Author response

On the effect of upwind emission controls on ozone in Sequoia National Park

Referee Comments (bold)

Author Response (italics)

Author Changes to Manuscript (standard)

We thank the reviewers for their feedback, which has improved the quality of the manuscript. We address each point below.

Anonymous Referee #1

This is a well-written manuscript, supported by ample and well-developed data analysis, that involves a topic of substantial interest. Thanks to the authors for preparing the work. A few minor suggestions and comments are provided simply for consideration.

General:

1) Am a little concerned about the conclusion regarding O3 sensitivity to downwind *distance* from Visalia/SJV given that (if i understood correctly) that this finding is based on just the two sites in SNP (one ~ 10km downwind of the other). Two general concerns here:

a) Do the authors intend for readers to extrapolate this conclusion beyond these two specific SNP sites? Would a hypothetical 3rd site further downwind by 10-20 km from SJV be expected to have shown even greater responsiveness? At some point downwind, this conclusion presumably breaks down as areas become less and less influenced by SJV. If this conclusion is intended to be limited to these two SNP sites, maybe those parts of the paper that cite downwind distance as a factor in responsiveness could be revised to limit this conclusion to the SEQ1 and SEQ2 sites.

We have adjusted the way we speak about downwind distance and limited our conclusions to SEQ1 and SEQ2. We also add text to address this comment directly in the text.

Page 3, Lines 31–32: "We describe these O_3 changes in Visalia and SNP as function of distance downwind of Visalia by way of data collected at two monitoring stations located on the western slope of the Sierra Nevada Mountains."

Page 8, Lines 32–33: "…O_3 decreased more rapidly in SNP versus Visalia and at SEQ2 versus SEQ1."

Page 9, Line 12: "Additionally, greater O_3 decreases were observed at SEQ1 than Visalia and at SEQ2 compared to SEQ1."

Page 10, Lines 2–3: "NO_x decreases have generally made greater improvements in O_3 in SEQ1 than Visalia and in SEQ2 than SEQ1, a trend that corresponds to increasing distance downwind of the SJV."

Page 11, Lines 14–20: "Downwind sites usually experience PO_3 chemistry that is more NO_x-limited than in the often NO_x-suppressed (or at least more NO_x-suppressed) urban core. As a result, we expect similar location-specific O_3 trends in other ecosystems and national parks downwind of major NO_x sources like cities. However, while the extent of observed O_3 improvements in SNP follows the pattern of increasing distance downwind of Visalia with sustained NO_x emission control in the SJV (Russell et al., 2010; Pusede and Cohen, 2012), PO_3 chemistry is non-linear and the direction of location-specific trends may vary. That said, at some distance downwind this conclusion breaks down, as areas become less and less influenced by the upwind source."

b) Is it possible, especially given complex flows in the region, that the elevation differences between these two sites is an equal or greater driver of the differential O3 decreases in the area than distance?

You are correct. The distance of airflow will be dictated by the mountain terrain and will travel a distance longer than determined as a straight-line path on a flat surface. Accounting for the change in elevation simply using the Pythagorean theorem, the horizontal change is greater than the vertical change to the extent that the horizontal distance is a reasonable approximation. We have added text to this point.

Page 6, Lines 9–16: "If O_3 attributed to local PO_3 in Visalia is greatest around 2 pm LT, typical of many urban locations, with mean winds at SEQ1 of 3 m s^{-1} and SEQ2 of 2 m s^{-1}, we expect O_3 to peak in SEQ1 at ~5 pm (45 km downwind of Visalia) and at SEQ2 shortly after (9.7 km downwind of SEQ1, which includes the change in elevation using the Pythagorean theorem). This is broadly what we observe. While the actual distance of airflow is dictated by the mountain terrain and a parcel of air will travel a distance longer than the straight-line path on a smooth surface, the timing of the O_3 diurnal patterns is consistent with airflow travel time roughly equal to that determined by the horizontal distance and mean wind speed. There has been no change in the hour of peak O_3 mixing ratio at either SEQ1 or SEQ2 over the 2001 to 2012 period."

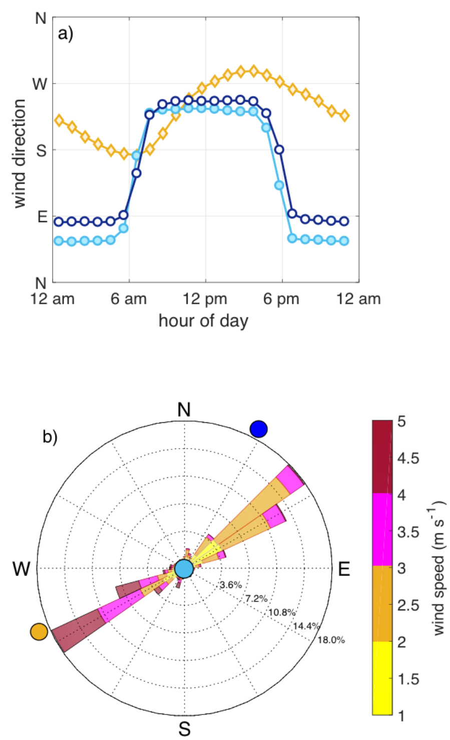
2) A little more detail on the approach used to project future exceedances would be useful (Table 5 [sic]). Clearly, the trend is assumed constant, but then did you also assume that the within-year variability would also stay the same?

We have removed future projections from our analysis in response to comments from anonymous Reviewer 2.

Specific:

3) Figure 2: Information about mean vector flow would be more informative than mean direction. The paper notes the mean wind speeds on page 6, line 3.

We have added wind rose plots over SEQ1 and SEQ2 with mean wind speeds shown. In the process, we also discovered an error, in which SEQ1 and SEQ2 were mislabeled. New Figure 2:



"**Figure 2.** Hourly mean wind directions in Visalia (orange diamonds), SEQ1 (cyan filled circles), and SEQ2 (dark blue open circles) in April–October, 2001–2012 (panel a). Wind rose for SEQ1 (panel b) with the direction of the neighboring sites of Visalia (orange), SEQ1 (cyan), and SEQ2 (dark blue) indicated."

4) Section 2: If possible, a schematic of the various flows and layers would be valuable.

Because we do not advance knowledge of airflow in the SJV, we prefer not include a schematic. We direct the reviewer to the excellent diagram in Zhong et al. (2004).

Page 4, Line 16: "Multiple airflow patterns influence O_3 in SNP and the SJV (see Zhong et al. (2004) for a diagram)."

Page 5, line 2: "during *the* ozone season"

Corrected.

Page 6, line 1: "Figure 2"

Corrected.

Page 7, line 18: The 8-hour ozone NAAQS is slightly more involved than described here. EPA defines a design value metric to determine an area's status relative to the NAAQS. For 8-hour ozone, the design value is the 3-year average of the 4th-highest max daily 8-hour ozone concentration at a site. Based on the description here, it's not entirely clear what metric was used for the trends in table 1. Clarification would be helpful.

We have added a definition of non-attainment, changed NAAQS to MD8A O_3 in numerous places throughout the text, and defined our use of exceedance.

Page 7, Lines 31–34; Page 8, Line 1: "The MD8A O_3 is a human health-based metric computed as the maximum unweighted daily 8-h average O_3 mixing ratio. A region is classified as in nonattainment of the NAAQS when the fourth-highest MD8A O_3 over a 3-yr period, known as the design value, exceeds a given standard. In this work, we utilize the seasonal mean MD8A and discuss O_3 exceedances as individual days in which MD8A O_3 > 70.4 ppb, the current 8-h NAAQS."

8) Page 7, lines 21-26: My (limited) understanding is that statistics like W126 are typically calculated over a specified period (e.g., 3 months for W126 as discussed on page 8). Does this paper follow those conventions for the Table 1 trends? Either way, may want to clarify.

We have changed our previous computations of daily SUM0 and W126 indices to consecutive 3-month summations, following EPA protocol for W126. We have clarified this in the text.

Page 8, Lines 8–18: "Here, SUM0 and W126 summations are computed following the W126 protocol (Environmental Protection Agency, 2016), affording straightforward comparisons between the metrics. First, in months with less than 75% of hourly data coverage in the 8 am–8 pm LT window, missing values are replaced with the lowest observed hourly measurement over the study period (i.e. April–October) only until the dataset is 75% complete. Second, monthly summations of daily indices, comprised of hourly data (8 am–7 pm), are computed; when data are missing, the summation is divided by the data completeness fraction.

Consecutive 3-month metrics are computed by adding monthly indices. In practice, SUM0 and W126 are computed as 3-yr averages of the highest 3-month summation; however, we define springtime SUM0 and W126 as the 3-month summation over April–June and O_3 season SUM0 and W126 as the mean of the 3-month summations over June–August, July–September, and August–October (not the highest of the three 3-month sums). Because less than 15% of data were available for August 2008 at SEQ1, O_3 season SUM0 and W126 were computed as the mean of 3-month summations over June, July, and September, and July, September, and October only for this site and year."

9) Page 8, lines 1 and 11: It's not immediately clear to me ... is the term "interannual variability" as used in the context of Table 1 referring to the year-to-year differences in these metrics (i.e., the standard deviation of yearly values over the 12-year period)? Or is it just referring to the trend itself as "interannual variability"? May want to clarify, especially if you mean the latter.

We have exchanged use of "variability" for "trends" to make this distinction clear and renamed the subsection.

10) Page 9, line 21: The paper tends to reserve the use of the term "impacts" for O_3 impacts on plants and use the term concentrations when talking about non-plant effects (e.g., human health). This is fine, but of course both are impacts.

We added clarification to the initial usage but retained the convention for clarity.

Page 5, Lines 18–20: "In this manuscript, for clarity we generally use the term *impacts* when discussing ecosystem metrics and *concentrations* when talking about human health metrics; O_3 ecosystem and human health effects are of course both O_3 impacts."

11) Table 5: Given the statement on page 10, lines 1-3 about the potential overly optimistic nature of the W126 metric relative to SUM0, why not include SUM0 in Table 5 instead of W126?

There are no protective thresholds for SUM0, only for W126. We have removed the values for O_3 season, which we state are the poorest predictor of plant O_3 uptake. We have clarified as follows:

Page 9, Line 26: "While there is no standard for SUM0, there are three time-integrated W126 protective thresholds."

12) Table 5: Very minor It might make it easier on the reader if this table was reconfigured such that directional changes were consistent across the two metrics (i.e.,

lower numbers indicate improvement). May want to consider switching from # of days required for an exceedance to something like the inverse of that.

We have removed the table, placing the values in the text. We were unable to think of a clear way to present the inverse of days until exceedance. However, now that the data and explanation are in paragraph form, we hope the distinction is improved. New text:

Page 9, Lines 29–32: "Rather than calculate W126 exceedances using a 3-month summation of monthly indices, we instead count the number of days required for an exceedance to occur, summing daily W126 indices from the first day of the springtime (1 April). A larger number of days indicates improved air quality. We do this to generate information in addition to exceedance frequency, as W126 O_3 at SEQ1 and SEQ2 is greater than all three standards in all years in both seasons."

13) Table 5: Per an earlier comment, it's not clear to me how you could have a value > 92 days (e.g., the value of 107 listed for 9 ppm h in 2021) if the W126 metric is calculated over 3 months. Wouldn't that be a "never"?

This has been removed.

14) Page 11, lines 13-14: May want to clarify these specific listed values in text are for SUM0.

Reference to ~18% reduction in O_3 season at SEQ2 encompasses the reductions across all three metrics of 19% (MD8A), 18% (SUM0), and 17% (Morning O_x). We have changed the text to make this clear:

Page 13, Lines 27–29: "The three metrics, MD8A, SUM0, and morning O_x, all indicate comparable reductions in O_3 over 2001–2012, with decreases of ~7% (springtime) and ~13% (O_3 season) at SEQ1 and 13–16% (springtime) and 15–19% (O_3 season) at SEQ2."

Anonymous Referee #2

General comments:

Overall, the paper is well written and is an easy read, but there are some fundamental issues that must be addressed before this paper can be published. In general, there are numerous, rather bold statements, that need to be substantiated. Most things are overstated in the manuscript, and the rudimentary analysis done in Section 3.4, past and future exceedances, is completely unacceptable for any paper that is going to be published in ACP, or any other scientific journal.

1) As far as overstating goes, the first sentence of the abstract is simply not correct:

"Abstract. Sequoia National Park (SNP) experiences the worst ozone (O_3) pollution of any national park in the U.S." [quotations added]

My response to the first sentence of the abstract is: NO – if you look at the NPS ozone data for all of their sites, you find that Joshua Tree is actually the worst, Sequoia and Kings Canyon is comparable at best. Even though they are only using data through 2012, the following is still relative and the patterns remain the same: Acadia and Joshua Tree recently reported the highest ozone levels in 2017 for all NPS sites, and Yosemite also beat out Sequoia and Kings Canyon for 2017. Moreover, Dinosaur National Monument has wintertime ozone levels can greatly exceed what is observed at Sequoia and Kings Canyon. My point, the information disseminated throughout the manuscript must be conveyed accurately, and not overstated. Simply changing the sentence to "some of the worst" is all that is needed, but these types of statements are too common throughout the manuscript.

Changed to:

Abstract, Line 1: "Ozone (O_3) pollution in Sequoia National Park (SNP) is among the worst of any national park in the U.S."

Page 1, Lines 25–26: "Sequoia National Park (SNP) is a unique and treasured ecosystem that is also one of the most ozone-polluted national parks in the U.S. (National Park Service, 2015a)."

National Park Service, 2009–2013 Ozone estimates for parks, http://www.nature.nps.gov/air/Maps/AirAtlas/IM_materials.cfm, last access 7 July 2018, 2015a.

2) Additionally, the authors don't really convey any new information – one of their main points, that the transport of NOx is more important than the transport of ozone to the sites in the park is something people already know and understand for this area. How else would you manage to get higher levels in the park if the precursors were not being transported out of the source region photochecmically processed along the way?

We agree past analyses have identified precursor transport as important and stated that fact in the initial submission with reference to Jacobson (2001). Our focus is on observed trends in O_3 over time and differences in those trends with season, which to our knowledge have not yet been published. Our main point is not that NO_x transport is more important than O_3 transport, but that because of this, O_3 chemistry in the SJV and SNP, and hence O_3 concentrations, are differently sensitive to NO_x emission control.

In addition, the influence of NO_x transport on SNP O_3 has not yet been shown empirically to our knowledge.

