

Thank you to the two reviewers for the positive feedback, detailed comments and occasional corrections. Below is our response to each comment (original in black, response in red).

RC1:

The manuscript is an excellent contribution to the scientific community for this subject. However, several technical corrections should be made for clarity and consistency. These are: 1) past work should be discussed in the past tense, not present tense; 2) PM2.5 should not be referred to as PM, since they are not equivalent (this problem appears in both text and figures); 3) "tracer" and "indicator" should not be used interchangeably. The authors should make sure the three listed items (above) are corrected for the entire manuscript. I have also suggested several technical corrections (below) that could help clarify the writing. **Agree. These changes have been made in the manuscript.**

Title: change "a tracer" to "an indicator"—**I agree. This is a better title.**

Abstract:

Line 9: remove commas **Done**

Line 10: change "lower" to "low" **Done**

Line 11: PM2.5 **Done**

line 12: specify this is in regards to wildfire smoke **Done.**

Line 14: "extent" is ambiguous; temporal or spatial extent? **Reworded.**

Line 14: change to "We use observations at a regulatory surface monitor site..." **Done**

Line 15: remove "During this time," **Done**

Line 16: PM to PM2.5; change to "ratio threshold of 30" **Done**

Line 19: "in fresh smoke plumes" - provide example range of values **Values added.**

Line 23: change to "...this ratio can also help quantify..." **Done**

1.Introduction:

Line 25: change to "... smoke impacts have become more prevalent due to..." **Original preferred**

Line 30: change to "diameter less than" **Done**

Line 31: change to "...(VOCs) which include many toxic..." **Done**

Line 32: change "In addition, secondary" to "Furthermore, atmospheric" **Done**.

Line 33: change "emissions" to "pollutants" **Original preferred**

Line 34: New paragraph **Done**

Line 34: change "be transported" to "originate" **Done**

Line 40: change "markers" to "tracers" **Done**

Line 41: change to "... at surface sites and also have some anthropogenic..." **Done**

Line 43: change "anthropogenic" to something like "commercial, institutional, residential". Otherwise it is a redundant statement since industrial and vehicular are already anthropogenic **Done**

Line 43: change "in having" to "with" **Original preferred**

Line 44: remove "For the US as a whole," **Original preferred**

Line 45: change to "... emission sources in the U.S., excluding..." **Original preferred**

Line 46: change "report" to "reported" **Done**

Lines 43 to 64: this paragraph could be summarized in a table. **This is a good suggestion. The paragraph will get shortened and the information summarized in a new table. See below:**

**Table 1. Emission ratios and observed NERs for non-smoke and smoke conditions.** Emission ratios are converted into NERs using a pressure of 1 atmosphere and temperature of 272K (STP). This calculation assumes no loss of either PM<sub>2.5</sub> or CO. For observed NERs, the study mean is given and range (if known) is shown in parentheses.

	PM <sub>2.5</sub> /CO E.R. (g/g)	PM <sub>2.5</sub> /CO NER (µg m <sup>-3</sup> ppm <sup>-1</sup> )
<b>Non-smoke emissions and observed NERs</b>		
US industrial and mobile emissions (excludes wildfires and residential wood combustion) <sup>1</sup>	0.076	95
U.S. Mobile sources only <sup>1</sup>	0.009	11
Observed NERs in urban areas with no smoke <sup>2</sup>		37 (21-66)
<b>Smoke emissions and observed NERs</b>		
Temperate wildfires ERs <sup>3</sup>	.142	177
Temperate wildfires ERs <sup>4</sup>	.176 (.07-.57)	220 (87-712)
Observed smoke NERs in urban areas <sup>2</sup>		128 (57-228)
Observed smoke NERs, surface sites <sup>5</sup>		103 (120-156)
Fresh plumes, high elevation site <sup>6</sup>		258 (66-377)

Fresh plumes, high elevation site and aircraft data <sup>7</sup>		299 (170-630)
Fresh plumes, aircraft data <sup>8</sup>		201 (80-400)
Fresh plumes, aircraft data <sup>9</sup>		339 (21—492)

<sup>1</sup>Data from the EPA's 2017 National Emission Inventory (EPA 2022).

<sup>2</sup>Data from Laing et al 2017.

