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Key Points:

- We found a strong relationship between long-term concentrations of nitrogen dioxide (NO_2) and peak O_3 in all U.S. non-attainment areas
- In 2020, O_3 in the eastern U.S. declined due to nitrogen oxide reductions, but increased in the western U.S. due to large wildfires
- Since 2010, inter-annual variations in wildland fires in the western U.S. are significantly correlated with policy relevant O_3 concentrations

Supporting Information:

Supporting Information may be found in the online version of this article.

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NO_x and O_3 Trends at U.S. Non-Attainment Areas for 1995–2020: Influence of COVID-19 Reductions and Wildland Fires on Policy-Relevant Concentrations

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Abstract We analyzed NO_2 and O_3 data from 32 U.S. non-attainment areas (NAAs) for 1995–2020. Since 1995, all regions have shown steady reductions in NO_2 and the weekend-weekday pattern indicates that the O_3 production regime in most NAAs has transitioned to a NO_x-limited regime, while a few NAAs remain NO_x-saturated. In the eastern U.S., all NAAs have made steady progress toward meeting the current (70 ppb) O_3 standard, but this is less true in midwestern and western NAAs, with most showing little improvement in peak O_3 concentrations since about 2010. Due to COVID-19 restrictions, NO_2 concentrations were substantially reduced in 2020. In the eastern NAAs, we see significant reductions in both NO_2 and peak O_3 concentrations. In the midwestern U.S., results were more variable, with both higher and lower O_3 values in 2020. In the western U.S. (WUS), we see variable reductions in NO_2 but substantial increases in O_3 at most sites, due to the influence from huge wildland fires. The recent pattern over the past decade shows that the large amount of wildland fires has a strong influence on the policy-relevant O_3 metric in the WUS, and this is making it more difficult for these regions to meet the O_3 standard.

Plain Language Summary O_3 exposure has serious health impacts up to and including premature mortality. In the U.S., more than 100 million people live in areas that do not meet the current National Ambient Air Quality Standards (NAAQS) for O_3. In this analysis, we used available air quality data from 32 major metropolitan areas that do meet the current O_3 standard. We find that in nearly all regions there is a significant long-term relationship between nitrogen oxides and the policy relevant peak O_3 concentrations. In addition, the pattern of weekend-weekday concentrations indicates that nearly all regions have transitioned to a NOx limited regime. Given the strong reductions in nitrogen oxide emissions in 2020 due to COVID-19 restrictions, we expected to see a significant reduction in peak O_3. This was seen at all sites in the eastern U.S. However, in the western U.S., we found a significant increase in peak O_3 associated with the large wildfires that burned in 2020. These results provide important data to show how nitrogen oxides control O_3 in most regions, but suggest an ongoing challenge for western regions to meet the O_3 standard, given the likelihood of continued climate driven wildfire increases.

1. Introduction

Surface ozone (O_3) is formed from photochemical reactions of nitrogen oxides ($NO_x = NO + NO_2$) and volatile organic compounds (VOCs). O_3 has serious health impacts up to and including premature mortality (Bell et al., 2004; Di et al., 2017). In the U.S., reductions in NO_x and VOCs over the past several decades have reduced peak O_3 concentrations considerably (Simon et al., 2015), but at present, there are still more than 100 million Americans living in areas that do not meet the O_3 standard (American Lung Association (ALA), 2020). For O_3, the U.S. National Ambient Air Quality Standards (NAAQS) is based on the annual fourth highest maximum daily 8-hr average (MDA8) concentration. To be in compliance with the 2015 8-hr O_3 standard, each monitor in a region must have a 3-year average of the fourth highest MDA8 of 70 ppb or less. Using data from 2018 to 2020, the U.S. Environmental Protection Agency (EPA) reports that there are 52 regions in the U.S. that are considered non-attainment areas (NAAs) for the 2015 standard (https://www.epa.gov/air-trends/air-quality-design-values). Most of these NAAs are in large metropolitan areas. However, some areas have relatively low emissions but are downwind of a major metropolitan area.

Simon et al. (2015) report downward trends for U.S. anthropogenic NO_x and VOCs emissions of 39% and 14%, respectively, for 2002–2011. Hidy and Blanchard (2015) report a near linear relationship between peak

O_3 concentrations and O_3 precursors at most sites using data through 2013. According to the U.S. EPA's 2017 National Emissions Inventory (https://www.epa.gov/air-emissions-inventories/2017-national-emissions-inventory-nei-data), industrial or vehicle sources account for 87% of the NO_x emissions in the U.S., but only 29% of the VOC emissions. This is due to large emissions of VOCs, such as isoprene, from biogenic sources. Using data from 2000 to 2018 from the Los Angeles region, Nussbaumer and Cohen (2020) show that NO_x concentrations are continuing to decline linearly, but the rate of decline in total VOC concentrations (anthropogenic plus biogenic) has slowed and appears to be approaching a lower limit of about 50 ppbC (ppbC = ppb × # carbon atoms). Chen et al. (2019) report that most regions of the U.S., even urban areas, have VOC reactivity that is now dominated by biogenic compounds. An additional source of VOCs that is not well captured by inventories and is probably not declining are emissions from volatile chemical products (McDonald et al., 2018).

The production of O_3 is known to be non-linear with the VOC and NO_x precursors at high NO_x/VOC ratios (Jin et al., 2020; Qian et al., 2019). Under these conditions, higher NO_x concentrations will lead to O_3 suppression. Given greater emissions of NO_x on weekdays, due to greater diesel sources (Pollack et al., 2012), many studies have reported lower O_3 concentrations on weekdays (Altshuler et al., 1995; Blanchard et al., 2008; Marr & Harley, 2002; Pollack et al., 2012). But as NO_x emissions have declined, several studies have reported weakening or reversal of this weekend-weekday pattern in some regions (Baidar et al., 2015; de Foy et al., 2020; Jin et al., 2020). The presence of this weekend-weekday effect is an important indicator of the NO_x sensitivity in each air shed, though this has never been examined in a systematic way for all U.S. NAAs. However, these patterns are not static, as the O_3 production can also vary by time of day, season, and location (Mazzuca et al., 2016). In particular, temperature and other meteorological parameters are key controls on the day-to-day variations in O_3 concentrations (e.g., Camalier et al., 2007; Gong et al., 2017; Ninneman & Jaffe, 2021a; Nussbaumer & Cohen, 2020; Wells et al., 2021).

