Importance of Background Ozone for Air Quality Management

by Daniel A. Jaffe, Arlene M. Fiore, and Terry J. Keating

An overview of the sources and variability of background ozone and how to quantify them for improved air quality management in the United States.

U.S. background ozone (BG O_3) is the quantity of O_3 resulting from all sources other than U.S. anthropogenic emissions.[1-4] In essence, it is the lowest level of O_3 that could be achieved if all domestic anthropogenic sources of nitrogen oxides (NO and NO_2 = NO_x) and volatile organic compounds (VOCs) were eliminated. While methane (CH_4) has both domestic and international sources, typically its contribution to O_3 is included with the background share.[2] BG O_3 is a construct that can be estimated with global or global-to-regional chemical transport models (CTMs), which should be informed by observations. In contrast, measured O_3 includes both BG O_3 and the U.S. anthropogenic contribution.

It is important to distinguish between BG O_3 and "baseline O_3," which is a measured quantity determined by observations at remote sites away from local anthropogenic sources.[2] The U.S. National Ambient Air Quality Standard (NAAQS) for O_3 is met if the three-year average of the annual fourth highest maximum daily 8-hr average (MDA8) is 0.070 parts per million (ppm; or 70 parts per billion, ppb) or lower. BG O_3 varies in space and time, and in some cases, can approach or exceed the current NAAQS threshold of 70 ppb. Since BG O_3 cannot be controlled by domestic regulations, it is important to understand the sources and variability in BG O_3 and our ability to quantify them.

BG O_3 includes O_3 produced by natural sources within the United States, such as O_3 produced by the interaction of NO_x from wildfires, lightning, or soils with biogenic VOCs (BVOCs). O_3 can also be produced from the NO_x and/or VOCs emitted from wildfires. Some O_3 is transported to the surface from the upper troposphere/lower stratosphere (UTLS) or produced by international anthropogenic precursor emissions or by international natural precursor emissions (wildfires, BVOC, soil and lightning NO_x). While in theory, one can account for O_3 produced during oxidation of CH_4 emitted outside U.S. borders, as well as the natural portion of U.S. CH_4 emissions (e.g., from U.S. wetlands), in practice, air quality models are rarely configured to separate out the sources of CH_4.

A major challenge for our attribution is that natural or international anthropogenic sources of NO_x or VOCs can interact with "domestic" O_3 or emissions from U.S. anthropogenic sources in complex ways through atmospheric chemistry. U.S. anthropogenic O_3 includes the sum of contributions from O_3 produced from local anthropogenic emissions (e.g., within urban airshed), as well as O_3 produced and transported from an upwind airshed (e.g., inter-state transport). The policy context for NAAQS implementation requires understanding the role of each component of BG O_3 and U.S. anthropogenic O_3 on days when observed MDA8 O_3 exceeds the NAAQS level (i.e., 0.070 ppm).

The U.S. Clean Air Act (CAA) provides a variety of mechanisms to account for separate components of BG O_3 in air quality planning and provides some regulatory relief, depending on the source. For example, international transport of anthropogenic air pollution is addressed in CAA §179B;5 stratospheric O_3 intrusion or wildfire smoke events may be addressed by the exceptional events policy under CAA §319. Rural areas with high O_3 that are downwind of major cities can be designated as Rural Transport Areas under CAA §182(h), since the benefit of local controls may be minimal. However, the ability to utilize these provisions in air quality planning depends on accurate and quantitative attribution of each BG O_3 component to specific processes.

