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Reply to RC1 and RC2

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Author comment on "The impacts of wildfires on ozone production and boundary layer dynamics in California's Central Valley" by Keming Pan and Ian C. Faloona, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-144-AC1, 2022

Response to Reviewers of "The Impacts of Wildfires on Ozone Production and Boundary Layer Dynamics in California's Central Valley" submitted to ACP by Pan & Faloona

Referee comments reproduced below in italics and our responses below.

Response to Anonymous Referee #1:

This manuscript describes an ambitious effort to characterize the effects of wildfire smoke on photochemical ozone production and boundary layer dynamics in the Central Valley of California between 2016 and 2020. The authors use HYSPLIT and HMS mapping to identify periods influenced by wildfires, and measurements from two Ameriflux sites to evaluate the boundary layer impacts. The paper is well written and organized to integrate these two themes. The greatest weakness of the paper is with the ozone production estimates

which rely heavily on NO_2 measurements based on the conversion to NO on a heated molybdenum surface, a technique fraught with problems that the authors recognize and discuss at length. As they note, it has long been known that heated molybdenum also converts other NO_y compounds (e.g., PAN and HNO_3) to NO, and they apply the correction developed by Steinbacher et al. (2007) from measurements at two sites in Switzerland to correct for this interference. This correction can be quite large outside of urban areas, however, and the study by Steinbacher et al. found that the apparent NO_2 measured using molybdenum converters was more than 2.5x the actual value on summer afternoons at a rural site. Newer studies (e.g., Xu et al., 2013) have reached similar conclusions, and the situation is made even worse in aged wildfire smoke where there is very little actual NO_x. Recent findings from WE-CAN (Peng et al., 2021) show that nearly all of the NOx in wildfire plumes is converted to PAN or HNO_3 within a few hours which makes the inferred fire-related increases in NOx and calculated ozone production efficiencies extremely uncertain to say the least. I'm not convinced that the CARB NO_x measurements can be corrected to the degree needed for this analysis and I recommend that these sections be removed from the manuscript. The remaining material is of sufficient interest to warrant

publication.

Specific comments.

L16-18. Delete.

L115-120. Replace with "We also present the enhancement ratios (ERs) for O3/T, PM2.5/CO during the wildfire influenced periods in the CV."

L150-166. Delete.

L209-238. Section 2.3 should be deleted.

L233. Perhaps moot, but there is no Eq. R(6) in the text. Should this be "…Eq. R(3) and R(4)…"?

L275 and Figure 3. The NOx panel can be deleted.

L285-286. Delete.

L348-387 and Figure 5. Delete.

L471-472. Delete.

L477-480. Delete.

Table C1. Delete.

We thank the reviewer for their considered and thoughtful reading of our manuscript. Referee #2 also expressed concern about this issue and recommended an uncertainty analysis of our $P(O_3)$ estimates, which we have carried out in the revised manuscript. While we understand the misgivings expressed in our analysis of the flawed NO_x measurements, we stand by our choice to use this commonly available data set and argue for its retention in our analysis, after the current revision, based on three primary reasons. We have tried to change the language of the text to assimilate the inherent uncertainty of these measurements, but we believe there is still something useful that can be gleaned from them. Our reasons for respectfully requesting that the referees and editors concede to this decision are the following.

First, other extant studies in the literature already report on the NO_x influences by wildfire effluents from the same flawed measurements without addressing these complexities. Examples include Buysse et al. (2019), Ninneman & Jaffe (2021), Cai et al. (2016), and Jaffe et al. (2013). Therefore, we feel that there is an established precedent that has been set, yet a thorough discussion of the drawbacks of these measurements, and possible uncertainties, have not been discussed vis-à-vis wildfire impacts, and there is a need to call out this important point in the scientific discussion.

Second, all measurements are flawed to some extent, and to disregard the information present in the long history and ubiquity of these heated Mo catalytic chemiluminescence measurements is to discard a lot of human effort and resources that went into installing and operating these systems for decades. Thus, we believe it is a worthwhile endeavor to discuss these measurements and press them into service whenever possible even when there is admittedly considerable uncertainty, which we have now attempted to quantify.