3) Additionally, it is stated in a couple places in the manuscript that emission controls are optimized for the hottest days in the summer, so the policies that have been implemented are not optimized for decreasing springtime ozone, when it is cooler. This is a rather bold and cavalier statement to make without providing any type of information on what the policies are, and how the seasonal differences in temperature affect the emissions control strategies. In my opinion, there needs to be a substantial discussion, that pulls in what the policies are, and how the overall emissions are affected by these seasonal temperature differences to justify their statement that these polices are less effective in the springtime during cooler weather. My guess is they are trying to rehash the points about temperature dependence as described in Pusede et al. (2015); however, they have stated that it's the emission control strategies that aren't optimized, so this means diving into the SIPs and seeing what and how emissions were/are controlled and correlating this to the seasonal temperature changes. The authors beat on policy not being appropriate for the seasonal changes, so this needs to be addressed. In particular, what part of the SIPs are not effective for the springtime emissions and how can they demonstrate this? What would be done differently to improve the effectiveness of the emission control policies to improve springtime ozone?

We state that controls are designed to address high O_3 as defined by the 8-h NAAQS. We also state that in the SJV, these exceedances are most frequent when temperatures are hottest. We do not say that controls are just optimized for hot days.

Pusede et al. (2015) is a review paper on the body of literature describing the O_3-temperature correlation, we are not sure in what respect such a paper can be rehashed.

We have added text and references to EPA guidance for modeling to be used for regulatory design to select O_3 episodes in which the MD8A is high. We have also elaborated on episode selection as relevant.

Page 12, Lines 3–24: "Over 2001–2012, O_3 declines have mostly been smaller in SNP when plant O_3 uptake is greatest (springtime), despite comparable NO_x decreases in both seasons. This may be in part because regulatory strategies prioritize attainment of the O_3 NAAQS in polluted urban areas like the SJV basin, where air parcels influenced by the results of these controls are then transported downwind to locations with different PO_3 chemistry. In the development of regulatory plans, agencies use models to hindcast past O_3 episodes, facilitating testing of the efficacy of specific NO_x and/or organic emissions reductions over that episode to meet the 8-h O_3 NAAQS or progress goals (Environmental Protection Agency, 2007; Environmental Protection Agency, 2014). In nonattainment areas, U.S. EPA guidance recommends modeling past time periods that meet a number of specific criteria, such as typifying the meteorological conditions that correspond to high O_3 days as defined by the MD8A greater than the NAAQS value and focusing on the ten highest modeled O_3 days (Environmental Protection Agency, 2007; Environmental Protection Agency, 2014). Regulatory modeling in the SJV (Visalia, SEQ1, and SEQ2 are included in this attainment demonstration) is more comprehensive, as it was recently updated to span the full O_3 season (defined as May–September); still potential reductions (known as relative reduction factors, RRFs) are based on the MD8A and restricted to high O_3 days (San Joaquin Valley Air Pollution Control District, 2007; San Joaquin Valley Air Pollution Control District, 2014). In the SJV, high O_3 days are most frequent in the late summer (O_3 season) and on the hottest days of the year (Pusede and Cohen, 2012). Even in SEQ1 and SEQ2, days with MD8A > 70.4 ppb are far more common in the summer. Because of chemical and meteorological differences between seasons, this may lead to policies not optimized to decrease O_3 in cooler springtime conditions, which in the SJV are more NO_x-suppressed and therefore more sensitive to controls on reactive organic compounds (Pusede et al., 2014). In addition, we observe greater year-to-year O_3 variability in the springtime than during O_3 season (Figure 6), suggestive of a larger relative role of interannual meteorological variability controlling O_3. Deeper cuts in emissions would be required in the springtime, as decreases in anthropogenic emissions have a proportionally smaller effect on the total O_3 abundance than during O_3 season."

Environmental Protection Agency: Guidance on the use of models and other analyses for demonstrating attainment of air quality goals for ozone, $PM_{2.5}$, and regional haze, EPA-454/B-07-002, Research Triangle Park, NC, 2007.

Environmental Protection Agency: Draft modeling guidance for demonstrating attainment of air quality goals for ozone, $PM_{2.5}$, and regional haze, Research Triangle Park, NC, 2014.

San Joaquin Valley Air Pollution Control District, 2016 Plan for 2008 8-hour ozone standard: http://www.valleyair.org/Air_Quality_Plans/Ozone-Plan-2016.htm, 2016.

San Joaquin Valley Unified Air Pollution Control District, 2007 Ozone plan: http://www.valleyair.org/Air_Quality_Plans/AQ_Final_Adopted_Ozone2007.htm, 2007.

4) Moreover, the authors discuss how precursor emission controls have been less effective at reducing O_3 concentrations in SNP in springtime, yet, there is no mention or discussion about other factors that may be influencing springtime ozone. For example, how do the springtime chemistry and dynamic processes of the widely observed springtime maximum of ozone in the Northern Hemisphere mid latitudes influence ozone levels in

this region? Are these processes influencing what the authors are referring to as less effective emission controls during the spring? Also, it's not actually clear in the paper how the authors get to the conclusion that ". . .precursor emission controls have been less effective at reducing O_3 concentrations in SNP in springtime. . .".

The initial submission included discussion of trans-Pacific transport and its influence on springtime O_3 trends. We have expanded the discussion to address trends in springtime background O_3 more broadly and included this text:

Page 12, Lines 24–34; Page 13, Lines 1–14: "An additional challenge to regulators is the contribution of background O_3 concentrations to O_3 levels (Cooper et al., 2015), as natural sources produce O_3 even in the absence of anthropogenic precursor emissions, O_3 can be transported over significant distances, and O_3 concentrations are influenced by large-scale meteorological and climatic events. Multiple studies have identified an increasing trend in O_3 at rural sites (often used as a proxy for background O_3) in the western U.S., particularly in the springtime (e.g., Cooper et al., 2012, Lin et al., 2017). Parrish et al. (2017) presented observational evidence of a slowdown and reversal of this trend on the California west coast since 2000, though the reversal was stronger in the summer than springtime. Using observations and the GFDL-AM3 model, Lin et al. (2017) computed that Asian anthropogenic emissions accounted for 50% of simulated springtime O_3 increases at western U.S. rural sites, followed by rising global methane (13%) and variability in biomass burning (6%). Northern mid-latitude transport of Asian pollution to the western U.S. is strongest during March–April and weakest in the summertime (e.g., Wild and Akimoto, 2001; Liu et al., 2003; Liu et al., 2005), with high-elevation locations in the Sierra Nevada Mountains being more vulnerable to reception of Asian O_3 and O_3 precursors (e.g., Vicars and Sickman, 2001; Heald et al., 2003; Hudman et al., 2004). Hudman et al. (2004) compared surface observations with GEOS-Chem-modeled O_3 enhancements in Asian pollution outflow, finding that, on average, transport events in April–May 2002 led to 8 ± 2 ppb higher MD8A O_3 concentrations at SEQ2. East Asian NO_x emissions have risen over our study window (e.g., Miyazaki et al., 2017), potentially causing an increase in the influence of trans-Pacific transport on O_3 concentrations at SEQ2 and reducing the efficacy of local NO_x control in springtime. Background O_3 concentrations are also responsive to large-scale climatic events, and elevated springtime O_3 at rural sites in the western U.S. has been linked to strong La Niña winters (Lin et al., 2015; Xu et al., 2017), which are associated with an increased frequency of deep tropopause folds that entrain O_3-rich stratospheric air into the troposphere (Lin et al., 2015). Over our study period, strong La Niña events occurred during the winter of 2007–2008 and 2010–2011. In general, transport of Asian pollution and tropopause folds are expected to have a greater impact in the springtime and at the higher-elevation SEQ2. While we do observe smaller decreases in O_3 in springtime at

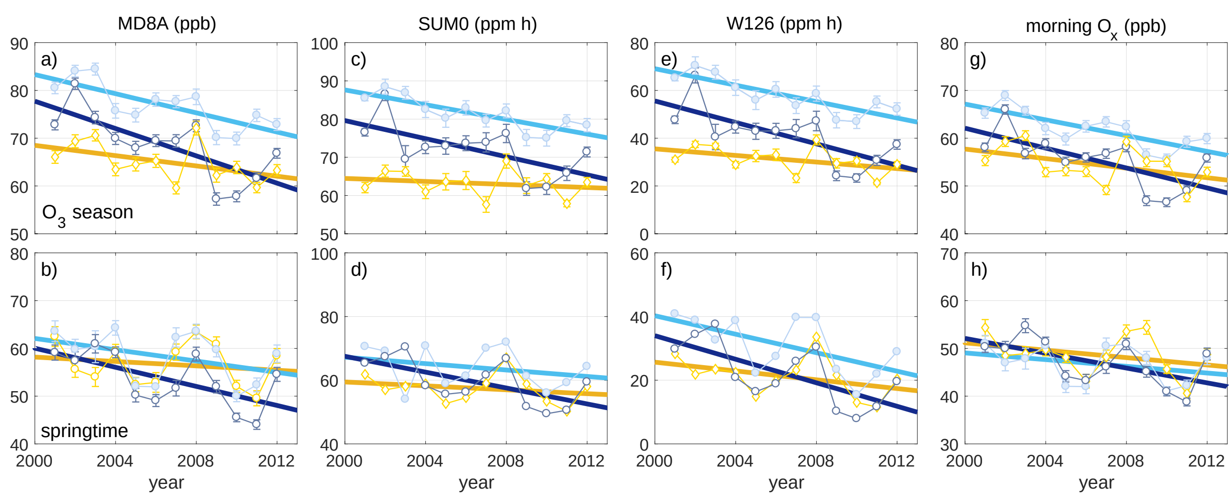
SEQ2 than during O_3 season, interannual trends have been more downward at SEQ2 than at the lower elevation sites, SEQ1 and Visalia, in both seasons. This suggests that these factors may impact surface O_3 at high-elevations in SNP during individual events (e.g., Hudman et al., 2004) but that interannual trends in seasonal averages are more influenced by chemistry during upslope outflow from the SJV."

5) Finally, the term "trend analysis" is used quite a bit in the paper; however, it would be useful if they included a figure of the full time series of ozone data from the sites, the annual 8-hr 4th high, a table of annual basic statistics to help set the stage for the analysis. What is presented is rather "thin" – the reader needs to be provided more information in order to better evaluate what is presented. . .which is very little. This is even that much more important for Sect. 3.4 – the authors should, at minimum, show the simple regression that was used to come up with the values in Table 2. I personally think this section should be removed or done in a much more rigorous manner, but the authors need to show how these values were derived.

We did not find any use of the term "trend analysis" in the paper.

We have removed Section 3.4.

We have added a figure with the regressions used to produce Table 1 that also displays the basic statistics we discuss. We do not include the design value because that is not a focus of our study, a point that should be clear in the updated version.



"**Figure 6.** O_3 trends in Visalia (orange diamonds), SEQ1 (cyan filled circles), and SEQ2 (dark blue open circles) computed using MD8A (a–b), SUM0 (c–d), W126 (e–f), and morning O_x (g–h) metrics during O_3 season (top row) and springtime (bottom row). Error bars in panels a–b and g–h are standard errors of the mean. Error bars in panels c and e are standards errors of the mean of the three O_3 season 3-month summations."

Specific comments:

6) P1, L21-22: If you are referring to the whole area (re Sierra Nevada forests), then you should use the 4 letter NPS designation for the site, SNP should be referred to as SEKI, as the measurements are representative of Sequoia and Kings Canyon NPs.

We use data from two monitoring stations in SNP and do not use data from Kings Canyon. We do not know if these measurements are representative of the full SEKI and believe SNP is a more accurate descriptor for our purpose.

7) P1, L25-26: The reference cited in this sentence does not make the statement that it Sequoia is the most ozone polluted park in the U.S. – please ensure that you accurately represent what a reference says, period. "Sequoia National Park (SNP) is a unique and treasured ecosystem that is also the most ozone-polluted national park in the U.S. (Meyer and Esperanza, 2016)."

We have updated the reference:

Page 1, Lines 25–26: "Sequoia National Park (SNP) is a unique and treasured ecosystem that is also one of the most ozone-polluted national parks in the U.S. (National Park Service, 2015a)."

National Park Service, 2009–2013 Ozone estimates for parks, http://www.nature.nps.gov/air/Maps/AirAtlas/IM_materials.cfm, last access 7 July 2018, 2015a.

8) P2,L7-9: Revise the following sentence – reads awkwardly: On multi-decadal timescales, O_3-resistant plants may thrive over O_3-sensitive species, system-level dynamics that would maintain forest productivity and carbon storage, but would induce changes in ecosystem composition (Wang et al., 2016).

We have revised the sentence to read:

Page 2, Lines 7–9: "On multi-decadal timescales, O_3-resistant plants may thrive over O_3-sensitive species, and these system-level dynamics would maintain forest productivity and carbon storage but would induce changes in ecosystem composition (Wang et al., 2016)."

9) P3, L3: there are additional references that should be included regarding the W126 Metric.

We have included two additional EPA references:

Page 3, Lines 5–6: “W126 is a 12-h daily 3-month summation weighted to emphasize higher O_3 concentrations (Environmental Protection Agency, 2006; Environmental Protection Agency, 2016) that is used by the U.S. National Park Service.”

Environmental Protection Agency, Air quality criteria for ozone and related photochemical oxidants, Final report EPA/600/R-05/004aF-cF, Washington, DC, 2006.

Environmental Protection Agency, Ozone W126 index: https://www.epa.gov/air-quality-analysis/ozone-w126-index, last access: 27 October 2016, 2016.

10) P3,L16; technically, the NPS started measuring ozone in the early 1980s, not the late 1980s. Shenandoah NP started in 1983 and Sequoia and Kings Canyon NP – Lower Kaweah started in 1984.

We removed the word “late” from this sentence.

11) P4, L25: Beginning a sentence with “Due to” is grammatically incorrect. The Chicago Manual of Style suggests using “due to” when you can replace it with “attributable to,” but not when you could use "because of"; if a sentence starts with “due to”, it is most likely incorrect. Therefore, please revise.

We have revised the sentence to read:

Page 4, Lines 29–32: “The prevalence of shallow nighttime surface inversions in the SJV means that evening downslope valley flow at higher elevations may be stored within nocturnal residual layers and entrained into the surface layer the following morning.”

12) P4, L30-31: “strongly” in “strongly temperature-dependent” really should be defined here – a counter to this statement is that in the Uintah Basin, during snow cover cold periods, ozone levels are usually higher there than in the Sequoia and Kings Canyon National Parks, yet the temperature is significantly lower. Temperature is only one factor, not the only factor. Moreover, high ozone episodes have occurred as early as March, but high ozone starting in the spring is fairly typical, so I would change “summer” to “spring” or through the fall. Ozone levels in Sequoia and Kings Canyon are comparable in April and September.

As stated in the paper, 90% of days with MD8A > 70 ppb occur during O_3 season, with just 10% occurring in spring (2001–2012). Over 2001–2012, we calculate mean MD8A of 51 ppb in April and 76 ppb in September. Therefore, we opt to keep “summer” in the text.

