CRC Report No. A-99

Contributions to US Ozone of US, non-US and Natural Emissions

Executive Summary

February 2017

COORDINATING RESEARCH COUNCIL, INC.

5755 NORTH POINT PARKWAY·SUITE 265·ALPHARETTA, GA 30022

The Coordinating Research Council, Inc. (CRC) is a non-profit corporation supported by the petroleum and automotive equipment industries. CRC operates through the committees made up of technical experts from industry and government who voluntarily participate. The four main areas of research within CRC are: air pollution (atmospheric and engineering studies); aviation fuels, lubricants, and equipment performance, heavy-duty vehicle fuels, lubricants, and equipment performance (e.g., diesel trucks); and light-duty vehicle fuels, lubricants, and equipment performance (e.g., passenger cars). CRC's function is to provide the mechanism for joint research conducted by the two industries that will help in determining the optimum combination of petroleum products and automotive equipment. CRC's work is limited to research that is mutually beneficial to the two industries involved, and all information is available to the public.

Contributions to US Ozone of US, non-US and Natural Emissions

CRC A-99
EXECUTIVE SUMMARY

Prepared for:
Coordinating Research Council, Inc.
5755 North Point Parkway, Suite 265
Alpharetta, GA 30022

Prepared by:
Ramboll Environ
773 San Marin Drive, Suite 2115
Novato, California, 94998
www.ramboll-environ.com
P-415-899-0700
F-415-899-0707

February 23, 2017

RAMBOLL ENVIRON

EXECUTIVE SUMMARY

In 2008, the US Environmental Protection Agency reduced the National Ambient Air Quality Standard (NAAQS) for 8-hour ozone to 75 ppb, and in October 2015, further reduced the NAAQS to 70 ppb. An important consideration for regulators and the regulated community is how difficult it will be to meet this standard by reducing US emissions alone. One issue is the magnitude of the ozone background in the absence of anthropogenic emissions. Another issue is the contribution of anthropogenic emissions from outside the US to US ozone. This can occur by the transport of ozone and other secondary pollutants formed outside the US from anthropogenic emissions but also by the transport of the anthropogenic emissions themselves. The objective of this project was to estimate the relative importance of US anthropogenic emissions, other anthropogenic emissions, and natural emissions to the total ozone concentrations.

We used the Goddard Earth Observing System global chemical transport (GEOS-Chem) model to simulate ozone in year 2010 with and without worldwide anthropogenic emissions (G-Base and G-Bkgd cases). These simulations provided boundary concentrations (BCs) for a North American domain encompassing 48 US states plus parts of Canada and Mexico. The Comprehensive Air-quality Model with Extensions (CAMx) was then applied to the domain for simulations of March to September, 2010 with and without North American anthropogenic emissions (NA-Base and NA-Bkgd cases). The NA-Base and NA-Bkgd cases used BCs from the G-Base and G-Bkgd cases, respectively. The NA-Bkgd case thus represents the natural ozone in the US, the NA-Base case represents ozone in year 2010, and the difference (NA-Base minus NA-Bkgd) is the anthropogenic increment to total ozone. Figure ES-1 shows the relationship among the simulations.

An alternative global model is the Geophysical Fluid Dynamics Laboratory's Atmospheric Model 3 (AM3). GEOS-Chem and AM3 have important differences in biogenic isoprene emissions and chemistry, lightning NO_x and wildfire emissions, and stratosphere–to–troposphere transport. Previous work found that CAMx simulations using BCs from GEOS-Chem had better performance for maximum daily average 8-hour (MDA8) ozone at rural sites in spring than simulations using BCs from AM3. Considering this work, we chose GEOS-Chem for the present study. However, GEOS-Chem may have less stratosphere-troposphere exchange than AM3, which may be the cause of the smaller background O_3 concentrations with GEOS-Chem than AM3 found in another study for the western US in spring. Hence, using BCs from AM3 for the CAMx simulations may give greater background (and base-case) O_3 at western sites in spring.