A number of examples of model performance for BG O_3 are discussed in Jaffe et al.[2] One recent study investigated BG O_3 across the United States using the GEOS-Chem global CTM.[6] While the model captured many of the key features of the surface O_3 distribution by season and region, this model—similar to previous work—demonstrated significant biases that need to be explored if we are to fully understand the sources of O_3 in the United States. Figure 1 shows the model calculated BG O_3 and domestic O_3 contributions for two U.S. Environmental Protection Agency (EPA) regions. These are quantified from a set of sensitivity simulations described by Guo et al.[6] We show the top 10 summer days (June–August), averaged over 2004–2012, across EPA Regions 2 (NY, NJ) and 8 (CO, MT, ND, SD, UT, WY), including all urban, rural, and more remote monitors in these regions. The figure shows the observed and modeled values averaged across all monitors on the same days. The averaged data presented here are not intended to reflect attainment status, which is determined based on measurements at individual monitors after screening for exceptional events.

While these two regions have similar mean concentrations for the top 10 days (67 ppb for Region 2 and 60 ppb for Region 8), the model indicates a remarkably different breakdown of BG O_3 and U.S. anthropogenic contributions on the 10 highest O_3 days in each region. For Region 2, the BG O_3 and U.S. anthropogenic values are 42 and 38 ppb, respectively, and for Region 8, the same values are 57 and 12 ppb, respectively. This difference reflects the generally higher altitudes in the Intermountain West and the larger population and anthropogenic emissions in the East. Also note that for Region 8, the "Other USBO" category, which includes both stratospheric and wildfire contributions, is the largest single share of O_3 on these days. We note that these two quantities are particularly challenging to resolve accurately with a global CTM.[2,7-9] Furthermore, we note that averaging across the region can mask the large contributions from anthropogenic sources that occur in metropolitan areas in Region 8.

