Response to Referee #1

We would like to thank Reviewer 1 for carefully reading the manuscript and providing thorough comments.

General comment 1: "While the scientific asset of using such an instrument to study wildfire is explicit, discussion about the limitation of this study is clearly missing."

Response to General Comment 1: A paragraph describing the limitations of this study and instrument were added to the discussion: lines 455-477.

General Comment 2: "There are some repetitions in the paper and authors should reorganize some parts to make it more concise. For instance, some similar sentences are shown in different parts throughout the paper: lines 78-80 similar to lines 240-244, line 557-559 is quasi similar to lines 94-96, and 26-28."

Response to General Comment 2: We deleted lines 78-80 to make introduction more concise and is no longer similar to lines 240-244. We modified the lines 26-28, 94-96 and 557-559 to:

Lines 26-28: Our work demonstrates a novel application of the ground based EM27/SUN solar spectrometers in wildfire monitoring ~~and contributes to the development of techniques for analyzing remotely sensed greenhouse gas measurements.~~ by integrating regional scale measurements of trace gases and aerosols from smoke plumes.

~~Lines 94-96: Our work demonstrates a novel application of the ground based EM27/SUN solar spectrometers in wildfire monitoring and contributes to the development of techniques for analyzing remotely sensed greenhouse gas measurements.~~

Lines 557-559 (now lines 553-554): Overall, our analysis ~~demonstrates a novel application of the EM27/SUN solar spectrometers and will~~ contributes to the development of techniques for analyzing remotely sensed greenhouse gas measurements from wildfires.

General Comment 3: "The discussion section is interesting and well written. The introduction section should be more concise and emphasize the research state of art. More appropriate references are needed. I would recommend not using references in the conclusion section and focusing on summarizing the main results of this study. A paragraph in the conclusion is missing to highlight the limitations and the perspectives of this work."

Response to General Comment 3: Thank you for the suggestions. References were removed from the conclusion section. A paragraph was added to the discussion section (lines 455-477) to highlight the limitations of the EM27/SUN measurement technique.

"While advantages of this technique allow for understanding regional scale emissions, limitations exist with this method. The EM27/SUN solar column observations are limited to daytime hours as the instrument requires the sun as the light source. For this reason, we were not able to capture nighttime observations despite the continued release of smoke emissions and growing concern of increasing nighttime wildfire activity in the continental United States (Freeborn et al., 2022). Additionally, optically thick smoke plumes obstruct the sunlight and prohibits continued measurements when the solar disk is not traceable by the instrument's solar tracker. Exposing the instrument's mirrors to harsh conditions such as ash depositing to ground observations on Sept. 12 decreases the instrument signal and may decrease the lifetime of mirrors. Although total column measurements are sensitive to larger scales than in situ stations, the FTIR is limited to the line of sight of the instrument and on occasions can miss the plume like we did on Sept. 13 and 14. Whereas aircraft observations have extensive spatial reach and more flexibility in locating and sampling plumes to obtain spatially rich information of the plume. However, when used in tandem with satellite observations our instrument collects continued temporal observations of a site of interest that a satellite does not, thus synchronous observations provide a better spatiotemporal understanding of the emission source. EM27/SUN instruments are also costly which can limit the number of instruments deployed. Unless instruments are secured properly as they have been done in long term network studies (Frey et al., 2019; Dietrich et al., 2021), measurements require personnel to set up and operate the instrument daily. The EFs, MCE, and their uncertainties fall within the range of expected values, thus lends confidence that this technique can be used for studying combustion phases of wildfires for other vegetation types. Despite the limitations of the EM27/SUN, we demonstrate the ability to gather new information of EF, MCE and AOD for understudied vegetation types and regions. Furthermore, the EM27/SUN observations can be used as a validation tool for orbiting satellites like TROPOMI, Orbiting Carbon Observatory-2 (OCO-2), OCO-3, and future satellites. The next generation weather forecasting, greenhouse gas, and air pollutant satellites such as Tropospheric Emissions: Monitoring of Pollution (TEMPO) will have more temporal frequency and greater spatial resolution allowing for continuous monitoring of burning activity and smoke emissions (Zoogman, 2017). This may allow remote sensing products to provide new insight into fuel properties of many types of vegetation in remote areas. It is will also be important to evaluate satellite-based observations with ground-based stations like the EM27/SUN as we did in this study."