O_3 is unique amongst the pollutants regulated by the Clean Air Act in that background concentrations, ranging from 30 to 60 ppb, are a large fraction of the health-based standard, 70 ppb (Jaffe et al., 2018; Pollack et al., 2012). The problem stemming from background O_3 is particularly acute in the western U.S. (WUS) due to high contributions from the stratosphere, Asian industrial emissions, and regional wildfire emissions (Altshuler et al., 1995; Jaffe et al., 2018, 2020; Pollack et al., 2012). At present, there is no evidence that stratospheric contributions to tropospheric O_3 has changed, and NO_x emissions in Asia have declined during the last decade, although O_3 concentrations have increased in some parts of Asia (Li et al., 2019). In contrast, there is substantial evidence that emissions from wildland fires have increased in the past decade (Baidar et al., 2015; Blanchard et al., 2008; de Foy et al., 2020; Higuera et al., 2021; Marr & Harley, 2002; McClure & Jaffe, 2018a).

Starting in the winter of 2020, the COVID-19 pandemic substantially reduced vehicle traffic and emissions (Gkatzelis et al., 2021; Sokhi et al., 2021). Many studies have identified reduced concentrations of primary air pollutants, especially those associated with vehicle emissions. Most studies have focused on NO_2, using satellite observations (e.g., He et al., 2020; Naeger & Murphy, 2020; Poetzscher & Isaifan, 2021), but a few also looked at particulate matter with diameters of less than 2.5 μm ($PM_{2.5}$; He et al., 2020) and/or used surface monitors (Amouei Torkmahalleh et al., 2021; Xiang et al., 2020). It is important to note that observations of the tropospheric NO_2 columns by satellite do not directly give surface concentrations due to an increasing contribution from background sources in the free tropospheric column, as surface sources have declined (Qu et al., 2021; Silvern et al., 2019). Additionally, meteorological variability can complicate satellite signals (Goldberg et al., 2020). For this reason, we use surface observations of NO_2 to give the best indication of local photochemistry. Kroll et al. (2020) make the point that assessments of secondary pollutants that ignore atmospheric chemistry may yield misleading or surprising results. Indeed, several studies have looked at O_3 changes early in 2020 and found little change (Bekbulat et al., 2021) or even an increase (Zhao et al., 2020), likely due to reduced NO_x suppression in winter or early spring conditions. Some studies have identified small reductions in free tropospheric O_3 concentrations over the Northern Hemisphere (Chang et al., 2022; Steinbrecht et al., 2021), which reverses the earlier trends (Gaudel et al., 2020). Campbell et al. (2021) used the NOAA forecasting system and observations to examine 2020 changes and found a complex pattern of increases and decreases, depending on location and time of year. Jing and Goldberg (2022) examined mean 2020 NO_2 and O_3 changes for the Chicago region and found no significant change in mean O_3 concentrations, which they attributed to the presence of hotter than normal conditions. However, despite great interest in the 2020 air quality changes, at present, no studies have examined the change in policy relevant fourth highest MDA8 O_3 at all non-attainment regions of the U.S.

The year 2020 was also a year of huge wildland fires in the WUS. The National Interagency Fire Center (NIFC) reports that the area burned in the U.S. was 4.2 million hectares, 3.8 million of which were in the conterminous WUS. This is the highest total for the WUS in at least the past 2 decades. (The NIFC reports that data prior to 2001 should not be compared to current data; https://www.nifc.gov/fire-information/statistics.) The 2020 area burned in the WUS is part of a long-term trend that is driven by forest management practices (Calkin et al., 2015), increasing human ignitions (Balch et al., 2017), and climate change (Parks & Abatzoglou, 2020). Wildfires emit significant amounts of O_3 precursors, and a number of studies have pointed out that the probability of an O_3 exceedance day in urban areas is much higher in the presence of smoke (Buysse et al., 2019; Jaffe et al., 2020; Kaulfus et al., 2017; McClure & Jaffe, 2018b).

The 2020 combination of reduced NO_x emissions due to the COVID-19 pandemic lockdowns and large wildfires means that we can use these data to learn a great deal about what controls O_3 in U.S. NAAs. In this study, we examine the long-term pattern of NO_2 and O_3 in 32 U.S. metropolitan regions that exceed the O_3 standard with a primary objective of understanding both the long-term and 2020 changes. Specifically, our goals are:

1. Evaluate the long-term and 2020 changes in NO_2 and O_3 during the primary O_3 season (May–September) in each NAA of the WUS
2. Examine the weekend-weekday pattern to ascertain the degree to which NO_x controls O_3 in each NAA
3. Evaluate the role of temperature on the 2019–2020 O_3 changes in each of the major NAA regions
4. Examine the role of COVID-19 NO_x reductions and wildland fires on the policy-relevant O_3 metric in the each of the major NAA regions

2. Methods

In acquiring data for this analysis, every effort was made to ensure that we used a consistent record for each metropolitan region. For surface air quality, the data were obtained from the EPA's Air Quality System (AQS) database via the Air Data interface (https://www.epa.gov/outdoor-air-quality-data).

The EPA has identified 52 O_3 NAAs, and many have NO_2 data. However, some do not have sufficient NO_2 data for a useful analysis. We focused on 32 regions that have consistent long-term (20+ year) records of both O_3 and NO_2 (see Table S1 in Supporting Information S1). This includes six regions in the eastern U.S., six in the midwestern U.S., and 20 in the WUS, of which 12 are in California. For each metropolitan region, we identified the individual O_3 monitor with the greatest annual fourth highest MDA8 in the recent 3-year period and that had at least a 10-year data record. For NO_2, we identified all monitors in each region with a long-term and near continuous record. In some cases, no single monitor had a long-term record of NO_2 and in these cases, we averaged multiple monitors to obtain a long-term record for that region. Table S1 in Supporting Information S1 shows the air monitors used in this analysis for each NAA. We use the MDA8 concentrations for O_3, and we use the 1-hr daily maximum concentrations for NO_2. We note that in some regions, the location of highest O_3 may have shifted over this decadal time period (Jin et al., 2020), and this could complicate interpretation of some of the long-term patterns in individual NAAs.

The 1-hr daily maximum NO_2 concentrations are most likely to occur at night or during the early morning hours, when the nocturnal inversion traps local emissions and elevates concentrations of primary emissions. At this time, nearly all emitted NO_x ($NO + NO_2$) will be in the form of NO_2, due to the absence of sunlight that would photolyze it. It is also important to note that the most common NO_2 analyzers used in the EPA network have a significant interference from other nitrogen oxide species (Dickerson et al., 2019). As such, these NO_2 concentrations are better thought of as the sum of all nitrogen oxides (NO_y). It is possible that long-term changes have redistributed the relative contribution from different species, but the measured NO_2 concentrations will still reflect the total NO_x emissions. In keeping with common usage, we will still refer to these observations as NO_2, as this is how the EPA refers to them.