Third, as we point out in the body of the manuscript any misidentification of wildfire-enhanced NO_z as NO_x in our analysis really just boils down to a matter of the timing of the smoke's impacts on ozone, which is uncertain in this work because of the stagnation of wildfire plumes in the weakly ventilated Central Valley. So, whether NO_x is elevated due to wildfire emissions or whether it is an interference in the measurement of a NO_z compound in the fire effluent, it still has, will, or is currently making ozone, and that fact is important. Furthermore, the reviewer points to the results of Peng et al. (2021) that suggest much combustion NO_x from wildfires is rapidly converted into PAN and nitric acid, and further propose that a large amount is likely to be further sequestered into other organic peroxynitrates. Given that the ozone impacts under study are in the valley floor where temperatures are expected to be highest (outside of the immediate vicinity of the fire itself), there is likely a fair bit of NO_x liberated from such peroxynitrates in the afternoon boundary layers and therefore contributing to in-situ (or very nearby) ozone production. Finally, the observed median enhancements of NO, an analyte that is in principle not subject to the NO_z interference of the Mo converter, are on average 0.07 ppb greater in the wildfire impacted site-days (Figure 5) which would indicate that there is in fact some additional NO_x in the aged plumes' contributions to the near surface air quality.

We would further like to clarify that the referee's comments seem to overemphasize the typical corrections reported by those who have measured the interferences of the Mo-catalyzed NO_2 chemiluminescence measurement. In their review they claim that Steinbacher et al. (2007) report a factor of 2.5 greater NO_2 measured on summer afternoons in a Swiss rural environment. However, Fig. 4 in Steinbacher et al. (2007) shows that for the Taenikon site (rural with a reasonably large sized urban area nearby, the site whose correction parameterizations we used in this study) the measurement excess is about 1.4 ppb over a true value of 5 ppb for June – Sept, amounting to an observed ratio of about 1.3, not 2.5. It is only at their remote site, Rigi, far from significant emissions where true NO_x levels are below 1-2 ppb that the ratios reach 2.5. This was also found to be the case in Xu et al. (2013) where their remote mountain site saw enhancements of up to 260%, but far from direct sources. The suburban sites reported in Xu et al. (2013) showed corrections much more in-line with what we used in this study, that is midday summertime values of 25%-40%. In general, it is found that the Mo-chemiluminescence interference is proportionally smallest in urban regions, moderate in suburban regions, and highest in remote regions. We believe that the Central Valley of California is somewhere between urban and suburban/rural in its air quality and therefore the Steinbacher et al. (2007) correction we use, which to reiterate is only derived from their semi-rural/urban site at Taenikon, not from their remote site at Rigi, is reasonably appropriate.

Having argued to retain this analysis, we now have to admit that in reviewing our analysis to determine $P(O_3)$ and its uncertainty, we found a unit conversion error in our initial calculation which led to much large values of ozone production than are actually present. Whereas the previous draft reported valley-wide average values of about 100 ppb/hr

(background) and 125 ppb/hr (under the influence of wildfire emissions), these numbers are, in fact, only 19 ppb/hr (background) and 28 ppb/hr (wildfire) and are revised in the current version of the manuscript. Furthermore, in dialogue with the Monitoring & Laboratory Division of the California Air Resources Board, we discovered that the NO_x monitor at the Sacramento site uses a photolytic NO_2 converter and therefore should not be subjected to the Mo-catalyst correction, so we leave those measurements uncorrected.

In an effort to understand the uncertainty of these $P(O_3)$ estimates, we performed an analysis calculating successive stages of the NO_2 correction, ranging from no correction (0%) to full correction (100%) illustrating the valley average (of each site's median value) at intervals of 20%. Our revised Figure 5b shows the uncertainty analysis of $P(O_3)$ as a function of each site's median NO_2 concentration separated into background and wildfire-influenced data. On average overall, the $P(O_3)$ changes linearly by about 30 ppb/h for 1 ppb of NO_2 correction, and there is no significant difference in slope between wildfire and background conditions (29.1 vs 31.0). In using the rural/suburban correction parameters reported in Steinbacher et al. (2007), the average correction of NO_2 is about 1.3 ppb (~30%) reduction, which results in about 40 ppb/h changes in $P(O_3)$. In the full correction, only one site, Bakersfield, shows a negative median value of $P(O_3)$ for background days, and the valley-wide average is 18.7 ppb/hr. While we acknowledge that this is an extremely difficult parameter to measure and Mannschreck et al. (2004) report that the photostationary state method used here is 2-3 times larger than when RO_x radicals are measured directly, we still find it instructive to summarize here the midday $P(O_3)$ values that have been reported in the literature for the Southern San Joaquin Valley (SSJV).