General Comment 4: "Although authors are very thorough in the sensitivity tests, the main concerns are error estimations and background measurements. The error estimations are missing in this study. There are no estimation of the measurement uncertainties of Xgas and AOD which should be incorporated and propagated in the calculation of ER, EF and MCE. The slopes of the linear fits should reflect errors propagation and be mentioned with an error bar (+-)."

Response to General Comment 4: Thank you for this comment. We have incorporated uncertainties in the AOD estimation and included errors in the CO/AOD linear fit (lines 273-276), see Figure 3. AOD errors are described in the Appendix B, lines 575-575.

Error propagation was described for ER (lines 190-192) and EFs (lines 337-338). For the averaged MCE we reported the standard deviation but clarified that in line 339. We also show the slope error in Figure 5.

General Comment 5: "The background values are very important in the ER computation. How can you ensure that the 2nd percentile of the daily measured mixing ratios represent the background at this location? Why comparing the background SJV values to the very remote location of Mauna Loa? Could you find appropriate background values located at closer sites (Caltech, Mont Wilson, Dryden, other)? The ER values greatly depend on background concentrations and measurement precision. Error bars should be added to these estimates."

Response to General Comment 5: We agree with the reviewer that a more appropriate background location closer to the measurement site should be used, however during the 2020 fire season several wildfires occurred during the same period in Southern California. We explored using the southern California TCCON sites (Caltech and Armstrong/Dryden) as background sites, but X_{CO} was elevated due to local wildfires in those areas and thus were not appropriate to use during this time. Mt. Wilson was also heavily fire impacted in September 2020, with flames reaching within 150 m of the observatory.

Leveraging the comparison between our ground-based instrument and TROPOMI, we compared the background measured from our site to the variation shown in the TROPOMI overpass. Figure S2 was added to the supplement where TROPOMI retrievals support the 2^{nd} percentile background. Text and figure added to supplements:

"Background estimation of EM27/SUN measurements

The enhancement over background (ΔXgas) was calculated by subtracting the background (Xgas, bkdg) determined as the 2nd percentile of the daily measured mixing ratios (Xgas). Due to ongoing wildfires throughout the state, TCCON stations in Southern California (Caltech or NASA Armstrong/Dryden) were inappropriate as background sites. We used TROPOMI satellite retrievals of X_{CO} on the Sept. 12 plume event as a case study to determine whether a 2nd percentile subtraction is appropriate.

TROPOMI satellite measurements can provide a better spatial understanding of heterogenous emissions during events like the wildfire plume. During the day on Sept. 12, the EM27/SUN measured a background of 220 ppb determined by the 2nd percentile (green line, Figure S2a). From TROPOMI observations, we can see that south of the plume was relatively "cleaner" while north of the plume X_{CO} levels were higher (Figure S2b) due to emissions of multiple wildfires burning in the Sierra Nevada flowing southward. On this day, the EM27/SUN did not reach lower X_{CO} levels as observed by TROPOMI and in Figure S2c we can see that the appropriate background for the EM27/SUN is determined by the instrument itself as the 2nd percentile.