<sup>3</sup>Data from Akagi et al 2011.

<sup>4</sup>Data from Anderea 2019.

<sup>5</sup>Data from Selimovic et al 2020.

<sup>6</sup>Data from Briggs et al 2016. Scattering values are reported at STP and converted to PM<sub>2.5</sub> using a dry mass scattering coefficient of 3.5 m<sup>2</sup>g<sup>-2</sup>

<sup>7</sup>Data from Collier et al 2016. This value includes refractory PM<sub>1</sub>. Values are reported at STP.

<sup>8</sup>Data from Garofolo et al 2019. This value includes only the organic, non-refractory fraction, however this is likely more than 90% of total PM<sub>2.5</sub> mass.

<sup>9</sup>Data from Kleinman et al 2020. This value includes only the non-refractory PM<sub>1</sub> mass.

\*\*\*\*\*

Line 65: change to "...smoke events should allow us to use the observed ratios to derive the smoke..." **Original preferred**

Line 68: remove "recently" **Done**

Line 69: change "develop" to "developed" **Done**

Line 70: include a reference to what a Monte Carlo simulation is **Done**.

*[Baez, J.C. and Tweed, D. Monte Carlo methods in Climate Science. Math Horizons, November 2013. \(available at: https://www.maa.org/sites/default/files/pdf/horizons/baeztweed\\_nov13.pdf, accessed May 2022.\)](https://www.maa.org/sites/default/files/pdf/horizons/baeztweed_nov13.pdf)*

2. Methods and data sources:

Line 73: remove "routine" **Done**

Line 74: remove (NV) **Original preferred**

Line 80: preliminary data should be finalized and this sentence should be removed **Done**.

Line 84: remove "Because there were some zero and very low values for" **Original preferred**

Line 84: change to "PM<sub>2.5</sub> concentrations less than the DL were set..." **Done**

Line 86: change to "...we use the daily smoke polygons from..." **Done**

Line 87: change to "The smoke polygon product is created by expert image analysts that digitize smoke plume extent a few times per day based on analysis of GOES-16 and GOES-17 ABI True Color Imagery available during daylight hours." **Done**.

### 3. Results:

Line 96-97: change to "Washoe County is located due east of the California-Nevada border, so smoke from fires in California..." **Done**

Line 98: remove comma at end of line **Done**

Table 1 caption last sentence: "ratios" should not be plural; "35" should include units **Done**

Line 111: change "(i)" to "(1)" **Done**

Line 115: change PM to PM2.5 **Done**

Lines 116 and 117: both sentences should be changed to past tense **Done**

Line 117: remove "So, while" **Done**

Line 118: change to "... lower ratio, but the large PM2.5 concentrations..." **Done**

Line 120: correct PM to PM2.5 and remove "alone" **Done**

Line 121: correct PM to PM2.5; change "as well as using" to "and"; change "alone" to "separately". **Done**

Line 122: change "change" to "difference" **Done**; change "smoke influenced and non-smoke data" to "segregated data methods" **Original preferred**

Line 123: remove "We note that" **Done**

Line 124: change "are" to "were" **Done**

Line 134: change "use" to "used" **Done**

Line 139: change "are only included" to "were non-zero" **Done**

Line 140: change "model" to "represent" **Done**

Line 145: change "For" to "During" **Done**

Line 147: remove "whereas" **Original preferred**

Line 162: remove "and" **Done**

Line 165: include values for comparison **Done**.

Line 167: remove "Here,"; change "is more" to "of R-urban was" **Done**

Line 168: change PM/CO to PM<sub>2.5</sub>/CO; change "this parameter" to "R-urban" **Done**

Line 169: include units for value of 20 **Done**

Line 170: remove first occurrence of "and" **Done**

Line 172: change Emission to Emissions **Done**

Lines 174-176: This sentence need to be consistent with plural and singular usage; change "this approach" to "the Monte Carlo approach" **Done**

Line 177: change "remaining" to "rest" **Done**; move "for the rest of this analysis" to end of sentence (and remove comma). **Original preferred**

Line 185: change "all had significant" to "all had a significant" **Done**

Line 191: change PM to PM<sub>2.5</sub> **Done**

#### 4. Summary:

The summary should specify that the conclusions are specific to warm weather (or May-Sept) where RWC is not a factor. Other details about the monitoring details could also be shared. **Done**.