Study	Year of data	Average [NO_x] (ppb)	$P(O_3)$ (ppb/hr)
Brune et al. (2016)	2010 (Bakersfield)	7	[7-12]
Trousdell et al. (2016)	2013/14 (Bakersfield)	4	8.2, [4-12]
Pusede et al. (2016)	2010 (Bakersfield)	2.3 (weekend)	[12-15]
Pusede et al. (2016)	2010 (Bakersfield)	5.3 (weekday)	[10-26]

Trousdell et al. (2019)	2016 (Fresno-Visalia)	8	7, [2-14]
Ninneman & Jaffe (2021)	2018 (Bakersfield)	6	7, 12 (wildfires)

Realizing that this study found the average NO_x to be only ~ 3 ppb, and that the NO_2/NO photostationary state method is known to overestimate $P(O_3)$ by about 2.5 times, we can crudely surmise that the values we estimate should be in the range of about 10 – 20 ppb/hr, and thus the correction of Steinbacher et al. (2007) is most likely accurate to within ~ 30%. In any event, the method is not meant to be quantitative so much as relatively informative of the changing photochemical environment under the influence of wildfire emissions. Assuming that the NO_2 corrections in the presence of fire smoke are most likely larger than those used here from average conditions of Steinbacher et al. (2007), we infer that the average influence of wildfires in the Central Valley is to enhance in-situ ozone production rates by at most 50% (18.7 ppb/hr to 28.3 ppb/hr), and consider this to be an upper limit of enhancement.

Although we have retained and refined the analysis of $P(O_3)$ in the revised manuscript, we have attempted to de-emphasize the quantitative inferences made about photochemical ozone production. Nevertheless, we believe the inclusion of a summary of such estimates in the literature is still worthwhile and important for other researchers to consider.

Specific Comments:

L16 - L18. We would prefer to retain this, but subject to the revisions and qualifications as explicated above.

L115 - L120. Ibid.

L150 – L166. Ibid.

L209 – L238. Ibid

L233. Thank you, but we had that part deleted.

L275, Fig.3. We would prefer to retain this. See discussion above. We also contacted the California Air Resource Board's Monitoring and Laboratory Division and confirmed that they operate a NO_2 photolytic converter at the Sacramento site, and thus it should be much less affected by the interference of other oxidized nitrogen compounds (NO_z). Therefore, we see this fact as confirmation of the impact of elevated NOx (0.9 ppb, or 36% valley wide) when influenced by wildfire effluents.

L285-286. Ibid.

L348 – L387, We have replaced Fig. 5b with a plot of the approximate uncertainty in $P(O_3)$ with respect to different magnitudes of NO_2 corrections. We have also included an expanded discussion about the results in the literature of $P(O_3)$ estimates in the Southern San Joaquin Valley (SSJV) to give context and a sense of uncertainty in our results. As for the ozone production efficiency (OPE), we believe the Fresno site data should be kept in the work because it has not been previously reported on in the literature and gives important clues as to the ozone production regime during the warm season in the SSJV. Although the NO_2 measurements are possibly still flawed after our correction, and an under-correction of the NO_2 interference would tend to overestimate the O_x vs. NO_z slope, we believe the additional uncertainty analysis of our correction method shows that it is likely within ~50% of the proper correction (in order to sustain the valley average $P(O_3)$ above zero) and therefore should not misrepresent the actual OPE by much.

L471 – L472. We have modified this to highlight the uncertainty in our NO_x estimates.

L477 – L480. We have revised this part to include the new analysis of $P(O_3)$ and explain the resultant enhancement in wildfire conditions to be an upper limit.

Table C1. We have kept the correction as defined by Steinbacher et al. (2007) for their suburban/rural site, which is most similar to the Central Valley of California.

Response to Anonymous Referee #2:

The paper presents a detailed analysis of surface measurements and radiosone observations on the impacts of wildfire smoke on surface ozone production and boundary layer dynamics. The authors demonstrate an overall difference between fire impacted and non-fire days with the former exhibiting enhanced surface ozone concentrations and reduced buoyancy fluxes and boundary layer height. This is a very well conducted analysis and the paper clearly and in detail discusses the approaches and results. I also want to comment positively on the comprehesinve review of and reference to other studies that puts this study well into context of previous works.