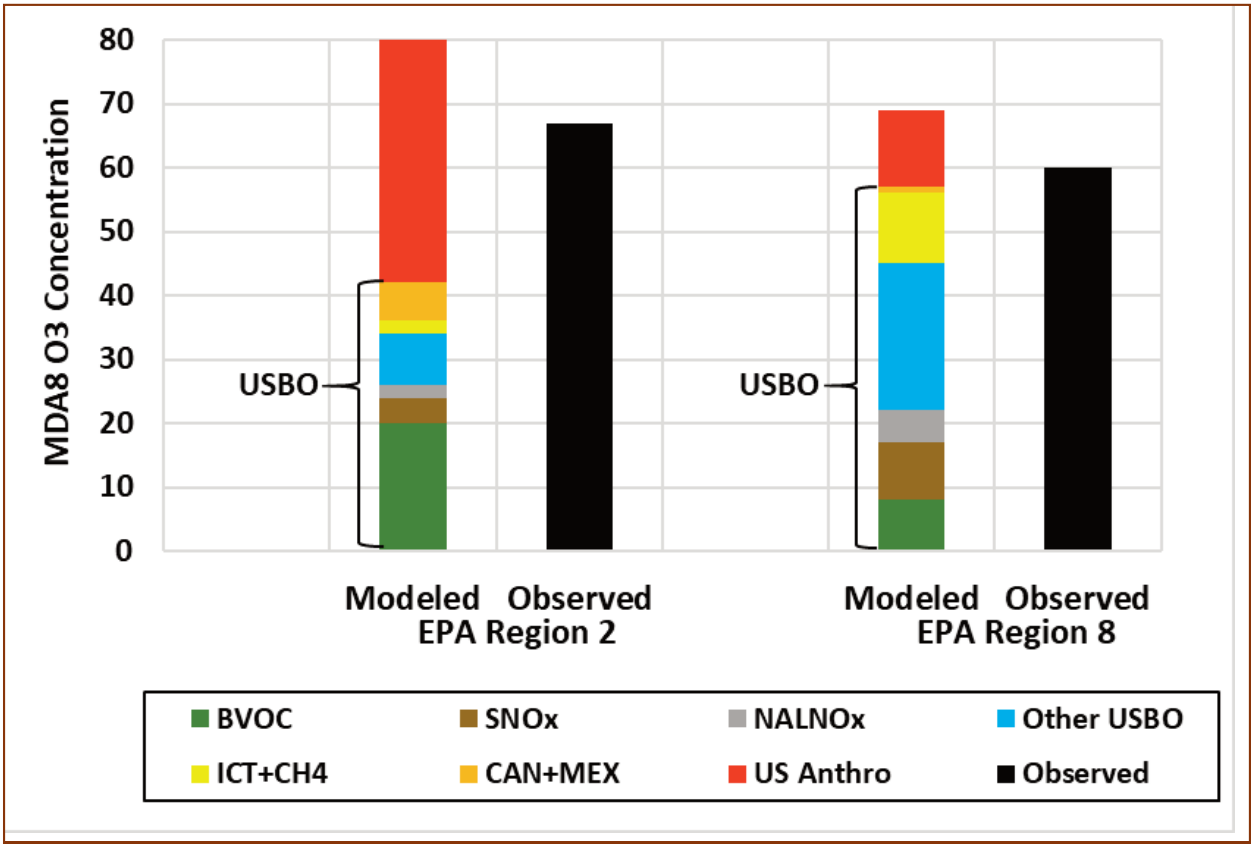


Figure 1. Model calculated contributions to the MDA8 O_3 and observed MDA8 O_3 for the 2004–2012 average of the 10 highest observed summertime (June–August) days each year for all sites in EPA Region 2 (NY, NJ; left side of figure) and EPA Region 8 (CO, MT, ND, SD, UT, WY; right side of figure).[6]

Notes: USBO = total U.S. BG O_3; ICT+CH_4 = contributions to O_3 from intercontinental anthropogenic emissions plus all CH_4; BVOC = contributions to O_3 from biogenic VOC emissions; CAN+MEX = contributions to O_3 from Canadian and Mexican anthropogenic emissions; SNO_x = contributions to O_3 from soil NO_x emissions; $NALNO_x$ = contributions to O_3 from lightning NO_x over North America; US Anthro = contributions to O_3 from U.S. anthropogenic emissions; Other USBO = all other BG O_3 sources, including wildfires, stratospheric O_3, lightning outside of North America, etc. Individual contributions are determined from a combination of separate zero-out emission perturbation simulations or from the difference of more than one sensitivity simulation. The BVOC contribution is not strictly a natural contribution, as it includes the interaction of BVOCs with all available NO_x, including anthropogenic NO_x. For details, refer to Guo et al., 2018 [ref. 6].

While the CTM provides estimates of the contributions from specific sources, the model also shows significant bias in MDA8 O_3 for these top 10 days: +13 ppb for Region 2 and +9 ppb in Region 8 (difference between modeled and observed values in Figure 1). These biases are a significant fraction of the total observed O_3; more importantly, we do not understand the cause(s) of these biases, and they must be understood to improve models or apply meaningful bias corrections. Observations from the U.S. ground-level monitoring network can confirm only the total amount of O_3 at the surface. Without additional chemical and meteorological tracers and significant research effort, it is very difficult to utilize the O_3 observations by themselves to quantify the sources of O_3.

Note here that while we tend to focus on sources, as this is what is required for policy implementation, errors in sinks, especially those that are spatially non-uniform, as is the case for O_3 dry deposition or reactions with halogens, can also be important contributors to model biases. Errors in the loss terms would not necessarily affect all sources of BG O_3 or U.S. anthropogenic O_3 proportionally. A recent review2 suggested that the uncertainty in model-calculated BG O_3 was approximately 10 ppb for seasonal mean estimates. The uncertainty for individual days or for the top 10 days could be even larger. This degree of uncertainty limits the ability to apply the regulatory relief provisions of the CAA discussed above.