We did not say PO_3 is only dependent on temperature, just that PO_3 is temperature dependent. We have rewritten the sentence to read:

Page 5, Lines 2–4: "High O_3 days are most frequent in SNP and the SJV in the summer through early fall (Pusede and Cohen, 2012; Meyer and Esperanza, 2016), as PO_3 chemistry is often temperature-dependent (reviewed in Pusede et al., 2015) and this effect is particularly strong in the SJV (Pusede and Cohen, 2012; Pusede et al., 2014)."

13) P5, L4: "due to" is inappropriate here – it is a result of the Mediterranean climate.

We have changed "due to" to "because of."

14) P5, L21: First off, there isn't a methods or experimental section – more information needs to be provided. This get to the more important point that you state that all data are provided by CARB, when in fact, they are not. The data are served up by CARB on their website, but the NPS data is provided by the NPS on their data page, which also gets uploaded to AQS, which is the main repository that houses all national ozone data – it is the single main repository. CARB either serves up the data directly from AQS or in their own database, where they have either obtained the data directly from the NPS or AQS. So, it should be clearly stated who is the proprietor of the data and where it was obtained from. These have been merged into one item, and what is disseminated in this sentence is incorrect.

While there is not a dedicated methods section, we believe we have provided readers will all relevant details on which data were used, where data were acquired, and how the calculations were done. We have adjusted the sentence to read:

Page 5, Lines 26–28: "The data are collected by various agencies, including the National Park Service, and are hosted by the California Air Resources Board and available for download at https://www.arb.ca.gov/aqmis2/aqdselect.php."

15) P7, L15: The NAAQS for ozone is an 8 hour average value; it is the annual 4th highest daily maximum 8 hour average ozone concentration, averaged over 3 consecutive years (the design value) – this must not exceed 70 ppb. So, what you are saying is repetitive and not conveyed properly. What you should say is the annual 4th highest daily maximum 8 hour average, or the DM8HA or DM8A, not the "8-h O3 NAAQS".

We have added a definition of non-attainment, changed NAAQS to MD8A O_3 throughout the text, and defined our use of exceedance.

Page 7, Lines 32–33; Page 8, Line 1: "The MD8A O_3 is a human health-based metric computed as the maximum unweighted daily 8-h average O_3 mixing ratio. A region is classified as in nonattainment of the NAAQS when the fourth-highest MD8A O_3 over a 3-yr period, known as the design value, exceeds a given standard. In this work, we utilize the seasonal mean MD8A and discuss O_3 exceedances as individual days in which MD8A $O_3 > 70.4$ ppb, the current 8-h NAAQS."

16) P7, L16: Why are trends only reported as a percent change? It would be more useful to include the ppb per yr trend in the table, along with the percent change. Moreover, why is only the 8-hr daily max being listed? I'm assuming it's the annual 4th high daily max 8-hr average, but it's not stated in the text. Please clarify.

We have updated the table to include the change in O_3 amount per year derived from the slope of the regression. We have clarified the meaning of the MD8A trend in the text, which is the seasonal mean MD8A and not the design value. See previous comment.

"**Table 1.** O_3 changes in Visalia, SEQ1, and SEQ2 over 2001–2012 according to MD8A, SUM0, W126, and morning O_x metrics based on a linear fit of annual mean data (shown in Figure 6) in the springtime and O_3 season. Each left column is the percent change with respect to fit value in 2001 at SEQ1 during O_3 season for comparison, which is the highest O_3 observed for each metric. Each right column is the fit slope with slope errors in O_3 abundance units per year."

O_3 metric	MD8A		SUM0		W126		Morning O_x	
O_3 season (June–October)								
	%	ppb y^{-1}	%	ppm h y^{-1}	%	ppm h y^{-1}	%	ppb y^{-1}
SEQ2	–19	–1.4 ± 0.41	–15	–1.2 ± 0.46	–37	–2.2 ± 0.72	–17	–1.0 ± 0.32
SEQ1	–13	–1.0 ± 0.27	–12	–0.96 ± 0.21	–28	–1.7 ± 0.36	–14	–0.83 ± 0.21
Visalia	–7	–0.54 ± 0.30	–3	–0.20 ± 0.28	–11	–0.69 ± 0.41	–6	–0.50 ± 0.30
Springtime (April–May)								
	%	ppb y^{-1}	%	ppm h y^{-1}	%	ppm h y^{-1}	%	ppb y^{-1}
SEQ2	–13	–1.0 ± 0.38	–16	–1.2 ± 0.47	–30	–1.8 ± 0.62	–13	–0.78 ± 0.34
SEQ1	–8	–0.59 ± 0.42	–6	–0.50 ± 0.53	–24	–1.5 ± 0.62	–6	–0.35 ± 0.32
Visalia	–3	–0.23 ± 0.39	–4	–0.31 ± 0.38	–11	–0.69 ± 0.49	–8	–0.39 ± 0.35

17) P7, L16: This sentence is not correct – see previous comment. "The 8-h O3 NAAQS is a human health-based metric computed as the maximum unweighted daily 8-h average O3 mixing ratio." As for SUM0 – you need to say why it's called SUM0 – as in, why is it a "0"?

We have added a definition of non-attainment, changed NAAQS to MD8A O_3 throughout the text, and defined our use of exceedance.

Page 7, Lines 32–33; Page 8, Line 1: "The MD8A O_3 is a human health-based metric computed as the maximum unweighted daily 8-h average O_3 mixing ratio. A region is classified as in nonattainment of the NAAQS when the fourth-highest MD8A O_3 over a 3-yr period, known as the design value, exceeds a given standard. In this work, we utilize the seasonal mean MD8A and discuss O_3 exceedances as individual days in which MD8A O_3 > 70.4 ppb, the current 8-h NAAQS."

We have added clarification for the SUM0 metric:

Page 8, Lines 1–3: "SUM0 is equal to the sum of hourly O_3 concentrations over a 12-h daylight period (8 am–8 pm LT), as opposed to SUM06, which is limited to hourly O_3 mixing ratios greater than 60 ppb."

18) P8, L1-11: For this paragraph, only percentages are reported – it is absolutely necessary to include what the corresponding values were on ppb and ppm hrs for W126 and SUM0. For this to have value to ecosystem and plant effects folks, numbers, not percentages, are needed.

The new Figure 6 does this and we have added numbers for each metric at SEQ1 to the text. See comment 5.

Page 9, Lines 2–11: "For context in SEQ1, during O_3 season the mean MD8A declined from 82.3 ppb (2001–2002) to 73.8 ppb (2011–2012), but in the springtime the MD8A fell from 61.7 ppb (2001–2002) to 55.6 ppb (2011–2012). SUM0 O_3 fell from 87.0 ppm h (2001–2002) to 79.0 ppm h (2011–2012) during O_3 season and from 69.9 ppm h (2001–2002) to 61.8 ppm h (2011–2012) in the springtime. W126 O_3 decreased from 67.8 ppm h (2001–2002) to 53.7 ppm h (2011–2012) during O_3 season and from 39.8 ppm h (2001–2002) to 25.4 ppm h (2011–2012) in springtime. Morning O_3 fell from 67.1 ppb (2001–2002) to 59.6 ppb (2011–2012) during O_3 season and from 49.0 ppb (2001–2002) to 45.1 ppb (2011–2012). This pattern was not observed in one instance: SUM0 in SEQ2. Here, seasonal differences were comparable; however, mean daily indices were observed to differ, where SUM0 O_3 decreased from 0.914 ppm h (2001–2002) to 0.816 ppm h (2011–2011) during O_3 season, and, in the springtime, fell from 0.673 ppm h (2001–2002) to 0.616 ppm h (2011–2012), which amount to a change of –11% during O_3 season –8% in the springtime."

19) Section 3.4 You state that you "predict future O_3 levels in the context of protective threshold"; however, it is not stated how your do this in this section – please provide necessary information and figures.

We have clarified this point as follows:

Page 9, Lines 20–34; Page 10, Lines 1–4: "High O_3, as defined by exceedances of protective thresholds, also became less frequent over the 12-yr record. The number of days in which MD8A O_3 was greater than 70.4 ppb in 2001–2002 (averages are rounded up) was 68 yr^{-1} (O_3 season) and 15 yr^{-1} (springtime) in Visalia. In 2011–2012, the number of exceedances fell to 42 yr^{-1} (O_3 season) and 6 yr^{-1} (springtime). At SEQ1 in 2001–2002, there were 121 exceedance days yr^{-1} (O_3 season) and 21 yr^{-1} (springtime), declining in 2011–2012 to 99 yr^{-1} (O_3 season) and 10 yr^{-1} (springtime). At SEQ2 in 2001–2002, there were 103 exceedance days yr^{-1} (O_3 season) and 13 yr^{-1} (springtime). In 2011–2012, this decreased to 63 exceedance days yr^{-1} (O_3 season) and 3 yr^{-1} in 2011–2012 (springtime).

While there is no standard for SUM0, there are three time-integrated W126 protective thresholds. These are: 5–9 ppm h to protect against visible foliar injury to natural ecosystems, 7–13 ppm h to protect against growth effects to tree seedlings in natural forest stands, and 9–14 ppm h to protect against growth effects to tree seedlings in plantations, known as the 5, 7, and 9 ppm h standards (Heck and Cowling 1997). Rather than calculate W126 exceedances using a 3-month summation of monthly indices, we instead count the number of days required for an exceedance to occur, summing daily W126 indices from the first day of the springtime (1 April). A larger number of days indicates improved air quality. We do this to generate information in addition to exceedance frequency, as W126 O_3 at SEQ1 and SEQ2 is greater than all three standards in all years in both seasons. We only consider springtime, as this is when W126 is reported to better correlate with plant O_3 uptake (Panek et al., 2002; Kurpius et al., 2002; Bauer et al., 2000). At SEQ1 from 1 April in 2001–2002, 37, 41, and 45 days of O_3 accumulation reached exceedances of the 5, 7, and 9 ppm h thresholds, respectively (averages are rounded up). In 2011–2012, 3 to 13 more days were needed at SEQ1, as 40, 49, and 58 days of O_3 accumulation were required to exceed the 5, 7, and 9 ppm h thresholds. At SEQ2 from 1 April in 2001–2002, 41, 46, and 49 days of accumulation led to exceedance of the 5, 7, and 9 ppm h thresholds, respectively. In 2011–2012, 59, 65, and 73 days were required at SEQ2, or 18–24 more days."

20) "Future exceedances are computed assuming individual daily indices continue to decline at the 2001–2012 rate and are projected from 2011–2012 values." Is this a reasonable assumption? I'm not convinced this is the case. There is ozone data beyond 2012 at these sites, so does the rate of decline hold true? The fact that you are predicting future ozone levels off of this would suggest it should be evaluated.

We have removed our projections of future exceedances and instead included a discussion of Val Martin et al. (2015) and known regulations.

Page 11, Lines 21–34; Page 12, Lines 1–2: "Because PO_3 in SNP is NO_x-limited, future NO_x reductions are expected to have at least as large an impact on local PO_3 as past reductions. Seasonal mean NO_2 concentrations have decreased by 58% and 53% in Visalia in springtime and O_3 season, respectively. Local NO_x emissions should continue to decline into the future, as there are significant controls currently ongoing or in the implementation phase, including more stringent national rules on heavy-duty diesel engines (Environmental Protection Agency, 2000), combined with California Air Resources Board (CARB) diesel engine retrofit-replacement requirements (California Air Resources Board, 2008), and more stringent CARB standards for gasoline-powered vehicles (California Air Resources Board, 2012). While O_3 declines near or greater than those that occurred from 2001 to 2012 are required to eliminate exceedances in SNP, modeling analysis by Lapina et al. (2014) suggests that W126 in the region would be well below these thresholds in the absence of anthropogenic precursor emissions, implying further emissions controls would be effective. Under the stringent precursor controls of RCP4.5, Val Martin et al. (2015) projected decreases of 11% and 67% for the MD8A and W126 in 2050, respectively, from the base year of 2000, with mean O_3 decreasing from 58.9 ppb (MD8A) and 45.5 ppm h (W126) in 2000 to 52.7 ppb (MD8A) and 15.1 ppm h (W126). Under the RCP8.5, smaller O_3 declines were predicted, with MD8A unchanged and W126 falling by 38% to 28.3 ppm h. Given that these scenarios represent a reasonable spread of possible future climatic conditions, Val Martin et al. (2015) suggest at least W126 will remain well above protective thresholds in 2050."

21) "If past decreases in O3 continue over the next two decades, we predict no exceedances of the 8-h O3 NAAQS at SEQ2 by 2021 in springtime and by 2031 during O3 season, no exceedance of the 9-ppm h W126 threshold by 2021, and no further exceedances of 5- and 7-ppm h thresholds by 2031."

Following suit, more information needs to be provided to make such a bold statement when using such a rudimentary method. How much is NOx going to go down? How are large scale circulation patterns (e.g., PDO, ENSO, etc.) going to change and influence what is being transported in? What about the different climate futures? There are an array of emissions scenarios that can lead to significant differences in what you are inaccurately and inappropriately conveying here. Also, climate change - This section either must be expanded upon significantly or simply removed from the paper. As an example that contradicts your statements about ozone exceedances, the following is pulled directly from Val Martin et al. (2015) for Sequoia and Kings Canyon. Here, the report the actual values using different climate futures in order to assess what the ozone and W126 values will be. According to their rigorous analysis, both ozone and W126

values will exceed the current NAAQS level of 70 ppb and W126 values will also increase, and be well above the 5-9 ppm hr range.

Summer MDA-8 Ozone (ppb)

2000 (Baseline): 71.3, 2050 (RCP4.5): 72.9, 2050 (RCP8.5): 73.8

O3 W126 (ppm-hr)

2000 (Baseline): 46.0, 2050 (RCP4.5): 50.6, 2050 (RCP8.5): 53.2

Val Martin, M., C. L. Heald, J.-F. Lamarque, S. Tilmes, L. K. Emmons, and B. A. Schichtel

How emissions, climate, and land use change will impact mid-century air quality over the United States: a focus on effects at national parks, Atmos. Chem. Phys., 15, 2805–2823, 2015, www.atmos-chem-phys.net/15/2805/2015/.