I recommend publications but believe that the paper could be strengthened if the following points would be addressed:

1) I agree with the other reviewer that the uncertainties associated with the surface NO2 measurements impact the calculations of ozone production. The authors do address this to some degree but wonder if an uncertainty analysis could be conducted assuming various degrees of corrections to the data

2) The authors combine measurements from different surface sites in their analysis, e.g. near-by meteorological data when colocated data are not available at chemistry measurement locations or the joint analysis of RASS and AmeriFlux measurements. Can the authors comment on the variabilty across the study region and what uncertainties these assumptions might introduce in their results?

3) The authors briefly mention that also climatogical conditions might contribute to the differences seen between fire and no-fire days. While I understand that a full analysis of

this sich as an assessment of large scale weather patterns is beyond the scope, it would be good to elaborate on this point some more.

3) Section 2.2: Do I understand correctly that for background days only cloud-free days are considered whereas for fire impacted days both cloud-free and cloudy days are considered? On fire-impacted days one would also expect that the high aerosol loading leads to more cloud formation which, in addition to aerosol loadings, would impact dynamics.

As for the use of NAM: The study region has rather complex terrain and I wonder if the 12km resolution is sufficient to resolve the transport well enough. Have the authors considered using the higher resolution HRRR product?

4) Figure 3: it would be informative to also for this figure state the number of data points available for each bin.

5) Figure 4: Label fire/no-fire is missing

6) Line 354: Do you have a hypothesis why these two sites show negaticve increments?

7) Figures B1 and B2 appear to be of fairly low quality.

We thank the reviewer for their thoughtful suggestions on our work. We address each in the numbered responses below.

1) Our response to referee #1 includes a detailed argument as to why we think retaining the NO_x analysis, subject to revisions as suggested by referee #2 in this work, is useful in spite of its uncertainty. We performed an uncertainty analysis of $P(O_3)$ with different stages of NO_2 correction, ranging from 0% to 100% at an interval of 20%, and show these changes in Figure 5b in the revised manuscript. Overall, the $P(O_3)$ will change almost linearly by about 30 ppb/h for every 1 ppb of NO_2 correction. The slope has no significant difference between the wildfire and background conditions (29.1 vs 31.0). In our case, the average correction of NO_2 is about 1.3 ppb (~25%) reduction, which results in a 40 ppb/h decrease in $P(O_3)$. In the full correction stage (100%), only one site shows a negative median value of $P(O_3)$ for the background days. The linear regressions of the $P(O_3)$ vs

NO2 under background conditions yields an intercept with $P(O_3)=0$ at about 1.4 ppb, which likely indicates a limit for the maximum NO_2 correction physically reasonable. We report the average for the entire CV in Figure 5b and the values at each site in Fig. 5c, with the medians and standard deviations shown by the error bars to a sense of the range of $P(O_3)$.

2) We have included the coordinates of each site in Table 1, and below we also present the distance in km between air quality sites and meteorological sites (though we do not add it to the manuscript because we already present latitude and longitude). Except for Sutter Buttes, Twitchell Wetland and Vaira Ranch, the distances for other sites are within 10 km. Given that the spatial extent of overhead wildfire plumes tend to be hundreds of kilometers, we do not think this significantly impacts our analyses. Moreover, the afternoon averages considered in this study (10:00 to 15:00 PST) will be made at each site as the valley air moves past the instruments (~ 3m/s) sampling a distance of about 55 km. So fixed-station measurements are still representative of a spatial distance upwind of the detection site. For example, Trousdell et al. (2019) report a decorrelation distance in the SJV derived from aircraft to be ~18 km for potential temperature, so we expect that the met sites are close enough to be representative of the conditions at the air quality sites. Only the Sutter Buttes site provides a measurement of CO, which we use in order to supplement the Yuba City site. The CO measurements in our study only provide insights on air quality degradation during wildfire days, thus the impact of the non-co-located measurement is believed to be negligible. The Twitchell Wetland and Vaira Ranch sites provide surface flux measurements using the results of Sacramento and Stockton as the binary wildfire indicator. Although they are farther away from the air quality sites than the meteorology sites are, we believe that when wildfire smoke impacts the CV it tends to stagnate across large portions of the valley, which has a width of 75 to 120 km across. We therefore expect that the flux sites are fairly well represented by overhead smoke detected by the HMS at their corresponding air quality sites.

