Author response

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

Referee Comments (bold)

5 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.

10 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:

15 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 O₃ 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."

30 Nevada Mountains."

Page 8, Lines 32–33: "...O3 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."

- 5 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,
- 10 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?

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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 O₃ attributed to local *P*O₃ 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 O₃ 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 O₃ 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 O₃ 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:

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3) Figure 2: Information about mean vector flow would be more informative than mean5 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:



10 **"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.

5 **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 8hour 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 O₃ is a human health-based metric computed as the maximum unweighted daily 8-h average O₃ mixing ratio. A region is classified as in nonattainment of the NAAQS when the fourth-highest MD8A O₃ 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 O₃ exceedances as individual days in which MD8A O₃ > 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 SUMO 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–

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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 O₃ 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, O₃ 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.

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

15 **10)** Page 9, line 21: The paper tends to reserve the use of the term "impacts" for O₃ 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

20 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?

25 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."

30 **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 O₃ at SEQ1 and SEQ2 is greater than all three standards in all years in both seconds."

10 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.

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15 **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:

20 Page 13, Lines 27–29: "The three metrics, MD8A, SUM0, and morning O_x, all indicate comparable reductions in O₃ over 2001–2012, with decreases of ~7% (springtime) and ~13% (O₃ season) at SEQ1 and 13–16% (springtime) and 15–19% (O₃ season) at SEQ2."

Anonymous Referee #2

25 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

30 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 (O₃) 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

- 5 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
- 10 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.
- 15 Changed to:

Abstract, Line 1: "Ozone (O₃) 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)."

20 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.

25 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

- ⁵ 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
- 10 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
- 15 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?
- 20 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.

25 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 O₃ 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 *P*O₃ chemistry. In the development of regulatory plans, agencies use models to hindcast past O₃ episodes, facilitating testing of the efficacy of specific NO_x and/or organic emissions reductions over that episode to meet the 8-h O₃ NAAQS or progress goals (Environmental Protection Agency, 2007; Environmental
 Protection Agency, 2014). In nonattainment areas, U.S. EPA guidance recommends modeling past time
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periods that meet a number of specific criteria, such as typifying the meteorological conditions that correspond to high O₃ days as defined by the MD8A greater than the NAAQS value and focusing on the ten highest modeled O₃ 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 O₃ 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 O₃. Deeper cuts in emissions would be required in the springtime, as decreases in anthropogenic emissions have a proportionally smaller effect on the total O₃ abundance than during O₃ season."

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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.

20 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 O₃ 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 O₃ concentrations in SNP in springtime...".

5 The initial submission included discussion of trans-Pacific transport and its influence on springtime O₃ trends. We have expanded the discussion to address trends in springtime background O₃ 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 10 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, 15 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 20 al., 2005), with high-elevation locations in the Sierra Nevada Mountains being more vulnerable to reception of Asian O₃ and O₃ 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 25 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
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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 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
- 10 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.

15 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.



20 **"Figure 6.** O₃ 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 O₃ 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.

10 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)."

15 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.

20 8) P2,L7-9: Revise the following sentence – reads awkwardly: On multi-decadal timescales, O₃-resistant plants may thrive over O₃-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:

25 Page 2, Lines 7–9: "On multi-decadal timescales, O₃-resistant plants may thrive over O₃-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.

30 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:

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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
- 25 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 O₃ days are most frequent in SNP and the SJV in the summer through early fall (Pusede and Cohen, 2012; Meyer and Esperanza, 2016), as *P*O₃ 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)."

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

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

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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

10 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

15 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:

20 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

25 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. 5 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.

10 "Table 1. O₃ 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 O₃ season. Each left column is the percent change with respect to fit value in 2001 at SEQ1 during O₃ season for comparison, which is the highest O₃ observed for each metric. Each right column is the fit slope with slope errors in O₃ 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

15 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"?