The Path-Integral Method (PIM) for source apportionment (developed in CRC Project A-90) has a unique capability to allocate the difference in ozone between two simulations (e.g., the anthropogenic increment) to portions of the emissions change between the two simulations. In Project A-90, we allocated the ozone increment due to US anthropogenic emissions to the major source categories, e.g., light-duty vehicles, area sources, etc. For this project, we divided the anthropogenic increment to US ozone into the four source contributions: 1) US anthropogenic emissions in the CAMx domain; 2) Canadian/Mexican anthropogenic emissions in the domain; 3) the anthropogenic component of the lateral BCs; 4) the anthropogenic component of the top BCs. The latter two sources represent pollutants from anthropogenic emissions outside the domain that arrive through the boundaries. Together, the four sources account for the impact of all anthropogenic emissions, worldwide, on the US ozone.

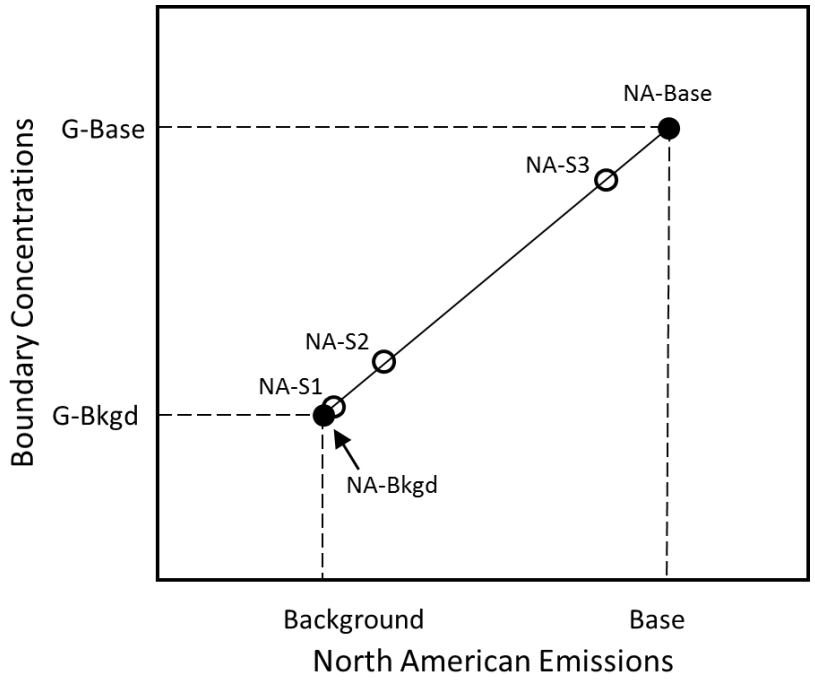


Figure ES-1. Schematic representation of the simulations with CAMx for the North American domain. The sensitivities are integrated numerically from the background case to the base case using values of the sensitivities at the points NA-S1, NA-S2, NA-S3 along the path. At these points, the boundary concentrations from GEOS-Chem and the anthropogenic emissions are reduced from those in the base case.

The PIM determines the source contributions by integrating first-order sensitivity coefficients over a range of emissions, a path, from the NA-Bkgd case to the NA-Base case. Viewed in

reverse, the path is a scenario for reducing emissions from the base case to achieve the background case. We chose a path on which all emissions are reduced by the same factor, which is an unbiased approach that assumes future controls on anthropogenic emissions will produce similar fractional reductions in different regions. (A different path could be chosen for the integration, but we know of no other assumption for emission controls that has a better justification.) We used the Decoupled Direct Method (DDM) in CAMx to calculate the first-order sensitivities of ozone to the four sources of anthropogenic emissions at three points, NA-S1, NA-S2, and NA-S3, shown in Figure ES-1. Each of these points represents a simulation with anthropogenic emissions and BCs between the NA-Bkgd and NA-Base simulations. The points were chosen to give accurate numerical integration of the DDM sensitivity coefficients via a Gauss–Legendre formula. The relationship of the PIM to the GEOS-Chem and CAMx simulations is illustrated in Figure ES-2.