Surface meteorological data were obtained from the NOAA National Centers for Environmental Information (https://www.ncdc.noaa.gov/cdo-web/). For this analysis, as we are interested in the day-to-day variations, it is important to choose long-term meteorological sites that provide the most consistent record for a region. Table S1 in Supporting Information S1 shows the meteorological source used for each NAA. Overhead smoke data were from the NOAA Hazard Mapping System Fire and Smoke Product (HMS; Kaulfus et al., 2017; Rolph

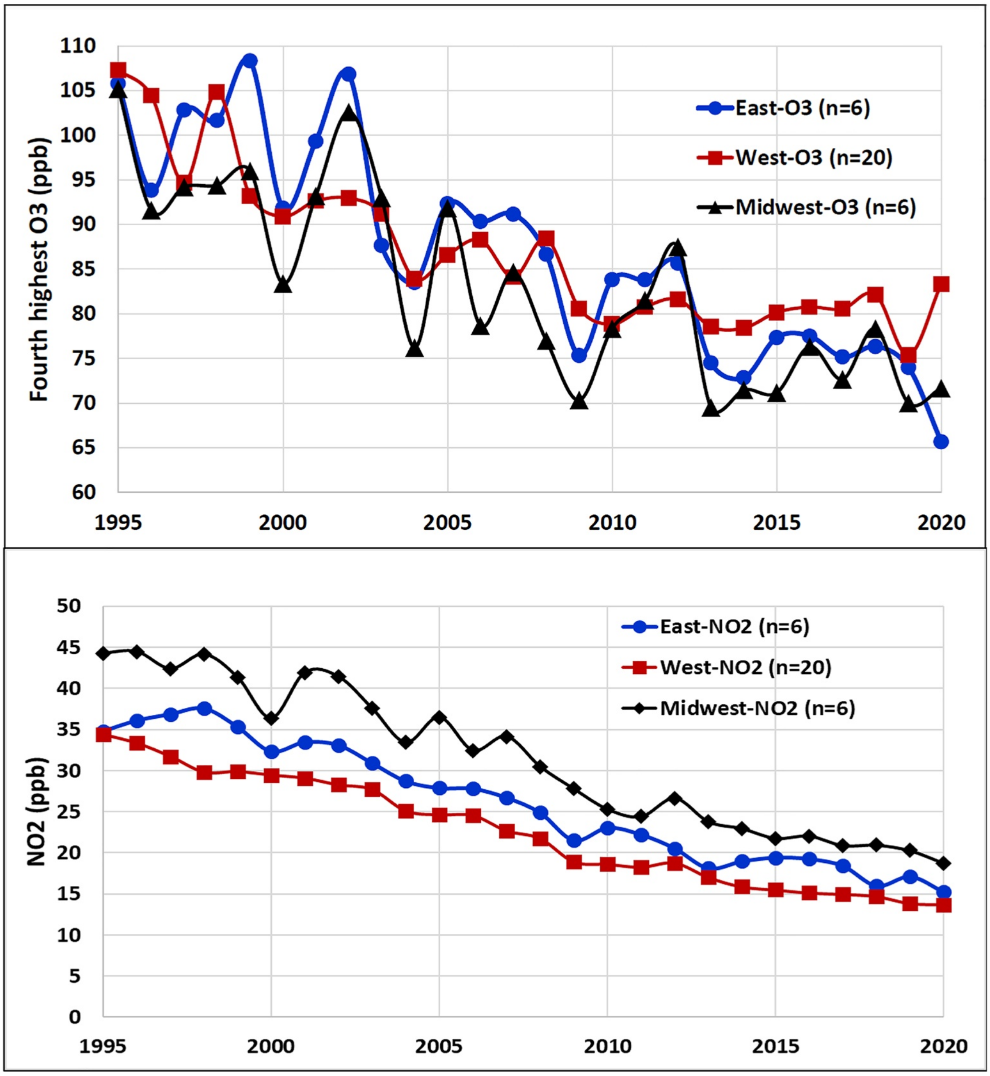

Figure 1. Annual fourth highest O_3 (a: top) and annual average daily 1-hr maximum NO_2 for May–September (b: bottom) in 32 metropolitan regions that do not meet the current O_3 standard. The value shown is the mean across the selected monitors in that region.

et al., 2009), and these data were obtained from the NOAA National Environmental Satellite, Data, and Information Service (NESDIS) HMS archive (https://www.ospo.noaa.gov/Products/land/hms.html). Imagery of the HMS data were also available from the EPA's AirNow-Tech database. The HMS product is derived from multiple satellite imagery and updated daily. Details on the HMS product are given in Kaulfus et al. (2017) and Rolph et al. (2009). For this work, we use the HMS data to identify overhead smoke for each O_3 monitoring station on a daily basis.

3. Results

Figure 1 shows the annual fourth highest MDA8 O_3 and May–September annual mean of the one-hour daily maximum NO_2 concentrations for eastern, midwestern, and western NAA regions going back to 1995. Table S1 in Supporting Information S1 gives the locations and monitor information. Table S2 in Supporting Information S1 shows the annual fourth highest MDA8 O_3 and annual averaged NO_2 values for 1995–2020 in each of the 32 NAA regions. For NO_2, all regions have shown steady declines in concentrations through 2020. All regions have shown declines in fourth highest MDA8 O_3 through 2013. From 2013 to 2019, O_3 concentrations have been variable, but not systematically declining in the midwestern or WUS. For the WUS, concentrations in the past