We thank the reviewer for the reference. We have included discussion of Val Martin et al. (2015) as shown below. However, the quoted numbers must come from another article. Val Martin et al. (2015) report the following for Sequoia, which do imply O_3 *declines:*

Summer MDA-8 Ozone (ppb)

2000 (Baseline): 58.9, 2050 (RCP4.5): 52.7, 2050 (RCP8.5): 58.9

O3 W126 (ppm-hr)

2000 (Baseline): 45.5, 2050 (RCP4.5): 15.1, 2050 (RCP8.5): 28.3

Page 10, Lines 26–31: "With the Community Earth System Model, Val Martin et al. (2015) modeled air quality in national parks under two Representative Concentration Pathway (RCP) scenarios, computing substantially larger decreases over a 50-yr period in W126 O_3 compared to the MD8A. Considering that the SUM0 metric has been shown to best correspond to plant O_3 uptake in Sierra Nevada forests using O_3 flux observations (Panek et al., 2002) and that we observe W126 O_3 has declined at approximately twice the rate of SUM0 over 2001–2012, W126 trends may provide an overly optimistic representation of past declines in ecosystem O_3 impacts in SNP."

22) P9, L30: "O_3 reductions predicted by W126 are almost twice those of SUM0." What does this statement mean? How are ozone reductions predicted by W126 or SUM0? Both of these metrics are determined from ozone levels – how are these used to predict ozone reductions?

Changed as follows:

Page 10, Line 23: "Reductions in ecosystem O_3 impacts as represented by declines in W126 are greater than those of SUM0."

Page 13, Line 3; Page 14, Lines 1–2: "O_3 decreases over 2001–2012 computed with W126 are almost double those for SUM0, with the W126 emphasis of higher O_3 concentrations giving the most optimistic evaluation of the efficacy of past emission controls."

23) P10,L2: Regarding the following statement: ". . .W126 likely provides an overly optimistic representation of past and future trends in O3 impacts in SNP.", this is a rather bold statement to make to summarize the paragraph, yet you provide no hard evidence of this – there is nothing in this section that supports this statement. Please address this in a more rigorous manner.

We clarified our logic. Greater decreases in W126 relative to other O_3 metrics have also been reported by two national parks-focused modelling studies: Lapina et al. (2014), as we mentioned in the initial submission, and Val Martin et al. (2015). We have added Val Martin et al. (2015) to the discussion on this point.

Page 10, Lines 23–31: "Reductions in ecosystem O_3 impacts as represented by declines in W126 are greater than those of SUM0. We attribute this difference to the W126 weighting algorithm that makes the metric most sensitive to changes in the highest O_3. Using the GEOS-Chem model with a focus on national parks, Lapina et al. (2014) also found W126 was more responsive to decreases in anthropogenic emissions than daily (8 am–7 pm, LT) average O_3 concentrations. With the Community Earth System Model, Val Martin et al. (2015) modeled air quality in national parks under two Representative Concentration Pathway (RCP) scenarios, computing substantially larger decreases over a 50-yr period in W126 O_3 compared to the MD8A. Considering that the SUM0 metric has been shown to best correspond to plant O_3 uptake in Sierra Nevada forests using O_3 flux observations (Panek et al., 2002) and that we observe W126 O_3 has declined at approximately twice the rate of SUM0 over 2001–2012, W126 trends may provide an overly optimistic representation of past declines in ecosystem O_3 impacts in SNP."

24) P10, L9: For the following statement: ". . .leading to policies not optimized to decrease O_3 in cooler springtime conditions." Please elaborate on this point - this needs to be shown quantitatively. How large or small of a difference are you suggesting? What are the policies? How are they not optimized for the cooler springtime conditions? What could/should be done to address this policies in order to optimize them for the springtime?

We have elaborated on why policies may not be optimized for springtime and given two examples of what the results of this might be. Without performing model simulations, we cannot quantify these effects, but we have widened the discussion to be more specific and we believe more useful. The new text is included in response to comment 3 and shown below:

Page 12, Lines 3–23: "Over 2001–2012, O_3 declines have mostly been smaller in SNP when plant O_3 uptake is greatest (springtime), despite comparable NO_x decreases in both seasons. This may be in part because regulatory strategies prioritize attainment of the O_3 NAAQS in polluted urban areas like the SJV basin, where air parcels influenced by the results of these controls are then transported downwind to locations with different PO_3 chemistry. In the development of regulatory plans, agencies use models to hindcast past O_3 episodes, facilitating testing of the efficacy of specific NO_x and/or organic emissions reductions over that episode to meet the 8-h O_3 NAAQS or progress goals (Environmental Protection Agency, 2007; Environmental Protection Agency, 2014). In nonattainment areas, U.S. EPA guidance recommends modeling past time periods that meet a number of specific criteria, such as typifying the meteorological conditions that correspond to high O_3 days as defined by the MD8A greater than the NAAQS value and focusing on the ten highest modeled O_3 days (Environmental Protection Agency, 2007; Environmental Protection Agency, 2014). Regulatory modeling in the SJV (Visalia, SEQ1, and SEQ2 are included in this attainment demonstration) is more comprehensive, as it was recently updated to span the full O_3 season (defined as May–September); still potential reductions (known as relative reduction factors, RRFs) are based on the MD8A and restricted to high O_3 days (San Joaquin Valley Air Pollution Control District, 2007; San Joaquin Valley Air Pollution Control District, 2014). In the SJV, high O_3 days are most frequent in the late summer (O_3 season) and on the hottest days of the year (Pusede and Cohen, 2012). Even in SEQ1 and SEQ2, days with MD8A > 70.4 ppb are far more common in the summer. Because of chemical and meteorological differences between seasons, this may lead to policies not optimized to decrease O_3 in cooler springtime conditions, which in the SJV are more NO_x-suppressed and therefore more sensitive to controls on reactive organic compounds (Pusede et al., 2014). In addition, we observe greater year-to-year O_3 variability in the springtime than during O_3 season (Figure 6), suggestive of a larger relative role of interannual meteorological variability controlling O_3. Deeper cuts in emissions would be required in the springtime, as decreases in anthropogenic emissions have a proportionally smaller effect on the total O_3 abundance than during O_3 season."

25) P10,L15: Regarding the following "Third, aircraft observations collected in the direction of daytime upslope flow from the SJV to Sierra Nevada foothills reveal substantial decreases in NOx concentrations relative to isoprene, a key contributor to

total organic reactivity (e.g., Beaver et al., 2012).” You are consding the 2001-2012 time frame, how representative is this single day? Can this be put in to greater context?

We have added this text:

Page 6, Lines 26–31: “While these data were collected on one day in a different year from our study, the relative pattern of NO_x to organic compound emissions is likely representative, as there have been no substantial changes in the locations of urban NO_x and biogenic organic emitters. This NO_x to organic compound gradient is consistent with observations over longer sampling periods downwind of the Central California city of Sacramento, where the NO_x-enriched Sacramento urban plume is transported up the western slope of the vegetated Sierra Nevada Mountains (e.g., Beaver et al., 2012; Dillion et la., 2002; Murphy et al., 2006).”

26) P10, L18: For the following sentence: “This implies that PO_3 in Visalia and SNP is differently sensitive to emission controls, with SNP more responsive to NO_x emissions control than Visalia.” This is only one aspect of the issue, the other is that you are sitting in a source region, so the regime you are in is different; also, there is mixing and dilution that occur with transport, so this is another major factor - it’s not simply response to emissions controls. This needs to be addressed and put into context.

We have changed the text as follows:

Page 11, Lines 11–14: “Distinct local PO_3 regimes lead to PO_3 chemistry in Visalia and SNP that is differently sensitive to emission controls, with NO_x-limited SNP historically more responsive to NO_x emission control than Visalia. SNP NO_x-limitation is enhanced by NO_x dilution during transport, which further decreases NO_x relative to the abundance of local organic compounds.”

27) P11, L15-16: “. . .day due the mixing. . .” please fix this sentence, and it would be best not to use due to. . .

Corrected:

Page 13, Line 31: “…which results from the mixing…”

28) As it currently stands, the data disseminated in the tables is not very useful, especially Table 2. What would be better to provide in Table 2 are the projected DM8HA values in ppb and the W126 values in ppm hrs, along with their corresponding #s of exceedances per year. However, the method used for this work is not suitable for providing any type of reasonable predicted value. As for Table 1, actual values should be included along with the percent change.

Table 1 has been updated and Figure 6 added to show actual values. Table 2 has been deleted.

In summary, before this paper is worthy of being published, there are significant issues that must be addressed.

On the effect of upwind emission controls on ozone in Sequoia National Park

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Abstract. Ozone (O_3) air pollution in Sequoia National Park (SNP) ~~experiences~~is among the worst ~~ozone (O_3) pollution~~ of any national park in the U.S. SNP is located on the western slope of the Sierra Nevada Mountains, downwind of the San Joaquin Valley (SJV), which is home to numerous cities ranked ~~among~~in the top ten most O_3-polluted in the U.S. Here, we investigate the influence of emission controls in the ~~directly~~ upwind SJV city of Visalia on O_3 concentrations in SNP over a 12-yr time period (2001–2012). We show that export of nitrogen oxides (NO_x) from the SJV ~~plays~~has played a larger role in driving high O_3 in SNP than ~~does~~ transport of O_3. As a result, O_3 in SNP has been more responsive to NO_x emission reductions ~~as~~at a ~~function of increasing downwind distance from~~higher elevation monitoring station than at a site nearer to the SJV. We report O_3 trends by various concentration metrics~~,~~ but do so separately for when environmental conditions are conducive to plant O_3 uptake and for when high O_3 is most common, which are time periods that occur at different times of day and year. We find that precursor emission controls have been less effective at reducing O_3 concentrations in SNP in springtime, which is when plant O_3 uptake in Sierra Nevada forests has been previously measured to be greatest. We discuss the implications of regulatory focus on high O_3 days in SJV cities on O_3 concentration trends and ecosystem impacts in SNP.

1 Introduction

Sequoia National Park (SNP) is a unique and treasured ecosystem that is also one of the most ozone-polluted national ~~park~~parks in the U.S. (~~Meyer and Esperanza, 2016).~~National Park Service, 2015a). Ozone (O_3) concentrations in SNP exceeded the current U.S. human health-based ~~8-h~~ O_3 National Ambient Air Quality Standard (NAAQS), defined as 8-h maximum daily average (MD8A) O_3 greater than 70 ppb, on an average of 119 days per year over the time period 2001–2012. At the same time, there were ~~on~~an average of 76 ~~8-h NAAQS exceedances~~days per year with MD8A O_3 greater than 70 ppb in Los Angeles,

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California, 36 per year in Denver, Colorado, and 55 per year in Phoenix, Arizona, cities which are three of the most O_3-polluted in the U.S. ~~cities~~ (American Lung Association, 2016).

While O_3 is harmful to humans, it is also damaging to plants and ecosystems (e.g., Reich, 1987), with visible O_3 injury observed in many forests across the U.S. (Costonis, 1970; Pronos and Vogler, 1981; Ashmore, 2005), including in SNP (Peterson et al., 1987; Peterson et al., 1991; Patterson and Rundel, 1995; Grulke et al., 1996; National Park Service, 2013). O_3 exposure also causes a variety of other effects such as decreased plant growth (Wittig et al., 2009), reduced photosynthesis and disrupted carbon assimilation (Wittig et al., 2007; Fares et al., 2013), diminished ecosystem gross and net primary productivity (Ainsworth et al., 2012; Wittig et al., 2009), modified plant resource allocation (Ashmore, 2005), and impaired stomatal response (Paoletti and Grulke, 2010; Hoshika et al., 2014). On multi-decadal timescales, O_3-resistant plants may thrive over O_3-sensitive species, and these system-level dynamics ~~that~~ would maintain forest productivity and carbon storage~~,~~ but would induce changes in ecosystem composition (Wang et al., 2016).

SNP is home to more than 1,550 plant taxa with numerous plant species found nowhere else on Earth (Schwartz et al., 2013). One endemic species is the giant sequoia (*Sequoiadendron giganteum*), the largest living tree in the world. Large-tree ecosystems like SNP have been shown to be more sensitive to perturbation (Lutz et al., 2012) because ecological functions are provided primarily by a few large trees, rather than many smaller species. Large-diameter trees disproportionately influence patterns of tree regeneration and forest succession (Keeton and Franklin, 2005), carbon and nutrient storage, forest structure and fuel deposition at death, arboreal wildlife habitats and epiphyte communities (Lutz et al., 2012), and water storage (Sillett and Pelt, 2007), which is of critical importance in drought-prone SNP. While mature sequoias are relatively resistant to O_3, seedlings are sensitive, and high O_3 has been demonstrated to cause both visible injury and altered plant-atmosphere light and gas exchange (Miller et al., 1994). Giant sequoias grow in mixed-conifer groves with companion species ponderosa pine (*Pinus ponderosa*) and Jeffrey pine (*Pinus jeffreyi*). O_3 impacts on these pines have been documented for decades in SNP (Duriscoe, 1987; Pronos and Vogler, 1981) and include early needle loss, reduced growth, decreased photosynthesis, and lowered annual ring width (Peterson et al., 1987; Peterson et al., 1991).

SNP is located in Central California on the western slope of the Sierra Nevada Mountains downwind of the O_3-polluted San Joaquin Valley (SJV) (Figure 1). Previous model estimates of a pollution episode in August 1990 suggest at least half of peak daytime O_3 in SNP is produced upwind from anthropogenic precursors (Jacobson, 2001). For the past two decades, regulations have reduced O_3 concentrations in the SJV (Pusede and Cohen, 2012). For example, in Fresno, high O_3 days, defined as days ~~exceeding~~when the MD8A exceeded 70 ppb ~~8-h O_3 NAAQS~~, were 50% less frequent in 2007–2010 than ten years earlier (on high temperature days). At the same time, in Bakersfield, high O_3 days were 15–40% less frequent (on high temperature days). NO_x emission controls contributed to these decreases (Pusede and Cohen, 2012), with summertime (April–October) daytime (10 am–3 pm local time, LT) nitrogen dioxide (NO_2) concentrations falling by 50% from 2001 to 2012, changing linearly by –0.5 ppb yr^{-1} in the SJV city of Visalia. The precursor reductions ~~in precursor emissions~~ that brought about these decreases in high O_3 are likely to have also affected O_3 in SNP.

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The success of O_3 regulatory strategies ~~is generally~~can be measured through attainment of human health-based NAAQS ~~rather than~~and ecosystem-impact metrics. ~~While~~However, while there is a secondary ~~standard~~NAAQS requirement aimed at vegetation protection, it has historically ~~been~~used the same metric (MD8A O_3) and been set at the same threshold as the primary NAAQS (Environmental Protection Agency, 2016). Plants and ecosystems have been shown to be sensitive to lower O_3 concentrations, over longer-term exposures, and at different times of day and year than when NAAQS exceedances are frequent (e.g., Kurpius et al., 2002; Panek 2004; Panek and Ustin, 2005; Fares et al., 2013). The U.S. Environmental Protection Agency (EPA) has considered redefining the secondary standard to reflect ecological systems, with the W126 metric put forth (Environmental Protection Agency, 2010). ~~W126 is a 12-h daily summation weighted to emphasize higher O_3 concentrations~~W126 is a 12-h daily 3-month summation weighted to emphasize higher O_3 concentrations (Environmental Protection Agency, 2006; Environmental Protection Agency, 2016) that is used by the U.S. National Park Service. There are a number of other concentration metrics used to quantify ecosystem O_3 impacts. In Europe, the AOT40 index is common, and is equal to all daytime (defined as solar radiation ≥ 50 W m^{-2}) hourly O_3 concentrations greater than 40 ppb. In the U.S., two widely used indices are the SUM0 and SUM06 (Panek et al., 2002), which are the sum of all daytime hourly O_3 mixing ratios greater than or equal to 0 ppb and 60 ppb, respectively.