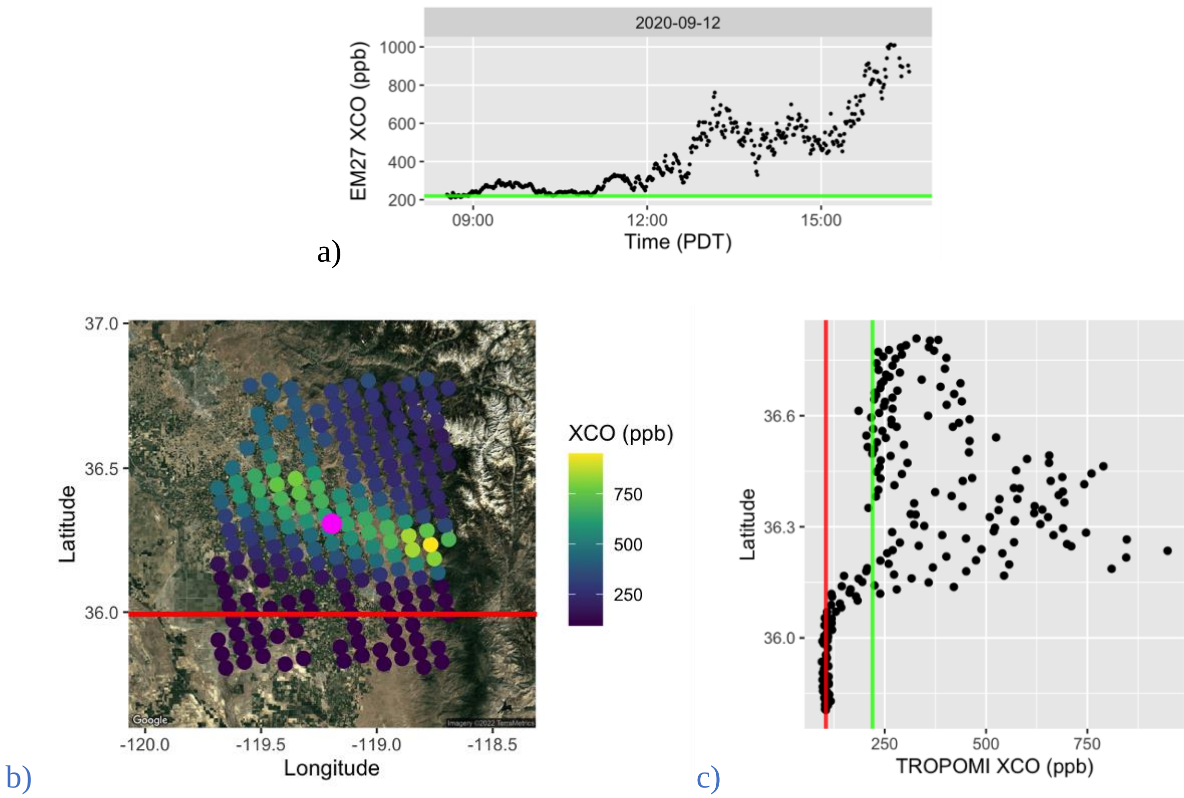

Figure S3. a) Timeseries of Sept. 12 plume event with green line representing the background (220 ppb) determined as the 2nd percentile of the daily measurements. TROPOMI satellite XCO retrievals of Sept 12 plume event with a) with location of EM27/SUN displayed with a magenta marker and a red line marking the 36° latitude line. b) Latitudinal TROPOMI X_{CO} with red line showing the average 105 ppb X_{CO} below the 36 latitudinal line and the green line displaying the background determined from the EM27/SUN 2nd percentile daily measurement.

General Comment 6: "Some figures could be improved for clarity purpose. Ex: the time series does not display well intraday variations."

Response to General Comment 6: Thank you for the suggestion. Based on similar comments made by reviewer #2, Figure 4 was added to Figure 2 and intraday variations are more visible.

General Comment 7: "The word "large scale" is recurrent in the manuscript and does not seem to be appropriated. Is regional more appropriated? The title should be modified since "large scales" is vague.

Response to General Comment 7: Thank you for the comment. We agree with the reviewer that the term is vague and have replaced the term "large scale" with "regional" throughout the text.

Specific comments:

1) Please clarify the differences between Emission Ratios and Enhancement Ratios.

Response: We thank the reviewers for this comment and agree that clarification is necessary between emission ratio and enhancement ratios. We follow the definitions described in Yokelson et al., 2013 where a distinction is made between both concepts based on the conservation of the source ratio after the plume mixes with background air. We further elaborate below, and, in the manuscript in lines 365-369.

Enhancement ratios are also known as the normalized excess mixing ratios. Excess mixing ratios are calculated by subtracting the mixing ratio of a species from a source plume minus a mixing ratio of the same species in background air. To correct for dilution, excess mixing ratios are then normalized by a stable tracer, such as CO or CO_2. When an enhancement ratio does not change with dilution and mixing with background air, the enhancement ratio is equal to the emission ratio of a source.