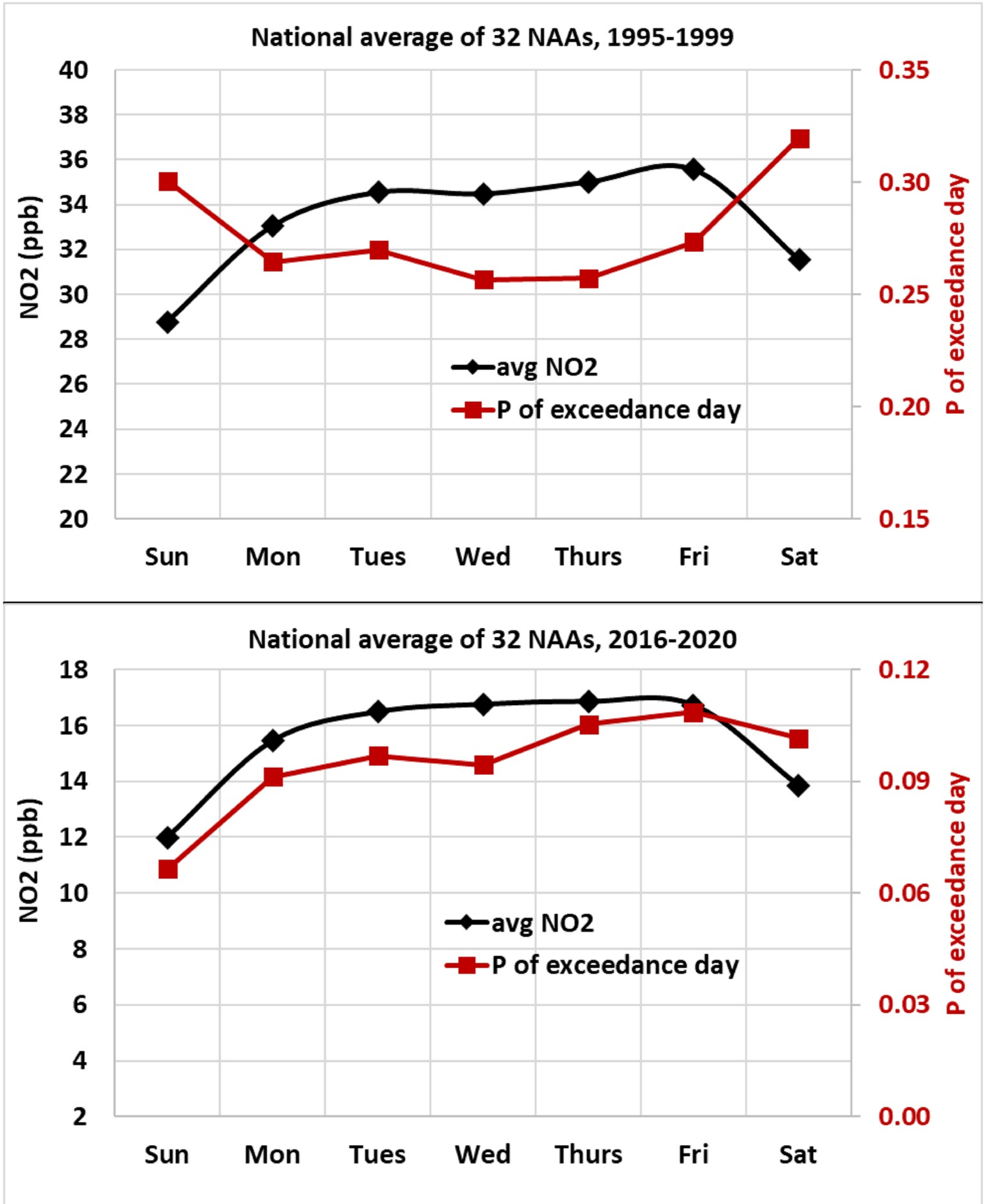

Figure 2. Average NO_2 concentration and probability (P) of an exceedance day (MDA8 > 70 ppb) for May–September for 32 U.S. NAAs. Top figure (a) shows 1995–1999 data, and bottom figure (b) shows 2016–2020 data.

decade have been highly variable, but not declining, although the 2019 values were the lowest values on record. In the eastern U.S., the lowest value on record was seen in 2020. The midwestern sites showed a more variable pattern, with both 2019 and 2020 being on the low end, compared to the previous decade. In a later part of this paper, we will focus specifically on the 2019 to 2020 differences.

3.1. Weekday-Weekend Pattern and NO_x Sensitivity

Given the strong association of O_3 and NO_2, we want to examine the weekend-weekday pattern as an indicator of the NO_x versus VOC sensitivity. Figure 2 shows the mean NO_2 concentration and probability of an exceedance day (MDA8 > 70 ppb) for May–September averaged across all 32 U.S. NAAs. The plot is divided by the early part of the data record (1995–1999) and more recent data record (2016–2020). Data for each individual NAA is given in Table S3 in Supporting Information S1. For all regions, NO_2 concentrations have declined considerably

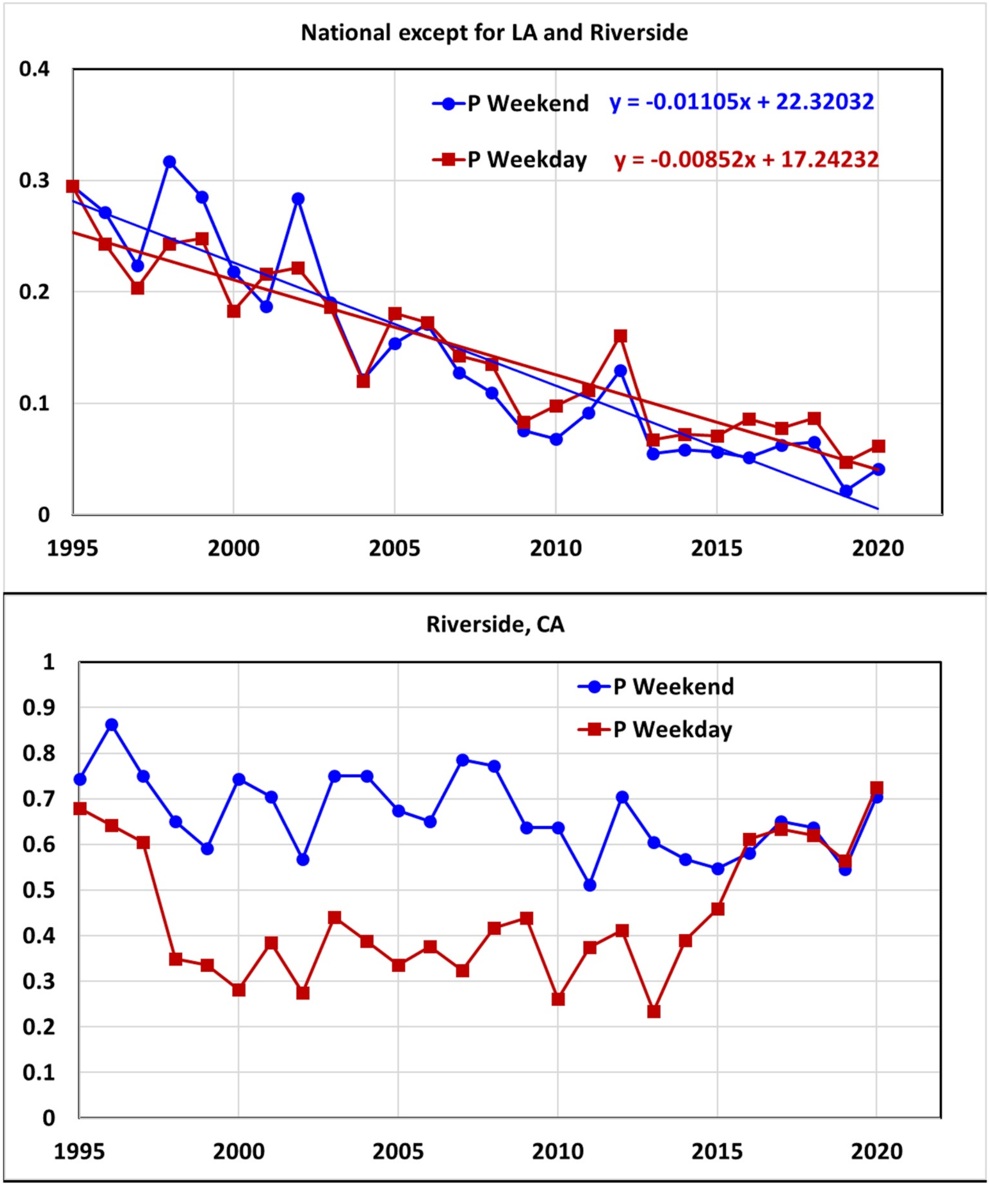

Figure 3. Probability of an O_3 exceedance day (MDA8 > 70 ppb) for weekends and weekdays for 1995–2020 during the primary O_3 season (May–September). The top figure (a) shows the values for 30 U.S. NAAs, and the bottom plot (b) shows the values for Riverside, CA.