Site Name	Site/Airport name (ICAO)	Distance (km)
Chico-East	KCIC	4.6
Yuba City	KMYV	6.4
Yuba City	Sutter Buttes (CO)	18.8

Arden Arcade - Del Paso Manor	Vaira Ranch	42.6
Stockton – Hazelton Street	KSCK	5.9
Stockton – Hazelton Street	Twitchell Wetland	37.1
Modesto - 14th Street	KMOD	3.6
Madera-City	Madera - Pump Yard	9.1
Fresno - Garland	KFAT	5.0
Visalia - N. Church Street	KVIS*	9.3
Bakersfield - California Ave	KBFL	6.4

3) We have added a composite mean of the 500 hPa geopotential heights for wildfire and background days in Figure B5 in the supplement. As expected there does seem to be a general difference in wildfire conditions, with the climatological coastal trough more dominant in the non-wildfire days, and the high-pressure bulge of the warm Southwestern US lower troposphere more dominant across California during wildfire days. However, we agree that this is beyond the scope of the current work, and we just briefly mention this in section 3.1 in our revised manuscript.

4) (Referee #2 used the same number as last comment, the following numbered bullet points are offset by one) Yes, the background days are selected to only be cloud-free because we wanted the $j(NO_2)$ estimates to be unimpacted by cloud cover. The wildfire days are identified by using the HMS product and HYSPLIT model, so it is possible to

include cloudy days for the wildfire conditions, in so far as the satellite retrieval algorithm can discern. The cloud-free days are identified by MODIS visible band images twice per day, so it's possible that there would be cloud formation between two observation windows of the MODIS satellites and the same also goes for wildfire scenario. Nevertheless, we do not expect the additional aerosols from the wildfires to significantly enhance the cloudiness aside from local pyrocumulus perhaps. As described by Koren et al. (2008) smoke invigoration is most effective in low CCN environments such as over the ocean, whereas the Central Valley of California has ample aerosol loading (average ~10 mg m^{-3}, Fig. 3) and thus is not expected to exhibit a large change in cloudiness from the additional aerosols. Moreover, fire aerosols are also known to inhibit convection and thus cloud cover by absorbing shortwave radiation aloft and cooling the surface increasing static stability and reducing cloud cover. Nonetheless, this region is typically quite cloud free during the warm season because of the persistent subsidence on the lee of the Pacific High as well as the mesoscale subsidence over the valley due to the valley-mountain circulation (Faloona et al., 2020). Further notice the clear-sky nature of the downwelling shortwave in Figures 5d and 7.

As for the resolution of HYSPLIT, we mention in the manuscript that the diurnal flow in the CV has been fairly well-characterized in the literature (Zhong et al., 2004; Bao et al., 2008; Bianco et al., 2011; Faloona et al., 2020). Because we are not studying the long-range transport of wildfire plumes and the wildfire sources in our study are typically located near the surrounding mountain ridges of the CV, and every dynamic model will have uncertainties to some degree, we would not expect the binary indicator of wildfire (0 or 1) to change. Furthermore, the HRRR products do not cover the entire period of our study (i.e., 2016-2020), and the NAM 12km is the highest resolution that AirNow-Tech currently provides. We thus think it is best to keep the consistency of HYSPLIT for all 5-years of our analysis. Moreover, we also mentioned in section 3.1 that the CO and 24-hr PM 2.5 have distinctive differences between wildfire and background days, as identified in other studies, so we believe that our method for wildfire identification has at least comparable effectiveness as previous studies that use only CO or PM2.5 as thresholds (McClure et al., 2018; Briggs et al.,2016).

5) We added the requested information in the captions and the numbers of background days at each station in Table C2. The number of data points are the same as the number of background days or wildfire days unless there are missing data. We mention these changes in section 2.1.

6) We added the labels, thank you for bringing that to our attention.

7) We have eliminated the $k*[RO_x]$ part of our analysis entirely.

8) We are not sure why these figures reproduced poorly. We have replotted Figure B2 with smaller scatter points and will be sure to upload the higher resolution version. Figure B1 was also replotted, in which the sites are now sorted into SV and SJV. Because the SV and SJV have significant differences in background O_3 levels, we separate them for the purpose of showing that the wildfire impacts significantly increase the chances of O_3

exceedances for both valleys.

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