CRC Project A-95, Modeling Inter-continental Transport of Ozone in North America with CAMx for the Air Quality Model Evaluation International Initiative (AQMEII) Phase 3, also estimated the contribution of boundary ozone to North American ozone. There are some key differences between A-95 and A-99, however. In A-95, the contribution of the total ozone concentration entering via the boundaries was determined. In A-99, we focused only on the anthropogenic component of the total ozone (and other pollutants) entering the domain. Also, the Composition-Integrated Forecast System model provided the BCs for A-95 whereas GEOS-Chem was used for A-99. Lastly, A-95 used reactive tracers to determine the boundary ozone contribution, and A-99 determined the contribution by the PIM.

We evaluated the GEOS-Chem model performance for ozone using surface and ozonesonde sites inside and outside the US. At sites outside the US, there is both over- and underprediction compared to the measurements, leading to some uncertainty in the BCs for the North American domain. GEOS-Chem consistently overpredicted ozone in summer at sites within the US but this should not directly influence our modeling for the North American domain, which used CAMx. Evaluation of CAMx predictions for MDA8 ozone at EPA's Clean Air Status and Trends Network (CASTNet) and Air Quality System (AQS) monitoring sites showed underprediction at the highest concentrations. However, with a 40-ppb cutoff, the normalized mean bias was < ±5% and the normalized mean error was < 15%. This performance is generally considered acceptable and is like that obtained in Project A-95.

The natural background MDA8 ozone predicted for North America is larger in the western US and Mexico than in the eastern US. The spatial pattern is similar in spring and summer with the exceptions that concentrations are smaller in Mexico and larger in Canada in summer. The largest background MDA8 ozone in the US is in the mountainous areas of Colorado, New Mexico, Arizona, and California. Averaging the background over the 10 days with the largest

base-case MDA8 ozone (T10Base average), the background ozone at Denver is 50 ppb, which is 69% of the corresponding base-case concentration (72 ppb). The T10Base background ozone exceeds 60 ppb in some other areas of Colorado, New Mexico, Arizona, and California.