and are lowest on weekends. Sunday has the lowest mean NO_2 in 32 out of 32 NAAs and the lowest probability of an exceedance day in 22 out of 32 NAAs. Of the remaining 10 locations, seven have P-values on Sundays that are very close to the next lowest probability day (with P-values within 0.01 of the lowest days). Only Cleveland, Bakersfield, and Los Angeles have P-values for Sunday that are not the lowest by more than 0.01, suggesting that these sites have not yet transitioned to a NO_x-limited regime, consistent with the work of Fujita et al. (2016) and Jin et al. (2020). So, we conclude that the significant emission reductions have led to most NAAs transitioning to a NO_x-limited regime. Figure 3 shows this transition for 30 of the 32 regions by examining a time series of the probability of an exceedance day in May–September. The analysis of the national data show that (a) the probability of an exceedance day has declined since 1995 and (b) weekdays started to have a higher probability around 2007, averaged nationally, although there is considerable variability across each individual NAA. For Los Angeles and Riverside, CA, the results are somewhat different. Figure 3b shows that starting in about 2016 in Riverside, the weekday and weekend probabilities are very close, suggesting that the recent NO_x reductions

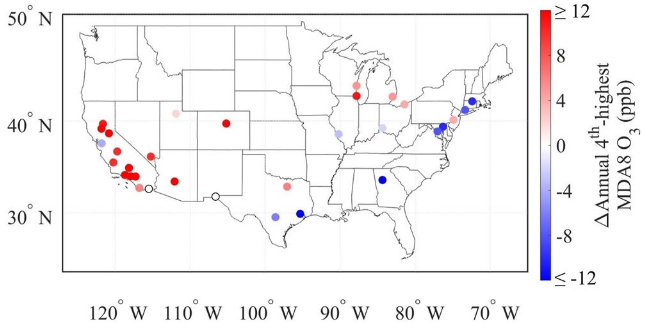

Figure 4. Difference (2020 minus 2019) in the annual fourth highest MDA8 O_3 for 32 NAAs.

have now brought this region into a near optimum O_3 production environment. This is further confirmed by the very high exceedance probabilities for Riverside, more than 50%. Figure S1 in Supporting Information S1 shows a similar plot for Los Angeles, which continues to have the highest probability of an exceedance day on weekend days.

While the day-of-week pattern confirms that most NAAs in the U.S. have transitioned to a NO_x-limited regime, this does not imply that VOCs are not also important in O_3 production. In fact, it is possible that some of this weekend-weekday effect in O_3 could be due to reductions of VOCs precursors on weekends. There are few reports on the VOC pattern and VOC reactivity on weekends versus weekdays. In one study, Blanchard et al. (2008) found a decline in VOCs of 19% on weekends compared to weekdays in several U.S. cities. In another study, which examined sites near Sacramento, Murphy et al. (2007) found that NO_x was reduced by 35% on weekends compared to weekdays, but VOC reactivity changed by less than 10%. Further characterization of the VOC pattern on weekends and weekdays would be useful; however, the current data across all NAAs is limited.

Despite the changes in NO_x-VOC sensitivity, we see a significant relationship in 30 out of 32 NAAs between the annual May–September mean daily maximum NO_2 and the fourth highest MDA8 O_3 for each NAA (Table S4 in Supporting Information S1). The R^2 values for the linear correlations are mostly above 0.5, and some can be as high as 0.87 (Ventura County, CA). The mean slopes are highest in the Midwest, followed by the East and lowest in the West. The correlation coefficients are highest in the Eastern U.S., followed by the Midwest and lowest in the West. For Denver and Las Vegas, this relationship is very weak and non-significant. The intercepts to these functions (O_3 value at 0 ppb NO_2) are highest in the West (mean value of 64 ppb) and lowest in the East (mean value of 48 ppb). The higher intercepts and lower correlation coefficients in the West likely result from the much greater influence of background O_3, including Asian pollution, stratospheric O_3 and wildfires, which is indicative of a more challenging problem for the western states in meeting the air quality standards for O_3 (Jaffe et al., 2018, 2020; Langford et al., 2017).

3.2. Difference in 2019–2020 Policy-Relevant O_3 Due To Temperature, COVID-19/Traffic Reductions (East), and Wildland Fires (West)

Figure 4 shows the change between 2020 and 2019 in the annual fourth highest MDA8 O_3 at the 32 U.S. NAAs. Table 1 shows a summary of the changes by region, and Table S5 in Supporting Information S1 shows the changes in each individual NAA. In the East, five of the six NAAs show a decline in the annual fourth highest MDA8 O_3, averaging 8 ppb, making 2020 the lowest O_3 year ever, averaged across the six sites. Only the Philadelphia site shows an increase of 3 ppb between 2019 and 2020. In the West, 15 of 20 sites showed an O_3 increase, averaging +12 ppb, making 2020 the highest O_3 year in the past decade, averaged across the 20 sites. Five sites in the West did not show an increase, including Houston, San Antonio and San Francisco, which declined between 2019 and 2020, and Dona Ana and Imperial counties, which showed no change. At the midwestern sites, changes in the annual fourth highest MDA8 O_3 were more variable. Three had elevated O_3 levels in 2020 compared to 2019, and three had lower O_3 levels in 2020 compared to 2019.

Table 1
Change in O_3 and Temperature (2020 −2019) for 32 NAAs by Region, Regression Slopes (MDA8 vs. Daily Max Temperature), and the Contribution of Temperature Change to the O_3 Change

	ΔO_3 (ppb)	ΔT 98th Perc. (°C)	Regression slope (ppb/°C)	ΔO_3 due to temp. (ppb)	Fraction due temp.
Eastern NAAs ($n = 6$)	−8.3	−0.10	1.4	−0.67	0.07
Midwestern NAAs ($n = 6$)	1.7	0.1	1.3	−0.01	0.20
Western NAAs ($n = 20$)	8.0	1.3	1.0	1.07	0.10

Note. Values in the table are averaged across all NAAs in that region. Individual values for each NAA are shown in Table S6 in Supporting Information S1.