The final paragraph should have at least one instance of "smoke" changed to "wildfire smoke". Last sentence: change "propose" to "conclude". **Done**

Figure 2: change "sorted" to "segregated"; two graphs in one figure should be denoted as (a) and (b) on the graphs or referred to as top/bottom in the text; PM to PM<sub>2.5</sub> in text **Done**

Figure 3: R-smoke should use subscript; "Monte Carlo" should come before "simulations". **Done**.

Figure 4: R-smoke should use subscript **Done**.

Figure 5: PM to PM<sub>2.5</sub> (text and figure) **Done**.

#### RC2:

This manuscript describes a new method to determine the relative contribution of smoke to observed PM<sub>2.5</sub> during wildland fire smoke season. Following Liang et al, 2017, the authors use PM<sub>2.5</sub>/CO to categorize smoke and non-smoke influenced days. In contrast to the overhead HMS

smoke product from satellite measurements that can misrepresent conditions on the ground due to inadequate (or nonexistent) retrieval of near-surface smoke concentrations, the PM<sub>2.5</sub>/CO method uses in-situ ground measurements typically present at regulatory surface monitoring sites. After determining the PM<sub>2.5</sub>/CO ratios for urban and smoke aerosol by comparing Monte Carlo simulations to observations, the authors estimate relative contribution of smoke to PM<sub>2.5</sub> for smoke-influenced days, finding that indeed all the PM<sub>2.5</sub> exceedance days during the period of study have high influence of smoke. Because the simulation is trained on local conditions, the values reported here may not be widely applicable, but the method can be applied to other sites to identify and estimate relative smoke influence. This manuscript describes the development of methods for interpretation of atmospheric data, but with a limited scope of one study location where all PM<sub>2.5</sub> exceedance days are from smoke, so its publication as a Technical Note is appropriate. I recommend publication with minor revisions below:

1. Discussion of previous work may be improved by description of the various units for normalized enhancement ratios (NERs).
  2.  $\Delta\text{PM}_{2.5}/\Delta\text{CO}$  in g/g vs  $\text{ug m}^{-3} \text{ppm}^{-1}$ : Inclusion of the scale factor may be appropriate. **This discussion has been completely re-done.**
  3. Ambient  $\text{ug m}^{-3}$  vs STP  $\text{ug sm}^{-3}$ : Confirm that all values are reported at standard volume to compare like-to-like. **We note this for each of the elevated site or aircraft studies, where it is reported.**
  4. PM<sub>2.5</sub> vs PM<sub>1</sub>: Studies using Aerosol Mass Spectrometers (e.g. Kleinmann et al., 2020, and Garofalo et al., 2019) will report non-refractory PM<sub>1</sub>. **So noted.**

I acknowledge that choosing a convention will not have any bearing on the analysis, since this manuscript recommends performing the complete analysis for a particular location. Therefore, any definitions or units of PM will be consistent. However, uniformity in discussion of previous results and between the abstract and main text is appropriate. **We have tried to maintain this uniformity as much as possible. We now consistently use the term PM<sub>2.5</sub>/CO, rather than a more general PM/CO.**

2. Ln 115: The authors state “Using the PM<sub>2.5</sub>/CO ratio to segregate the data, we find an improved correlation of PM and CO in the lower range of ratios, compared with using the HMS alone as an indicator (Figure 2).”