Even ecosystem-based concentration metrics are proxies of variable quality for O_3 impacts, if O_3 concentrations are not well-~~-~~correlated with plant O_3 uptake (e.g., Emberson et al., 2000; Panek et al., 2002; Panek, 2004; Fares et al., 2010a). This is because of temporal mismatches between when O_3 is high and when plants uptake O_3 from the atmosphere, with differences in high O_3 and efficient O_3 uptake occurring on both diurnal and seasonal timescales. While ecosystem O_3 impacts are best represented by direct measurements of the O_3 stomatal flux (e.g., Musselman et al., 2006; Fares et al., 2010a; Fares et al., 2010b), exceedances of flux-based standards are difficult to operationalize, as there are few long-term O_3 flux observational records and because reported thresholds, when available, are highly species-specific (Mills et al., 2011).

Under the 1977 Clean Air Act Amendments, selected national parks were designated as Class I Federal areas and, as part of this, the National Park Service began measuring O_3 concentrations in the ~~late~~ 1980s, prioritizing national parks downwind of cities and polluted areas, including SNP (National Park Service, ~~2015a~~2015b). Data from these monitors can be used to compute various O_3 concentration metrics; however, direct flux measurements do not exist in SNP, or other national parks, over long enough timescales to assess the effects of multi-year emissions controls. Forest survey data, which assess O_3 impacts by monitoring changes in plants and forests from visible injury records and species population estimates, are limited, as they are labor- and time-~~-~~intensive, requiring the evaluation of at least dozens of trees per stand to distinguish moderate levels of injury (Duriscoe et al., 1996). These studies occur at some time interval after exposure, making correlation to specific O_3 concentrations not possible. As a result, there is a need to assess trends using concentration metrics, but to do so with knowledge of when plant O_3 uptake is greatest.

In this paper, we report O_3 trends from 2001 to 2012 in SNP and the upwind SJV city of Visalia to study the ~~effect~~effects of SJV emission controls on SNP O_3. We compute trends in human health- and ecosystem-based concentration metrics separately when regional environmental conditions favor plant O_3 uptake (springtime) and when high O_3 is most frequent (O_3

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season). We describe these O_3 changes in Visalia and SNP as function of distance downwind of Visalia~~.~~ by way of data collected at two monitoring stations located on the western slope of the Sierra Nevada Mountains. We demonstrate the importance of transport of urban NO_x from the SJV on trends in O_3 production (PO_3) chemistry in SNP. Finally, we discuss the descriptive power of various O_3 metrics~~, considering the~~ and consider implications of a regulatory focus on human health-based standards to ~~improve O_3 air pollution and to~~ reduce ecosystem O_3 impacts in SNP ~~and polluted downwind ecosystems more broadly~~.

2 Sequoia National Park (SNP) and the San Joaquin Valley (SJV)

SNP is located in the southern Sierra Nevada Mountains (Figure 1) and is part of the largest continuous wilderness in the contiguous U.S., which includes Kings Canyon NP and Yosemite NP. The SJV extends 250 miles in length and is situated between the Southern Coast Ranges to the west, the Sierra Nevada Mountains to the east, and the Tehachapi Mountains to the south. The southern SJV is the most productive agricultural region in the U.S., an oil and gas development area, and home to the cities of Fresno, Visalia, and Bakersfield. The same climatic conditions that support agriculture in the region, especially the numerous sunny days, are also favorable for PO_3. The high rates of local PO_3 (Pusede and Cohen, 2012; Pusede et al., 2014), diverse local emission sources outside historical regulatory focus, e.g., agricultural and energy development activities (e.g., Gentner et al., 2014a; Gentner et al., 2014b; Pusede and Cohen, 2012; Park et al., 2013), and surrounding mountain ranges that impede air flow out of the valley, have resulted in severe regional O_3 pollution. Four SJV cities rank among the ten most-O_3-polluted cities in the U.S.: Bakersfield (ranked 2), Fresno (3), Visalia (4) and Modesto-Merced (6) (American Lung Association, 2016).

Multiple airflow patterns influence O_3 in SNP and the SJV (see Zhong et al~~.,~~. (2004) for a diagram). First, summertime (April–October) afternoon low-level winds in the southern SJV are generally from the west-northwest (represented by Visalia in Figure ~~2~~2a). These winds are strengthened by an extended land-sea breeze, with onshore flow entering central California through the Carquinez Strait near the San Francisco Bay and diverging to the south into the SJV and north to the Sacramento Valley (e.g., Zaremba and Carroll, 1999; Dillon et al., 2002; Beaver and Palazoglu, 2009; Bianco et al., 2011). Second, at night, a recurring local flow pattern in the SJV, known as the Fresno eddy, recirculates air in the southern region of the valley around Bakersfield in the counterclockwise direction back to Fresno and Visalia, further enhancing O_3 pollution and precursors in these cities (e.g., Ewell et al., 1989; Beaver and Palazoglu, 2009). Third, the most populous and O_3~~-~~polluted cities in the southern SJV, Fresno, Visalia, and Bakersfield, are located along the eastern valley edge. Here, air movement is also affected by mountain-valley flow (e.g., Lamanna and Goldstein 1999; Zhong et al., 2004; Trousdell et al., 2016). During the day, thermally-driven upslope flow brings air from the valley floor to higher mountain elevations from the west-southwest (Figure 2). In Figure 3, a high elevation SNP site (Moro Rock, 36.5469 N, 118.7656 W, 2050 m ASL) is visibly above the SJV surface layer in the late morning, but within this polluted layer in late afternoon. At night, the direction of flow reverses and air moves downslope from the east-northeast (Figure 2). ~~Due to the~~The prevalence of shallow nighttime surface inversions~~,~~ in the SJV

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means that evening downslope valley flow at higher elevations may be stored within nocturnal residual layers and entrained into the surface layer the following morning.

3 Results

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High O_3 days are most frequent in SNP and the SJV in the summer ~~and~~through early fall (Pusede and Cohen, 2012; Meyer and Esperanza, 2016), as PO_3 chemistry is ~~strongly~~often temperature-dependent (reviewed in Pusede et al., 2015~~),~~) and this effect is particularly strong in the SJV (Pusede and Cohen, 2012; Pusede et al., 2014). The O_3 season is defined here as June–October and 90% of annual O_3 8-h ~~O_3~~ NAAQS exceedances in SNP occur during O_3 season (2001–2012).

In the Sierra Nevada foothills, high rates of plant O_3 uptake are asynchronous with O_3 season ~~due to~~because of the Mediterranean climate (e.g., Kurpius et al., 2002; Kurpius et al., 2003; Panek, 2004). ~~Because plants~~Plants also capture carbon dioxide required for photosynthesis and transpire through stomata; therefore, O_3 uptake is not only a function of the atmospheric O_3 concentration, but also of photosynthetically-active radiation (PAR), the inverse of the atmospheric vapour pressure deficit (VPD) (Kavassalis and Murphy, 2017), and soil moisture (e.g., Reich, 1987; Bauer et al., 2000; Fares et al., 2013). In SNP, PAR is highest in the late spring through early fall and VPD is at a minimum in winter and spring. In the Sierra Nevada Mountains, plant water status (VPD and soil moisture) has been shown to explain up to 80% of day-to-day variability in stomatal conductance, with conductance decreasing with increasing water stress from mid-May to September and remaining low until soils are resaturated by wintertime precipitation. Plant O_3 uptake in Sierra Nevada forests has been reported to be greatest in April–May (Kurpius et al., 2002; Panek, 2004; Panek and Ustin, 2005).

In this context, we separately consider O_3 trends in springtime (April–May), which is when plant O_3 uptake best correlates with variability in atmospheric O_3 concentrations in the region, and during O_3 season (June–October), which is when O_3 concentrations are highest. In this manuscript, for clarity we generally use the term *impacts* when discussing ecosystem metrics and *concentrations* when talking about human health metrics; O_3 ecosystem and human health effects are of course both O_3 impacts.

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3.1 Diurnal O_3 variability

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Diurnal O_3 and O_x ($O_x \equiv O_3 + NO_2$) concentrations are shown in Figure 4 in springtime (panel a) and O_3 season (panel b) ~~from~~over the 2001–2012~~.~~ time period. Hourly O_3 data in SNP are collected at two monitoring stations, a lower elevation site, SNP-Ash Mountain (36.489 N, 118.829 W), at 515 m above sea level (ASL) and a higher elevation site, SNP-Lower Kaweah (36.566 N, 118.778 W), at 1926 m ASL (Figure 1). We refer to these stations as SEQ1 and SEQ2, respectively. O_3 and NO_2 data are measured in Visalia (36.333 N, 119.291 W), ~~directly~~ which is in the upwind direction of SNP at 102 m ASL~~. All~~ (Figure 2). The data are ~~provided~~collected by various agencies, including the National Park Service, and are hosted by the California Air Resources Board and ~~are~~ available for download at https://www.arb.ca.gov/aqmis2/aqdselect.php. In Figure 4, Visalia data are shown as O_x to account for the portion of O_3 stored as NO_2, which can be substantial in the nearfield of fresh

NO_x emissions and at night. NO_2 data are not available in SEQ1 and SEQ2; however, these sites are removed from large NO_x sources (Figure 1) and $O_3 \approx O_x$ is a reasonable approximation.

In Visalia, O_x concentrations increase sharply beginning in early morning (5 am LT) until 2 pm LT, continuing to rise slightly until 4–5 pm LT (Figure 4). This diurnal pattern reflects a combination of local PO_3 (the initial rise) and advection of O_x from the upwind source region (late afternoon maximum). In the morning (8 am LT) 30–40% of O_x is NO_2 and at 12 pm LT ~10% of O_x is NO_2.

Diurnal O_3 variability at SEQ1 and SEQ2 is characterized by two features, an early morning rise (6 am LT) and an increase in the late afternoon (3–4 pm LT). The timing of this morning O_3 increase is consistent with entrainment of O_3 in nocturnal residual layers aloft during morning boundary layer growth. The influence is substantial, as morning O_3 accounts for 50% (springtime and O_3 season) of the daily change in O_3 at SEQ1 and 50% (springtime) and 37% (O_3 season) of the daily change in O_3 at SEQ2. The timing of afternoon peak O_3 is consistent with upslope air transport from the SJV (~~Figures~~Figure 2). If O_3 attributed to local PO_3 in Visalia is greatest around 2 pm LT, typical of many urban locations, ~~then~~ with mean winds at SEQ1 of 3 m s^{-1} and SEQ2 of 2 m s^{-1}, we expect O_3 to peak in SEQ1 at ~5 pm (45 km downwind of Visalia) and at SEQ2 shortly after (9.~~6~~7 km downwind of SEQ1~~),~~, which includes the change in elevation using the Pythagorean theorem). This is ~~generally~~broadly what we observe. ~~Data in Figure 4 are averaged over 2001–2012.~~ While the actual distance of airflow is dictated by the mountain terrain and ~~there~~a parcel of air will travel a distance longer than the straight-line path on a smooth surface, the timing of the O_3 diurnal patterns is consistent with airflow travel time roughly equal to that determined by the horizontal distance and mean wind speed. There has been no change in the hour of peak O_3 mixing ratio at either ~~SNP site from~~SEQ1 or SEQ2 over the 2001– to 2012 period.

3.2 Weekday-weekend O_3 variability

SNP and the SJV are in close geographic proximity but their local PO_3 regimes are different. In 2016, as part of the Korea-U.S. Air Quality (KORUS-AQ) experiment (https://www-air.larc.nasa.gov/missions/korus-aq/index.html) and Student Airborne Research Program (SARP), the NASA DC-8 sampled a low-altitude transect (~130 m above ground level) along the trajectory of SJV mountain-valley outflow. The DC-8 flew at ~10 am LT from Orange Cove, an SJV town 35 km north of Visalia, 24 km up the western slope of the Sierra Nevada Mountains to an elevation of ~1000 m ASL. In Figure 5, the change in NO_x and isoprene along this transect is shown as a function of change in surface elevation. Boundary layer NO_x is observed to decrease with increasing distance downwind of the SJV, while isoprene concentrations increase. Isoprene is a large source of reactivity in the Sierra Nevada foothills (e.g., Beaver et al., 2012~~) and the combined NO_x and isoprene gradients suggest distinct PO_3 regimes in the SJV and SNP.~~; Dreyfus et al., 2002) and the combined NO_x and isoprene gradients suggest potentially distinct PO_3 regimes in the SJV and SNP. While these data were collected on one day in a different year from our study, the relative pattern of NO_x to organic compound emissions is likely representative, as there have been no substantial changes in the locations of urban NO_x and biogenic organic emitters. This NO_x to organic compound gradient is consistent with observations over longer sampling periods downwind of the Central California city of Sacramento, where the NO_x-

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enriched Sacramento urban plume is transported up the western slope of the vegetated Sierra Nevada Mountains (e.g., Beaver et al., 2012; Dillion et la., 2002; Murphy et al., 2006).

If the major source of O_3 in SNP is O_3 produced in the SJV and transported downwind, then the observed NO_x dependence of PO_3 in SNP and the SJV would be the same even if PO_3 regimes in the two locations were different. To test this hypothesis, we consider O_3 in SNP and O_x in the SJV separately on weekdays and weekends. Weekday-weekend NO_x concentration differences are well-documented across the U.S. (e.g., Russell et al., 2012) and California (e.g., Marr and Harley, 2002; Russell et al., 2010), and are caused by reduced weekend heavy-duty diesel truck traffic, where heavy-duty diesel trucks are large sources of NO_x but not O_3~~-~~-forming organic gases. As a result, NO_x concentrations are typically 30–60% lower on weekends than weekdays and these NO_x changes occur without comparably large decreases in reactive organic compounds (e.g., Pusede et al., 2014). PO_3 is the only term in the O_3 derivative expected to exhibit NO_x dependence.