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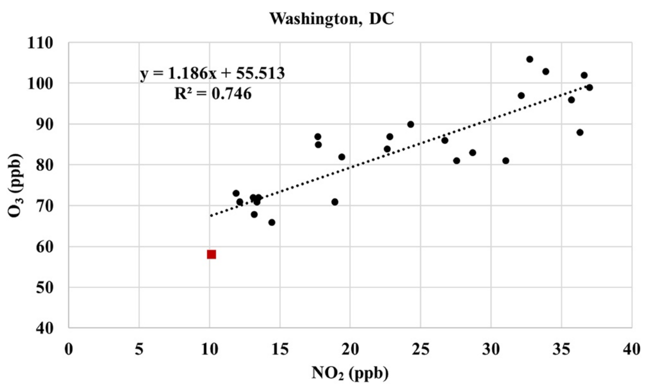

Figure 5. Linear fit of the annual fourth highest MDA8 O_3 versus May–September mean daily 1-hr maximum NO_2 concentration for Washington, DC, with the 2020 point highlighted (red square). Similar plots for Atlanta, Baltimore, Hartford, New York and Philadelphia are included in Figure S3 in Supporting Information S1. All annual data are shown in Table S2 in Supporting Information S1.

As temperature is one of the most important meteorological controls on O_3 production (Camalier et al., 2007; Jaffe, 2021; Wells et al., 2021) we first examine the role that temperature may have played on the 2019–2020 changes. For this, we examined the relationship between daily maximum temperature (DMT) and MDA8 O_3 for the primary O_3 season (May–September) in each of the 32 NAAs. We used robust linear regression with bisquare weights in Matrix Laboratory (MATLAB) version R2020a to define a temperature sensitivity for each NAA using data from 2018 to 2020. We then examined the degree to which temperature changes may have contributed to the O_3 changes by comparing the difference in 98th percentile temperatures in 2019 and 2020 in each region. Results by region (East, Midwest, West) are shown in Table 1 and results for each NAA are shown in Table S6 in Supporting Information S1. As noted previously (Ninneman & Jaffe, 2021a), the O_3-temperature sensitivity, or slopes, have declined in the last decade from between 2 and 3 ppb per ºC to between 1 and 1.5 ppb per ºC, as emissions and peak O_3 concentrations have declined. The results in Table S6 in Supporting Information S1 are slightly higher than those reported in Ninneman and Jaffe (2021a), due to inclusion of a larger number of locations in this analysis. Using the change in the 98th percentile of DMT, we can estimate a contribution for each NAA due to the 2019–2020 temperature changes from Equation (1) below:

$$\Delta O_3 \text{ due to temperature} = \text{Slope (ppb per°C)} * \Delta 98\text{th perc. DMT (°C)} \tag{1}$$

This would then account for any increase or decrease in temperature on the 2% warmest days, which are generally the highest O_3 days. We note that while temperature is usually the most important predictor for variations in MDA8 O_3, other meteorological factors can also play a role (Jaffe, 2021; Wells et al., 2021). The results, shown in Table 1, indicate that temperature changes were a small contributor to changes in the fourth highest MDA8 in these NAAs, but there is considerable variability from city to city. Table S6 in Supporting Information S1 shows the results for each individual NAA. Repeating this analysis using the change in the 99th percentile temperatures yields almost no change in the results. Given this result, we next explore other causes for the changes in O_3.

The 2020 reductions in O_3 and NO_2 for Washington, DC, are typical of the eastern sites, so we use it as an indicator of the pattern seen for the region. Figure S2 in Supporting Information S1 shows the 2019 and 2020 daily 1-hr maximum NO_2 concentration as a 10-day running mean. The smoothing allows for a better visualization of the changes, given the significant day-to-day variations driven by local meteorology. Concentrations of NO_2 in 2020 were down the greatest in March (−28%) and July (−23%). For the whole O_3 season (May–September), concentrations of NO_2 in 2020 were down by an average of 16%. Table S7 in Supporting Information S1 shows the ratio of surface NO_2 for 2019 compared with 2020 in each of the eastern NAAs by month. Averaged across all six eastern sites, NO_2 concentrations were down by 12% for May–September.

Figure 5 shows the annual fourth highest MDA8 O_3 versus annual NO_2 for Washington, DC, for 1995–2020, and Figure S3 in Supporting Information S1 shows similar plots for the five other eastern CBSAs. In four of the six regions, the lowest concentrations of fourth highest MDA8 O_3 and annual averaged NO_2 simultaneously occurred in 2020. Table S4 in Supporting Information S1 shows results of this linear fit for each of the 32 NAAs. For Washington, D.C., a 16% reduction (2.0 ppb) in mean NO_2 concentrations led to a 13 ppb reduction in the annual fourth highest MDA8 O_3. This decline in O_3 was larger than would be suggested from the long-term linear relationship. The 2020 annual fourth highest MDA8 O_3 was 58 ppb, whereas the linear fit would suggest a value of 67 ppb at the 2020 level of NO_2. While meteorological variability can impact the day-to-day and seasonal O_3 values (e.g., Camalier et al., 2007; Wells et al., 2021), temperatures in the Washington, DC, area were very close to normal in the summer of 2020. The 98th percentile daily maximum temperature at Washington, DC/Reagan National Airport for 2020 was 35.4°C, compared to a 25-year mean of 35.1°C and as indicated above, this had little impact on peak O_3 concentrations. An alternate hypothesis for the strong O_3 reduction in 2020 is the non-linear nature of O_3 production, especially in a NO_x-limited regime. Several studies have found that as NO_x concentrations decline, the O_3 production efficiency (OPE) increases, where OPE is defined as the number of O_3 molecules produced per NO_x molecule reacted to other products (Henneman et al., 2017; Kleinman et al., 2002).

