 9-01-03 with different anthropogenic emissions for each emission year (1970, 1980, etc.) but meteorology and natural emissions for 2005, as for CAMx. Anthropogenic emissions for the U.S. are from the NEI and elsewhere from the Emissions Database for Global Atmospheric Research version 4.1 (EDGAR, 2011) and other inventories with 2020 future emissions estimated using RCP 8.5 (Riahi et al., 2011).

We use the CAMx Ozone Source Apportionment Technology (OSAT) and Particulate Source Apportionment Technology (PSAT) tagged species methods for O_3 and $PM_{2.5}$ (Dunker et al., 2002; Yarwood et al., 2007) to compute contributions from 6 source sectors (Table S1) in 7 source regions (Figure S3) plus 6 other groups for a total of 48 tagged contributions. The OSAT and PSAT methods have been used in many other studies including EPA (2011), Fann et al. (2013) and Dolwick et al. (2015). Tagged species methods have been developed for other photochemical grid models such as ISAM (Kwok et al., 2013). Our 7 source regions simplify 9 climate regions defined by the National Oceanic and Atmospheric Administration (NOAA) and are named west (W), north rockies and plains (NRP), southwest (SW), south (S), southeast (SE), Midwest (MW), and northeast (NE). The 6 emission categories tracked from each of 7 U.S. regions are: on-road mobile sources; off-road mobile sources (e.g., rail, air, construction, agriculture); electrical generating units (EGU); non-EGU point sources; area sources (i.e. stationary sources that are estimated collectively such as domestic heating or natural gas wells), and; agricultural ammonia (NH_3). The 6 remaining tagged groups are: Canadian anthropogenic emissions; Mexican anthropogenic emissions; international shipping; natural emissions from vegetation and all fires, and; the CAMx BCs.

Relationships between O_3 and precursors are non-linear causing apportionment results to depend upon methodology (Dunker et al., 2002, 2015) and we expect our results from the OSAT tagged species method to be similar but not identical to alternatives of setting selected emissions to zero sequentially (zero-out) or integrated