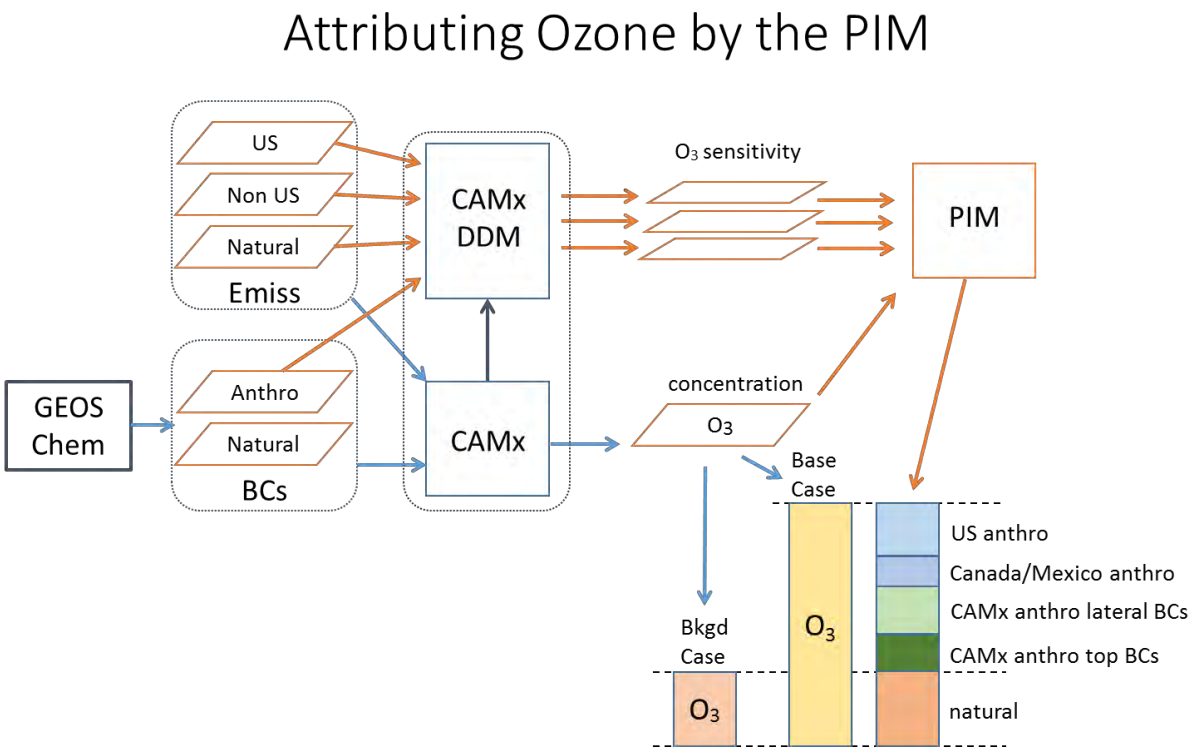


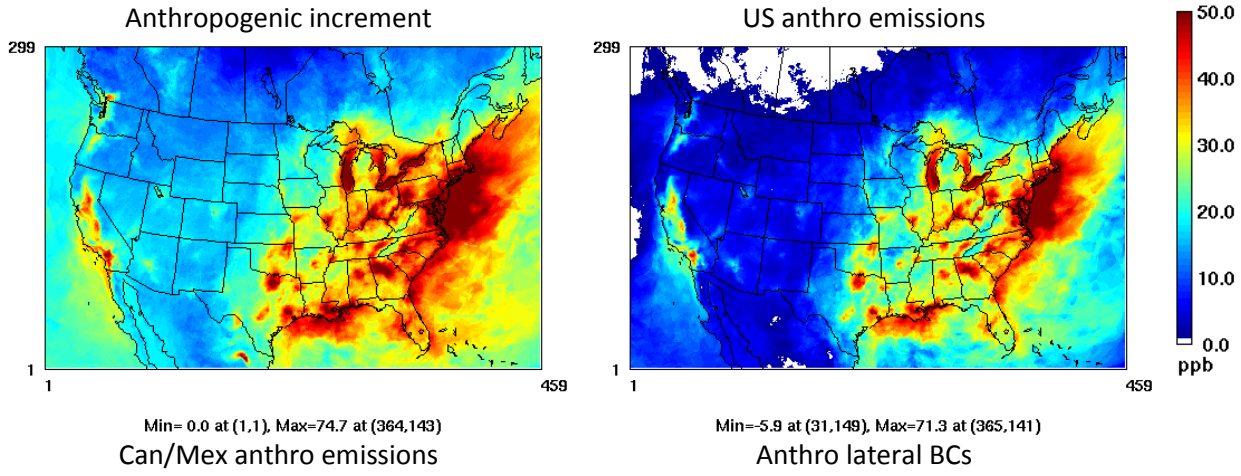
Figure ES-2. Schematic diagram of source apportionment by the Path-Integral Method (PIM). Two worldwide GEOS-Chem simulations with and without anthropogenic emissions provide boundary concentrations (BCs) for the corresponding CAMx simulations. Two CAMx simulations with and without anthropogenic emissions give the base and background cases for North America. Three CAMx simulations with emissions and BCs between the base and background cases, using the decoupled direct method (DDM), provide the sensitivities needed to allocate the anthropogenic increment to ozone (base minus background cases) to four source categories.