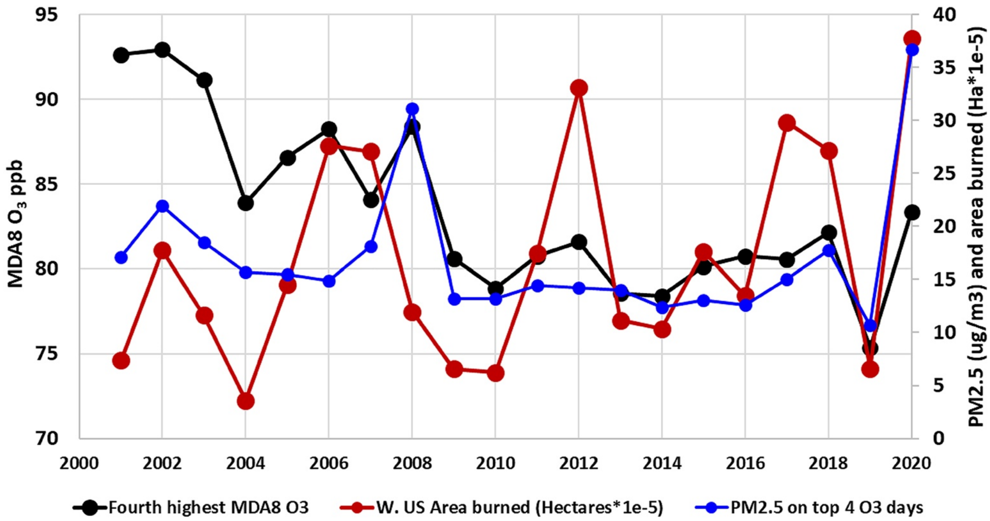

Figure 6. Annual fourth highest MDA8 O_3 (ppb, left axis), area burned in the western U.S. (hectares × $1e^{-5}$, right axis), and $PM_{2.5}$ ($\mu g/m^3$, right axis) on top four O_3 days for 20 sites in the western U.S.

This non-linearity reduces the annual fourth highest MDA8 O_3 in 2020 by an average of 5.2 ppb at these sites and allows four of the five NAAs to reach levels that are consistent with meeting the U.S. NAAQS (70 ppb or below) for the first time.

Similar NO_2 reductions were seen in the other eastern U.S. cities, ranging from −6% in Hartford to −18% in Atlanta (Table S5 in Supporting Information S1). Reductions in the annual fourth highest MDA8 O_3 were between 8 and 13 ppb, except for Philadelphia, which had a slight increase in 2020. The O_3 reduction for Hartford was quite strong (−10 ppb), despite a modest reduction in NO_2 (−6%). This probably reflects reductions in the New York metropolitan area, which is typically upwind of Hartford. The average NO_2 reduction across the six eastern U.S. cities was 12.1% or 2.1 ppb, with a corresponding reduction in the annual fourth highest MDA8 O_3 of 8.3 ppb.

While anthropogenic emissions are clearly the primary cause for elevated surface O_3 in most regions, an increasingly important factor is wildland fires. In 2020, wildland fires burned 3.7 million hectares (ha) in the WUS, the largest area burned in at least two decades. The largest fires were in California (1.6 million hectares), but nearly all regions of the WUS experienced extensive fires in 2020. Most of the fires burned during August and September, and this period also saw extreme levels of air quality degradation, with daily $PM_{2.5}$ values exceeding 600 $\mu g/m^3$ in some locations, similar to the extreme values seen in the 2018 wildfire season (Laing & Jaffe, 2019). However, the extreme $PM_{2.5}$ values are not generally associated with the highest O_3 days for multiple reasons, including reductions in photolysis and/or heterogeneous chemistry (Buysse et al., 2019). Instead, the highest O_3 values are typically seen on moderately smoky days, with $PM_{2.5}$ concentrations between about 15 and 100 $\mu g/m^3$ (Buysse et al., 2019). On average, smoke adds between 3 and 15 ppb to the MDA8 O_3 in an urban area, although this can be higher on individual days (Gong et al., 2017; McClure & Jaffe, 2018b).

Figure S4 in Supporting Information S1 shows a plot of MDA8 O_3, fire locations and the NOAA Hazard Mapping System (HMS) smoke product for 21 August 2020, a particularly bad day for smoke in the WUS. Huge fires were burning in many parts of the West, especially in California. Some of the largest fires burning in California at that time include the August Complex, the CZU Lightning Complex, and the SCU Lightning Complex, which together burned 6.14 × 10^5 ha. On this date, many sites in the WUS had elevated MDA8 O_3 values, including some of the highest values of the year at sites in California, Nevada, Utah, and Colorado, all of which were influenced by smoke from the California fires.

Figure 6 shows the annual fourth highest MDA8 O_3, the area burned in the WUS, and the average $PM_{2.5}$ on the top four O_3 days for 20 NAA regions in the WUS for 2001–2020. The decline in the policy relevant fourth highest, averaged across these NAAs, is apparent until about 2010. While 2008 was not a large fire year for the entire WUS, it was a large fire year in California ($6e^5$ ha burned, third highest after 2020 and 2018) and this likely had

10.1029/2021JD036385

a significant impact on O_3 concentrations throughout the state. The average increase in the annual fourth highest MDA8 O_3 across all sites for 2020 compared to 2019 is 8 ppb, with a maximum increase of 21 ppb (Bakersfield, CA). The mean $PM_{2.5}$ on the top four highest O_3 days averaged across all sites was 35 μg/m^3, a value that is higher than all previous years, with 2008 being second in this category due to fires that year (Washoe County, 2022; Wilkins et al., 2018). In comparison, 2019 – a year with low area burned – was associated with significantly reduced MDA8 O_3 values at these western sites. The year 2018 was also a year with major wildfires in the WUS and significantly enhanced O_3. The patterns for the 2018–2020 time period give a clear indication of the degree of influence from fires on the policy-relevant O_3 metric in the 20 western NAAs. Since 2010, there is a strong and significant correlation ($P < 0.05$) between WUS area burned and fourth highest MDA8 O_3 for the 20 western NAAs ($R = 0.83$). Prior to 2010, this relationship was not significant, likely due to a stronger role for O_3 production from local anthropogenic precursors and lesser amounts of area burned. However, it is important to note there are regional variations in each of the 20 NAAs due to timing, fire locations, transport and meteorology. Nonetheless, Figure 6 demonstrates the important role that wildland fires now play on policy relevant O_3 in the WUS.

Given the large amount of burning in California in 2020, we can examine one site that was strongly influenced by these fires. Figure S5 in Supporting Information S1 shows a scatter plot of MDA8 O_3 versus DMT for May–September 2018–2020, and Figure S6 in Supporting Information S1 shows a time series of the 2020 O_3 and $\mathbf{PM_{2.5}}$ data, both for Sacramento. The heavy smoke period started in mid-August and extended through September. The highest MDA8 O_3 days of the year were 20, 21, 22, and 23 August, with 20 August (100 ppb) being the highest value in the last decade. Figure S6 in Supporting Information S1 clearly shows that these dates were outliers from the usual relationship, due to additional O_3 formed from smoke precursors. A similar approach has been used in previous studies (e.g., Gong et al., 2017; Jaffe, 2021; Lindaas et al., 2017; Pollack et al., 2021).