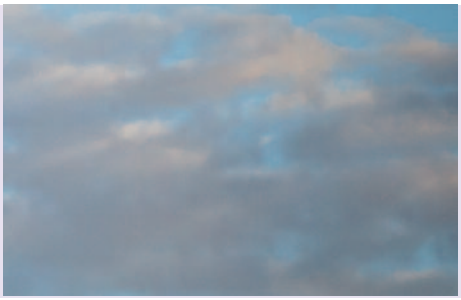
In addition to "routine" contributions to BG O_3, there can also be, on occasion, substantial contributions from a specific

source. For O_3, the most important and most common non-domestic sources are from UTLS intrusions or wildfire smoke, either of which can add up to 40 ppb to the MDA8 ppb on specific days.[2] In addition, O_3 from anthropogenic sources in Mexico or Canada in the adjacent border regions can contribute significantly on some days.[10] In some regions, dry stratospheric air can enhance the occurrence of wildfires, making attribution especially complex.[11] Modeling of UTLS and smoke events is very challenging[8,9] and detailed observations of tracers are needed to identify the sources of enhanced O_3 and improve their representation in models.

The combination of routine BG O_3 contributions and exceptional events from wildfire and stratospheric sources result in a challenging dynamic for air quality management. Similar challenges apply to particulate matter and implementation of the Regional Haze Rule (https://www.epa.gov/visibility/regional-haze-program). Addressing these challenges will require new investments in focused research and monitoring programs to better understand all O_3 sources and reduce the uncertainty in BG O_3 estimates (as well as estimates of regional haze contributions.[12]

Specifically, a program of targeted observations coordinated with an ensemble of high-resolution CTM simulations that track the various BG O_3 and U.S. anthropogenic source contributions is needed. Targeted observations could include strategic locations that would better inform BG O_3 (e.g., aircraft, mountain top, sondes) and with specific tracers (e.g., CO, NO_y, water vapor, chemically speciated PM) to help elucidate O_3 and haze sources. Model simulations would need to compute and archive high-resolution fields of O_3, related chemical and meteorological tracers, track source contributions—for example, through online source apportionment tools or a set of emission perturbation simulations or adjoint methods—to examine daily and hourly variations in these source contributions and their connection to related, observable atmospheric constituents and meteorology. Model intercomparisons of BG O_3 and its components would allow us to understand how model formulation impacts results. Future intercomparisons can build on those organized under the Task Force on Hemispheric Transport of Air Pollution (HTAP).[13,14]

While model intercomparisons are an important activity to inform our understanding, agreement by two models does not necessarily indicate a high degree of understanding, unless informed by clear observational constraints. Greater application of sensitivity studies would identify parameters with a high degree of influence on the results and uncertainty that could be identified for focused studies (e.g., dry deposition). With respect to long-term, routine monitoring networks, these should be augmented with continuous observations of key tracers at strategically selected sites affected by processes contributing to BG O_3 to give source identification and attribution for exceptional event identification and to help develop long-term strategies for NAAQS attainment.



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The activities above offer a foundation from which to develop an ensemble reanalysis modeling framework to provide probabilistic ranges for not only air pollutants and related species, but also their source contributions. This effort should parallel approaches taken for long-term meteorological reanalysis,[15] where models and observations are used together through data assimilation techniques and locally applied bias corrections.

In all aspects of research and monitoring, it is critical that the modeling and observational communities work together to optimize research investments. While ongoing research will likely make continued, albeit slow, progress in understanding BG O_3, a more targeted investment in observations and modeling would go much further and generate more rapid progress in reducing and quantifying the uncertainties. The investments needed for this research and monitoring network upgrades would be a tiny fraction of the billions of dollars spent on air quality compliance and health impacts from air pollution. We also note that many of these source attribution aspects are also relevant to the implementation of the Regional Haze Rule, such that if an investment were to be made, it could be most effective to include particulate matter and O_3 together. **em**

Daniel A. Jaffe is Professor and Chair of the Physical Sciences Division, School of STEM at the University of Washington, Bothell, and a Professor in the Department of Atmospheric Sciences at the University of Washington, Seattle. E-mail: **djaffe@uw.edu**. **Arlene M. Fiore** is with the Department of Earth and Environmental Sciences and Lamont–Doherty Earth Observatory of Columbia University. **Terry J. Keating** is with the U.S. Environmental Protection Agency's Office of Research and Development in Washington, DC.

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