In this study we measure stable compounds (CH_4, CO, and CO_2) that are not expected to chemically react in atmosphere during the duration of the local atmospheric transport. However, during this fire period many wildfires were burning throughout the state and smoke plume funneled into the SJV from other fires changing the background composition. Because of the SJV valley topography, air becomes stagnant, and pollution builds up, thus a true background was never reached for Sept. and Oct. 2020 measurements. We use Sept. 12 as a case study, where remote sensing instrument was directly underneath a thick smoke plume. We subtract the local background to isolate the plume in order to calculate an emission ratio for the fire.

2) Abstract line 19: please define at “10km scales”. Is it vertical or horizontal scale?

Response: Thank you for pointing this out. We have changed “10 km scales” to “10 km horizontal scales.”

3) Line 57: “fire conditions”, please explain what conditions.

Response: Thank you for the comment. We have changed the vague term “fire conditions” to “wildfire combustion phases.”

4) Figure 3b: the error bars are the standard deviation. Errors on both TROPOMI and EM27/SUN measurements should be included in the linear fit.

Response: We used the York linear fit that incorporated errors in the x and y to calculate the slope and error. We have modified the figure (now Figure 4b) to show the slope with error.

5) Section 3.3: authors state that FTIR and AERONET AOD are in agreement. What is the R value? How can you prove it?

Response: A scatterplot was added to the supplements (Figure S3) displaying a R value of hourly average comparisons.

6) Figure 4: reduce point size or find a solution to better display intraday variations.

Response: Figure 4 was merged with Figure 2 and focused on days Sept. 8 -15.

7) Figure 5: What are the measurement errors? Could you propagate the errors to obtain slopes values with all uncertainties?

Response: We used York linear regression to calculate the slope and error. The instrument errors for X_{CO}, X_{CO2} and X_{CH4} were used for this calculation and the Figure 5 was modified to show the slope with error. Text was added to clarify this in lines 190-193.

8) Figure 9: what is the error bar on the Top 20 CA wildfires emissions?

Response: The error shown on the Top 20 CA wildfire CH_4 emissions (now Figure 8) was calculated by propagating the ER_{CH4} error from Table E1 into each individual wildfire CH4 estimate and added in quadrature to obtain a total error. More detail was added in the text in lines 424 - 426.

9) Please verify the order of the references in the parenthesis throughout the manuscript. It should follow the ACP journal recommendations: https://www.atmospheric-chemistry-and physics.net/submission.html#references (ex: line 72; lines 110-111; …)

Response: We have verified the order of references throughout the manuscript.

10) Title of section 3.5 should be more specific. SJV GHG's sources are only dairy farms?

Response: Section 3.5 was split into two sections: "Enhancement ratios of livestock and wildfire emissions" and "Total methane emissions from wildfires in California."

Technical Corrections

1) Line 34-35: Rephrase this sentence and define particulate matter 2.5.

Response: This sentence was rephrased and particulate matter 2.5 was defined.

2) Line 56: Is reference "CARB 2018" appropriate?

Response: We have updated the reference to an appropriate reference: Lasslop et al., 2019.

3) Line 51: Is IPCC 2014 correct? Can you refer to a more recent report?

Response: Thank you for your comment. We have updated "IPCC 2014" to "IPCC 2021".

4) Line 65-66, 69, 70, 71 and more: add references.

Response: More references were added.

Lines 65-66: Schneising et al., 2020, Whitburn et al., 2015, Adams et al., 2019, Griffin et al., 2021, and Jin et al., 2021

Line 69 (now line 72): Chen et al., 2016 and Heerah et al., 2021

Line 70 (now line 73): Frey et al., 2019, Vogel et al., 2019, Alberti et al., 2022a, and Alberti et al., 2022b

Line 71 (now line 74-75): Bader et al., 2017 and De Mazière et al., 2018

5) Figure 5, 8: Change dots color or size to clearly display all the points.

Response: Points on Figures 5 and 8 (now Figure 3 and 7) were reduced in size to clearly display all points.

6) Figure 3b, 6, D1: R2 should be R2

Response: Thank you for your suggestion. The "R2" in Figures 3b, 6, D1 was switched to R^2.