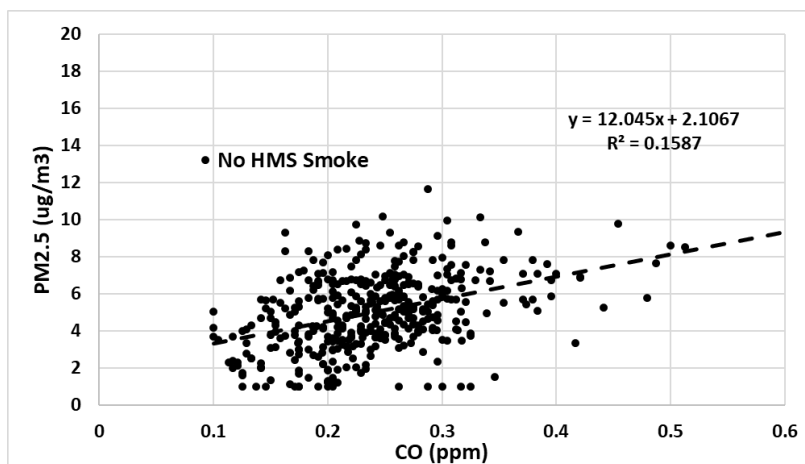
In Fig. 2, the R<sup>2</sup> values for the smoke days indicated by HMS smoke and PM<sub>2.5</sub>/CO>30 for the entire range seem comparable, while the R<sup>2</sup> values for the non-smoke days are less comparable, indicating the main difference between these methods is in the lower PM<sub>2.5</sub> concentration range, well below the NAAQS. At these lower concentrations, the HMS smoke product is less likely to capture conditions at the surface and produces false negatives and positives for smoke-influence. To me, a major strength of the ratio method is the improved sensitivity and specificity in identifying smoke days at these lower concentrations. To highlight this, an SI figure explicitly showing the PM to CO correlations or an inset of PM<sub>2.5</sub>/CO vs CO that better shows this lower range would be helpful. Additionally, or alternatively, making the dots smaller or with some

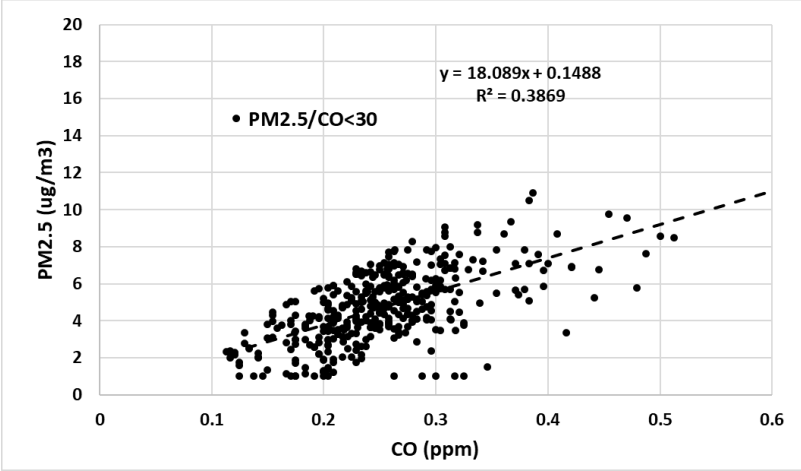
transparency might allow the reader to better see the differences in the two methods at low concentrations in Fig. 2. Table 2 indicates that only a net change of 2 days between methods, but it seems that more than 2 dots have changed color between Fig. 2a and 2b. Can you add how many days switch categorization (and in which direction)?

I have added this line:

There are 612 days in the analysis. 200 have a positive HMS smoke identification and 220 have  $PM_{2.5}/CO$  ratios  $>30$ . In total, 73 days with  $PM_{2.5}/CO$  ratios  $>30$  do not have a positive HMS smoke identification. As noted in Table 1, using a criteria of  $PM_{2.5}/CO >35$ , there are 27 days with identified smoke, but no HMS indication.