We focus on the earliest 3-yr time period in our record, 2001–2003, which is when differences in PO_3 chemical sensitivity in the SJV and SNP are expected to be most pronounced (Pusede and Cohen, 2012). We define weekdays as Tuesdays–Fridays and weekends as Sundays to avoid atmospheric memory effects. Statistics were sufficient to minimize any co-occurring variation in meteorology, with no significant weekday-weekend differences observed in daily maximum temperature, wind speed, or wind direction. We focus on afternoon (12–6 pm LT) O_x, when O_3 concentrations in SNP are most influenced by the SJV~~.~~ (from Figure 4). We also compare weekday-weekend O_x at high and moderate temperatures, with temperature regimes defined as days above and below the 2001–2012 seasonal mean daily maximum average temperature in Visalia. Temperatures in Visalia are well correlated ($R^2 = 0.98$) with temperatures in SEQ1 over 2001–2012. During springtime and O_3 season, mean maximum average temperatures in Visalia were 25.1 ± 5.9 and 32.0 ± 5.3 °C (ranges are 1σ variability), respectively.

At high temperatures, weekday-weekend differences in O_x in Visalia and O_3 at SEQ1 and SEQ2 were not statistically distinct in either springtime or during O_3 season. Averaged across sites, percent differences in weekdays and weekends (relative to weekends) were 9.4 ± 5.4% in the springtime and 4.1 ± 2.4% during O_3 season, with greater weekday concentrations implying NO_x-limited chemistry. Errors are the average standard errors of the 3-yr means.

At moderate temperatures, statistically significant weekday-weekend differences were observed. During O_3 season, O_x was 6.3 ± 3.5% higher on weekends than weekdays in Visalia, indicating local PO_3 was NO_x suppressed. At the same time, O_3 was 4.6 ± 3.3% and 4.9 ± 3.9% higher on weekdays than weekends at SEQ1 and SEQ2, respectively, implying PO_3 in SNP was NO_x limited. A similar pattern was observed ~~at moderate temperatures~~ during springtime~~,~~ as O_x was 7.4 ± 4.6% higher on weekends than weekdays in Visalia and O_3 was 3.5 ± 7.4% and 4.7 ± 5.5% higher on weekdays than weekends in SEQ1 and SEQ2. These weekday-weekend patterns indicate a substantial portion of O_3 in SNP is produced by low-NO_x PO_3 chemistry during air transport from the SJV. At high temperatures, PO_3 during upslope transport is ~~likely occurring, but is~~ not apparent by this method because PO_3 is also NO_x limited in Visalia.

3.3 ~~Interannual~~ O_3 ~~variability~~trends over time

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In Figure 6 and Table 1, we report 12-yr O_3 trends (2001–2012) in SNP and the SJV in springtime and during O_3 season using four concentration metrics: ~~8-h O_3 NAAQS~~MD8A; two common vegetative-based indices, SUM0 and W126; and a morning average metric. ~~Trends are represented as the percent change from 2001 through 2012 divided by the fit value in SEQ1 in 2001 (the highest O_3 observed for each metric).~~

The ~~8-h~~MD8A O_3 ~~NAAQS~~ is a human health-based metric computed as the maximum unweighted daily 8-h average O_3 mixing ratio. A region is classified as in nonattainment of the NAAQS when the fourth-highest MD8A O_3 over a 3-yr period, known as the design value, exceeds a given standard. In this work, we utilize the seasonal mean MD8A and discuss O_3 exceedances as individual days in which MD8A O_3 > 70.4 ppb, the current 8-h NAAQS. SUM0 is equal to the sum of hourly O_3 concentrations over a 12-h daylight period (8 am–8 pm LT~~).~~), as opposed to SUM06, which is limited to hourly O_3 mixing ratios greater than 60 ppb. SUM0 is based on the assumption that the total O_3 dose has a greater impact on plants than shorter duration high O_3 exposures (Kurpius et al., 2002). The summation is unweighted, attributing equal significance to high and low O_3 concentrations (Musselman et al., 2006). SUM0 averaging is restricted to time periods when stomata are open (daylight), a condition not required for the ~~8-h O_3 NAAQS. In our SUM0 calculation, we only include days in which there were at least 11 hourly daytime measurements.~~MD8A. W126 is a weighted summation (8 am–8 pm LT), assuming higher O_3 is more damaging to plants than lower O_3 levels. W126 weighting is sigmoidal, with hourly O_3 weights equal to $(1 + 4403e^{-126[O3]})^{-1}$ ~~(~~(U.S. ~~U.S.~~ Environmental Protection Agency, 2015~~). We have followed the protocol for computing W126, which replaces missing data with the minimum measured concentration in the 8 am–8 pm LT time window. We also~~). Here, SUM0 and W126 summations are computed following the W126 protocol (Environmental Protection Agency, 2016), affording straightforward comparisons between the metrics. First, in months with less than 75% of hourly data coverage in the 8 am–8 pm LT window, missing values are replaced with the lowest observed hourly measurement over the study period (i.e. April–October) only until the dataset is 75% complete. Second, monthly summations of daily indices, comprised of hourly data (8 am–7 pm), are computed; when data are missing, the summation is divided by the data completeness fraction. Consecutive 3-month metrics are computed by adding monthly indices. In practice, SUM0 and W126 are computed as 3-yr averages of the highest 3-month summation; however, we define springtime SUM0 and W126 as the 3-month summation over April–June and O_3 season SUM0 and W126 as the mean of the 3-month summations over June–August, July–September, and August–October (not the highest of the three 3-month sums). Because less than 15% of data were available for August 2008 at SEQ1, O_3 season SUM0 and W126 were computed as the mean of 3-month summations over June, July, and September, and July, September, and October only for this site and year. We compute morning (7 am–12 pm LT) trends (O_x in Visalia and O_3 in SNP), as high O_3 plant uptake rates (in the morning) and high O_3 concentrations (in the afternoon) are out of phase within daily timeframes in the Sierra Nevada Mountains. Plant O_3 uptake typically follows a pattern of rapid morning uptake, relatively constant flux through midday, and a decrease in uptake in afternoon as plants close their stomata to prevent water loss in the hot, dry afternoon (Kurpius et al., 2002; Fares et al., 2013). Efficient morning uptake occurs because plants recharge their water supply overnight, which with low morning temperatures and VPD, results in high stomatal conductance (Bauer et al., 2000). Morning uptake in the Sierra Nevada maximizes in springtime around 8 am LT (Kurpius et al., 2002; Panek and Ustin, 2005; Fares et

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al., 2013). In Figure 6, mean seasonal daily MD8A and morning metrics and cumulative SUM0 and W126 metrics are shown for Visalia, SEQ1, and SEQ2 with their fit derived using a simple linear regression. Table 1 reports both the regression slope value (right columns) and the change in O_3 relative to the O_3 season fit value in SEQ1 in 2001 reported as a percent (left columns). SEQ1 experiences the highest O_3 observed for each metric and using a standard denominator facilitates comparison between monitoring sites and between seasons.

Three patterns ~~in interannual O_3 variability~~ emerge in SNP ~~emerge (Table 1):~~O_3 trends over time: (1) O_3 decreased everywhere over the 12-yr record by all metrics in both seasons; (2) O_3 decreased at a slower rate in the springtime than during O_3 season by ~~all~~most metrics; and (3) O_3 decreased more rapidly in SNP ~~than in~~versus Visalia and ~~as a function of downwind distance.~~ at SEQ2 versus SEQ1.

Seasonal differences in O_3 trends are prominent at each site~~; for~~. For example, O_3 at SEQ1 generally decreased ~~by 40–60%~~ less in springtime than during O_3 season~~.~~ (Table 1). For context in SEQ1, during O_3 season the mean MD8A declined from 82.3 ppb (2001–2002) to 73.8 ppb (2011–2012), but in the springtime the MD8A fell from 61.7 ppb (2001–2002) to 55.6 ppb (2011–2012). SUM0 O_3 fell from 87.0 ppm h (2001–2002) to 79.0 ppm h (2011–2012) during O_3 season and from 69.9 ppm h (2001–2002) to 61.8 ppm h (2011–2012) in the springtime. W126 O_3 decreased from 67.8 ppm h (2001–2002) to 53.7 ppm h (2011–2012) during O_3 season and from 39.8 ppm h (2001–2002) to 25.4 ppm h (2011–2012) in springtime. Morning O_3 fell from 67.1 ppb (2001–2002) to 59.6 ppb (2011–2012) during O_3 season and from 49.0 ppb (2001–2002) to 45.1 ppb (2011–2012). This pattern was not observed in one instance: SUM0 in SEQ2. Here, seasonal differences were comparable; however, mean daily indices were observed to differ, where SUM0 O_3 decreased from 0.914 ppm h (2001–2002) to 0.816 ppm h (2011–2011) during O_3 season, and, in the springtime, fell from 0.673 ppm h (2001–2002) to 0.616 ppm h (2011–2012), which amount to a change of –11% during O_3 season –8% in the springtime.

Additionally, greater O_3 decreases were ~~generally greater~~ observed at SEQ1 than Visalia and at SEQ2 compared to SEQ1. Over the 12-yr period, MD8A O_3 declined at a rate of 50% (O_3 season) and 61% (springtime) faster at SEQ1 than in Visalia, and ~~always greater~~29% (O_3 season) and 41% (springtime) faster at SEQ2 than SEQ1~~. Over~~ (based on the ~~12-yr period, the 8-h~~slopes reported in Table 1). SUM0 and W126 O_3 ~~NAAQS declined 63~~decreased 79% and 59% (O_3 season) and 38% and 54% (springtime) ~~and 46% (O_3 season) more~~faster at SEQ1 than in Visalia and ~~77~~20% and 23% (O_3 season) and 58% and 17% (springtime) ~~and 63% (O_3 season) more~~faster at SEQ2 than ~~Visalia. SUM0 and W126 O_3 decreased 86% and 52% (springtime) and 69% and 43% (O_3 season) more at SEQ1 than Visalia, and 77% and 92% (springtime) and 63% and 78% (O_3 season) more at SEQ2 than Visalia. In the morning,~~SEQ1. Morning O_x trends at SEQ1 and Visalia were similar in springtime, but ~~O_3 fell by 57~~O_x decreased 40% more rapidly at SEQ1 ~~than O_x in Visalia~~ during O_3 season~~. At SEQ2, morning O_3 decreased 38% (springtime)~~ and ~~65~~faster at SEQ2 than SEQ1 by 17% (O_3 season) ~~more than O_x in Visalia.~~and 55% (springtime). For each metric, we observe greater interannual variability relative to the net decline in springtime than during O_3 season.

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~~3.4 Past and future exceedances~~

~~High O_3 is often defined by exceedances of O_3 thresholds. To better understand the effects of regulatory strategies in SNP, we examine past trends and predict future O_3 levels in the context of protective thresholds. Currently, the human-health based 8-h O_3 NAAQS is 70 ppb and, while~~High O_3, as defined by exceedances of protective thresholds, also became less frequent over the 12-yr record. The number of days in which MD8A O_3 was greater than 70.4 ppb in 2001–2002 (averages are rounded up) was 68 yr^{-1} (O_3 season) and 15 yr^{-1} (springtime) in Visalia. In 2011–2012, the number of exceedances fell to 42 yr^{-1} (O_3 season) and 6 yr^{-1} (springtime). At SEQ1 in 2001–2002, there were 121 exceedance days yr^{-1} (O_3 season) and 21 yr^{-1} (springtime), declining in 2011–2012 to 99 yr^{-1} (O_3 season) and 10 yr^{-1} (springtime). At SEQ2 in 2001–2002, there were 103 exceedance days yr^{-1} (O_3 season) and 13 yr^{-1} (springtime). In 2011–2012, this decreased to 63 exceedance days yr^{-1} (O_3 season) and 3 yr^{-1} in 2011–2012 (springtime).

While there is no standard for SUM0, there are three time-integrated W126 protective thresholds. These are: 5–9 ppm h to protect against visible foliar injury to natural ecosystems, 7–13 ppm h to protect against growth effects to tree seedlings in natural forest stands, and 9–14 ppm h to protect against growth effects to tree seedlings in plantations, ~~with exceedances recorded over a rolling 3-month time window~~known as the 5, 7, and 9 ppm h standards (Heck and Cowling 1997). ~~In Table 2~~Rather than calculate W126 exceedances using a 3-month summation of monthly indices, we ~~show the mean number of 8-h O_3 NAAQS exceedances per year (rounded up) in 2001–2002 and 2011–2012 and~~ instead count the ~~predicted exceedances in 2021–2022 and 2031–2032 in springtime and during O_3 season. We also report the average number of days (rounded up) required to exceed the three W126 protective thresholds (5, 7, and 9 ppm h),~~number of days required for an exceedance to occur, summing daily W126 indices ~~starting with~~from the first day of the springtime ~~and O_3 season~~ (1 April ~~and 1 June) over the next three months. We only present springtime data, as the W126 metric has been shown~~). A larger number of days indicates improved air quality. We do this to ~~poorly correspond~~generate information in addition to exceedance frequency, as W126 O_3 at SEQ1 and SEQ2 is greater than all three standards in all years in both seasons. We only consider springtime, as this is when W126 is reported to better correlate with plant O_3 uptake ~~in late summer in Sierra Nevada forests~~ (Panek et al., 2002; Kurpius et al., 2002; Bauer et al., 2000). ~~Future exceedances~~At SEQ1 from 1 April in 2001–2002, 37, 41, and 45 days of O_3 accumulation reached exceedances of the 5, 7, and 9 ppm h thresholds, respectively (averages are ~~computed assuming individual daily indices continue to decline at the~~ rounded up). In 2011–2012, 3 to 13 more days were needed at SEQ1, as 40, 49, and 58 days of O_3 accumulation were required to exceed the 5, 7, and 9 ppm h thresholds. At SEQ2 from 1 April in 2001–~~2012 rate~~2002, 41, 46, and 49 days of accumulation led to exceedance of the 5, 7, and ~~are projected from~~ 9 ppm h thresholds, respectively. In 2011–2012 ~~values.~~ , 59, 65, and 73 days were required at SEQ2, or 18–24 more days.