For the larger MDA8 ozone concentrations in the base case, the relative importance of the anthropogenic sources is generally US emissions > anthropogenic lateral BCs > Canadian/Mexican emissions >> anthropogenic top BCs. The US anthropogenic emissions are the largest contributor in ppb to the anthropogenic ozone increments in the eastern US and California, as seen in Figure ES-3. The contributions of the anthropogenic lateral BCs are largest for the higher elevation US sites in the Intermountain West and sites closest to the boundaries. The Canadian/Mexican emissions are third in importance, affecting the northern, east-coast, and southwest US and some interior states. The contribution of the anthropogenic top BCs is

always very small, ≤ 0.5 ppb or ≤ 3% of the anthropogenic increment. For three AQS and two CASTNet sites, the relative contributions of the sources to the ozone increment are shown on the left side of Figure ES-4, as calculated with the T10Base average. Denver and Big Bend are at higher elevation (> 1 km).

We also examined results for the 10 days with the largest MDA8 ozone in the background case (T10Bkgd average). The anthropogenic ozone increment is smaller with the T10Bkgd average than the T10Base average due to a reduced contribution from US emissions. The contribution of the Canadian/Mexican emissions remains about the same, and the contribution from the lateral BCs increases by up to 5 ppb at lower elevation urban sites. The net effect is that the relative importance of the lateral BCs is significantly increased for the days with the largest background concentrations, as shown on the right side of Figure ES-4. At Denver and Big Bend, the lateral BCs account for 71% and 67%, respectively, of the anthropogenic increment using the T10Bkgd average.

Global and regional models are continuing to evolve as new data and analyses become available. There have been numerous studies published in the past two years proposing changes to lightning NO_x and US anthropogenic NO_x emissions and revisions to the chemistry, e.g., the addition of halogen reactions. Implementing such changes in GEOS-Chem and/or CAMx will likely bring changes to the model results for ozone and the source contributions.



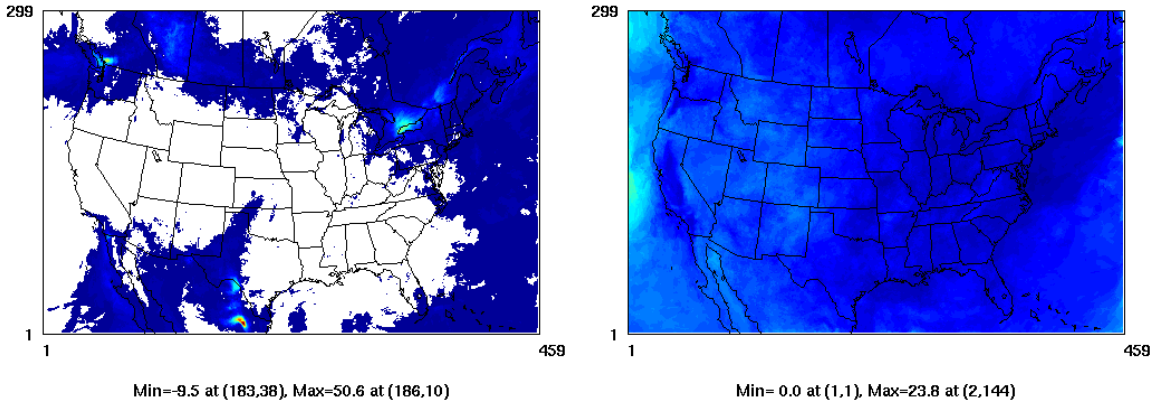


Figure ES-3. The anthropogenic increment averaged over the 10 largest MDA8 O_3 concentrations in the base case and the contributions to this increment. The contribution from the anthropogenic component of the top BCs (not shown) is ≤ 0.5 ppb.