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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:

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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 O₃ 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 O₃ fell from 87.0 ppm h (2001–2002) to 79.0 ppm h (2011–2012) during O₃ season and from 69.9 ppm h (2001–2002) to 61.8 ppm h (2011–2012) in the springtime. W126 O₃ decreased from 67.8 ppm h (2001–2002) to 53.7 ppm h (2011–2012) during O₃ season and from 39.8 ppm h (2001–2002) to 53.7 ppm h (2011–2012) during O₃ season and from 39.8 ppm h (2001–2002) to 25.4 ppm h (2011–2012) in springtime. Morning O₃ fell from 67.1 ppb (2001–2002) to 59.6 ppb (2011–2012) during O₃ 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 O₃ decreased from 0.914 ppm h (2001–2002) to 0.816 ppm h (2011–2011) during O₃ season, and, in the springtime, fell from 0.673 ppm h (2001–2002) to 0.616 ppm h

19) Section 3.4 You state that you "predict future O₃ 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 O₃, as defined by exceedances of protective thresholds, also became less frequent over the 12-yr record. The number of days in which MD8A O₃ was greater than 70.4 ppb in 2001–2002 (averages are rounded up) was 68 yr⁻¹ (O₃ season) and 15 yr⁻¹ (springtime) in Visalia. In 2011–2012, the number of exceedances fell to 42 yr⁻¹ (O₃ season) and 6 yr⁻¹ (springtime). At SEQ1 in 2001–2002, there were 121 exceedance days yr⁻¹ (O₃ season) and 21 yr⁻¹ (springtime), declining in 2011–2012 to 99 yr⁻¹ (O₃ season) and 10 yr⁻¹ (springtime). At SEQ2 in 2001–2002, there were 103 exceedance days yr⁻¹ (O₃ season) and 13 yr⁻¹ (springtime). In 2011–2012, this decreased to 63 exceedance days yr⁻¹ (O₃ 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 O₃ 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 O₃ 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 O₃ 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 O₃ 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.

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Page 11, Lines 21–34; Page 12, Lines 1–2: "Because PO₃ 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 O₃ 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 O₃ 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."

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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

- 25 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
- 30 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

5 **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 O₃ declines:

15 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 O₃ compared to the MD8A. Considering that the SUM0 metric has been shown to best correspond to plant O₃ uptake in Sierra Nevada forests using O₃ flux observations (Panek et al., 2002) and that we observe W126 O₃ has declined at approximately twice the rate of SUM0 over 2001–2012, W126 trends may provide an overly optimistic representation of past declines in

25 ecosystem O₃ impacts in SNP."

22) P9, L30: "O₃ 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?

30 *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: "O₃ decreases over 2001–2012 computed with W126 are almost double those for SUM0, with the W126 emphasis of higher O₃ concentrations giving the most optimistic evaluation

5 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.

- 15 Page 10, Lines 23–31: "Reductions in ecosystem O₃ 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 O₃. 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 O₃ 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 O₃ compared to the MD8A. Considering that the SUM0 metric has been shown to best correspond to plant O₃ uptake in Sierra Nevada forests using O₃ flux observations (Panek et al., 2002) and that we observe W126 O₃ 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 O₃ impacts in SNP."
 - 24) P10, L9: For the following statement: ". . .leading to policies not optimized to decrease O₃ 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
- 30

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:

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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 O₃ 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 10 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 O₃ 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 15 correspond to high O_3 days as defined by the MD8A greater than the NAAQS value and focusing on the ten highest modeled O₃ 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 20 high O₃ 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 SEO1 and SEO2, 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 25 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 O₃. Deeper cuts in emissions would be required in the springtime, as decreases in anthropogenic emissions have a proportionally smaller effect on the total O₃ abundance than during O₃ season."

30 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)."

10 Murphy et al., 2006)."

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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 PO₃ regimes lead to PO₃ chemistry in Visalia and SNP that is differently sensitive to emission controls, with NO_x-limited SNP historically more responsive to NO_x

20 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:

25 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

30 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.