~~In each case, we find there were fewer exceedances (8-h O_3 NAAQS) or more days until exceedance (W126) in 2011–2012 than at the start of the record, with declines generally linear over the 12-yr period. As with trends in the mean metrics, we observe greater declines at SEQ2 than SEQ1, with 24% (springtime) and 45% (O_3 season) larger reductions in the frequency of NAAQS exceedances at SEQ2 than SEQ1. For comparison, in Visalia there were 15 8-h O_3 NAAQS exceedances in 2001–~~

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~~2002 and 6 in 2011–2012 in springtime and 68 (2001–2002) and 42 (2011–2012) exceedances during O_3 season. Eighteen to 24 more days were required in 2011–2012 than in 2001–2002 for the cumulative daily W126 index to exceed the three protective thresholds at SEQ2 in springtime. Only 3–13 more days were required at SEQ1 and changes at SEQ1 were 20–36% smaller than observed at SEQ2.~~

~~If past decreases in O_3 continue over the next two decades, we predict no exceedances of the 8-h O_3 NAAQS at SEQ2 by 2021 in springtime and by 2031 during O_3 season, no exceedance of the 9-ppm h W126 threshold by 2021, and no further exceedances of 5- and 7-ppm h thresholds by 2031. O_3 reductions at past rates are not sizable enough to eliminate future exceedances of either 8-h O_3 NAAQS or W126 thresholds at SEQ1. Models suggest that W126 in the region would be well below these thresholds in the absence of anthropogenic precursor emissions (Lapina et al., 2013), implying further controls would be effective. Because PO_3 in SNP is NO_x-limited, future NO_x reductions are expected to have as large an impact on local PO_3 as past reductions. NO_x emissions should continue to decline, as there are significant controls currently in the implementation phase, including more stringent national rules on heavy-duty diesel engines (Environmental Protection Agency, 2000) combined with California Air Resources Board (CARB) diesel engine retrofit-replacement requirements (California Air Resources Board, 2008), and more stringent CARB standards for gasoline-powered vehicles (California Air Resources Board, 2012).~~

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4 Discussion

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4.1 O_3 metrics

Long-term measurements of O_3 fluxes rather than O_3 concentrations are required to fully understand the effects of upwind emission controls on ecosystem O_3 impacts. This is particularly true in Mediterranean ecosystems like SNP and under drought conditions (e.g., Panek et al., 2002), which is where and when plant O_3 uptake and high atmospheric O_3 concentrations may ~~not~~ be ~~correlated~~uncorrelated. We have based our analysis on results from years of O_3 flux data collected in forests on the ~~eastern~~western slope of the Sierra Nevada Mountains (Bauer et al., 2000; Panek and Goldstein, 2001; Panek et al., 2002; Kurpius et al., 2002; Fares et al., 2010; Fares et al., 2013); however, there are few other O_3 flux datasets that span multiyear timescales and no flux observations in SNP. In California, flux measurements suggest springtime SUM0 trends offer the most insight into trends in ecosystem O_3 impacts in SNP; that said, we find similar conclusions would be drawn regarding multiyear O_3 variability by location by assessing trends in SUM0, ~~8-h~~MD8A O_3 ~~NAAQS~~, and ~~our~~the morning O_x metric. This can be explained by the upslope-downslope air flow in our study region and is evident in SNP diurnal O_3 patterns (Figure 4), which show considerable O_3 entrained into the boundary layer in the morning. ~~As a result,~~ O_3 concentrations are strongly influenced by afternoon concentrations on the previous day. Comparable trends in morning, afternoon, and daily average O_3 would then arise under conditions of persistence, which are common in Central California, but these results ~~are unlikely to~~may not extend to other downwind ecosystems in the absence of an upslope-downslope flow pattern. ~~Dynamically~~The dynamically-driven elevated morning O_3 concentrations have important consequences for plants~~. Vegetation~~, as vegetation in SNP may be particularly vulnerable because plant O_3 uptake rates are often highest in the morning.

~~O_3 reductions predicted~~Reductions in ecosystem O_3 impacts as represented by declines in W126 are ~~almost twice~~greater than those of SUM0. We attribute this difference to the W126 weighting algorithm that makes the metric most sensitive to changes in the highest O_3. ~~A similar result was modeled using~~ Using the GEOS-Chem~~, which~~ model with a focus on national parks, Lapina et al. (2014) also found W126 was more responsive to decreases in anthropogenic emissions than daily (8 am–7 pm, LT) average O_3 concentrations ~~to decreases in anthropogenic emissions (Lapina et al., 2013).~~. With the Community Earth System Model, Val Martin et al. (2015) modeled air quality in national parks under two Representative Concentration Pathway (RCP) scenarios, computing substantially larger decreases over a 50-yr period in W126 O_3 compared to the MD8A. Considering that the SUM0 metric has been shown to best correspond to plant O_3 uptake in Sierra Nevada forests using O_3 flux observations (Panek et al., 2002~~), W126 likely provides~~) and that we observe W126 O_3 has declined at approximately twice the rate of SUM0 over 2001–2012, W126 trends may provide an overly optimistic representation of past ~~and future trends~~declines in ecosystem O_3 impacts in SNP.

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4.2 Reducing high O_3 in SNP and polluted downwind ecosystems

~~Emission controls have been less effective in SNP when plant O_3 uptake is greatest (springtime), despite comparable NO_x decreases in both seasons. This is in part because regulatory agencies prioritize attainment of the 8-h O_3 NAAQS, with controls developed using models to hindcast past high O_3 episodes and efficacy of NO_x and/or organic emissions reductions tested under conditions typifying O_3 season. In the SJV, high O_3 episodes occur on the hottest days during O_3 season (Pusede and Cohen, 2012; Pusede et al., 2014), leading to policies not optimized to decrease O_3 in cooler springtime conditions.~~

NO_x decreases have generally made greater improvements in O_3 ~~with~~ in SEQ1 than Visalia and in SEQ2 than SEQ1, a trend that corresponds to increasing distance downwind of the SJV. ~~This is because~~We attribute this to the importance of export of NO_x from the SJV ~~has a larger impact~~ on ~~SNP~~ O_3 ~~than does transport of O_3 produced~~ in ~~urban SJV~~SNP, combined with distinct PO_3 chemical regimes in SNP versus Visalia. Evidence for this is four-fold. First, O_3 at SEQ1 is greater than O_x in Visalia, at least during O_3 season, suggesting net O_3 formation as air travels from the SJV to SNP. Second, according to observations of O_x (Visalia) and O_3 (SNP) on weekdays versus weekends, PO_3 was simultaneously NO_x-suppressed in Visalia and NO_x-limited in SNP, with the weekday-weekend dependence of O_3 reflecting the chemical regime in which it is produced. Third, aircraft observations collected in the direction of daytime upslope flow from the SJV to Sierra Nevada foothills reveal substantial decreases in NO_x concentrations relative to isoprene, a key contributor to total organic reactivity (e.g., Beaver et al., 2012). Fourth, O_3 decreases (2001–2012) are observed to be greater in SNP than Visalia, and greater with increasing distance downwind. ~~This implies that PO_3 in Visalia and SNP is differently sensitive to emission controls, with SNP more responsive to NO_x emissions control than Visalia~~Distinct local PO_3 regimes lead to PO_3 chemistry in Visalia and SNP that is differently sensitive to emission controls, with NO_x-limited SNP historically more responsive to NO_x emission control than Visalia. SNP NO_x-limitation is enhanced by NO_x dilution during transport, which further decreases NO_x relative to the

Formatted: Indent: First line: 0", No widow/orphan control abundance of local organic compounds. Downwind sites usually experience PO_3 chemistry that is more NO_x-limited than in the often NO_x-suppressed (or at least more NO_x-suppressed) urban core. As a result, we expect similar location-specific O_3 trends in other ecosystems and national parks downwind of major NO_x sources like cities. However, while the extent of observed O_3 improvements in SNP follows the pattern of increasing distance downwind of Visalia with sustained NO_x emission control in the SJV (Russell et al., 2010; Pusede and Cohen, 2012), PO_3 chemistry is non-linear and the direction of location-specific trends may vary. That said, at some distance downwind this conclusion breaks down, as areas become less and less influenced by the upwind source.

~~An additional challenge to regulators is that high-elevation locations in the Sierra Nevada Mountains are also receptor sites for O_3 and O_3 precursors transported across the Pacific Ocean from east Asia (e.g.,~~ Because PO_3 in SNP is NO_x-limited, future NO_x reductions are expected to have at least as large an impact on local PO_3 as past reductions. Seasonal mean NO_2 concentrations have decreased by 58% and 53% in Visalia in springtime and O_3 season, respectively. Local NO_x emissions should continue to decline into the future, as there are significant controls currently ongoing or in the implementation phase, including more stringent national rules on heavy-duty diesel engines (Environmental Protection Agency, 2000), combined with California Air Resources Board (CARB) diesel engine retrofit-replacement requirements (California Air Resources Board, 2008), and more stringent CARB standards for gasoline-powered vehicles (California Air Resources Board, 2012). While O_3 declines near or greater than those that occurred from 2001 to 2012 are required to eliminate exceedances in SNP, modeling analysis by Lapina et al. (2014) suggests that W126 in the region would be well below these thresholds in the absence of anthropogenic precursor emissions, implying further emissions controls would be effective. Under the stringent precursor controls of RCP4.5, Val Martin et al. (2015) projected decreases of 11% and 67% for the MD8A and W126 in 2050, respectively, from the base year of 2000, with mean O_3 decreasing from 58.9 ppb (MD8A) and 45.5 ppm h (W126) in 2000 to 52.7 ppb (MD8A) and 15.1 ppm h (W126). Under the RCP8.5, smaller O_3 declines were predicted, with MD8A unchanged and W126 falling by 38% to 28.3 ppm h. Given that these scenarios represent a reasonable spread of possible future climatic conditions, Val Martin et al. (2015) suggest at least W126 will remain well above protective thresholds in 2050.

Over 2001–2012, O_3 declines have mostly been smaller in SNP when plant O_3 uptake is greatest (springtime), despite comparable NO_x decreases in both seasons. This may be in part because regulatory strategies prioritize attainment of the O_3 NAAQS in polluted urban areas like the SJV basin, where air parcels influenced by the results of these controls are then transported downwind to locations with different PO_3 chemistry. In the development of regulatory plans, agencies use models to hindcast past O_3 episodes, facilitating testing of the efficacy of specific NO_x and/or organic emissions reductions over that episode to meet the 8-h O_3 NAAQS or progress goals (Environmental Protection Agency, 2007; Environmental Protection Agency, 2014). In nonattainment areas, U.S. EPA guidance recommends modeling past time periods that meet a number of specific criteria, such as typifying the meteorological conditions that correspond to high O_3 days as defined by the MD8A greater than the NAAQS value and focusing on the ten highest modeled O_3 days (Environmental Protection Agency, 2007; Environmental Protection Agency, 2014). Regulatory modeling in the SJV (Visalia, SEQ1, and SEQ2 are included in this attainment demonstration) is more comprehensive, as it was recently updated to span the full O_3 season (defined as May–

September); still potential reductions (known as relative reduction factors, RRFs) are based on the MD8A and restricted to high O_3 days (San Joaquin Valley Air Pollution Control District, 2007; San Joaquin Valley Air Pollution Control District, 2014). In the SJV, high O_3 days are most frequent in the late summer (O_3 season) and on the hottest days of the year (Pusede and Cohen, 2012). Even in SEQ1 and SEQ2, days with MD8A > 70.4 ppb are far more common in the summer. Because of chemical and meteorological differences between seasons, this may lead to policies not optimized to decrease O_3 in cooler springtime conditions, which in the SJV are more NO_x-suppressed and therefore more sensitive to controls on reactive organic compounds (Pusede et al., 2014). In addition, we observe greater year-to-year O_3 variability in the springtime than during O_3 season (Figure 6), suggestive of a larger relative role of interannual meteorological variability controlling O_3. Deeper cuts in emissions would be required in the springtime, as decreases in anthropogenic emissions have a proportionally smaller effect on the total O_3 abundance than during O_3 season.

An additional challenge to regulators is the contribution of background O_3 concentrations to O_3 levels (Cooper et al., 2015), as natural sources produce O_3 even in the absence of anthropogenic precursor emissions, O_3 can be transported over significant distances, and O_3 concentrations are influenced by large-scale meteorological and climatic events. Multiple studies have identified an increasing trend in O_3 at rural sites (often used as a proxy for background O_3) in the western U.S., particularly in the springtime (e.g., Cooper et al., 2012, Lin et al., 2017). Parrish et al. (2017) presented observational evidence of a slowdown and reversal of this trend on the California west coast since 2000, though the reversal was stronger in the summer than springtime. Using observations and the GFDL-AM3 model, Lin et al. (2017) computed that Asian anthropogenic emissions accounted for 50% of simulated springtime O_3 increases at western U.S. rural sites, followed by rising global methane (13%) and variability in biomass burning (6%).~~Vicars and Sickman, 2001; Heald et al., 2003; Hudman et al., 2004).~~ Northern mid-latitude transport of Asian pollution to the western U.S. is strongest during March–April and weakest in the summertime (e.g., Wild and Akimoto, 2001; Liu et al., 2003; Liu et al., 2005), with high-elevation locations in the Sierra Nevada Mountains being more vulnerable to reception of Asian O_3 and O_3 precursors (e.g., Vicars and Sickman, 2001; Heald et al., 2003;~~)~~. Hudman et al~~.~~., 2004). Hudman et al. (2004) compared surface observations with GEOS-Chem-modeled O_3 enhancements in Asian pollution outflow, finding that, on average, transport events in April–May 2002 led to 8 ± 2 ppb higher ~~8-h~~MD8A O_3 concentrations at SEQ2. East Asian NO_x emissions have risen over our study window (e.g., Miyazaki et al., 2017), potentially causing an increase in the influence of trans-Pacific transport on O_3 concentrations at SEQ2~~,~~ and reducing the efficacy of local NO_x control in springtime. Background O_3 concentrations are also responsive to large-scale climatic events, and elevated springtime O_3 at rural sites in the western U.S. has been linked to strong La Niña winters (Lin et al., 2015; Xu et al., 2017), which are associated with an increased frequency of deep tropopause folds that entrain O_3-rich stratospheric air into the troposphere (Lin et al., 2015). Over our study period, strong La Niña events occurred during the winter of 2007–2008 and 2010–2011. In general, transport of Asian pollution and tropopause folds are expected to have a greater impact in the springtime and at the higher-elevation SEQ2. While we do observe smaller decreases in O_3 in springtime at SEQ2 than during O_3 season, interannual trends have been more downward at SEQ2 than at the lower elevation sites, SEQ1 and Visalia, in both seasons. This suggests that ~~while east-Asian pollution impacts~~these factors may impact surface O_3 at high-elevations in SNP during individual events~~,~~ (e.g., Hudman et al., 2004) but that interannual trends in seasonal averages are more influenced by chemistry during upslope outflow from the SJV~~, and, as such, O_3 can be regulated accordingly~~.