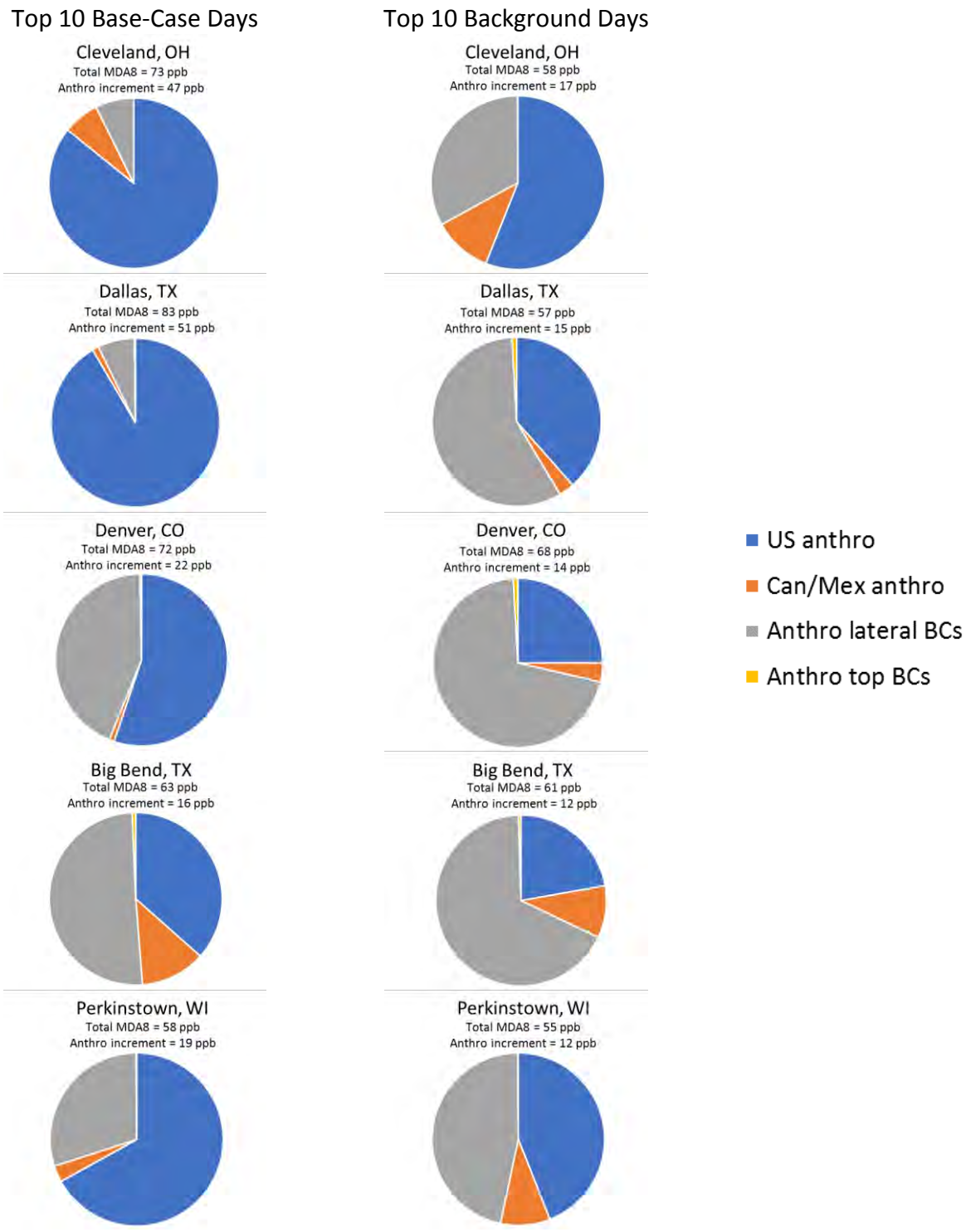


Figure ES-4. Relative contributions of anthropogenic sources to the anthropogenic MDA8 ozone increment (base case minus background case). Results are averages over the 10 days with the largest ozone in the base case (left) and averages over the 10 days with the largest ozone in the background case (right). Also shown is the total MDA8 ozone in the base case.