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OBSERVATION-BASED QUANTIFICATION OF BACKGROUND CONTRIBUTIONS TO MAXIMUM US O₃ CONCENTRATIONS

Executive Summary

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**Observation-based Quantification of Background Contributions to
Maximum US O₃ Concentrations**

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Over the past two centuries, humans have conducted a grand tropospheric chemistry experiment. Industrial and societal development, intended to improve human well-being, was accompanied by a massive increase in emissions of air pollutants, including precursors for photochemical production of ozone in the troposphere. These precursors are oxides of nitrogen ($\text{NO}_x = \text{NO} + \text{NO}_2$), volatile organic compounds (VOCs), methane and carbon monoxide (CO). These emissions were concentrated at northern midlatitudes, where they drove large increases in ambient ozone concentrations, most notably in urban areas such as Los Angeles, but also throughout the zonal troposphere. On rare occasions in Los Angeles in the 1960s, ozone concentrations exceeded 600 parts per billion (ppb, representing nanomoles per mole ambient air), and 8-hour averages exceeded 300 ppb. By the year 2000 background ozone had increased by approximately a factor of 2 above natural levels throughout northern midlatitudes. Since the 1960s emission control efforts have been implemented in North America and Europe, and more recently in China, in order to reduce urban ozone concentrations. These efforts have been extraordinarily successful, leading to continual decreases in ozone in US urban areas, and by the mid-2000s, northern midlatitude background ozone had reached a maximum and since has begun to slowly decrease.

Routine ozone monitoring began in the US in the early 1970s and continues to the present. The accumulated data set provides the basis for an observational-based quantification of US ozone air quality changes over 4 to 5 decades. This time period includes the later part of the urban ozone increase, much of the zonal background ozone increase, and the following ozone decreases; thus these changes in the temporal and spatial distribution of US ozone concentrations have been documented by observations of high reliability and relatively good spatial resolution. This archive of measurements constitutes an extremely detailed record of the response of tropospheric ozone concentrations within the US to this grand, human-caused tropospheric chemistry experiment. The overall goal of our research is to examine this record to improve our understanding of the ozone sources responsible for the largest ozone concentrations observed at urban and rural locations in the western US.

Approach

Although our analysis could be applied to any percentile of the measured surface ozone distribution, this project focuses on Ozone Design Values (ODVs) based on the 2015 ozone National Ambient Air Quality Standard (NAAQS). An ODV is the 3-year average of the annual fourth-highest daily maximum 8-hour average (MDA8) O_3 concentration. Since the four highest MDA8 O_3 concentrations are generally recorded in the warmer half of the year (i.e., the ozone season) the ODV corresponds to $\sim 98^{\text{th}}$ percentile of MDA8 values recorded at a site during the ozone season. This focus is chosen to maximize the policy relevance of the results, since it is the maximum ozone concentrations that are associated with the strongest health, crop and ecosystem impacts, and is hence the statistic upon which the NAAQS is based. Our analysis proceeds by fitting ODV time series to simple mathematical functions developed from a conceptual model that represent the very different long-term changes that have occurred in the important contributions to the tabulated ODVs.

This project comprises two tasks:

- 1. Analysis of ODVs recorded in inland California (CA).** The primary goal of this task is to quantify the contributions from background ozone, urban ozone transport, agriculture and wildfires to maximum ozone concentrations in inland CA air basins; these basins include one (San Joaquin Valley) of the two extreme US ozone nonattainment areas (the Los Angeles-South Coast Air Basin (AB) is the other). This

analysis utilizes contrasts in ODV time series in two pairs of CA air basins: the Mojave Desert AB (with minimal agricultural activity) versus the Salton Sea AB (which contains the intense irrigated agriculture activity of the Imperial Valley) and the Sacramento Valley AB (with less agricultural activity) versus the San Joaquin Valley AB (with more intense agricultural activity). Urban ozone transport strongly affects the Mojave Desert and Salton Sea ABs, but not the other two ABs. Wildfire emissions affect the entire region. Time series of ODVs recorded in further downwind areas of CA (Mountain Counties AB and Great Basin Valleys AB) provide additional information.