5 Conclusions

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We describe O_3 trends at two ~~locations~~monitoring stations in SNP and in the ~~upwind~~ SJV city of Visalia, which is located in the upwind direction from SNP. We show that a major portion of the O_3 concentration in SNP is formed during transport from NO_x emitted in the SJV, rather than from O_3 produced in Visalia and subsequently transported downwind. This has contributed to reductions in O_3 in SNP over the 12-yr period of 2001–2012, even while PO_3 in Visalia was NO_x suppressed. Evidence for this includes greater O_3 at SEQ1 than O_x in Visalia during O_3 season (Figure 4), distinct weekday-weekend O_3 differences in SNP and Visalia, steep gradients in NO_x and isoprene measured in the direction of upslope airflow out of the SJV within the boundary layer (Figure 5), and larger O_3 decreases over 2001–2012 ~~with increasing distance from Visalia (Table 1). NO_x emission controls in Visalia have reduced O_3 in SNP even when PO_3 in Visalia was NO_x suppressed.~~at SEQ1 versus Visalia and at SEQ2 versus SEQ1 (Table 1).

We compute interannual O_3 trends using human health- and ecosystem-based concentration metrics in springtime and O_3 season separately~~,~~ in order to distinguish between ecosystem O_3 impacts (plant O_3 uptake) and high O_3 concentrations. We find that O_3 has decreased in SNP and Visalia by all metrics in both seasons~~, but that~~ consistent with ongoing NO_x emission controls ~~have been less effective~~but observe smaller O_3 declines in springtime when plant uptake is greatest. The three metrics, MD8A, SUM0, ~~8-h O_3 NAAQS,~~ and morning O_x, all indicate comparable reductions in O_3 over 2001–2012, with decreases of ~7% (springtime) and ~13% (O_3 season) at SEQ1 and ~~∼~~13–16% (springtime) and ~~∼18~~15–19% (O_3 season) at SEQ2. We attribute similarity across these three metrics to upslope-downslope airflow at the ~~western~~eastern edge of the SJV, as morning O_x and SUM0 are strongly affected by high afternoon O_3 concentrations on the previous day ~~due~~which results from the mixing of O_3-polluted nocturnal residual layers into the surface boundary layer. Past O_3 flux ~~observations~~measurements in the region indicate the highest plant O_3 uptake in the springtime morning, therefore SNP vegetation experiences greater O_3 exposure than in locations without this memory effect. ~~W126 has predicted~~ O_3 decreases over 2001–2012 ~~that~~ computed with W126 are almost double those for SUM0~~. While~~, with the W126 emphasis of higher O_3 concentrations ~~gives~~giving the most optimistic evaluation of the efficacy of past emission controls~~, our future projections of days required to exceed W126 protective thresholds suggests that much larger decreases in NO_x emissions than took place 2001–2012 are required to eliminate O_3 impacts on vegetation in SNP over the next two decades~~.

Diurnal and seasonal mismatches between plant O_3 uptake rates and O_3 concentration-based metrics make it challenging to accurately assess vegetative O_3 damage and to quantitatively evaluate the success of regulatory action~~.~~ on ecosystems. Future work would benefit from the development of an environmentally- and biologically-relevant metric that captures patterns in plant O_3 uptake over daily and seasonal timescales, especially in Mediterranean ecosystems, where conditions conducive to plant O_3 uptake are asynchronous with conditions that lead to high O_3 concentrations.

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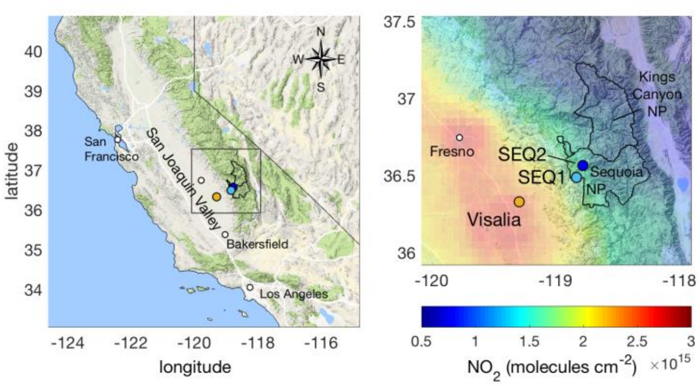

Figure 1. Map of California **(left)** with study region detail **(right)** indicating the locations of the SJV station, Visalia (orange), and two monitoring sites in SNP, SEQ1 (cyan) and SEQ2 (dark blue), with mean April–October, 2010–2012 OMI NO_2 columns using the BEHR (Berkeley High-Resolution) product (Russell et al., 2011).

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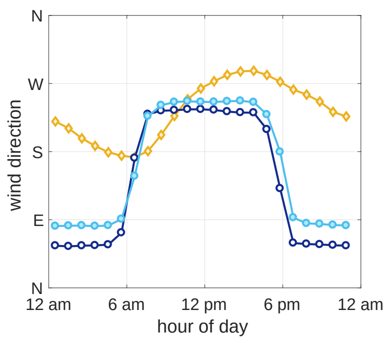
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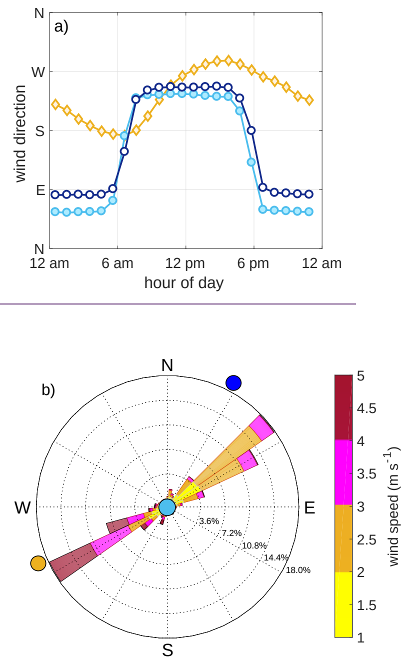
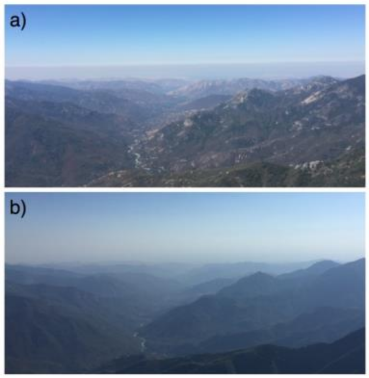


Figure 2. Hourly mean wind directions in Visalia (orange diamonds), SEQ1 (cyan filled circles), and SEQ2 (dark blue open circles) in April–October, 2001–2012 (panel a). Wind rose for SEQ1 (panel b) with the direction of the neighboring sites of Visalia (orange), SEQ1 (cyan), and SEQ2 (dark blue) indicated.



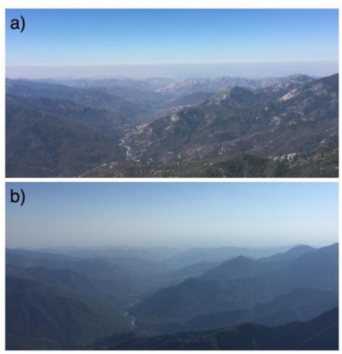


Figure 3. Looking toward the SJV from Moro Rock in SNP (36.5469 N, 118.7656 W; 2050 m ASL) at 11 am LT (panel a) and 5:30 pm LT (panel b). Photographs were taken by the authors on 29 June 2017.

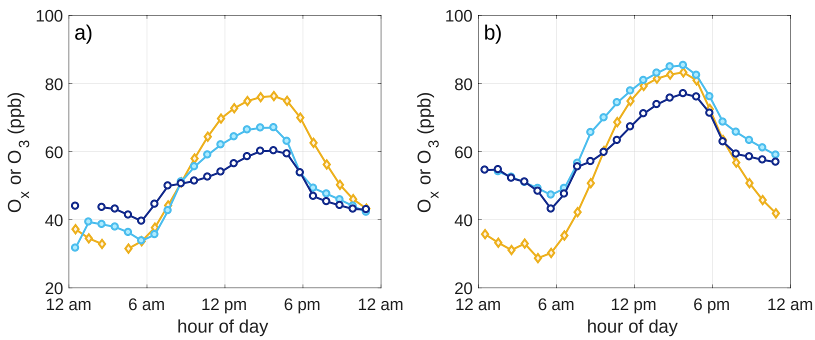


Figure 4. Hourly mean O_x in Visalia (orange diamonds), SEQ1 (cyan filled circles), and SEQ2 (dark blue open circles) in springtime (panel a) and during O_3 season (panel b) 2001–2012. Data gaps are due to routine calibrations.

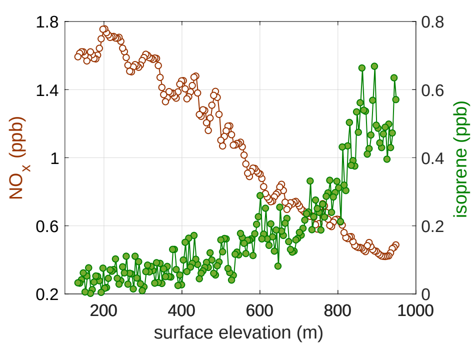


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Figure 5. NO_x (brown open circles) and isoprene (green filled circles) measured onboard the NASA DC-8 at ~10 am LT at the Sierra Nevada western slope from a mean altitude of 130 m AGL to 1000 m AGL on 19 June, 2016. The surface elevation is estimated by linearly interpolating across the total elevation change.

Table 1. ~~Percent change~~ O$_3$ changes in Visalia, SEQ1, and SEQ2 over 2001–2012 ~~in 8-h O$_3$ NAAQS~~according to MD8A, SUM0, W126, and morning O$_x$ metrics based on a linear fit of annual mean data (shown in Figure 6) in the springtime and O$_3$ season~~.~~. Each left column is the percent change with respect to fit value in ~~2000~~2001 at SEQ1 during O$_3$ season~~.~~ for comparison, which is the highest O$_3$ observed for each metric. Each right column is the fit slope with slope errors in O$_3$ abundance units per year.

O$_3$ metric	~~8-h NAAQS~~MD8A		SUM0		W126		Morning O$_x$	
O$_3$ season (June–October)								
	%	ppb y^{-1}	%	ppm h y^{-1}	%	ppm h y^{-1}	%	ppb y^{-1}
SEQ2	–19	–1.4 ± 0.41	–15	–1.2 ± 0.46	–37	–2.2 ± 0.72	–17	–1.0 ± 0.32
SEQ1	–13	–1.0 ± 0.27	–12	–0.96 ± 0.21	–28	–1.7 ± 0.36	–14	–0.83 ± 0.21
Visalia	–7	–0.54 ± 0.30	–3	–0.20 ± 0.28	–11	–0.69 ± 0.41	–6	–0.50 ± 0.30
Springtime (April–May)								
	%	ppb y^{-1}	%	ppm h y^{-1}	%	ppm h y^{-1}	%	ppb y^{-1}
SEQ2	–13	–~~13~~1.0 ± 0.38	–~~22~~16	–1.2 ± 0.47	–30	–1.8 ± 0.62	–13	–0.78 ± 0.34
SEQ1	–8	–~~7~~0.59 ± 0.42	–~~14~~6	–0.50 ± 0.53	–24	–1.5 ± 0.62	–6	–0.35 ± 0.32
Visalia	–3	–~~1~~0.23 ± 0.39	–~~8~~4	–0.31 ± 0.38	–11	–0.69 ± 0.49	–8	–0.39 ± 0.35
~~**O$_3$ season (June–October)**~~								
~~SEQ2~~	~~–19~~	~~–18~~			~~–43~~		~~–17~~	
~~SEQ1~~	~~–13~~	~~–13~~			~~–29~~		~~–14~~	
~~Visalia~~	~~–7~~	~~–4~~			~~–14~~		~~–6~~	

~~**Table 2.** Mean exceedances per year (rounded up) of the springtime and O$_3$ season 8-h O$_3$ NAAQS and days required for an exceedance of the springtime 5-, 7-, and 9-ppm h W126 protective thresholds at SEQ1 and SEQ2. Data are averaged over 2001–2002 and 2011–2012 and projected (*) for years 2021–2022 and 2031–2032 assuming past 12-yr trends continue (from 2011–2012 levels). Italicization indicates no exceedance of the W126 occurred (within 3-months) and *never* indicates the threshold would not be exceeded over the course of the year.~~

~~O$_3$ metric~~	~~8-h NAAQS~~		~~W126 (springtime)~~		
	~~Springtime~~	~~O$_3$ season~~	~~5-ppm h~~	~~7-ppm h~~	~~9-ppm h~~
~~SEQ2~~					
~~2001–2002~~	~~13~~	~~103~~	~~41~~	~~46~~	~~49~~

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~~2011–2012~~	~~3~~	~~63~~	~~59~~	~~65~~	~~73~~
~~2021–2022*~~	~~0~~	~~4~~	~~81~~	~~91~~	~~*107*~~
~~2031–2032*~~	~~0~~	~~0~~	~~*never*~~	~~*never*~~	~~*never*~~
~~**SEQ1**~~					
~~2001–2002~~	~~21~~	~~121~~	~~37~~	~~41~~	~~45~~
~~2011–2012~~	~~10~~	~~99~~	~~40~~	~~49~~	~~58~~
~~2021–2022*~~	~~6~~	~~67~~	~~48~~	~~61~~	~~68~~
~~2031–2032*~~	~~2~~	~~15~~	~~65~~	~~75~~	~~82~~

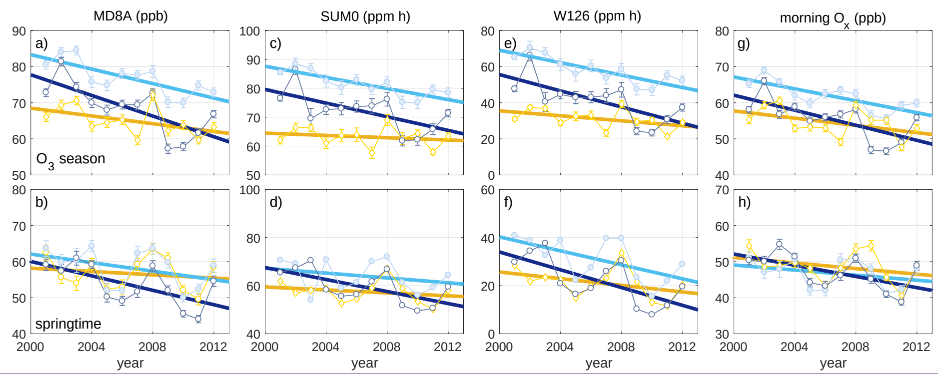

Figure 6. O_3 trends in Visalia (orange diamonds), SEQ1 (cyan filled circles), and SEQ2 (dark blue open circles) computed using MD8A (a–b), SUM0 (c–d), W126 (e–f), and morning O_x (g–h) metrics during O_3 season (top row) and springtime (bottom row). Error bars in panels a–b and g–h are standard errors of the mean. Error bars in panels c and e are standards errors of the mean of the three O_3 season 3-month summations.

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