In the midwestern NAAs, the pattern is not as clear. In 2020, five of the six cities showed a decrease in NO_2 (ranging from −3 to −20%), but only three showed decreased O_3 (Cincinnati, Detroit, and St. Louis). The three midwestern cities with enhanced 2020 O_3 (Chicago, Milwaukee, and Cleveland) had average June–July daily maximum temperatures that were 1.9°C higher than the 1995–2020 mean. Atmospheric chemistry, transport and the changing NO_x-VOC relationship may be complicating these relationships (Stanier et al., 2021; Vermeuel et al., 2019). Also, there were a number of days that likely had additional O_3 due to smoke from several areas of large fire activity. Figure S7 in Supporting Information S1 shows the fire locations, HMS smoke, MDA8 O_3, and daily $PM_{2.5}$ values on 19 June 2020. On that date, smoke came to the Midwest from two regions, Arizona and Missouri. The large Arizona fires included the Bush, Big Horn, and Magnum fires, which, combined, burned more than 155,000 ha from early June to mid-July. There is also significant burning occurring at this time in the midwestern states, particularly in southeastern Missouri. These fires are in heavy agricultural regions. The HMS data show that the smoke from both the Arizona fires and the midwestern agricultural fires was transported to the Midwest. Surface $PM_{2.5}$ is enhanced around the region, including in rural areas, and smoke can be seen in the HMS product over the Midwest. The daily $PM_{2.5}$ values in the Chicago area on June 19 are between 14 and 21 μg/m^3, which is consistent with surface impacts from smoke. Figure S8 in Supporting Information S1 shows the MDA8 O_3 concentrations versus DMT for Chicago for May-September 2018–2020 with the four highest days in 2020 highlighted. The 2020 highest days have significantly more O_3 than would be expected from the temperature relationship, which is consistent with earlier studies demonstrating impacts of smoke on the MDA8 O_3 (e.g., Gong et al., 2017; Jaffe, 2021; Lindaas et al., 2017; Pollack et al., 2021). The 19 June MDA8 O_3 concentration (104 ppb) is the highest value in the Chicago region since 2002 and emissions from distant sources likely contributed to this high value.

The combined impact of regional anthropogenic emissions and emissions from these fires, combined with warm temperatures on 18 and 19 June, led to the highest MDA8 O_3 values of the year in Chicago (93 ppb) and Milwaukee (76 ppb). The combination of above average temperatures and smoke are clear co-factors that enhance O_3 production (Jaffe, 2021), more than either one alone. In addition, other meteorological factors, for example, land/lake breeze effects or boundary layer exchange will also impact local O_3 levels and can complicate this interpretation.

3.3. Long-Term Relationship Between O_3 and NO_2

Over the 26-year period we examined, there is a strong relationship between the annual NO_2 and fourth highest MDA8 O_3 for most NAAs; the data show a significant and linear relationship. Table S2 in Supporting

Information S1 gives the annual NO_2 and fourth highest MDA8 O_3 data, and Table S4 in Supporting Information S1 gives the parameters for the linear fits. The R^2 values for the linear correlations are mostly above 0.5, and some can be as high as 0.87 (Ventura County, CA). The correlation coefficients are highest in the East, followed by the Midwest and lowest in the West. A few locations in the West have very weak or insignificant relationships including Denver ($R^2 = 0.15$), Las Vegas ($R^2 = 0.01$), and Salt Lake City ($R^2 = 0.27$). These weaker correlations are likely due to substantial year-to-year variations in influence from wildfires and smoke, contributions from the stratosphere (Langford et al., 2017) and generally higher background O_3 (Jaffe et al., 2020). The O_3-NO_2 slopes (ppb O_3 per ppb NO_2) tend to be highest and the intercepts (O_3 value at 0 ppb NO_2) lowest in the eastern U.S. In the WUS, the intercepts are relatively higher (mean value of 63 ppb) and this likely reflects the higher background O_3 in the WUS (e.g., Jaffe et al., 2020; Langford et al., 2017) or the ongoing transition to a NO_x limited regime (e.g., Riverside and Los Angeles, CA).

4. Summary and Recommendations for Future Research

O_3 is a key air pollutant. In the U.S., more than 100 million people live in areas that exceed the current NAAQS. While most NAAs in the eastern and midwestern regions are making good progress toward meeting the standard, most regions in the West are not. This is primarily due to the combined influences of high background O_3 and the recent large fire years.

In this study, we have shown that nearly all NAAs show a significant correlation between the observed peak O_3 and annual NO_2 concentrations. In addition, most regions have now transitioned to a NO_x-limited regime, as indicated by the weekend-weekday pattern of NO_2 and probability of an O_3 exceedance. In eastern U.S. NAAs, reductions of NO_x emissions of 10%–20%, as occurred in summer 2020 due to pandemic-related traffic reductions, were sufficient for these regions to achieve O_3 concentrations that are consistent with meeting the U.S. O_3 standard for the first time. The O_3 response was greater than expected from the long-term O_3-NO_2 relationship, suggesting that NO_x emission reductions become more effective in reducing O_3 at lower NO_2 concentrations. In the midwestern NAAs, the 2020 changes were not as consistent as in the eastern region. These sites are not far above the standard, and so continued emission reductions will likely lead to further O_3 reductions. For the WUS, the NAAs have a much more challenging problem in meeting the O_3 standard. While the O_3 versus NO_2 data still suggest a linear relationship, in many western NAAs, the relationship is weaker and the intercept is higher, indicating a greater contribution from background O_3 (Jaffe et al., 2020) or a slower transition to a NO_x limited regime. In addition, large fire seasons have become more frequent in the WUS, and these contribute substantially to the O_3 levels. Smoke influence on peak O_3 concentrations was especially apparent in 2018 and 2020. For smoke influenced days, there is significant uncertainty in the complex chemical processing. If an elevated pollution event can be attributed to a "natural" cause, then these data can be excluded from regulatory consideration under the EPA's "exceptional event" rules (U.S. Environmental Protection Agency (U.S. EPA, 2016). However, this does not mitigate the health impacts. One study suggests that the large amount of VOCs in smoke plumes, combined with urban anthropogenic NO_x, drive enhanced photochemical O_3 production (Ninneman & Jaffe, 2021b). This can be true even if the photochemistry in the region is generally NO_x-sensitive. If this is correct, then continued reductions in NO_x emissions would be beneficial to reduce O_3 for both smoke and non-smoke days.

Data Availability Statement

All data used in this analysis are publicly available. Air quality data (O_3, $PM_{2.5}$, and NO_2) are from the U.S. EPA's AQS database via the Air Data interface (U.S. EPA, 2022). Meteorological data are from the NOAA National Centers for Environmental Information database (NOAA, 2022a) and the NOAA Hazard Mapping System (HMS) Fire and Smoke Products are from the National Environmental Satellite, Data, and Information Service archive (NOAA, 2022b).

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