The results of this task are presented in a paper in the *Journal of the Air & Waste Management Association*, which is included as Appendix A of this Report. In this paper we quantitatively estimate temporal and spatial distributions of ODV contributions from different ozone sources; in order of importance they are 1) US background ozone, by far the largest contribution, 2) production from US anthropogenic precursor emissions, primarily transported from California coastal urban areas but also produced locally, and 3) significant contributions from wildfire and agricultural emissions interacting with each other and with urban and industrial anthropogenic emissions. Because coastal mountain ranges largely block direct transport of low altitude, low background ozone marine air from the Pacific Ocean into the inland areas, except at large passes like the San Francisco Bay, the transport of higher altitude, higher background ozone air is much more significant. This results in US background ODVs greater than 60 ppb throughout inland California, approaching 70 ppb in many areas. US anthropogenic emissions from urban and industrial sectors presently contribute enhancements of ~4 to 15 ppb to the maximum ODVs throughout inland California, except for larger contributions of 30 – 35 ppb transported to the sites immediately adjacent to the South Coast Air Basin (SoCAB). This transported contribution decreases rapidly with downwind distance from SoCAB with an e-folding distance of 70 (\pm 40) km. Wildfire contributions to ODVs are significant only in regions where those emissions interact with pre-existing emissions of oxides of nitrogen (NO_x), either from urban or agricultural sources. Wildfire contributions have increased markedly in the past decade, are highly sporadic, but are estimated to average ~10 to 15 ppb in the year 2020 in the San Joaquin Valley and in the Sacramento urban plume. This contribution is expected to increase in the future so long as sufficient NO_x emissions continue to be available.

2. Analysis of ODVs recorded in the southwestern US, including Texas. As air masses move inland from the Pacific, continental sources and sinks modify the marine ozone concentration distribution. The goal of this task is to develop an observational-based quantification of the relative contributions of local and regional photochemical ozone production, regional transport (primarily from CA), and US background ozone transport (both from the Pacific and, in Texas, from the Gulf of Mexico) throughout the southwestern US.

The results of this task are presented in a second paper in the *Journal of the Air & Waste Management Association*, which is included as Appendix B of this Report. In this paper we quantitatively estimate the various (background, regional anthropogenic, etc.) contributions to the temporal and spatial distributions of Ozone Design Value (ODV) throughout the southwestern US. We find that in 2000, background ODVs (primarily due to transported baseline ozone) contributed 64 to 70 ppb to the region's ODVs, and wildfire emissions (also considered a background contribution) enhanced ODVs by an additional 2 to 4 ppb. Together, these background contributions approach the national air ambient quality standards (NAAQS) for ozone of 75 ppb (implemented in 2008) and 70 ppb (implemented in 2015). US anthropogenic emissions from urban and industrial sectors produced additional, relatively modest ODV enhancements (less than ~16 ppb in 2000). Since 2000, regional US anthropogenic ODV enhancements have decreased

by ~60%. However, although baseline contributions have decreased by ~3 ppb, wildfire emissions have increased, so that ODVs have not greatly decreased. In contrast, northeastern (e.g., New York City) and southern (e.g., Atlanta GA) US urban areas have lower US background ODV contributions (~52 and 49 ppb, respectively) so that despite their higher US anthropogenic ODV enhancements in 2000 (~40 and 54 ppb, respectively), ODVs in these regions no longer exceed the 70 ppb standard. As a result, there has been a pronounced regional shift of the occurrence of the highest US ozone concentrations. Excluding California (which accounts for the large majority of high ozone episodes) over the most recent 5-year period (2017-2021) the southwestern US (including Texas) recorded 64% of the nation's ODVs above 75 ppb, while 20 years earlier that fraction was only 11%. In the southwestern urban areas the remaining US anthropogenic ODV enhancements are in the range of 2 to 6 ppb; ODVs in Denver, Phoenix and Salt Lake City have exceeded 75 ppb in each of the past 5 years. Our analysis shows that there is little potential for reducing these ODVs to the 70 ppb NAAQS by current emission control efforts that only address reductions in the US anthropogenic ODV enhancement. Possible alternate control strategies consistent with this analysis are discussed.

Summary

This project has examined the existing record of ozone concentrations measured in the southwestern US, including the interior portions of southern California and the states of Nevada, Utah, Arizona, Colorado, New Mexico and Texas. By far the largest contribution to ODVs throughout this region is US background ozone, with significant enhancements due to photochemical production from US anthropogenic precursor emissions; the accompanying figure shows contour maps giving the estimated spatial distributions (of these two primary ODV contributions across the entire western US. (The maps include results from earlier analyses).