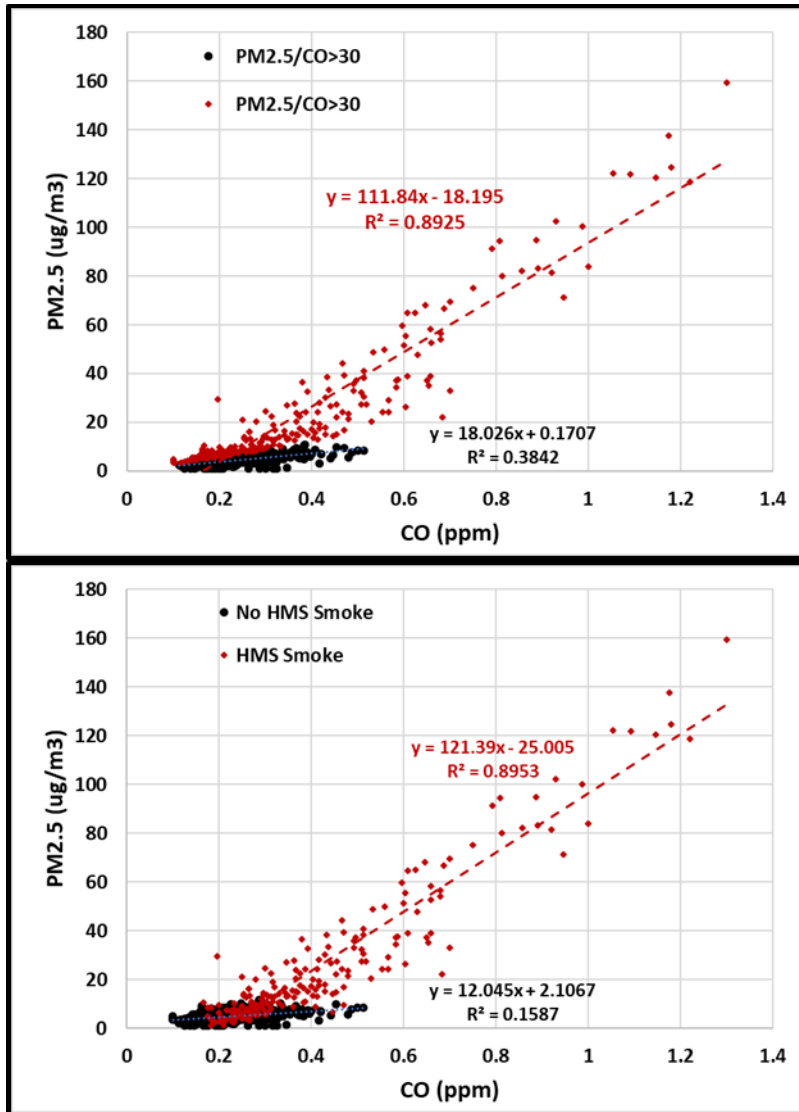
I also suggest adding the NAAQS to Fig. 2 to show that both methods successfully identify exceedance days. Further explanation and slight tweaks to the figures for the low concentration data will further support the authors' assertion that the  $\Delta PM_{2.5}/\Delta CO$  method is generally a more robust indicator of surface smoke than satellite-based measurements. Here are the two figures focusing on the lower end of the scale. As noted, the correlation is much better using the  $PM_{2.5}/CO$  ratio, rather than the HMS data alone. However, I don't really find that the figures are very compelling or showing anything that is not in the original figures. So I choose not to include these.







I did redo Figure 2 to change the dot size and this seems to address the reviewers comment. New versions are below. These will be swapped out in the final manuscript.



A new development is the use of a Monte Carlo simulation to estimate PM<sub>2.5</sub>/CO ratios for smoke and urban influence separately in order to estimate the relative contribution of smoke to observed PM<sub>2.5</sub>.

3. How sensitive are the Monte Carlo results to the chosen PM<sub>2.5</sub> and CO backgrounds and how do they compare to the non-smoke days (from either and both methods) from the Sparks site in 2019?

There is really a limited range of background values to use. CO might have a background from 0.1-0.2 ppm. PM<sub>2.5</sub> could have a background of 1-3  $\mu\text{g m}^{-3}$ . From the observations, we find an observed PM<sub>2.5</sub>/CO=21.7 for all PM<sub>2.5</sub> <10  $\mu\text{g m}^{-3}$

The table below shows the results of the Monte Carlo simulations results for the PM<sub>2.5</sub> range of 0-10  $\mu\text{g m}^{-3}$  using this range of background concentrations. There is not a lot of discrimination in the results based on the background concentrations. Given especially the significant uncertainty in the measurements at these lower concentrations I do not see a lot of value in adding this discussion.

<b>Background CO (ppm)</b>	<b>0.1</b>	<b>0.2</b>
<b>Background PM<sub>2.5</sub> (<math>\mu\text{g m}^{-3}</math>)</b>		
<b>1</b>	22.4	14.2
<b>2</b>	28.1	17.7
<b>3</b>	33.7	21.0

4. The ozone discussion is limited and the numbers in line 190 do not seem to match the numbers in Table 1. Thank you. The values in Table 1 and at line 190 have been corrected.

Careful reading for grammatical errors and missing references (e.g. Briggs et al., 2016) will improve readability. Added missing references.