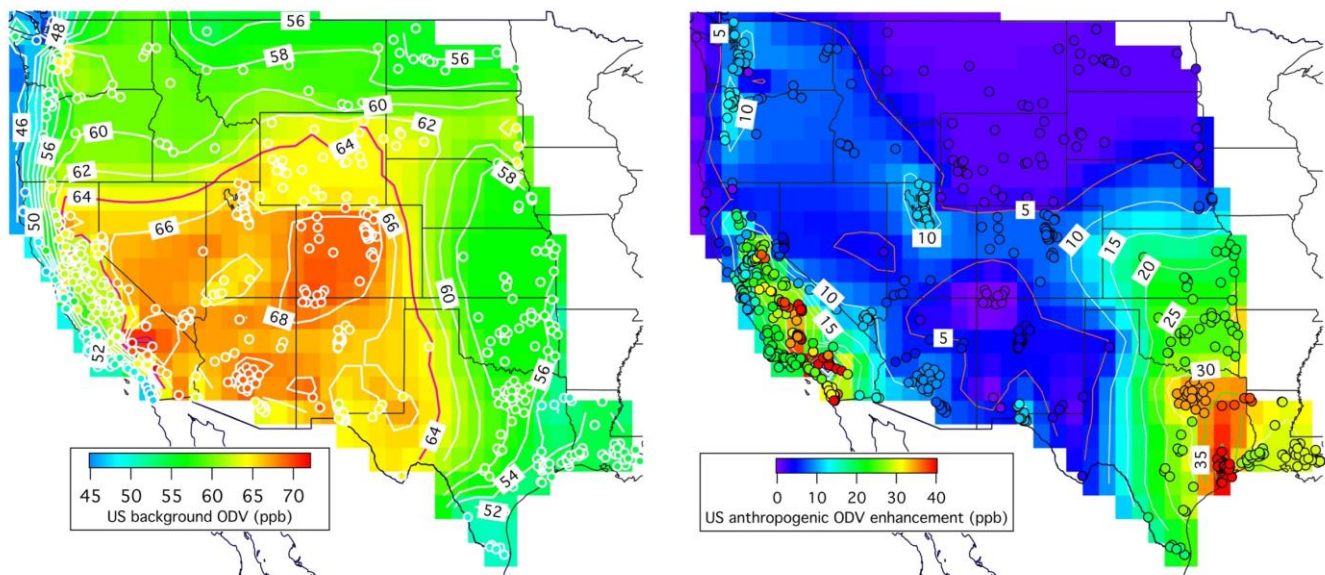


Figure. Contour maps of estimated US background ODV (left) and US anthropogenic ODV enhancement (right) for year 2000 over the western US. The 64 ppb contour is colored violet in the left map to indicate the extensive area of largest values. Symbols in the contour maps indicate individual monitoring sites included in the analysis with the same color coding as the contour map.

Implications of this work

The results of this analysis have important implications for US air quality control policies. In 2020 the ODVs that would result from transported baseline ozone alone are estimated to be in the range of 58 to 66 ppb throughout inland California. Wildfire and/or agricultural emissions enhance ODVs above this background by as much as ~7 to 15 ppb in some areas of inland California. Throughout the southwestern US, transported baseline ozone contributions and those from wildfire emissions together totaled ~65 to 73 ppb in 2000. These ODV contributions are so large that there remains little or no margin for additional ODV enhancements due to photochemical ozone production from urban and industrial emissions before exceeding the 70 ppb (implemented in 2015) and 75 ppb (implemented in 2008) ozone National Ambient Air Quality Standards.

Between 1980 to 2020 emission controls reduced the ODV enhancements from US anthropogenic urban and industrial sources by more than a factor of 6, so there remains relatively little potential for further reducing this ODV contribution by cost effective approaches, although it is the contribution primarily addressed by current emission control efforts. The US anthropogenic ODV enhancements are now quite small: 4 to 15 ppb throughout most of inland California (with larger contributions due to transport at sites adjacent to the South Coast Air Basin) and < 7 ppb throughout both urban and rural southwestern US areas. Overall, throughout inland California and the southwestern US, there is little practical likelihood of achieving the necessary ozone reductions to decrease the ODVs below the 70 ppb NAAQS through additional local and regional emission controls alone. Importantly, wildfire emissions have increased over the past decades, and are expected to continue increasing into the future, which further adds to the difficulty of attaining the ozone NAAQS. Further reductions in the standard would make attainment an even more distant possibility.

We briefly discuss alternative policy approaches that could be implemented to relieve this situation, including: 1) a regionally varying ambient ozone concentration standard that accounts for the varying background, 2) implementation of a standard based on limiting the anthropogenic increment of ambient ozone above the background concentrations (much as the approach chosen for limiting human exposure to ionizing radiation), and 3) a cooperative, international emission control agreement aimed at continuing or even accelerating the presently occurring decrease in baseline ozone throughout northern midlatitudes.